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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 #3

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1 General Remarks

In their manuscript J. R. Pierce et al. present an analysis of the contribution of carbonaceous aerosol to cloud condensation nuclei based on global modeling studies with the TOMAS sectional aerosol scheme coupled to the GISS climate model. For this purpose the authors extend their sectional aerosol model of sulfate and sea salt by primary carbonaceous aerosols (black carbon and organic matter).

The analysis of anthropogenic contributions to CCN is crucial for the understanding of the indirect aerosol effects and therefore a highly relevant research topic well in the scope of ACP. The manuscript is well written and the analysis is innovative - in particular the evaluation of the importance of size vs. composition for CCN predictions is an important quantitative extension of previous work.

However, and in this case unfortunately, to my understanding the manuscript has some major issues that will need a substantial revision of the manuscript before publication. The introduced changes demand for a careful evaluation of the model, in particular as the limited presented evaluation reveals some significant problems: if the simulated CN values are biased high by a factor of 4 the reader needs to be convinced that the (unevaluated) CCN values are reliable. Thus, I would recommend to either significantly extend the manuscript by a basic evaluation, as outlined below, or to split the manuscript into a introductory paper with detailed evaluation and an scientific application of the evaluated model.

2 Major Issues

Evaluation

The authors extended a microphysical model of sulfate and sea salt by carbonaceous aerosols, a substantial modification from their previous work that increased globally averaged surface CCN by 65-90%. This is essentially a new model - with completely new results that demand for a careful evaluation. To my understanding the authors somewhat leapfroged this step (that typically is a publication on its own) and jump right away into the scientific analysis.

This is worrisome as the limited three evaluations performed (surface mass concentrations of BC and OC; surface aerosol number concentration (CN$>$10nm); large scale

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average marine size distributions) show non-negligible deviations that ask for a more detailed analysis. The model over-predicts CN on average by a factor of 4, so I am missing a supportive analysis why the (unevaluated) CCN predictions should be a reliable basis for their subsequent scientific analysis. The model predicts also very high CCN surface concentrations, e.g. CCN(0.2%) are in the annual mean $>$ 1000 cm^{-3} over large parts of the continents and even $>$ 2000 cm^{-3} in polluted areas. These values seem high given that e.g. Hudson and Yum (2002) report CCN concentrations for polluted air masses as CCN(1%): 1190 cm^{-3}, CCN(0.1%): 580 cm^{-3} and demand for a more detailed evaluation. It is true that CCN measurements are not generally available on an operational basis. Therefore, an evaluation of the aerosol optical depth, that is in these areas typically dominated by the CCN relevant accumulation mode size range, could be an essential first evaluation step. However, also the limitation of the evaluation of the size-distributions to marine areas that, according to Fig. 3 and Fig. 6, are almost unaffected by anthropogenic aerosols is insufficient. Suitable datasets are available (e.g. Putaud et al., 2003) and have been used as benchmark in previous modelling studies. Finally, I would have expected some effect of the introduction of carbonaceous aerosols on the distribution of the other species, as modifications in the size distribution and composition directly affect the sinks. It would be a surprising and interesting results if the other species are unchanged. However, if they changed, the changes would need to be discussed in the manuscript and the species maybe re-evaluated.

2.1 Calculation of CCN

The basis of the calculation of CCN in the manuscript is somewhat unclear. In section 3.4 (page 7740, line 25) the authors write:

"The CCN(0.2%) concentrations are found using modified Köhler theory as discussed earlier with the **annually averaged size distributions and chemical compositions**."

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I could not find any information if this applies for all calculated CCN concentrations in the manuscript or not - thus I have to assume it does apply. This would to my understanding significantly affect the (interesting) analysis of the effect of aerosol size vs. composition in Section 3.5. If all CCN are derived from annual mean size and composition data, the model effectively uses a (locally refined) bulk scheme for the calculation of CCN. In this case the comparison in Section 3.5 and Fig. 7 would not be a comparison of a bulk scheme vs. an average of CCN from an exact instantaneous activation calculation but rather a comparison with a local bulk scheme for each grid box. Given the non-linearity of the activation process, I would be surprised if this yields the same result.

3 Specific Issues

- **Title, Abstract, Conclusions**

  I think it is important to point out that SOA's are not included (also not as proxy), thus I would recommend to consistently use "primary carbonaceous" instead of "carbonaceous".

- **page 7726**

  "cloud brightness forcing" is an unusual term. I assume you mean cloud albedo effect? Otherwise please explain.

- **page 7727**

  "The modal representation has an inherent disadvantage, however, in treating processes such as activation and cloud chemistry that create discontinuities in the size distribution."

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This is certainly true on a local basis. In the grid box average over more than 100x100 km, that global models represent, these discontinuities are rarely observed. This is for example evident in the Hoppel gap of the size-distributions in Fig. 5 that is very sharp in the simulation but log-normal in the averaged observations.

- **page 7728**

  "Carbonaceous particles may affect the CCN concentrations through two different pathways."

  This is a simplified view on this issue as other effects, such as surfactants, are neglected.

- **Section 2.3**

  The description of the calculation of CCN is very limited and should be extended.

- **Section 2.1, page 7731**

  "we neglect interstitial scavenging in clouds"

  Could this contribute to the overestimation of CN as it leaves turbulent dry deposition as the only sink for small particles?

- **page 7731**

  "During microphysics, all aerosols are treated as internally mixed."

  This is a serious limitation of this study as it will affect the growth of particles to the relevant CCN size. I think this requires some more detailed discussion of the potential implications for the results.

- **Section 2.2, page 7731**

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Please give the base years of the emission datasets. Given the availability of significantly improved satellite based wildfire emission datasets, the provide the observed seasonal distribution, the used inventories seem somewhat outdated.

- **Emissions, page 7732**

  I am somewhat surprised by the choice of the emission size-distributions from near source measurements. Given that the initial evolution of aerosols occurs on much smaller scales than the 5 degree grid boxes of the model, the assumption of somewhat aged size distributions would appear more appropriate. This could be the reason for the strong overestimation of CN in the model. Also, in particular downwind biomass burning size distributions have typically much larger sizes, a factor that has been taken into account in the recommendations for e.g. the AeroCom emission size distributions Dentener et al. (2006).

- **Mixing state, page 7733**

  Are hydrophobic and hydrophilic OM really always assumed internally mixed as has to be assumed from this description, dividing the carbonaceous aerosols into a pure EC population and an internal mixture of all other components? How can the internal mixture remain hydrophobic?

- **Mixing state, page 7733**

  The choice of the BC aging timescale seems to be based on a relatively old source. Recent model based estimates, e.g. Riemer et al., yield shorter timescales. This could also contribute to the simulated relatively long BC residence time.

- **Section 2.4, page 7734**

  I was missing a description of the simulation setup here. How long has the model been integrated, resolutions, boundary conditions, such as SST need to be described.

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- **Section 3.1, page 7736**

  It would be important to show the budgets of all components here.

- **Section 3.2, page 7736**

  Please be more specific about the sampling of the data (also in the captions). Is the evaluation also done on an annual mean basis? Why not as monthly means that are available at least from IMPROVE?

- **Section 3.3, page 7738**

  Please be more specific about the evaluation. How was the sampling performed? Do the measurement instruments have the same lower cut-off?

  I am somewhat puzzled by the strong overestimation of CN and surprised that they authors have not repeated the simulations with updated emission size distributions after the significant overestimation of CN by a factor of 4.

- **Figure 5, page 7740**

  The simulated marine size-distributions seem to have a distinct discontinuity, probably at the critical radius of activation, and do not look log-normally distributed - while the observations show smooth log-normal distributions. I am surprised that this is the case in the annual mean values. Is this due to the assumption of fixed supersaturation in clouds?

- **Section 3.4, page 7740**

  As discussed before, I see issues with the calculation of CCN from annually averaged aerosol data.

- **Section 3.4, page 7740**

  I think the authors need to make significant effort to evaluate the predictive skill of the model in terms of CCN - as CN have a significant bias of a factor of 4.

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- **Section 3.4.1, page 7742**

  It might be interesting to show ratios here, as would be easier to identify the regions of importance.

- **Section 3.5, page 7744**

  As discussed before, I see issues with the calculation of CCN from annually averaged aerosol data.

- **Conclusions, page 7746**

  They authors attribute the overestimation of CN on the primary sulfate emission size distribution. On what basis? How do the primary particle number compare to the carbonaceous flux?

- **Conclusions, page 7746**

  I was missing a reference to the caveat of the simplifying assumption of internal mixing in the microphysics and the potential implications here, and in the abstract.

- **Conclusions, page 7746**

  I was missing a discussion of the sensitivity of their results to the emission size distributions.

- **Figure Captions**

  Most of the figure captions are not self explaining and do not give basic information, such as sampling periods, etc.

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