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.

Anonymous Referee #1

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This paper presents the results of meticulously conducted cloud condensation nuclei (CCN) activity experiments of 7 organic species using both water and methanol as atomization solvents with or without heating treatment at 3 different supersaturations. The paper also provides a description of ADDEM on predicting CCN activities of particles. The authors clearly show the importance of the choice of atomization solvent in CCN activation measurements of some organic species. The solvent and heating can have significant effects on the residual solvent, particle morphology, and chemical reactions between the particle and gas phases, which are important factors in CCN activity measurements. The authors have also raised an issue that a thorough experimental design is important for conducting and interpreting the results of laboratory
CCN measurements. This is a very interesting paper on organic CCN measurements and is recommended for the publication in Atmospheric Chemistry and Physics. My comments below are mainly for clarification purposes. I have also put down some suggestions on further discussions of specific issues and possible future work for the consideration of the authors.

General comments

1) The authors have measured the CCN activity of different organic species using water and methanol as atomization solvents. Can the authors further elaborate why methanol was chosen as an atomization solvent?

2) An important issue pointed out by the authors on interpreting CCN measurements is the morphology of the particles. In this paper, the discussion focuses on the shape factor of the particles, as corrections used in the DMA measurements. Morphological characteristics of solid particles from evaporation of droplets can also include crystalline structure (amorphous vs. crystalline, crystallite size) and “physical” structure (hollow vs. solid particles), which may also affect the CCN measurements. I wonder if the authors have any comments on these factors. Also, it appears that ADDEM has not considered hollow particles. It may be useful to discuss these issues when the model predictions are compared with measurements.

3) The authors used the state of the art tools for CCN measurements and used typical inorganic particles as a calibration standard. While acknowledging that it is a common practice to use NaCl or (NH4)2SO4 as calibration standards, I wonder if the authors wish to comment on the need to have a suitable standard for CCN activity measurements of organic compounds in light of new results reported in this paper.

4) The authors have acknowledged that most of the studied organic species have been investigated by other researchers. The literature results are well summarized in the Figures and Tables. However, there is little comparison of their results with previous studies in the text.
5) The authors wanted to ascertain the effects of heating the particles on their CCN behaviors. A heater was placed after the atomizer for some experiments in an attempt to facilitate the evaporation of the atomization solvents from the particles. The heater was controlled at 40°C. The authors have also pointed out that the temperature at which the particles experienced could cause particle shape difference. Can the authors give more information of the heat treatment such as heating duration and heating rate? What is effect of heating temperature? Would this yield different results? I suppose that a natural extension of the current work would include how these parameters and the heating temperature would affect morphology and the resulting CCN measurements.

6) P. 13271: “The addition of the heater could affect the resulting morphology of the generated particles and, consequently, the size selection in the DMA. Esterification reactions (see Sect. 3.5.4) could also be driven by the presence of heat. Differences observed in the CCN activity for particles atomized in the presence versus absence of the heater could be explained by these phenomena.” Any explanation for why heating significantly changes the particle morphology for some organic species but not all?

7) P. 13272: Size distribution changes were observed after size selection in the classification DMA for malonic acid and glutaric acid generated from both aqueous and methanol solutions, with distributions broadened or multipeaked by the time they were measured in the DACAD. The authors suggested that solvent was trapped in some of the particles and it subsequently “escaped” from those particles between the DMA and DACAD/CCN instruments, effectively causing those particles to “shrink” more than the particles that did not contain solvent at the point of classification. Smaller particles exhibited greater size distribution changes and shrinking. I am puzzled at the state of the particles leaving the first DMA that caused the multipeak distribution downstream. Are these particles leaving the first DMA truly monodisperse in terms of morphology/composition, in addition to their mobility? Malonic acid and glutaric acid are widely studied in the field. What do the literature suggest in terms of their ability to trap solvents during evaporation?
8) P. 13275: “Discrepancies between the ADDEM predictions and the measured activation diameters are greatest for adipic acid, although the results for adipic acid particles generated from a methanol solution without the heater fall nicely within the ADDEM predictions.” Can the esterification reactions in adipic acid particles explain the difference? Esterification reactions favors for acid particles generated from methanol solution with heater. The authors may want to caution readers that such agreement between ADDEM and the results of methanol/heating particles can be coincidental. The products of the reactions may have properties different from adipic acid, that are not considered in ADDEM. Furthermore, why are the esterification products of glutaric and mlaonic acids as hygroscopic as parent compounds but those of adipic acid are less hygroscopic than parent compound?

9) P. 13303: In Figure 10, using water as solvent, it is observed that activation diameter of adipic acid particles with heating are smaller when compared to that of adipic acid particles without heating at different degree of supersaturations. Using methanol as solvent, the activation diameter of adipic acid particles with heating are larger when compared to that of adipic acid particles without heating. Any explanation for this observation?

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