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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Slower CCN growth kinetics of anthropogenic aerosol compared to biogenic aerosol observed at a rural site" *by* N. C. Shantz et al.

Anonymous Referee #1

Received and published: 15 August 2009

The manuscript submitted to ACPC by Shantz et al., 2009 "Slower CCN growth kinetics of anthropogenic aerosol compared to biogenic aerosol observed at a rural site" is an interesting study interpreting measurements of particle activation with respect to possible growth limitations, and carrying the obtained results further to estimate their effect on clouds by doing cloud parcel model simulations. The study is based on atmospheric aerosol particles, measurements were done on a rural site that sometimes was influenced by polluted anthropogenic aerosol.

Summarizing, the authors find that the results from the simulations of the signal produced in their CCNc can be adjusted to the measurements by changing two variables,

the hygroscopicity of the organic fraction of the aerosol or the mass accommodation coefficient of water. While hygroscopicity of the organic fraction can be k=0 or larger, the mass accommodation coefficient (a(c)) can be 1 or lower. These limited ranges allow constraining the possible parameter combinations that lead to an improved agreement between measurement and simulation. The obtained results are somewhat unexpected, as more oxygenated compounds that were observed for anthropogenically influenced aerosol seem to delay particle activation, while this could not be observed for the less oxygenated biogenic aerosol, which, on the other hand, seem to be more hygroscopic than the former.

While error bars are shown for an uncertainty of the super-saturation in the CCNc, an estimation of possible errors originating from measured number concentrations is not given and should be added.

The same is true for a further parameter, the surface tension, assumed in the simulations. How would changing this value influence your results? Could this explain the unexpected differences between anthropogenic and biogenic aerosol?

Also, when using the obtained data on particle hygroscopicity and a(c) for the biogenic and anthropogenic cases in order to do cloud parcel simulations, a new (good) argument appears, saying that it is likely that a(c) changes with the particles getting more and more dilute during the activation process. This possible effect largely influences the results from the cloud parcel model. Therefore, it should be tested in the sections 3.2. and 3.3, in which the growth kinetics were examined, to give the reader an idea of how this influences the agreement between measured and simulated CCNc response, i.e. of how probable this effect is.

One general remark about the Figures: The authors are strongly encouraged to ensure that the Figures will be large enough in the final draft, so that readers will be able to decipher the labels, legends, numbers, etc. .

Some minor and more detailed suggestions for improvements of the manuscript are

given below. ___

page 13777, line 1: It might be considered "textbook-knowledge" by now, but I would still cite Twomey, 1974 and Albrecht, 1989 here, for the change in radiative properties and lifetime, respetively. (Twomey, S. (1974), Pollution and the planetary albedo, Atmos. Environ., 8, 1251-1256. Albrecht, B. A. (1989), Aerosols, cloud microphysics, and fractional cloudiness, Science, 245, 1227-1230.)

page 13777, line 17: There are also studies showing, that the relative humidity (RH) present during the formation of organic biogenic aerosols can increase the particles hygroscopicity: Vesna, O., S. Sjogren, E. Weingartner, V. Samburova, M. Kalberer, H. W. Gaeggeler, and M. Ammann (2008), Changes of fatty acid aerosol hygroscopicity induced by ozonolysis under humid conditions, Atmos. Chem. Phys., 8, 4683-4690. Wex, H., M. D. Petters, C. M. Carrico, E. Hallbauer, A. Massling, G. R. McMeeking, L. Poulain, Z. Wu, S. M. Kreidenweis, and F. Stratmann (2009), Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol - Part 1 - Evidence from measurements, Atmos. Chem. Phys., 9, 3987-3997. The latter also showed that the particles hygroscopicity changes with changing dilution, i.e. with particle growth, and that surface tension no lower than 0.055 J/m2 should be used. All of these are important parameters concerning organic particulate mass, so you might want to add these citations. And, as said above: please add, which surface tension you used in your simulations (at an adequate place in your manuscript), and how the use of different surface tensions influences your results.

page 13778, line 14: There is an even newer reference on this topic: Voigtlaender et al. (2007) examined mass accommodation coefficients for water by comparing measured and simulated droplet growth, and found a value close to 1 (particles consisted of an inorganic salt): Voigtlaender, J., F. Stratmann, D. Niedermeier, and H. Wex (2007), Mass accommodation coefficient of water: a combined computational fluid dynamics and experimental data analysis, J. Geophys. Res., 112(D20208), doi:10.1029/2007JD008604.

page 13779, line 24: Add, if and how you dried the ambient aerosol and the AS aerosol prior to the measurements.

page 13782, line 12: A bit more information about the model would be of use, here.

page 13782, line 3-6: This sentence is difficult to understand. E.g. "... the ambient simulations ..." mean simulations representing the ambient aerosol with a(c)=1 and k(org)=0. Also, the conclusion you give us here ("suggesting that the organic compounds ... inhibited water uptake ...") can only be understood after having read your next argument (that an increase in k shifts the simulation further away from the measurements). Please modify this text passage.

page 13782, lines 8-10: You should add an extra Figure or a graph to Figure 3, showing how an increase in k would influence the simulation – this will give the reader a better understanding of the sensitivities.

page 13782, line 16: Try to find a better expression for "anthropogenic monodisperse cases".

page 13782, line 29: "For simulations of the ambient aerosols, ..." - Did you always use this composition when doing the simulations for the ambient aerosols, or only for the case you show in Figure 4 (13 June)?

page 13783, line 3: "(not shown)" – Please show this simulation in a Figure (e.g. added to Fig. 4), too. Also, add to Figure 4 the simulations for k=0.2 and a(c)=1 and k=0.05 and a(c)=1, similar to what you show for the other cases.

page 13783, line 15ff: More oxygenated compounds are thought to have shorter chain lengths and to be more soluble (as you indicate by citing Kanakidou et al., 2005), so your results are counterintuitive. Do you have any suggestion of what these compounds that cause the growth inhibition might be? Then add this. If not, stress somewhat more that the suggestion you make here is opposite to the up-to-date understanding, and that it is not yet clear as to which compounds could cause this.

page 13784, line 24: The aerosol number and chemical size distributions from when (i.e. which date) were you using?

page 13784, line 24ff: You use the outcome from your monodisperse simulations for the anthropogenic simulation, but for the biogenic case you use your results from the polydisperse simulations. Mention the reasons for this.

page 13785, line 9ff: Referring to what I said in the opening of this review: Please add one more graph to Fig. 3 and/or Fig. 5, that shows how the simulation of the voltage in the CCNc is influenced if you use values for a(c) changing from 0.044 to 1 during particle activation.

page 13786, line 10: Add a "," between "both" and "air"

page 13786, line 10ff: You give a(c) of 0.044 for the anthropogenic case, which comes from examining the monodisperse case, and do not mention the 0.07 from the polydisperse case. Likewise, you do not mention k=0.36, that you used as the upper margin when simulating the polydisperse biogenic case. Add these values in the conclusions, too.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 13775, 2009.