

Interactive comment on “Size dependence of volume and surface nucleation rates for homogeneous freezing of supercooled water droplets” by T. Kuhn et al.

Anonymous Referee #1

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Earle et al. and Kuhn et al. report on a series of flow tube experiments to investigate homogeneous freezing of liquid water. The first paper introduces the methodology and explores the approach based on just a volume nucleation mechanism dominating the freezing process. The second paper expands the discussion to include the effects of a surface (or near surface) homogeneous freezing mechanism. The results indicate that for crystals smaller than 5 microns a surface freezing mechanism dominates. This result is interesting and is likely to motivate other groups to repeat and extend these observations.

I feel that the papers are publishable in ACP. I have a few comments below that should

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be responded to. The papers cover very similar ground, so most of the comments apply to both.

Earle et al.

The evolution of the particles in the experiment is complex. Some haze particles are exposed to colder conditions before others, and the conversion of these particles to ice results in mixed-phase evolution that the model approach tries to capture. It would seem simpler in terms of the modelling and interpretation of results to limit the aerosol to a thin region of the aerosol flow tube. This would narrow down the conditions that the particles are exposed to.

22890 section 2.2. The IR retrieval method has been tested in Zsazsazsky et al. 2007 against particle by particle measurements of the size distributions of different phases. I was unable to get hold of the full paper in time, but the abstract indicates that uncertainties were derived. Please could you state those uncertainties so that the reader can assess what affect those would have on the subsequent analysis. It should be possible to propagate those uncertainties to show the impact on the final results shown in fig. 12.

22895 18. The function X is undefined. Provide link to later section, or describe in one sentence.

22898 Eq 10. The second term on the RHS is potentially more empirical than described in the text. What are the relative values of the 1st and 2nd term on the RHS? If the walls of the tube are exposed to water saturated conditions for an extended interval frost can grow and lead to a roughening of the surface. I feel that this would lead to a change in the Sherwood number through a change in the boundary layer depth near the walls. It would be good to assess the sensitivity of the results to variability in the 2nd term on the RHS of eq 10.

22903 3. I am a bit worried by this removal of part of the initial size distribution. Was an

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inertial impactor used when the aerosol was introduced to the chamber? This would solve the problem of large insoluble aerosols entering the flow tube. Could this secondary maximum be due to an error in the retrieval? This could potentially be more of a problem to the analysis and results.

22908 26. (fig 12 results). If there are systematic errors in the size distribution retrieval (see 22903 3) then this could effect the results plotted in fig 12. It would be good to generate 'sensitivity bars' to add to each line so that it is possible to assess the significance of the different results.

Figures - units and values missing from the size distribution figures.

Kuhn et al. (to points above also apply to this paper).

Although the authors could argue that it would be for future work to do this, I would be very interested in seeing a simple parcel model ascent to show the effect of the surface nucleation in the formation of cirrus type clouds. Would the action of J_s affect the numbers of ice crystals produced when compared to J_v ? Such an example would be a valuable addition to this paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 22929, 2009.