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Interactive comment on "A global off-line model of size-resolved aerosol microphysics: II. Identification of key uncertainties" *by* D. V. Spracklen et al.

Anonymous Referee #3

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General Comments

The manuscript "A global off-line model of size-resolved aerosol microphysics: II Identification of key uncertainties" by D. V. Spracklen et al. describes a variety of sensitivity studies performed with the offline GLOMAP aerosol microphysics model for sulfate and sea salt. A number of generally under-determined parameters in the aerosol microphysics are varied in estimated uncertainty ranges and the effects on the global distributions of condensation nuclei (CN)

and cloud condensation nuclei (CCN) of the simulated sulfate/sea salt system are investigated. With the upcoming availability of microphysical aerosol modules on the global scale, the presented systematic sensitivity analysis is of clear relevance well in the scope of ACP. The manuscript is well organised and clearly written. However, in my opinion the manuscript has shortcomings that should be addressed before publication in ACP.

I understand that the manuscript is only part two of a three part series and that the third paper is planned to be an evaluation of the model. However, the presented second manuscript goes beyond a pure sensitivity study and draws strong scientific conclusions. Due to the absence of a model evaluation, I cannot examine if the drawn conclusions are correct or of relevance for the scientific community. In particular the strong general conclusions about the effects on the global CCN distribution would require a careful evaluation.

I acknowledge that global microphysical aerosol models are still in their infancy and that small steps are a valid approach on the way to a full microphysical description of the global aerosol system. However, although the treatment of the global aerosol system in the manuscript is limited to the system of sulfate and sea salt, almost no attempts are made to analyse or describe how this limitation might affect the results. In fact, to my understanding this limitation might be the key uncertainty in the prediction of some of the discussed parameters that cannot be left out in an explicit uncertainty analysis and should be discussed in the concluding sections and abstract. The authors state that their treatment is analogue to the traditional independent treatment of the different aerosol components in global aerosol models. However, in contrast to the traditional approach with prescribed size-distributions, the explicit simulation of

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the size distribution can only lead to correct results if the relevant components are included. I assume an evaluation would show that the simulated system could be representative in the remote marine regions and this is confirmed by the limited evaluation in the first paper of this series (Spracklen et al., 2005). Therefore I suggest to focus on the regions where the simulated aerosol system is representative and present global and zonal means as means over the oceans.

Specific Comments

Abstract and other parts of the manuscript: Many parts of the manuscript refer to vertical shifts of the aerosol number concentrations given in cm^{-3} STP. In the abstract it concluded for one sensitivity study that "changes in the binary nucleation rate... cause a shift of the upper tropospheric CN layer by as much as 3 km while changes in the absolute concentration are relatively small". In fact, a vertically constant concentration at STP implies an exponential change of the concentration at ambient concentrations due to the exponential decrease of the air density. Vertical shifts at constant STP also imply changes in the total vertical integral of aerosol numbers. This should be considered in the argumentation throughout the paper.

p 3442, l 21: Please explain what is meant by aqueous phase processing. Addition of in-cloud produced sulfate?

p 3444, l 3: Does "dry deposition" include sedimentation or is this only turbulent dry deposition? Actually, switching off dry deposition does not seem to have a large effect on particle in the accumulation mode - indicating a good representation of the size-resolved dry deposition parameterisation.

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p 3447, l 9 + Fig. 5: Are this profiles for a single grid box? Please give coordinates.

p 3448, l 28: The small effect of the accommodation coefficient could also be a result of the small available surface area due to the neglect of other aerosol components. How would you expect this to affect your results?

p 3449, l 28: What do you mean by "are more likely to be coagulation scavenged"? This would speed up their growth to the CN size range.

p 3450, Section 5.2 + 5.3: The definition of activation does not become clear to me nor is it described in the Spracklen et al. (2005) paper. What does it imply and how does it relate to scavenging? How did you choose the effective scavenging radius? Is addition of in-cloud produced sulfate the only growth mechanism in the liquid phase?

p 3453, Section 5.4: The emphasised strong sensitivity of the CN and CCN distribution to the emission of primary sulfate might well be an artifact due to the neglect of other, observably predominant, sources of primary aerosol. While for example the cited reference Adams and Seinfeld (2002) discussed a similar effect of primary sulfate, these authors clearly stated that in their simulation the primary sulfate could act as surrogate for other primary emissions. As the used binary nucleation scheme does simulate little particle formation in the lower troposphere, the inclusion of primary sulfate emissions is the only source of primary particles in these simulations and therefore has to result in a strong effect. From Figure 12 it is discernible that the largest changes occur over the continents where other aerosol components are not negligible and constitute a significant part of the surface area available as condensational sink. Therefore, a more critical analysis and discussion seems appropriate here, in the conclusions, and in the abstract.

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Interactive Comment

p 3455, Section 5.4: You state that with the inclusion of primary sulfate emissions your simulated CN become comparable to measurements in polluted regions. Would you expect CN in polluted regions to be solely determined by sulfate?

p 3456, Section 5.4: I am a bit surprised to see this linear relation between emission rates and "CCN'" in a microphysical aerosol module. Could this be a result that you concurrently change the amount of primary emissions so that the primary particles and therefore primary surface area linearly increases with the emissions? What effect would you expect when the primary particles and surface areas remained constant or if you had a realistic background aerosol population as condensational sink?

p 3460, l 15: First sentence sounds like you concurrently changed the processes. Please clarify.

Section 7: To my understanding a critical reflection of the limitations of this study would be helpful in this section. You again point out that primary sulfate particles have a large effect and bring your results in better agreement with measurements. I missed a statement about the important of other relevant aerosol components and primary sources here.

Technical Corrections

Figures: In some of the zonal means, e.g. Fig 3a,b and Fig. 4a,b, the contour levels are hard to read. Maybe an additional colour bar could improve the readability.

Fig.1: The colours for the condensation and dry deposition cases appear almost identical on my printout.

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Fig.5: Captions: what are the differences of Fig. a)-d)?

Fig.5c,d and Fig. 7: It would be nice if the standard values were also given as in Fig. 9.

Fig. 16: Caption: split "runhas".

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3437, 2005.