

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

Anonymous Referee #1

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General Comments

I feel it necessary to provide a quick initial review of this paper due to my perception that some of the experiments described may have serious technical errors. I was immediately drawn to the section of the paper describing how it was possible to conduct a deposition ice nucleation experiment in a flow tube and concluded that the authors do not appear to have seriously considered thermodynamic issues that must apply. The paper takes a method that, with care, is useful for qualitative indication of ice formation by liquid particles under specific compositional and temperature conditions and violates its primary useful principle to try to apply it toward measuring deposition ice nucleation. This is what half of the paper is about. The other half is about experiments into how to assuredly study homogeneous freezing of liquid ammonium sulfate parti-

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cles with a flow tube. This is an interesting perturbation on previous work by Hung et al. (2002). The authors use the results from procedures that vary the apparent liquid/solid proportion of particles as support for the role of heterogeneous ice nucleation when some effloresced particles enter the cold observation flow tube (in contrast to the conclusions of Hung et al. suggesting that a mixed aerosol phase state leads to a mis-attribution of freezing composition). Nevertheless, the basic first problem I see in the present paper is that I do not believe that the authors have truly resolved the RH to which fully dry particles are exposed in the transition of air between the conditioning and observation tubes in their experiments to confirm the role of deposition nucleation on ammonium sulfate. This is a highly non-equilibrium situation. The RH attributed to ice nucleation cannot possibly have been measured in my present opinion. What should be measured at the base of the observation tube, within the uncertainty of measurements, is ice saturation. Figure 7 seems to prove that. Furthermore, why would such a large temperature difference between the conditioning and observation tubes be required to achieve such a low RH for ice activation if the particles cannot take up water?

I conclude that if the authors are to support their conclusions and publish this work, they need to go through some numerical modeling studies or detailed calculations that prove that the methods used to interpret RH for ice nucleation by “deposition” have any basis in reality. Simple considerations suggest to me that the air enters the observation flow tube and achieves a transient high supersaturation (probably non-homogeneous) that could even lead to water condensation and homogeneous freezing in an unknown proportion of the particles. In the experiments to measure RH without particles present, most of the vapor should still be lost to the walls by diffusion, again over a transient period. The authors seem to acknowledge this fact, then ignore it and state that the hygrometer is measuring the activation RH. Why would a steady-state supersaturation be sustained when humid air enters a constant-temperature cold tube, unless there is a temperature gradient and steady vapor source on the walls? Consequently, even if heterogeneous nucleation is being observed, I have significant doubts that the RH for

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freezing is being accurately attributed.

If the authors can show that I am incorrect in these conclusions, then I will provide a more complete review of the paper.

References

Hung, H., Malinowski, A., and Martin, S. T.: Ice nucleation kinetics of aerosols containing aqueous and solid ammonium sulfate particles, *J. Phys. Chem. A*, **106**, 293-306, 2002.

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