Responses to Reviewer 1 comments on "Intercomparison and evaluation of aerosol microphysical properties among AeroCom global models of a range of complexity" by G.W. Mann et al.,

Reviewer 1

This paper is an initial comparison of the global aerosol microphysics models in the AeroCom comparison. The authors approach the comparison by evaluating the group of models as a whole, rather than focusing on individual models, which would make for a cumbersome paper. The variability between the middle-2/3s models is particularly informative in this paper.

A considerable amount of work went into the comparisons in the paper, so the lead authors should be commended. The paper is very useful for understanding regions and species where the models need improvement. I feel that it should be published once several issues have been addressed.

We thank the reviewer for their constructive comments and provide responses to each below, with reference to changes in the manuscript that have been made. Reviewer comments are shown in italics and numbered for reference. Excerpts of the manuscript shown in bold-italics.

1) Throughout: The discussion and figures jump between diameter and radius (seemingly randomly). For example, the cuttoffs for N30 etc are in diameter while the size distributions are plotted as a function of radius. This may likely cause confusion for those readers integrating size distributions by eye (I frequently do this). I find it tedious to switch between them within a paper, I highly recommend using only diameter.

For consistency with equations 4 and 5 we have changed the size distribution Figures (14, 15, 16, 18 and 19) to use dry diameter rather than dry radius. References to radius values within the text have been changed to diameter values.

2) P30852 and Table 1: GEOS-Chem-APM is a hybrid microphysics scheme. Sulfate and SOA is sectional, POA and BC is modal, and sea-salt and dust is sectional (but different sections from sulfate and SOA). http://wiki.seas.harvard.edu/geoschem/index.php/APM_aerosol_microphysics describes the scheme fully. It is possible that the Albany folks did something different for AeroCom, but I've never seen an APM paper with size sections for all species. The Yu and Luo (2009) paper that is cited as the APM reference uses this hybrid scheme.

We have corrected the description of GEOS-Chem-APM in Table 1 and have clarified the text in section 2.2. referring to the two schemes which compare bin and mode replacing:

ECHAM5-HAM2, EMAC, TM5, CAM5-MAM3, GISS-MATRIX and HadGEM-UKCA) and four use sectional schemes (GISS-TOMAS, GLOMAP-bin, ECHAM5-SALSA and GEOS-Chem-APM), whilst CanAM4-PAM is a~hybrid approach combining bin and mode.

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ECHAM5-HAM2, EMAC, TM5, CAM5-MAM3, GISS-MATRIX and HadGEM-UKCA), three use sectional schemes (GISS-TOMAS, GLOMAP-bin and ECHAM5-SALSA), whilst GEOS-Chem-APM uses a modal approach for black carbon and primary organic particles, with sectional approach for other particle types. CanAM4-PAM uses the piecewise log-normal approach, which applies sectional and modal methods for different parts of the particle size spectrum (see Von Salzen, 2006).

3) P30854: I don't think Dthresh is defined anywhere.

The text has Dthresh in parentheses after the phrase "These threshold dry diameters". We therefore assert that it is already clear that Dthresh is the threshold for the different size-resolved particle concentrations derived from the bin/mode schemes.

4) Figures 1 and 2: Please make it clear that panels a and b are the means and c and d are the diversity. Please say in the caption that the diversity is a ratio (you say this in the text, but many readers will skim figures).

We have added to the caption for Figures 1-4 the following text:

Diversity here is the ratio of the maximum and minimum values over the central 8 of the 12 models (defined locally, as described in section 2.4).

5) Figure 3 and 4: Are these STP or ambient concentrations? This need to be specified (will make more than a factor of 5 difference in the concentrations in the UT).

All particle concentrations are at ambient temperature and pressure except for Figure 21 where the measurements were concentrations at standard temperature and pressure. We have added the following text to the caption of Figure 21:

Note that model particle concentrations have been converted to values at standard temperature (300K) and pressure (1000 hPa) to be consistent with these u-CPC measurements. In all other Figures measured and model values are at ambient conditions.}

6) P30868 L20 (and elsewhere): What is "b"? Is it the bias. If it's bias, is it in log space? Is it normalized? Please define it clearly, later you use "b" but there is less context to figure out that it is probably a form of bias.

We have defined "b" as the normalised mean bias in lines 19-20 of page 30867. In response to comments from Reviewer 2 we have added, to each of the Figures where model and observations are compared, values of the normalised mean bias and the Pearson correlation coefficient.

7) P30871 L5: It took me a long time to figure out that Ispra is "JRC" (still not positive). I'm not sure why the full name and 3-letter abbreviation have nothing in common. Would it be possible to always put the 3-letter abbreviations in parentheses when discussing the sites throughout the text?

Yes JRC is Ispra. The abbreviation is the acronym for the European Union "Joint Research Centre", which is based in Ispra. The abbreviation is already explained in Figures 11 and 16 and is also given in the text in section 3.2 on page 30865 lines 24-29. We have added the abbreviation JRC in parentheses after first mention of Joint Research Centre in section 3.2 (page 30864, line 15).

8) P30871 L28-29: "...only one of the models includes boundary layer nucleation...". Only one of the models includes "activation-type nucleation" or "kinetic-type nucleation" that have been confusingly referred to as "boundary-layer nucleation". The ternary and IMN simulations will also have nucleation in the boundary layer (and the ternary schemes have been shown to be better at predicting the occurrence and non-occurrence of nucleation and the magnitude of nucleation rates [when scaled] than activation nucleation [see Westervelt et al., ACP 2013 and Jung et al., AS&T, 2008]). Thus several models have nucleation in the boundary layer. Activation and kinetic nucleation schemes need to stop being referred to as "boundary layer nucleation" as if they are the only schemes that predict nucleation in the boundary layer; it leads to confusion such as in this sentence in the manuscript

We have revised that part of the text replacing:

A possible explanation for the poor seasonal variation of small CCN is that only one of the models includes boundary layer nucleation, which generates small CCN effectively during summer when photochemical production of \chem{H_2SO_4} and organic vapours is stronger. As already noted, Spracklen et~al.~(2010) found that although adjustments to the assumed size distribution for primary emissions could reproduce observed annual mean concentrations of the finest particles, better agreement with the observed seasonal cycle could be achieved when a~boundary layer nucleation mechanism was included.

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For many of the models (see Table 2), binary homogeneous nucleation is the only new particle formation mechanism, and this may explain the poor seasonal variation of N_{30} in the models. As already noted, Spracklen et~al.~(2010) found that, in model simulations with only binary nucleation, although adjustments to the assumed size distribution for primary emissions could reproduce observed annual mean concentrations of the finest particles at Pallas, better agreement with the observed seasonal cycle could be achieved by also including a nucleation mechanism effective in the boundary layer.

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9) Figure 14 and 15 and section 3.2.3: I'm not sure that these add benefit to the paper since the distribution statistics (e.g. N10, 30 100) at the same sites are given in the previous figures. If you wanted to make the paper a little more sleek, I recommend removing these figures and section 3.2.3.

We do not agree with Referee 1 here. The size distribution comparisons shown in Figures 14 and 15 give much more information than the N10, N30 and N100 comparisons in the earlier Figures. As we have tried to explain in section 3.2.3 the size distribution Figures also help interpret the cause of some of the N10/N30/N100 biases (e.g. whether error could be due to too little number or too little growth).

10) P30874 L27: I think "month" should be "season" here.

Corrected.

11) P30877 L14: "concentrations" is misspelled.

Corrected.

12) Figures 17 and 21 and P30878 L5: Figure 17 is ambient concentrations, and Figure 21 is STP concentrations (thanks for labeling these here). Also, the y-axes are different (pressure vs. altitude, respectively). Please make these consistent (e.g. both STP with pressure axes). I believe these inconsistencies between the concentrations in the figures leads to authors thinking that there are bigger differences between figures than there are. For example, at P30878 L5, the authors claim that UT concentrations above the north pacific reach 5000 cm-3 (in Figure 21), compared to only 800 cm-3 above Europe (Figure 17). However, at 200 hPa, the difference between STP concentrations and ambient concentrations are about a factor of 4, so the concentrations measured over Europe are more like 3000-3500 cm-3 when corrected to STP, which is much closer to the concentrations over the north Pacific. Thus, while some of the difference may be attributable to differences between condensation sinks between continental and oceanic regions, the differences are not as large as currently claimed in the manuscript.

Many thanks for this point. That was a mistake to compare the LACE-98 ambient N5 concentrations from LACE-98 with the STP N3 concentrations from Clarke and Kapustin (2002). We now realize that, at 200 hPa, the 5000 per cc at STP corresponds to an ambient concentration of approximately 1000 per cc, which is quite close to the 800 per cc observed in the UT over Europe. We have therefore deleted the following text (from the ACPD paper) in the revised paper for ACP (and also removed Heintzenberg et al. (2011) and Hermann et al (2003) from the bibliography as this was the only text to refer to those papers):

In contrast, as we show below, aircraft measurements of N_3 over the northern Pacific (Clarke and Kapustin, 2002) show much higher concentrations in the upper troposphere, up to 5000\,\unit{cm^{-3}}. A~recently compiled database (Heintzenberg et~al., 2011) of over 10\,\unit{yr} (1997 to 2008) of CPC measurements in the upper troposphere aboard two commercial aircraft as part of the CARIBIC project (e.g. Hermann et~al., 2003) shows generally lower concentrations over Europe than over the North Atlantic, potentially due to the higher condensation sink over continents leading to lower ambient sulphuric acid vapour concentrations and thus lower nucleation rates.

13) P30882 L6-8: The authors claim that models have a low variability in CCN because of the low variability in N30, but there is a higher variability in N100. Whether N30 or N100 is more representative of CCN very much depends on the situation.

In the revised manuscript, the current sentence

The relatively low N30 diversity in remote environments suggests that current global aerosol microphysics models are fairly consistent in their simulations of ``natural'' background CCN concentrations.

Is replaced by

The relatively low N30 diversity in remote environments suggests that current global aerosol microphysics models are fairly consistent in their simulation of ``natural'' background concentrations of particles in the 30 to 100nm dry diameter range.

14) P30883 L22: Ammonium nitrate has a high-enough volatility that it tends to stay close to equilibrium as it condenses (i.e. quasi-equilibrium growth http://www.tandfonline.com/doi/abs/10.1080/02786826.2012.679344). This means that its size-resolved condensation is proportional to mass (rather than condensation to the Fuchs-corrected surface area as low/non-volatile species, such as with sulfate and low-vol organics). Thus, unless nitrate is reacting in a way to make it less volatile than ammonium nitrate (e.g. organic nitrates perhaps), it will have little effect on nucleation and growth (e.g. see the "thermodynamic" cases in http://www.atmos-chemphys.net/11/3865/2011/acp-11-3865-2011.html).

In the revised manuscript, the current text

which is likely due to inadequately constrained particle number sources (both primary and secondary) or underprediction of growth due to a~general underprediction of wintertime mass sources of nitrate and secondary organic aerosol, or both.

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which is likely due to inadequately constrained particle number sources (both primary and secondary) or too little growth due to a~general underprediction of wintertime sources of mass (for example from secondary organic aerosol), or both.