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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Volume nucleation rates for homogeneous freezing in supercooled water microdroplets: results from a combined experimental and modelling approach" *by* M. E. Earle et al.

Anonymous Referee #2

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This is the first of a set of two consecutive papers addressing the homogeneous freezing of water droplets. It is a valuable contribution to a problem of great relevance, as there is a long standing and controversial discussion on the importance of surface nucleation of atmospheric aerosols. The authors report rates for the homogeneous nucleation of water in the 1-3 micron size range and derive mass accomodation coefficients for water vapor on ice. The experimental procedures are described in detail and the results are presented clearly. I have major concerns regarding the interpretation of the data, however:

The authors derive J(T) and the mass accommodation coefficients <alpha> by fitting a process model to the experimental results. This is by no means a direct measurement of these quantities so great caution has to be applied to assess the limits of error. I feel that this has not been done sufficiently in the manuscript.

There are several possible caveats regarding the experimental and modeling methods, which should be addressed, including: It is unclear to me, how droplet position was converted to droplet residence time. It seems that the radial velocity (and temperature) distribution of the laminar flow was not taken into account. As freezing is strongly nonlinear with temperature, even small temperature variations may dominate the ice signal. In Fig. 7, the temperature has a minimum around t=10s which is almost 1K below the temperature at 23s. Where does this minimum come from? How does the temperature profile depend on the radial position? The ice detection is not very accurate, so the raw data had to be manipulated before the fitting procedure. There seems to be substantial evaporative cooling of the liquid droplets as soon as the first ice is formed. This should be included in the process model, as even small temperature fluctuations can have a large effect.

There are several features in the experimental results that indicate problems with the data evaluation: In Fig. 4, water and ice seem to coexist over a temperature range of almost 5K. This is much too large for any reasonable parameterization of homogeneous freezing. In Fig. 5, small and medium sized droplets freeze more readily than large droplets. This is opposite to what is expected from homogeneous nucleation. Large droplets should freeze first, small droplets last upon cooling. Apart from the shift, the shape of the respective curves should be the same however. What is the origin of the minimum in Fig. 7, upper panel? As can be seen in Fig. 10, the fits converge to completely unphysical slopes of ln J(T) (parameter a_v in Eq. 17), if applied to a single temperature. The individual points are off by up to almost one order of magnitude from the group fit and no difference was found in J(T) for the two warmest temperatures.

Despite of these problematic findings, no error bars were given in Fig. 12 for the results

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obtained. Rather strong conclusions with respect to a possible contribution of surface nucleation were drawn especially in the follow up manuscript.

My suggestion is to revise this paper adressing the remarks above and to present reliable limits of uncertainty to the obtained results. Only if these limits allow inferring a significant contribution of surface nucleation, the second manuscript should be submitted.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 22883, 2009.

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Interactive Comment