Atmos. Chem. Phys. Discuss., 7, S2539–S2542, 2007
www.atmos-chem-phys-discuss.net/7/S2539/2007/

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.

Anonymous Referee #4

Received and published: 20 June 2007

1 General remarks

The manuscript by Pierce et al. introduces the modeling of carbonaceous aerosols to the sectional aerosol model by Adams and Seinfeld (2002) applied in the GISS GCM. Compared to previous studies investigating the influence of carbonaceous aerosols on climate, this offers the advantage that more degrees of freedom are allowed for the evolution of the aerosol size distribution. The higher degree of sophistication allows to

test simpler models. Using this capability, the authors infer their most important conclusions of this paper, namely that the information about aerosol size distribution is more important than an accurate knowledge of aerosol chemical composition in order to infer the cloud condensation nuclei (CCN) concentration, and that models with a simpler description of the aerosol size distribution can indeed very well describe CCN concentrations. The authors also find that according to their model, the impact of the increased aerosol number concentration by carbonaceous aerosols is very large even if carbonaceous aerosols are considered entirely hydrophobic.
These qualitative findings are of high interest.
Quantitative results, however, need to be seen with caution due to the many assumptions in the aerosol model.
Also, very few model evaluations with observational data are shown, and these show relatively modest results. Indeed, evaluating a coarsely-resolved global model with point measurements as attempted here is difficult. The credibility of the model results would be much improved if comparisons to satellite data were shown allowing to assess the distributions simulated by the model. Aerosol optical depth for total aerosol concentration and Angstrom exponent for aerosol size distribution evaluations would be options.

2 Specific remarks

- The term "solute effect" seems very unclear to me, in particular when it is opposed to the term "seeding effect". In fact, it is not the pure solubility of carbonaceous aerosols which is of importance here (as it were, e.g., if by coating of insoluble aerosols by carbonaceous aerosols there became potential CCN), but the combination of the solubility and the increased number concentration. I think a simple straightforward formulation of the finding would be more understandable (e.g.: "The impact of carbonaceous aerosols on CCN concentrations is very large

ACPD
7, S2539–S2542, 2007

even if this aerosol type is considered entirely hydrophobic. The increase in CCN concentration amounts to about half the increase computed for the more realistic assumption of partially soluble carbonaceous aerosols.").

- (p 7726, l 14): Do you mean "locally dominant"? Or do you mean that indeed the effect of carbonaceous aerosols might dominate the one by sea salt and sulphate?
- (p 7726, l 21): Why "must" such studies use empirical relations to estimate CDNC? There are other approaches which don't use such simple formulations.
- (p 7726, l 27): Please be more specific about what you mean by "the aerosol general dynamic equation".
- (p 7728, l 10): "Highly accurate": The approach itself may in principle allow for more accurate simulations than simpler approaches. However, whether or not a simulation using this approach is more accurate than using a simpler formulation depends on whether or not the assumptions and input fields are realistic. A very sophisticated model is in some respect even more likely to fail than a simpler one.
- Do you have any estimate of the uncertainty in modeled CCN concentrations due to the assumptions considered in your manuscript as the main model uncertainties (the OM:OC ratio, p 7732, l 7; the assumed emission size distribution, p 7732, l 25; and the assumed aging of hydrophobic aerosols, p 7734, l 18)?
- Please add references to justify the assumptions on the densities of hydrophilic and hydrophobic OM (p 7733, l 23 and 28).
- Might here be some problem in the model, given that you simulate too little mass (Fig. 2) but too large numbers (Fig. 4)? Or is a bias in the simulated size distribution the reason (which, however, can't be told conclusively from the comparison of Figs. 2, 4, and 5)?

ACPD

7, S2539–S2542, 2007

- (p 7736, l 8 and Tab. 2): The lifetimes of EC and (for IBASE) OC are not marginally outside the range of the other studies cited here. They are rather 50
- In Fig. 6, parts (a) and (b) seem identical, and certainly don't allow to conclude a 65

3 Technical remarks

- The entire Introduction paragraph seems quickly written and would merit some revision of the formulations.
- p 7724, l 6: The "(0.2
- p 7725, l 1: Please cite the report of 2007, and do so in the recommended way by citing the pertinent chapter.
- p 7727, l 9: "in global models"
- p 7730, l 18: drop "-" from "in which"

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 7723, 2007.