Response to Anonymous Referee #1

We thank Referee #1 for their constructive comments, which are identified by italicized text in the following. We respond first to comments pertinent to this manuscript and then to those for the companion manuscript by Kuhn et al..

Comments for 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.

The reason for the formation of mixed-phase aerosol is the probabilistic nature of the freezing process rather than non-uniform temperature in the flow tube. We purposefully selected a range of experimental temperatures within which only a fraction of droplets nucleate ice and freeze. It is by observing the evolution in the composition of this mixed-phase aerosol that we were able to retrieve the freezing rates and mass accommodation coefficients.

With regard to the experimental configuration, the presence of cooled cross-shaped copper fins in each section of the flow tube (Sect. 2.1) ensures very high thermal transfer rates, which eliminate radial temperature gradients. In addition, any small residual radial gradients of

temperature and/or mass distribution are accounted for through the use of mass-weighted average axial temperature profiles determined using CFD (Sect. 2.4). These profiles are used when computing the size- and phase-evolution of the aerosol in the microphysics model. These aspects of the experiment have been discussed previously in [A. F. Khalizov, M. E. Earle, W. Johnson, G. D. Stubley and J. J. Sloan, " Development and Characterization of a Laminar Aerosol Flow Tube," Review of Scientific Instruments, 77(3), 2006].

22890 section 2.2. The IR retrieval method has been tested in Zasetsky 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.

Zasetsky et al. 2007 reported the measurement of commercial SiO_2 microspheres. The result (radius range of 1.1–1.25 μm) agreed well with the manufacturer's specified radius range (1.11–1.35 μm). On the basis of this result, we reported that our method of size distribution retrieval from infrared extinction measurements is accurate to within +/- 5%. We have added a statement of these uncertainties in Sect. 2.2. For further discussion of errors in model-derived nucleation rates, please see our response to 22908 26.

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

We have added a reference to Eq. 16.

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.

The second term on the RHS of equation 10 is subject to the stated assumption of steady state conditions for the water vapour evaporation and condensation. The validity of this assumption can be argued, but nevertheless it defines the N_V values, which are thus not empirical. At steady state, the first term is small, but individual components of the sum (i.e., dm(ice)/dt and dm(water)/dt) are large - significantly larger than the wall loss term. Ultimately, the total mass of water vapour lost to the wall in sections C and D is a factor of 10 to 20 smaller than the total mass of water present in aerosol particles for experiments using medium and large particles. For experiments using small particles, the mass of vapour lost to the wall is more significant – a factor of two to five smaller than the total water mass of aerosol particles (the water mass of particles is lower on account of the generation method).

We do not expect any variability in the wall-loss term (2nd term on the RHS of Eq. 10) due to frost. Visual inspection of the bottom sections C and D showed no signs of frost. The absolute amount of ice formed on the walls of the bottom sections is negligible because of the low water

saturation vapour pressure at the temperature of freezing onset. Furthermore, all convective mass transfer is suppressed because the only temperature gradient is caused by thermal transfer from the incoming gas and this occurs at the top of each flow tube section. (See [A. F. Khalizov, M. E. Earle, W. Johnson, G. D. Stubley and J. J. Sloan, "Development and Characterization of a Laminar Aerosol Flow Tube" Review of Scientific Instruments, 77(3), 2006] and [A. F. Khalizov, M. E. Earle, W. Johnson, G. D. Stubley and J. J. Sloan, "Modelling of Flow Dynamics in Laminar Aerosol Flow Tubes" Journal of Aerosol Science, vol. 37(2006), 1174-1187]).

To address the reviewer's comment regarding the sensitivity of the results to variability in the wall-loss term, we conducted a model sensitivity analysis using the small particle group, for which the wall-loss term is most significant. For variations in the Sherwood number of up to 25%, the nucleation rate coefficients remained within the limits of uncertainty (see response to 22908 26 below). We reiterate, however, that variations in the Sherwood number are not expected based on information derived from our previous characterization of the flow tube apparatus, including the absence of convection, the restriction of the observations to the central core of the flow and self-consistency of the freezing experiments, as well as the visual observations mentioned above.

22903 3. I am a bit worried by this removal of part of the initial size distribution. Was an 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 problem to the analysis and results.

The composition and size of the laboratory-generated aerosol were carefully controlled in freezing experiments. While no inertial impactor was used at the flow tube inlet, our method of generating the aerosol does not produce large, insoluble particles (see Sect. 2.1 for a discussion of the possible origins of the secondary mode). Even if these secondary modes are not removed in the model input files, the results are essentially the same because the large particle feature evolves independently of, and remains small compared to, the primary mode. For instance, when the secondary mode was not removed from the initial size distribution in experiments performed at 235.5 K, the volume nucleation coefficient changed by only 0.2%; α_w by about 2% and α_{ice} by 0.6%.

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.

As is evident from our response to 22903 3 (see above), the volume nucleation rate coefficient is not sensitive to these features in the size distribution (or their removal). The reviewer raises an excellent point, however, regarding the need for 'sensitivity bars' to assess the significance of the results. Initial estimates of the uncertainty for J_V values were derived by comparing model results from replicate freezing experiments conducted using the same temperature and droplet size. The J_V values varied within a factor of three to five, which we attributed to small-scale variations in the axial temperature profiles for the different experiments. The group fits, however, are far less sensitive to these variations than single fits. For example, compare the temperature dependence

of the single fits to that of the group fit in Fig. 10. To estimate how a single freezing experiment (and its inherent uncertainty) can influence the nucleation rate coefficients determined from group fits, an additional series of model runs was carried out for each group of small, medium, and large droplets. One freezing experiment was removed from the group fit in each of these model runs and the $J_V(T)$ curves were compared. The resulting temperature-dependent errors for each size group are represented by the sensitivity bars in the attached figure.

The sensitivity bars for small and medium groups indicate that the $J_V(T)$ values overlap slightly at warmer temperatures, but the size-dependent trends upon which our conclusions regarding the importance of surface nucleation are based are preserved. In addition, there is almost no overlap of the sensitivity bars for the large and medium groups, indicating that the observed difference in nucleation rate coefficients is significant. The above conclusions regarding the significance of the size-dependent results justify the further investigation of the role of surface nucleation in the companion paper.

We propose that the attached figure illustrating the sensitivity bars and related discussion be included in the manuscript, with this figure becoming Fig. 12 and the existing Fig. 12 (comparison of $J_V(T)$ curves with previous studies) changing to Fig. 13.

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

The Y-axis in the volume size distributions is the ratio of the volume of the condensed phase in μm^3 to each cm^3 of gas. We have added these units to the axis labels for clarity. We do not use

absolute volume concentration values in the figures, because only the relative values are important for the analyses and the addition of these values would clutter the size distribution figures. Furthermore, different experiments are considered independently in the model analyses. To provide readers with some perspective, we have noted the range of typical values in the text.

Comments for Kuhn et al. (the 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 Js affect the numbers of ice crystals produced when compared to Jv? Such an example would be a valuable addition to this paper.

We agree that it would be interesting to apply these results to the simulation of cirrus formation in the atmosphere. The amount of work involved in developing a model for this purpose, however, is considerably beyond the scope of this work, which is a laboratory study intended to provide information for other researchers who are equipped to carry out such modelling studies.