**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on **"Contribution of carbonaceous aerosol to cloud condensation nuclei: processes and uncertainties evaluated with a global aerosol microphysics model"** *by* **J. R. Pierce et al.**

J. R. Pierce et al.

Received and published: 29 August 2007

Response to Anonymous Referee 1

Referee comments in *italics*

Referee 1, thank you for the detailed read that you gave our paper and the comments that you made on it.

The authors present calculations of cloud condensation nuclei concentrations active at 0.2GCM II-prime global model. They contrast results from the Bond et al. and IPCC emission inventories. Then they evaluate the effect of solubility and of internal vs. external mixing state. They demonstrate (1) that the inclusion of carbonaceous aerosol results in a sharp increase in available CCN, (2) varying 'solubility'

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does affect the CCN budget but is not dominant, and (3) that mixing state has little effect on the modeled CCN budget.

The results from this study are important and of interest to the ACP audience. I recommend that the manuscript be published subject to a few clarifications and corrections, and offer the following comments for consideration in revisions to the manuscript.

Comments: The comparison of modeled vs. observed size distribution shown in Fig. 5 is encouraging. However, it should be acknowledged that aerosol (and CCN) number budgets are highly sensitive to the accuracy of the modeled size distribution. It is therefore not surprising that a globally averaged size distribution is in sharp disagreement with results from the grid size averaged size distributions. It is unclear that what resolution is really necessary to reliably predict CCN number and thus quantitative conclusions drawn in thus study are only permissible with extreme qualifications.

We are unsure of what the reviewer would like to be changed here. We have added the sentence "It is not certain what model resolution is necessary to predict accurately CCN concentrations."

The authors distinguish between hydrophilic OM and hydrophobic OM. Under the 'hydrophilic category' they list glutamic acid, pinic acid, norpinic acid, gasoline, glutaric acid, limonene, adipic acid, cholesterol, pinonic acid and α-pinene. Gasoline, limonene, and α-pinene are usually not found in the aerosol phase but are gaseous precursors for secondary organic aerosol formation. According to the Table 1 in Petters and Kreidenweis (2007) the hygroscopicity for most of the listed substances is $\kappa < 0.1$ Similarly the authors list for the hydrophobic OM component oxalic acid, α-pinene, diesel fuel, leucine, hexadecane, myristic acid, hexadecanol, palmitic acid, and stearic acid. I don't understand what the authors mean by referring to diesel fuel. Oxalic acid (Giebl et al., 2002) and particles generated from the ozonolysis of α-pinene (Huff-Hartz et al., 2005: VanReken et al., 2005) are actually hygroscopic and should fall into the hydrophilic category. Since none of the compounds are actually used in the model I am not sure if the amount of detail given in the manuscript is relevant. Nevertheless, the assumed values $\kappa = 0.12$ (critical dry diameter of 140nm at 0.2%, the value $\kappa = 0.18$ given in the paper seems to large) for hydrophilic OM and $\kappa = 0$ for hydrophobic OM are reasonable.

This is correct, we have removed this discussion from the manuscript and the paragraph now reads, "Hydrophobic and hydrophilic OM each represent a mixture of organic components with varying activation behaviors. We assume that hydrophilic OM has a critical dry diameter of activation of 140 nm at 0.2% supersaturation (the cor-

responding value of the κ parameter discussed in Petters and Kreidenweis (2006) is 0.12), a value representative of more hygroscopic organic compounds. The hydrophobic OM was assumed to be insoluble (κ =0). Model simulations that assumed a low solubility (0.01 g per 100 cm^3 H2O) as opposed to no solubility were performed, and the resulting CCN(0.2%) concentrations differed by <1%. The assumed density of hydrophilic OM is 1.4 g cm^{-3} and hydrophobic OM is 1.8 g cm^{-3}. These values are within the range used in (Kinne et al., 2003) and the CCN predictions do not depend strongly on the assumed density (it depends more strongly on the moles of solute)."

The distinction between 'solute' and 'seeding' effect is fuzzy. First I believe it is more appropriate to use the term hygroscopic vs. non-hygroscopic rather than soluble vs. insoluble. True, insoluble particles are also non-hygroscopic but generally solubility does not predict hygroscopicity. Since the distinction between hydrophobic and hydrophilic OM is really one in hygroscopicity I suggest to change 'solute effect' to 'hygroscopicity effect'. Furthermore, I would consider a 'seeding' effect something that results from the nucleation (or direct emission) of particles, which then grow to sizes where they are CCN active. Since your model assumes that primary emissions fit a lognormal size distribution function with mass median diameter of 100nm and a geometric standard deviation of 2 for both EC and OM, and because secondary organic aerosol is not considered, your model is always seeding the carbonaceous aerosol. The only difference between the OM insoluble (non-hygroscopic) and OM soluble (hygroscopic) simulations is the diameter when they become CCN active. Thus in either case CCN production are caused by 'seeding' and distinguishing them seems artificial.

It is correct that in either case the CCN increase from carbonaceous comes from the addition of new particles (because we have not included SOA). We have made the paper more clear that we are using primary carbonaceous only by changing the reference to carbonaceous aerosol in the model to primary carbonaceous. Distinguishing 'seeding' and 'solute' is artificial, but it allows us to quantify the sensitivity of CCN to the OC solubility. We prefer the "solute" effect to "hygroscpicity" effect because it is the number of solute molecules in Kohler theory that are used to determine the activation diameter. This was not clear in the abstract and we have changed the text toĚ "Sensitivity studies were performed to determine the relative importance of the organic solubility/hygroscopicity in predicting CCN. In a sensitivity study where carbonaceous

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aerosol was assumed to be completely insoluble, concentrations of CCN(0.2

We have also changed "solubility" to "solubility/hydroscopicity" throughout the paper.

Reference Chung and Seinfeld (2001) should be (Chung and Seinfeld, 2002)

Done, Thanks.

References Chung, S. H. and J. H. Seinfeld, 2002: Global distribution and climate forcing of carbonaceous aerosols. J Geophys Res, 107, D4407. Giebl, H., A. Berner, G. Reischl, H. Puxbaum, A. Kasper-Giebl, and R. Hitzenberger, 2002: CCN activation of oxalic and malonic acid test aerosols with the University of Vienna cloud condensation nuclei counter. Journal of Aerosol Science, 33, 1623-1634. Huff-Hartz, K. E. H., T. Rosenorn, S. R. Ferchak, T. M. Raymond, M. Bilde, N. M. Donahue, and S. N. Pandis, 2005: Cloud condensation nuclei activation of monoterpene and sesquiterpene secondary organic aerosol. J Geophys Res, 110, D14208. Petters, M. D. and S. M. Kreidenweis, 2007: A single parameter representation of aerosol hygroscopicity and cloud condensation nucleus activity. Atmos Chem and S3513 Phys, 7, 1961-1971. VanReken, T. M., N. L. Ng, R. C. Flagan, and J. H. Seinfeld, 2005: Cloud condensation nucleus activation properties of biogenic secondary organic aerosol. J Geophy Res, 110, D07206.

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