

Interactive comment on “Heterogeneous ice nucleation on phase-separated organic-sulfate particles: effect of liquid vs. glassy coatings” by G. P. Schill and M. A. Tolbert

Anonymous Referee #2

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The authors report about laboratory experiments on ice nucleation of single droplets, exhibiting liquid-liquid phase separation. Experiments on droplets using a Raman microscope have been performed on similar systems before, but the authors extend those measurements to lower temperatures and use a clever experimental approach to distinguish different mechanisms of heterogeneous ice nucleation in phase separated organic-inorganic mixtures. Besides showing that depending on the phase state of the organic phase, nucleation occurs either at the ammonium sulfate core or on the organic surface of these particles, they also show that phase separation has only a minor effect, if any, for deliquescence and efflorescence of ammonium sulfate over the whole temperature range encountered in the atmosphere.

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Interactive Discussion

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This is an excellent paper, well written and with conclusions appropriately supported by experimental data. The paper should be published as is, but the authors may consider the following comments/questions before submitting the final version.

page 30954, lines 17-22: The statement that mixed organic-sulfate particles always exhibit a fully engulfed core-shell morphology is not true in general. As pointed out by e.g. Kwamena et al. (2010) this depends on the relative magnitudes of the different surface/interfacial tensions. For example Song et al. (2012) showed that their C6/AS/H₂O mixture (C6 being a mixture of three C6 dicarboxylic acids) exhibits partially engulfing. Please reformulate your statement. Also, I wonder if the conclusion of your experiments does not imply that even a partially engulfed particle at low temperatures would nucleate at similar supersaturations, since it seems not to matter so much whether nucleation occurs at the surface of a semisolid organic or the AS core?

page 30961, line 2: compare with the estimate in Ciobanu et al. (2010). This seems very fast to me, also the diffusion constant will change drastically with temperature (long before becoming glassy).

page 30968, line 9: the data point at 210 K lays significantly above the homogeneous freezing line, as you also point out. What do you think is the reason for the deviation: Could it be that your calculated Scrit is too high below 215 K? Or do you believe that this is an indication for the Koop line being wrong below 215 K? Since also the 210 K points in Fig. 9 are higher than immediately obvious (I cannot see a reason for the apparent increase in Scrit from 215 K to 210 K in Fig. 9); I tend to believe in a problem of the setup at these very low temperatures. Please comment.

page 30965, line 14-22: this is very speculative and cannot inferred directly from the experiments. I feel the speculative character of this paragraph should be pointed out more clearly.

page 30967, line 1-4: your experiments indicated that the critical diffusion time for the particles of the size in your setup is reached between 220 K and 215 K. You could

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apply eq. (1) to give a rough estimate of the relevant viscosities.

References:

Kwamena et al., J. Phys. Chem. A, 114, 5787–5795 (2010).

Song et al., Atmos. Chem. Phys., 12, 2691-2712 (2012).

Ciobanu et al., J. Phys. Chem. A, 114, 9486-9495 (2010).

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 30951, 2012.

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