

Interactive comment on “Classifying organic materials by oxygen-to-carbon elemental ratio to predict the activation regime of cloud condensation nuclei (CCN)” by M. Kuwata et al.

Anonymous Referee #1

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This paper relates the CCN-derived hygroscopicity of organic-rich particles, composed either of pinonic acid, a C₆ – C₉ diacid, or secondary organic matter (SOM, either isoprene or α -pinene precursor), to the bulk water solubility of the OM. Many previous studies have dealt with this issue, and the major contribution of this paper is to relate the transitions between insoluble, slightly soluble, and highly soluble compounds to O:C ratios of 0.2 and 0.6. This subject will be of interest to ACP readers, and this data set and analysis are sufficiently extensive to allow more firm conclusions to be drawn than in previous studies of the relationship between solubility and CCN activity. I recommend publication in ACP after the following concerns are addressed.

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Major comments:

The authors found that pimelic acid was more CCN-active than predicted. In lines 16-27 on page 31839, the authors hypothesize that an increase in solubility with temperature accounts for this difference. I had several issues with this paragraph – first, due to its speculative nature, I feel it belongs in the discussion section (if anywhere), and not in the results. Furthermore, and more importantly, the authors cite similar results from Hartz et al. (2006) and Frosch et al. (2010) – but both of these studies used a static thermal-gradient CCN instrument, which involves active cooling and therefore temperatures at activation below room temperature. For example, for the Wyoming CCN instrument used in Frosch (2010), the temperature at activation is < 293 K (Snider et al., 2010). Therefore it seems that the enhanced CCN activity of pimelic acid occurs at lower temperature as well. This is in contrast to the actively heated CCNC used in this work, which as the authors point out involved activation at $T=303$ K. This difference in temperature between static and continuous-flow CCN instruments has been used in a previous study to constrain the influence of T on CCN activity (Asa-Awuku et al., 2009). Finally, as the authors point out, temperature effects were not observed for other compounds, and it is not clear why if temperature effects are important. Given all this, I feel that the authors should either test their hypothesis by presenting results taken at a lower temperature, for example 293K instead of 303K, or omit this paragraph.

The authors use small inorganic seed particles when generating SOM. However, they report κ_{CCN} without making it clear if this value is for the mixed particle or if they have used, e.g., the ZSR rule to isolate the organic fraction of the particle. Please include the details of how (if at all) the calculations accounted for the inorganic portion of the dry particles.

Table 2: Along with observed κ , the volume fraction organic in the dry SOM particle (or alternatively, the diameters of the SOM-coated particles) would be of interest. I recommend adding it to this table. The text mentions SOM diameters from 30 to 300 nm, but I'm assuming this wide range was not used for each experiment.

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Table 4: The water activities at deliquescence ($a_{w,D}$) cited from Chan et al. (2008) are actually the water activities of saturated macroscopic solutions (a_{sat}), so it is not correct to say that “Chan et al. (2008) measured $a_{w,D}$ ”. The authors explain for some of the other $a_{w,D}$ values how they were calculated from bulk solubility data; they should add that Chan et al. (2008) used the same approach. There are other places in the manuscript that imply that Chan et al. (2008) measured ($a_{w,D}$) directly (see below); they should also be corrected.

Minor comments:

P 31835, L 21: Recommend that “highly soluble” be changed to “NaCl” or “salts with high ion densities”, since “highly soluble” has already been used (on page 31834) to describe organic compounds with aqueous saturation concentrations greater than 0.1 v/v. Such highly soluble organic compounds have κ much lower than 1.3.

P 31839, L 3-4: As stated above, Chan et al. (2008) did not directly measure $a_{w,D}$, but rather estimated it based on bulk solubility data. The authors should change the text to indicate this.

P 31840, L 7-11: the authors state that for azelaic acid, the measured κ of 0.02 to 0.03 is “close to the theoretic result for limiting solubility”, but then that it is “higher than the theoretically predicted value of 0.001-0.002”. This seems contradictory, please clarify.

P 31840, L 12-13: the authors state that $a_{w,D}$ for azelaic acid is “0.999 as reported in the literature and as predicted from solubility data”. Please include a citation if this has in fact been measured directly. If it has not, I recommend omitting the phrase “as reported in the literature” and leaving “as predicted from solubility data.”

P 31840, L 24: “ $a_{w,D} = 0.999$ as reported in the literature” – again, if this has been reported in the literature, please provide a citation.

P 31845, L 10-13: I don’t think this is a satisfactory explanation, for reasons given above. I recommend either addition of pimelic acid CCN data at lower temperatures,

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or that these lines be removed.

References:

Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics of β -caryophyllene secondary organic aerosol, *Atmos. Chem. Phys.*, 9, 795–812, doi:10.5194/acp-9-795-2009, 2009.

Chan, M. N., Kreidenweis, S. M., and Chan, C. K.: Organic Compounds of Different Solubilities in Water and Their Relationship with Cloud Condensation Nuclei Activities, *Environ. Sci. Technol.*, 42, 3602-3608, 2008.

Frosch, M., Zardini, A. A., Platt, S. M., Müller, L., Reinnig, M.-C., Hoffmann, T., and Bilde, M.: Thermodynamic properties and cloud droplet activation of a series of oxoacids, *Atmos. Chem. Phys.*, 10, 5873–5890, doi:10.5194/acp-10-5873-2010, 2010.

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Snider, J. R., et al. (2010), Intercomparison of cloud condensation nuclei and hygroscopic fraction measurements: Coated soot particles investigated during the LACIS Experiment in November (LEXNo), *J. Geophys. Res.*, 115, D11205, doi:10.1029/2009JD012618.

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