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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.

**N. C. Shantz et al.**

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Response to Referee #1: First of all, we would like to thank Referee #1 for the detailed review of our manuscript, with constructive comments and suggestions. Below are our replies to the specific comments provided by the Referee. Please note that section numbers have changed in the newest version of the manuscript and the new section numbers are referred to below. All page and line numbers below refer to the original manuscript in ACPD.

Referee general comment: While error bars are shown for an uncertainty of the super-saturation in the CCNc, an estimation of possible errors originating from measured



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number concentrations is not given and should be added.

Response: The uncertainties in the number concentrations have been added to the manuscript. The number concentration is limited by the flow in the CPCs and this has been added to the instrumentation section on Page 13779 line 27. The error in the average number concentrations, evaluated based on 2 x standard error of the mean (2 x stdev/sqrt(N) where N is the number in the sample), was provided in the following locations in the manuscript: Monodisperse ambient anthropogenic and AS observations in the text on page 13781 line 7-8, Monodisperse ambient biogenic and AS observations in the text on page 13782 line 22-23, Polydisperse ambient anthropogenic observations figure 5b, Polydisperse ambient biogenic observations figure 6b,

Referee general comment: 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?

Response: Thanks for the suggestion. We performed sensitivity tests to the surface tension and found that the growth curves go to higher values as we decreased the surface tension, as expected. In order to match with the observations, a decreased organic kappa would be required. We have decided to not include these simulations in the manuscript. We have included a short discussion that kappa captures the variations in the CCN activity that arises from the differences in chemical composition and that would include any reductions in surface tension, even though surface tension is not explicitly in the kappa formulation (shown in the new Section 3). We have also added an explanation to the end of Section 4.3 (page 13784 line 17-18), including the sentence "We do not attempt to specify which component(s) are responsible for changes in the kappa."

Referee general comment: Also, when using the obtained data on particle hygroscopicity and a(c) for the biogenic and anthropogenic cases in order to do cloud parcel sim-



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ulations, 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.

Response: Thanks for the suggestion, great idea. We have added this to Section 4.2 (previously Section 3.2) and figure 3b. We have expanded on the discussion about this possibility with literature references (discussing how likely this would be, including the Wex et al. (2009) reference mentioned later in your comments) on page 13782 after line 10 and included a description of the new simulation to page 13782 line 15.

Referee general comment: 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.

Response: Thanks for the suggestion, we will definitely ensure that in the final draft, the fonts are large enough.

Referee detailed suggestions: 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.)

Response: Thank-you for the suggestion, we have added the 2 suggested references as well as one other (Liou and Ou, JGR, 1989) to page 13777 line 1.

Referee detailed suggestions: 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. Weingart-



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ner, 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.

Response: Thanks for these suggestions, we have added the 2 suggested references to the introduction (page 13777 line 16). We have pointed out that the Wex et al. article demonstrates a change in hygroscopicity with changing dilution to page 13777 line 18. As discussed above, we have decided to not include the surface tension sensitivity test simulations in the manuscript but we have included a short discussion that kappa captures the variations in the CCN activity that arises from the differences in chemical composition, including any reductions in surface tension (the new Section 3 that includes a more complete description of the model(s) used here, as well as a new paragraph added to page 13784 line 18).

Referee detailed suggestions: 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.



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Response: Thanks for pointing out this reference, we have added it to page 13778 line 15 as well as page 13780 in the new model description, Section 3.

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

Response: We did not actively dry the sample aerosol but did dry the AS aerosol from the atomizer. In particular, we did not want to experience ambient aerosol loss by taking the particles through a dryer. Also, it was generally cooler outside than inside the sampling building, so that ambient particles will have dried out after entering the warmer indoor conditions. It is also the case, for the monodisperse cases, that the sheath flow is dried in the DMA and we have observed that this tends to dry the aerosol flow considerably compared to the incoming sample flow. Thus, we feel that the particles being sampled will have little water content. Additionally, the relative humidity and temperature were very similar for the anthropogenic and biogenic cases so this would not explain the difference between the two. This has been added to page 13779, line 24.

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

Response: A new Section 3 describing the model has been added to the manuscript on page 13780 line 11. Thanks for the suggestion.

Referee detailed suggestions: 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.

Response: This has been modified. We agree that this was confusing. The first part ".



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. . the ambient simulations . . .” has been reworded and the second part (“suggesting that the organic compounds . . . inhibited water uptake . . .” has been moved to page 13782 line 11.

Referee detailed suggestions: 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.

Response: This change has been made to Figure 3, thanks. We now show in this figure kappa=0, 0.1 and 0.2.

Referee detailed suggestions: page 13782, line 16: Try to find a better expression for “anthropogenic monodisperse cases”.

Response: Thanks for the suggestion, this expression has been reworded.

Referee detailed suggestions: 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)?

Response: You are absolutely right but we have now removed this entire discussion (monodisperse biogenic simulations- see next suggestion/response).

Referee detailed suggestions: 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.

Response: We have decided to remove all mention of the monodisperse simulations. The explanation remains (page 13783 lines 5-6) that the number concentrations are just too low so that this work is highly uncertain and kappa_org could not be determined in this case. We refer the reader to look at Section 4.3 for analysis of the biogenic scenario, for a case that is much less uncertain.



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Referee detailed suggestions: 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.

Response: We do not have suggestions about which kind of compounds would cause this growth inhibition. We suggest that the anthropogenic aerosol, with more OOA-1, slows down the water uptake compared to the biogenic aerosol and "is counter to the more common concepts of organic CCN activation" and that it is not clear as to which compounds lead to this result (page 13783 line 15). This paragraph, starting on page 13783 line 9, has been moved to the end of the following section, Section 4.3.

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

Response: This correction has been made to page 13784 line 24 as follows: "...anthropogenic (7 June and 1 June) and biogenic (11 June)..."

Referee detailed suggestions: 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.

Response: We have now included both anthropogenic simulations (monodisperse and polydisperse cases) as well as the polydisperse biogenic case, at your suggestion.

Referee detailed suggestions: 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.



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Response: This correction has been made to Figure 3, Section 4.2.

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

Response: The words in this sentence were rearranged slightly so that it makes more sense.

Referee detailed suggestions: 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.

Response: This change has been made here as well as in the abstract. Thanks for all of the detailed suggestions.

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