

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

**M. E. Wise et al.**

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Primary reviewer 1 comment: 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.

Author response: We believe that the flow tubes are useful for the quantitative indication of ice formation from liquid particles. Many researchers in the past have used the flow tubes for the quantitative measurement of homogeneous ice nucleation and the measurements in most cases agree with other methods. In the present work, we first evaluate the method using liquid ammonium sulfate particles. We find agreement with other flow tube studies, and extend the measurements to explore why the flow tube studies show higher freezing temperatures than expected. We then extend the use to

a new area and also include for the first time studies of depositional nucleation using the flow tube. Since this has never been done before, it does not seem valid to simply dismiss the technique as invalid. We do not agree that it violates any principle.

We do, however, agree with the reviewer that the flow tube apparatus has some experimental limitations. Other techniques used to study ice formation in atmospheric particles also have drawbacks. For instance, techniques that employ emulsions to study ice formation encase particles in an organic liquid. Koop et al. used data from these experiments to develop their homogeneous ice nucleation theory. One could argue that because atmospheric particles are not incased in an organic liquid in nature that these studies are not useful. However, the atmospheric community accepts the Koop et al. model.

Primary reviewer 1 comment: 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.

Author response: The major problem reviewer 1 has with the manuscript is the method in which we determined the ice saturation ratio ( $S_{ice}$ ) in the flow tubes during depositional ice nucleation experiments. Without using numerical modeling studies (which the reviewer suggests that we do), we have the burden of proving that our experimental measurements determine  $S_{ice}$  correctly. In order to determine  $S_{ice}$  correctly, the vapor pressure in the observation region must be known with certainty. We have shown that the hygrometer correctly measures the vapor pressure created by the ice in the conditioning region (line 273). Therefore, we are confident that the hygrometer correctly measures the vapor pressure exiting the observation region.

The question then remains whether or not we are measuring the vapor pressure in the correct portion of the flow tube system. The reviewer inspired us to answer the question of whether measuring the vapor pressure from the observation tube exit was

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valid. Therefore, a new experiment was performed in which  $S_{ice}$  was measured half way down the observation tube. Given our experimental setup, this position was as far away as possible from the exit of the flow tube apparatus. In this experiment, the conditions of one of the heterogeneous ice nucleation experiments conducted in the original manuscript were duplicated.  $S_{ice}$  was then measured at the flow tube exit and at the middle of the observation region. It was found that the  $S_{ice}$  calculated at the flow tube exit was less than 5 % different than that reported in the original manuscript.  $S_{ice}$  measured at the middle of the observation region was less than 5 % higher than that measured at the flow tube exit. Therefore, it was not high enough to change the conclusions of the original manuscript.

Reviewer 1 comment: What should be measured at the base of the observation tube, within the uncertainty of measurements, is ice saturation.

Author response: We experimentally determined the value of  $S_{ice}$  with the hygrometer. The procedure is explained starting on line 250.

Reviewer 1 comment: 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?

Author response: There is a large temperature difference because water vapor is condensing on the tube walls (not onto the particle surface). This is explained on line 405,  $S_{ice}$  could not simply be calculated using the temperature of the two cells because water vapor was lost to the walls of the observation region. This is because the observation tube walls were much colder than the conditioning region. Thus water vapor pressure must be measured directly using the hygrometer.

Reviewer 1 comment: 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.

Author response: We agree that when the air enters the observation flow tube a transient higher supersaturation value is reached. However, we do not believe that water has condensed on the particles. The IR spectra do not show that the particles have deliquesced. Thus homogeneous freezing could not have occurred in an unknown portion of the particles. Furthermore, we do not believe that ice particles homogeneously condense from the water vapor in observation region. Without particles present we have never seen ice appear in the IR spectra.

Reviewer 1 comment: 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.

Author response: We acknowledge that water vapor is lost to the walls of the observations region (line 405).

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

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