

Interactive comment on “Cloud condensation nucleus (CCN) behavior of organic aerosol particles generated by atomization of water and methanol solutions” by T. A. Rissman et al.

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Received and published: 10 February 2007

The authors have done an excellent job of carefully considering the importance of solvent choice in the measurement of CCN activity of small organic acids. These results will provide a new framework in which to consider future CCN activation measurements. However, I believe that there are a few areas in which it would be beneficial for the authors to expand and clarify some of their discussion.

1. (Page 13270-13271) The CCN activity of the aerosols was considered for water and methanol solutions, both with and without inclusion of a heating step. In addition to the suggestions of Referee #1 (that further discussion of this heating step is desirable),

I believe it would also be useful if the authors provided some discussion of why they chose to include the heating stage prior to drying the aerosols in a series of diffusion driers. As a result, the heating occurred when the samples were still relatively wet (at least compared to after the drier) which may have significantly influenced role of heating on the relative importance of chemistry vs. morphological changes vs. a direct influence of residual solvent molecules on the observed CCN. Can the authors comment on this choice? Were experiments also tried where the heater was placed after the drier? I suspect that this might lead to quite different results.

2. (Page 13274) The authors assert that esterification might occur to a larger extent in smaller rather than larger particles for the methanol+heater+adipic acid studies. Although such a conclusion appears consistent with the observations, the authors should provide a physical rationale for this suggested phenomenon, i.e. why would smaller particles have greater reactivity upon heating. This conclusion does not seem self-evident and should be justified with further discussion. Ideally, size resolved composition measurements would have been made to validate this statement, however composition measurements were only made on the total of 300 nm, 200 nm, 100 nm and 50 nm particles. Such measurements could provide direct support for the claim that a greater fraction of the small particles are converted to esters.

3. (Page 13274-13275) In relation to the occurrence of esterification reactions in the methanol+heater+adipic acid studies, it is suggested that for 250 nm particles perhaps 18% of the sample is (non CCN-active) esters because $AR = 0.82$. The origin of this conclusion should be clarified and the discussion expanded. Do the authors assume that the heated trial should be directly compared to the not heated trial and the difference between the two is the approximate ester fraction? If this is the case, it would then seem that, for example, 150 nm particles are actually then 60-80% ester (because $AR = 0.2-0.4$), at least for the two highest supersaturations. If this were true, then the ester peak in the LC/ESI-MS analysis should be quite large. Can the authors comment on the quantitative nature of the LC/ESI-MS method used in this study and

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whether the data are consistent with a significant fraction of the collected sample being ester derivatives? Additionally, were analyses performed on non-heated aerosol samples generated from methanol solutions to determine whether the heating step was responsible for the ester formation?

4. (Page 13274-13275) Strong changes in adipic acid CCN activity are observed upon heating both water and methanol solutions. Do the authors believe that the change in the water-based CCN activity is due only to morphological changes and solvent removal, but that for methanol-based measurements the changes also derive from esterification? In such a case, it would be very interesting to see a measurement of the adipic acid methyl ester CCN activity compared to that of pure adipic acid in the water-based system. Additionally, measurements of CCN activity of adipic acid/adipic acid methyl ester mixtures might be particularly useful in deconvolving the competing influences.

5. (Page 13269) The authors hypothesize that the observation of multiple peaks in the DACAD scans after first sending the particles through a DMA (i.e. size selecting the particles) results from loss of solvent from some particles but not from others, apparently because only some fraction of the particles (at a given size) retained solvent. No reasons were given as to why the authors believe that only some of the particles might retain solvent while others do not, but it seems as if this should be considered in more detail as this conclusion is not intuitive.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 13251, 2006.

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