

Interactive comment on “Ice nucleation in sulfuric acid/organic aerosols: implications for cirrus cloud formation” by M. R. Beaver et al.

Anonymous Referee #1

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General Comments:

This manuscript investigates ice nucleation in sulfuric acid/organic aerosols, with possible implications for cirrus cloud formation. Ice nucleation in sulfuric acid and ammonium sulfate particles have been studied extensively, but the effect of organics on the ice nucleation properties of sulfate particles has received little attention. Hence, the subject of this paper is timely and may be important for predicting the formation of cirrus clouds.

The authors show that sulfuric acid particles exposed to organic compounds of eight carbons and greater nucleate ice at temperatures above aqueous sulfuric acid aerosols, and organic compounds of intermediate carbon chain length, C4–C7, nucle-

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ate ice at the same temperature as aqueous sulfuric acid aerosols.

This manuscript presents new data in an important area of research. Hence, I suggest publication after the authors have addressed the following comments sufficiently:

1. Page 2062, line 9. “there is much less data available for ice nucleation from other inorganic and organic species”. Many studies on ammonium sulfate have been carried out previously, so I would change this statement to reflect this fact.
2. Page 2064, line 20. “Although some infrared absorption cross-sections were available in the literature, infrared absorption cross sections were separately determined for all compounds in the present study and used to calculate partial pressures.” Later the authors state, “Cross-sections could not be determined experimentally for 2-nonanone or decanal due to vapor pressure limitations.” Are these two statements consistent with each other?
3. Page 2064, line 15. “This conditioning tube served to maintain the sulfate to water ratio of the aerosols and to pre-cool the aerosols.” How does this work? When the temperature decreases the sulfuric acid particles should take up water, and thus the sulfate to water ratio should change? Why does the concentration of the sulfuric acid solutions stay at 15 % when the temperature decreases? Please include more details here. Also other experimental details such as the aerosol concentration (number of particles/cm³) and the size of the aerosol particles would be beneficial.
4. Related to the above, on page 2066, line 9, the authors state: “all experiments were conducted using 15 wt% sulfuric acid composition as the precursor aqueous sulfuric acid aerosols”. The concentration of the sulfuric acid aerosol was also 15 wt % in the observation region? Correct?
5. Did the authors determined the sulfate to water ratio (from the spectrum) in the presence of organics or only before the organics were added.
6. Page 2071, Equations 1 and 2. In the literature these equations are used to predict

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how the freezing point and melting point of solutions vary from pure water. Are the authors also trying to do the same thing here? What “m” is used in Equation 2? Please expand or delete this section. I am not sure of the point of this section.

7. Page 2072, line 5. “Because the partial pressure of the organics is measured and the aerosol molarity is determined using the Mie scattering analysis, it is possible to carry out a Henry’s law analysis to determine the solubilities.” For the Henry’s law measurements, did the authors measure the partial pressure of the gas in equilibrium with the particles at low temperatures? This is the measurement needed for a Henry’s law analysis, so I assume this is what was measured, but it was unclear from the manuscript.

8. Page 2072, line 24. I don’t think the results demonstrate a depression of the water activity.

9. Page 2073, line 23. For the longer chained organics, which have a low solubility, could the organics homogeneously nucleate to form pure organic particles in the flow system?

10. Page 2067, line 5. “The difference in aqueous sulfuric acid aerosol mean diameters, before and after organic addition was converted to an aerosol volume change, which also allowed the calculating of organic aerosol molarity ($M = \text{moles solute/L of solution}$.)” Please give more details on how volume change was converted into M (e.g. what assumptions were used?).

11. Page 2068, line 14. “To determine if the organic uptake was due to acid-catalyzed chemistry, control experiments were performed with aqueous ammonium sulfate (approximately 30 wt %). Why did the authors use 30 wt % rather than some other composition?”

12. Figure 8. It would be good to have error bars in Figure 8, to better appreciate the significance of the results.

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13. The final question is related to the implications for this research to cirrus clouds. The authors show that at some concentration a solubility limit will be reached and a second organic phase will begin to form on the aqueous aerosol, and this second phase can lead to heterogeneous freezing. However, in the atmosphere will this solubility limit be reached? Marcolli et al.¹ suggested that in most cases a second organic phase will not form because the solubility limit of one specific organic molecule will not be reached. The authors should address this point.

(1) Marcolli, C. et al. J. Phys. Chem. A 2004, 108, 2216.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 2059, 2006.

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