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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Laboratory studies of ice formation pathways from ammonium sulfate particles" *by* M. E. Wise et al.

Anonymous Referee #2

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The paper by Wise et al. presents new measurements on the freezing properties of ammonium sulphate (AS) aerosols. In particular, the RH/temperature conditions for homogeneous freezing are measured, and there is an attempt at measuring the conditions for deposition nucleation onto solid AS particles. Numerous attempts to define homogeneous freezing conditions have been made in the past, and there has been a considerable variability in the reported results.

This paper makes two contributions. First, it shows that if the homogeneous freezing experiment is done carefully in a flow tube, the results match closely those predicted by the widely recognized model of Koop et al. In particular, it appears to be important that all the particles have to be fully deliquesced for the appropriate homogeneous freezing conditions to be observed. This is an important new contribution and merits publica-

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Discussion Paper

tion in its own right because it illustrates that having a minor fraction of particles not deliquesced can lower the apparent supersaturation required for the onset of freezing as observed in a flow tube/IR apparatus. The techniques used are well described, and build upon past studies by this research group and others.

The second part of the paper addresses the conditions for ice formation onto solid AS particles. Results are presented that indicate that the onset is close to the relative humidity of ice. There is no doubt that ice is forming in these experiments and that the AS particles are solid but more justification needs to be given that the quantitative values of the onset saturation ratios are correct. In particular, the values rely upon a hygrometer measurement of the water partial pressure exiting the flow tube. However, if there are supersaturated conditions in a flow tube, the maximum supersaturation at the entrance to the flow tube may be higher than that calculated from the exiting water flow, if there is considerable loss of water vapour to the walls of the flow tube. And so, it would be valuable to know whether the hygrometer measurements track the increasing input water vapor amounts arising from the steadily increasing conditioning flow tube temperatures. In particular, as the conditioner temperatures rise, is there a steady increase in the measured amounts of water exiting the flow tube? If these two correspond, particularly under conditions that are not supersaturated, and then if it is shown that moving into just slightly supersaturated conditions brings about deposition freezing, then I would tend to trust the results. However, if the hygrometer partial pressure is always measured to be close to that of the vapour pressure of ice in the observation flow tube independent of the temperature of the conditioning flow tube, then there may be problems.

Alternatively, can the authors provide an upper limit to the supersaturation conditions in the observation flow tube, from the temperatures of the conditioning and observation tubes? Identifying that the conditions for nucleation onto solid AS particles lie below those of the Koop et al. model would be an important result in its own right, even if the precise values for onset are harder to pin down.

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Interactive Comment

Overall, the homogeneous freezing result is solid and merits publication. The deposition freezing result may be correct, but I would like to see a bit more validation that the quantitative values for ice onset are correct. At a minimum, it would be valuable to state an upper limit to the saturation ratios at which deposition nucleation occurs.

Small points:

1. Section 2.1, 2nd paragraph. Are the baths at room temperature?\

2. Section 2.1, 5th paragraph. Can you justify that the particle composition remains the same during the homogeneous freezing expts?

3. Section 3.1, 5th paragraph. Is there any possibility that sulfuric acid vapor from the 98% solutions condenses on the particles?

4. Throughout: The authors need to better point out that a small fraction of particles freezing may be apparent as a large ice signal in the FTIR spectra.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 15101, 2008.

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