Atmos. Chem. Phys. Discuss., 7, S4496–S4503, 2007
www.atmos-chem-phys-discuss.net/7/S4496/2007/

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.

J. R. Pierce et al.

Received and published: 29 August 2007

Response to Anonymous Referee 4

Referee comments in *italics*

Referee 4, thank you for the detailed read that you gave our paper and the comments that you made on it. We appreciate you helping us make a better paper.

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.

We have not done a comparison of the model results here with aerosol optical depth or angstrom exponent measurements because in general they are not yet good indicators of aerosol size distributions, CN, or even CCN (Kapustin et al., 2006). Although we have used point measurements (or a compilation of point measurements in the case of the (Heintzenberg et al., 2000) marine distributions), these are actual measurements of the aerosol size distribution or CN. For the revised paper we have added comparison of the size distributions over Europe with measurements published in (Putaud, 2003).

We are currently performing a comparison of the model to measured aerosol optical properties in a model that includes of the species included here, plus dust. This will be published in a future paper.

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 even if this aerosol type is considered entirely hydrophobic. The increase in CCN concentration amounts to about half the increase computed

ACPD

7, S4496–S4503, 2007

for the more realistic assumption of partially soluble carbonaceous aerosols.").

You are correct that because we are dealing with primary carbonaceous aerosol the increase of CCN due to the carbonaceous aerosol is both due to seeding and solute. We found that it was helpful to break the increases of CCN into these two effects in order to quantify the importance of the solubility properties of primary organic aerosol on CCN. By assuming that all organic aerosol is insoluble during the sensitivity runs it allowed us to determine how sensitive CCN concentrations were to organic aerosol solubility (an uncertain parameter). We appreciate you pointing out the confusing nature of our discussion and have modified the text. In the abstract, the discussion of solubility now reads, "Sensitivity studies were performed to determine the relative importance of the organic solubility/hygroscopicity in predicting CCN. In a sensitivity study where carbonaceous aerosol was assumed to be completely insoluble, concentrations of CCN(0.2%) still increased by 40-50% globally over the no carbonaceous simulation because primary carbonaceous emissions were able to become CCN via condensation of sulfuric acid. This shows that approximately half of the contribution of primary carbonaceous particles to CCN in our model comes from the addition of new particles (seeding effect) and half from the contribution of organic solute (solute effect). The solute effect tends to dominate more in areas where there is less inorganic aerosol than organic aerosol and the seeding effect tends to dominate in areas where is more inorganic aerosol than organic aerosol." Also, in Section 3.4, "However, it is unclear from the base case simulations alone how much the increase in CCN from primary carbonaceous aerosol comes from the addition of new particles "carbonaceous seeding effect" verses the addition of more solute "organic solute effect". To understand this, we look at the sensitivity of our predicted CCN to organic solubility. The sensitivity of the number of CCN to the mixing assumptions is also explored."

(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?

We meant globally dominant based on the work cited in that paragraph, but because of

ACPD

7, S4496–S4503, 2007

large uncertainties we have changed the sentence to read, “Based on these studies, it seems likely that carbonaceous aerosol plays a significant role in the tropospheric CCN budget.”

(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.

These studies are only simulating aerosol mass or presume an existing aerosol size distribution and use anthropogenic sulfate to shift this size distribution (e.g. (Chuang et al., 1997)). In order to determine CCN and therefore CDNC from first principles, the aerosol size distribution must be known/predicted. Otherwise, one has to be assumed to calculate the CCN or the CDNC must be parameterized as a function of the aerosol mass. We have changed the sentence to read, “However, these studies must make assumptions about the aerosol size distribution or use empirical relations to predict CDNC from their predicted aerosol mass.”

(p 7726, l 27): Please be more specific about what you mean by "the aerosol general dynamic equation".

Now reads, “The most fundamental, albeit computationally intensive, method for predicting aerosol size distributions results is solving aerosol microphysics explicitly using the aerosol general dynamic equation (Seinfeld and Pandis, 1998), which governs how the aerosol size distribution evolves as a result of the microphysical processes of nucleation, condensation, and coagulation.”

(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.

We agree that more sophisticated models generally require better input data to realize their potential. The intent of the original sentence was more narrow: to comment on the inherent numerical accuracy of the algorithm. To be more clear the sentence now reads, “Two-moment sectional approaches (Tzivion et al., 1989; Tzivion et al., 1987) and the similar “moving-center” approach (Jacobson, 2002) represent a flexible

ACPD
7, S4496–S4503, 2007

EGU

treatment of aerosol microphysics that reduce the effect of numerical diffusion."

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)?

Changing the OM:OC ratio will scale the Bond (2004) emissions inventory evenly, which should change the mass linearly and the CCN sublinearly. We are using two emissions inventories already, so this explores the sensitivity of CCN to changes in emissions. The sensitivity of CCN to the emissions size distribution is much more complicated and is currently being done in present work. See, for example (Pierce and Adams, 2007).

Please add references to justify the assumptions on the densities of hydrophilic and hydrophobic OM (p 7733, l 23 and 28).

Now reads, "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)."

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)?

We have added another comparison, where we compare the simulated aerosol number size distributions versus four observed European locations where the number was overpredicted in Figure 4. It shows that the number overpredictions in are almost entirely from diameters $<$ 100 nm and that the CCN size are predicted more accurately. It is likely that there is an issue with the primary sulfate emissions and emissions size distribution and perhaps the size distribution of carbonaceous species. The issue of the underprediction of organic mass is consistent with other global modeling studies and is likely due to the lack of SOA in our model.

(p 7736, l 8 and Tab. 2): The lifetimes of EC and (for IBASE) OC are not marginally outside the range of

ACPD

7, S4496–S4503, 2007

the other studies cited here. They are rather 50

This comment, as well as the next is cut off. I assume this was due to the use of a “%” sign when converting to LATEX. We have changed the sentence to, “The burden and lifetime values for the BBASE and IBASE simulations are generally within the range of values presented in the previous work with the EC lifetime being longer in our model.”

We have also added a more detailed budget table that includes the effect of the addition of primary carbonaceous species on sulfate and sea-salt.

In Fig. 6, parts (a) and (b) seem identical, and certainly don’t allow to conclude a 65

Thank you very much for noticing this! This was a mistake that we have fixed.

3 Technical remarks The entire Introduction paragraph seems quickly written and would merit some revision of the formulations.

We have read through the introduction and made some updates.

p 7724, l 6: The "(0.2

Unfortunately, I think this got cut off too.

p 7725, l 1: Please cite the report of 2007, and do so in the recommended way by citing the pertinent chapter.

Done

p 7727, l 9: "in global models"

Done

p 7730, l 18: drop "-" from "in which"

Done

References

Chuang, C. C., Penner, J. E., Taylor, K. E., Grossman, A. S. and Walton, J. J.: An assessment of the radiative effects of anthropogenic sulfate, Journal of Geophysical

ACPD

7, S4496–S4503, 2007

Research-Atmospheres, 102, 3761-3778, ISI:A1997WK26600018, 1997.

Heintzenberg, J., Covert, D. C. and Van Dingenen, R. V.: Size distribution and chemical composition of marine aerosols: A compilation and review, Tellus, 52B, 1104-1122, 2000.

Jacobson, M. Z.: Analysis of aerosol interactions with numerical techniques for solving coagulation, nucleation, condensation, dissolution, and reversible chemistry among multiple size distributions, Journal of Geophysical Research-Atmospheres, 107, ISI:000180428300033, 2002.

Kapustin, V. N., Clarke, A. D., Shinozuka, Y., Howell, S., Brekhovskikh, V., Nakajima, T. and Higurashi, A.: On the determination of a cloud condensation nuclei from satellite: Challenges and possibilities, Journal of Geophysical Research-Atmospheres, 111, ISI:000235966500001, 2006.

Kinne, S., Lohmann, U., Feichter, J., Schulz, M., Timmreck, C., Ghan, S., Easter, R., Chin, M., Ginoux, P., Takemura, T., Tegen, I., Koch, D., Herzog, M., Penner, J., Pitari, G., Holben, B., Eck, T., Smirnov, A., Dubovik, O., Slutsker, I., Tanre, D., Torres, O., Mishchenko, M., Geogdzhayev, I., Chu, D. A. and Kaufman, Y.: Monthly averages of aerosol properties: A global comparison among models, satellite data, and AERONET ground data, Journal of Geophysical Research-Atmospheres, 108, ISI:000186198500001, 2003.

Pierce, J. R. and Adams, P. J.: Efficiency of cloud condensation nuclei formation from ultrafine particles, Atmospheric Chemistry and Physics, 7, 1367-1379, ISI:000244712400004, 2007.

Putaud, J. P.: A European aerosol phenomenology; physical and chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, 2003, European Commission, EUR 20411 EN., http://ies.jrc.cec.eu.int/Download/cc Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics, John Wiley

ACPD

7, S4496–S4503, 2007

and Sons., New York, 1998.

Tzivion, S., Feingold, G. and Levin, Z.: The evolution of raindrop spectra. Part II: Collisional collection/breakup and evaporation in a rainshaft, Journal of the Atmospheric Sciences, 46, 3312-3327, 1989. Tzivion, S., Fiengold, G. and Levin, Z.: An Efficient Numerical Solution to the Stochastic Collection Equation, Journal of the Atmospheric Sciences, 44, 3139-3149, 1987.

ACPD

7, S4496–S4503, 2007