

Interactive comment on “The effect of organic compounds on the growth rate of clouddroplets in marine and forest settings” by N. C. Shantz et al.

Anonymous Referee #1

Received and published: 21 May 2008

In this paper, measurements of growth rates of aerosol particles are presented. The technique has been applied previously in lab experiments but now it is the first time that ambient particles are used. The particles were samples at two distinctly different locations (marine, forest). Whereas many prior studies addressed the ability of organics as cloud condensation nuclei (CCN), only very few studies have been focused so far on the kinetic behavior, i.e. growth rates, of organic (or mixed inorganic/organic) particles. The present study addresses this topic by comparing growth rates of particles of different composition. The growth rates that are observed in the CCN counter are compared to those that are predicted by a model. The model is initialized with measured number concentration and aerosol composition whereas several assumption have been made for the organic fraction. The only parameter that is varied in order to match observed

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growth rates is the solubility of the organics; all other parameters are kept constant. based on their model studies, the authors conclude that the organic fraction in marine aerosols does not contribute to water uptake whereas in particles that have been collected above forests its contribution may be important. Whereas the conclusions - that are in agreement with prior studies using different methodologies – are important for our current understanding of the role of organic in aerosol/cloud-interactions, there are several points in the paper that should be clarified prior to publication.

General comments - The paper includes two different concepts that should be distinguished more clearly: (1) The water uptake is a dynamic process and the organics are slowly dissolved and, according two Koehler theory, the hygroscopicity (κ) changes over the course of particle/drop growth (kinetic approach). (2) The organics have a defined hygroscopicity (κ) that is lower than the inorganic fraction (thermodynamic approach) (Section 5). Whereas the second approach has been used in many previous studies, the effect of 'delayed growth'; by organics has not been discussed in many studies yet. However, it has been shown that mixed inorganic/organic particles usually have a very low efflorescence relative humidity (ERH), e.g., [Marcolli et al., 2004]. Even at a few % RH all organic might have been dissolved even though in a pure state they might have a lower solubility. Given such results, I am not sure whether reporting the solubility of organics (even though the authors point out that it probably does not reflect the 'true solubility'; anyway) is a useful parameter. The comparison of figure 7 and 13 suggests that assuming either a dissolving organic compound or a organic compound with a smaller κ leads to the same result in terms of the predicted growth rate. It should be made clear why the predicted solubility might be a better/equally useful parameter to represent organic properties.

- The difference between 'solubility' and 'hygroscopicity'; should be made clearer throughout the paper. Whereas inorganic salts (and possibly some organics) are very soluble, their hygroscopicity (proportional to κ and the solute term in the Koehler equation) and, thus, CCN ability can differ significantly.

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- The water soluble organic carbon (WSOC) measurements have been used as a measure to determine the soluble vs. insoluble OC fraction. However, recent CCN studies suggest that WSOC might not be dissolved at the relatively small amounts of water as present in the CCNc [Ervens et al., 2007]. Only at high dilution as used in order to dissolve filter samples, this OC fraction might be really dissolved. Can you give an estimate of the water on the activated particles? I.e. organics of which solubility can be dissolved in this water volume?

Specific comments p. 8206, l. 11: What is the range of assumed solubilities?

p. 8208, l. 9-11: Is the conclusion here that the organics only act as 'carrier'; and increase the size of particles? Do larger particles grow faster than smaller ones?

p. 8209, l. 20: Are the 17% and 4% the sulfate fractions? If so, you should reword this sentence as you talk here about sulfate/organic ratio.

p. 8210, l. 8: Is the molecular weight of the insoluble (BC) fraction used at all? In the Koehler equation usually only the insoluble fraction is used.

p. 8210, l. 19-22: Where would be the activation diameter if you assumed purely inorganic particles? Can mode 1 ever be activated at these S?

p. 8210, l. 22: You should remind the reader here (once more) that the observed growth rates are a combination of particle number, and size (and composition).

p. 8210, last line: Replace 'insoluble curves' by 'curves representing insoluble organics' or something like that (check also the remainder of the manuscript for this term).

p. 8214, l. 25: Mixing processes usually increase the CCN ability of particles since any soluble material added to a previously insoluble particle increases its hygroscopicity. Thus, mixing is unlikely to be the reason of decreasing kappa. The argument you give at the end of the 'summary and conclusion section', i.e. increased organic mass fraction seems more reasonable.

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p. 8215, l. 10: Replace 'the organic is of no consequence' by something like 'organics do not contribute to water uptake and appear to be insoluble'.

p. 8215, l. 23-26: You should also add 'molecular weight' to the list of these assumed parameters since it determines also the organic hygroscopicity.

Figures 2, 4, etc: Add either in the caption or in the figures itself that the numbers on top of the plot are the supersaturations.

Additional references

Ervens, B., M. J. Cubison, E. Andrews, G. Feingold, J. A. Ogren, J. L. Jimenez, P. DeCarlo, and A. Nenes (2007), Prediction of cloud condensation nucleus number concentration using measurements of aerosol size distributions and composition and light scattering enhancement due to humidity, *J. Geophys. Res.*, 112, doi: 10.1029/2006JD007426.

Marcolli, C., B. Luo, and T. Peter (2004), Mixing of organic aerosol fractions: liquids as the thermodynamically stable phases, *J. Phys. Chem. A*, 108, 2216-2224.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 8, 8193, 2008.

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