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Atmospheric Chemistry and Physics Discussions

Interactive comment on "First comparison of a global microphysical aerosol model with size-resolved observational aerosol statistics" *by* D. V. Spracklen et al.

Anonymous Referee #2

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The manuscript "First comparison of a global microphysical aerosol model with size-resolved observational aerosol statistics" by D. V. Spracklen et al. presents an evaluation of the aerosol size distribution statistics simulated with the sectional GLOMAP aerosol scheme with two compilations of measured size distribution statistics for the marine boundary layer. In addition, sensitivity studies are performed to investigate the role of primary emissions and cloud processing on the simulated aerosol concentrations and size distribution. The authors make a nice (and rare) effort to sample the model in analogy to the measurement data by sampling it according to the age of the airmass.

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Interactive Discussion
Discussion Paper

The increasing complexity of global aerosol models with embedded microphysics schemes demands for a careful evaluation. Hence, the presented analysis is clearly of relevance and well in the scope of ACP. The manuscript is generally well organised, clearly written and the analysis is scientifically sound. Thus, I can recommend this manuscript for publication in ACP after the following issues have been addressed.

General Comments

Title The usage of "First..." is always a challenge. In this case, if "first" refers to "first comparison of this model" it might be appropriate. But this should be made clear. In general, however, a number of datasets of statistical parameters of the aerosol size distribution have been available for quite a while, such as the European aerosol phenomenology (van Dingenen et al., Putaud et al, 2004) and the Heinzenberg et al. analyses used in this study. These datasets, including a statistical description of the aerosol size-distribution parameters such as percentiles, have been used before for the evaluation of a number of other microphysical aerosol models (e.g. Lauer et al, 2005; Stier et al., 2005). Thus, in this general sense "first" does not apply here and should be omitted.

Abstract The abstract contains a number of strong conclusions that are not sufficiently backed by the analyses in the paper. First, the statement "We show that a physically based cloud drop scheme is needed to explain the observed change in the accumulation mode geometric mean diameter with particle number" seems to be too strong to me. What has shown in the analysis is that the results improve when the highly simplistic treatment in the standard model is replaced by a more physically based scheme. No schemes of intermediate complexity have been assessed.

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Second, some of the sensitivity analyses performed are based on assumptions, such as a specific choice of the primary emission size distribution and the effective usage of primary sulfate as surrogate for black and organic carbon. The absence of suitable data might justify the usage of these assumptions, however, the conclusions should include this uncertainties and be less unambiguous.

Model description, p8876 The model description is to my understanding not detailed enough. In particular two aspects should be presented in more detail:

First, the presented results depend crucially on the treatment of the aerosol nucleation. The used Kulmala nucleation scheme has issues, in particular at low temperatures. Thus, the authors fix the nucleation rate at low temperatures, as described in their Spracklen et al. (2005b) paper. In addition, the temperature dependence of the used binary nucleation scheme predicts very little particle formation in the lower troposphere. This should be mentioned here and requires a brief discussion.

Second, the description of the introduction of the different components into the model is very limited. Although used in the result section later, the procedures to introduce black and organic carbon into the model are not described here. As far as I understand from the description, the model internally only treats one size distribution with sulfate as the only component. The other components (sea salt, black carbon, organic carbon) are introduced by increasing sulfate as surrogate for the respective species and leaving all physical properties identical to sulfate itself. If that is the case it should be clearly stated in the model description, as this is different from other multicomponent sectional aerosol models that explicitly treat several components for each size bin and calculate effective physical properties of the mixtures. The authors justify

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this approach for sulfate and sea salt by stating “Sea salt and sulfate aerosol activate at similar diameters so the impact of this assumption will be small.” However, things are not so easy. Berg et al., JGR, 1998 report from the used ACE 1 experiment that they could clearly find externally mixed populations of sea salt and sulfate with distinctly different hygroscopic properties. Growth factors of sulfate were around 1.7 and of sea salt around 2.1. Although I agree that this simplification can be justified, a bit more discussion seems appropriate. If I misunderstood this treatment from the limited description please explain in more detail.

Third, also the simplified treatment of the fixed activation diameter, the consequences for the cloud processing, and the replacement by the explicit activation scheme should be described appropriately in this section.

Observations, p8878 For the comparison of the aerosols size distributions the relative humidity plays a crucial role. Have the compiled measurements been performed at dry or ambient relative humidities? I could not find anything on that in the text. Only the figure labels show “dry diameter”, thus I assume the analysis is based solely on dry radii. This should be mentioned as a large part of the uncertainty in aerosol forcing estimates stems from the water uptake. How does this affect the general conclusions?

Focus The study focuses strongly on the aerosol number size distribution, thus on the smaller aerosol sizes. However, in the MBL, the coarse mode aerosol plays a significant role, also for the aerosol radiative properties. It would be nice to mention that - at the moment coarse mode sea salt is not even referred to in the manuscript. Ideally, it would be interesting to additionally take into

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account measures that are also representative for the coarse mode, such as the mass distribution or the total mass. Otherwise it should be stated clearly that this study focuses on sub-micron aerosol.

Specific Comments

p8874, l 15: "Most previous studies have also assumed that the aerosol entrained from the FT is composed entirely of sulfate, derived entirely from natural emissions." This does generally not apply to the many available global multicomponent aerosol modelling studies.

p8875, l12: DMS not defined.

p8876, l 9: "This is the first detailed comparison between a global sectional aerosol process model and remote MBL aerosol size distributions." Again, the usage of "first" is neglecting previous work. For example, Pierce and Adams (2006) also analysed their size distribution simulated with their sectional aerosol model in the MBL. If necessary, specify exactly what has been done first or (to my understanding better) omit.

Sections 5.1 and 5.2, p8881-8882: The description of the shape of the size distributions could be extended. What role does the (not so certain) emission size distribution of sea salt play? What about sub-micron sea salt emissions that are discussed later? Could they contribute to fill the gap between the two modes? The observed size distributions from ACE2 and ACE Asia show indication of tri-modal size distribution that is not captured in the model, why?

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Section 5.3 The analysis of the anthropogenic influence is interesting. It would be nice to see if this is in agreement with previous studies that are available.

Section 5.4.1, p 8886 It is not given how much of the total sulfate is actually emitted as primary particles - I assume 3%? The assumed size distributions for primary sulfate are very small, thus the primary particle flux has to be very large if 15% of the primary sulfate by mass is emitted in the nucleation mode size. How do the primary number fluxes compare to the number flux introduced by nucleation?

The authors conclude “Including primary emissions has relatively little impact on the size of the Aitken mode and does not help to explain model under-prediction of mode diameter.” First, this refers only to primary SO4 emissions and this should be clear in that sentence. Second, this is a very general statement given the fact that only one emission size distribution has been tested.

Section 5.4.2, p 8887 The chosen size distributions for primary emissions are very small, representative for fresh emissions close to the source. The initial growth processes are likely sub-grid scale (Jacobson and Seinfeld, 2004) and cannot be captured in large scale models. Thus, for the application in global models normally the size distribution of of aged plumes is prescribed, as discussed in Dentener et al. (2006). In particular also the radius of 40 nm for biomass burning seems to be on the very lower end of observations, see Fig. C1 in Dentener et al. (2006). How will this affect your results?

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Again, it is seems important to state here that primary sulfate is emitted here as surrogate for BC and OC as this makes clear how the model apparently treats these species.

Is all OC emitted as primary particles or is it partly condensed on pre-existing particles? This would shift the size-distribution to larger sizes.

You state that the complete removal of in-cloud scavenging only slightly increases the CN by 10%. This is intereting as this process is in most models the dominant sink. How are the sinks distributed among the different reomoval processes in GLOMAP? Are below cloud scavenging or dry deposition important contributors? I was surprised by this small number as in particular the very small particles below the Greenfield gap have non-negligible scavenging efficiencies. How is the size-depended scavenging dealt with in the model? This should also be briefly included in the model description. It migh be also interesting to state how the CCN concentration is affected by this.

The discussion on the aerosol numbers seems to be somewhat decoupled from the discussion of the size distribution. From Fig. 4 it seems that a over-prediction of the number seems to be associated with an underestimation of the radius. Thus the question arises if the model is really over-predicting the total aerosol mass or if the over-prediction of the number is just a consequence of the low bias in the radii? Are there any other constraints from the measurements that could be used to gain more insight?

It would be nice to use diameters not radii also for the emissions to be consistent with the rest of the manuscript.

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p8889, l17: “A further explanation for the under-prediction in the N. Atlantic could be that emissions inventories for anthropogenic primary particles (BC and OC) are too low in terms of particle number at Aitken mode sizes.” Couldn’t also a possible overestimation of the removal mechanism play a role? You introduce all anthropogenic emissions effectively as sulfate, thus instantly hydrophilic, this will certainly lead to a certain overestimation of the wet removal, or?

p8890, l17: “In these model runs sulfur is the only condensible species.” I am not sure what exactly this sentence refers to.

Section 6 I wonder: could the difficulty to represent sub-grid scale growth processes, with higher sub-grid scale aerosol concentrations, not also play a role in the underestimated growth?

p8891, l10: “...may be due to bad counting statistics...” Once could explicitly state that this selective sampling potentially leads to a high bias in the observations.

Section 7.2 This section could be improved by a few more explanations. What exactly is shown in the Figures? Further, the statement “The good comparison between modelled and observed persistence at this site suggests that the model is correctly calculating the source of secondary particles to the MBL (i.e., from the FT) and would appear to rule out a local particle formation source.” seems a bit strong for me given the dramatic uncertainties in the

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nucleation theory, mechanisms, and the known limitation of the nucleation parameterisation. Thus, it seems a bit too brave for me to state that the model correctly calculates the source of secondary particles to the MBL based on this analysis. Other species, such as marine organics could also play a role that might not necessarily have the same photochemical diurnal cycle as expected for sulfate.

p 8893, l 4: "... reduces the depth of the minimum between..." Insert minimum IN THE NUMBER SIZE DISTRIBUTION.

p 8893, l 25: "The NS03 scheme worsens the comparison..." It sounds like the activation scheme worsens the comparison. It is most likely the setup of this simulations and the chosen updraft speeds that worsen this comparison.

Conclusions, p 8895, l1: "Accurate average MBL aerosol number, 'closed' size distributions and a good comparison between model and observed persistence suggests that a binary homogeneous nucleation scheme correctly calculates the secondary source of particles to the MBL, at least as a global mean." I do not agree with this strong statement. First, to my understanding a global mean value does not make too much sense in this discussion as the conditions have a distinct spatio-temporal variability. Second, measurements indicate the contribution of other species than sulfate in the nucleation process or the initial growth that are just not understood sufficiently. Thus I would suggest to make this statement less absolute.

Figure 1 It would be nice to shade the area covered by the observations in the respective campaigns. Or include the lon/lat ranges in Table 1.

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I looked hard, but I could not find any diamonds in this figure - maybe increase the size or use colour.

Figure 3, 4, 5, 6 Please make the captions self explaining. It is not clear what e.g. the label BC/OC means from the caption of the figures.

Figure 7 Is it possible to use the same diameter range for the model as for the observations? This would guide the eye.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 8871, 2006.