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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Laboratory evidence for volume-dominated nucleation of ice in supercooled water microdroplets" *by* D. Duft and T. Leisner

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We thank the referee for the careful reading of the manuscript and his valuable comments which will certainly help to improve our manuscript. We respond as follows:

General comments:

We thank the referee for pointing us to the instructive comment on the Tabazadeh papers by Kay et al. We will include the reference in a final manuscript.

We agree that the droplet size used in our experiments is rather large compared to the droplets of interest in the atmosphere. We therefore were only able to give an upper limit (4 μ m) for the droplet radius below which surface nucleation might be important. Nevertheless we believe that our experiments are relevant, as the analysis

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of Tabazadeh et al. (refs. 14- 17) is based largely on experimental work which was performed just in the same size range. In future work we might be able to investigate somewhat smaller droplet sizes, with our experimental approach we are limited to droplets larger than about 5 μ m. We have demonstrated in the text that our method allows considerable extrapolation to smaller size droplets.

Specific Comments:

We have not stated in the introduction that homogeneous freezing is important in thunderstorm formation but wanted to use the latter as an example for the importance of phase transitions in atmospheric dynamics.

1We assumed that it is evident from the text that the formation of tree-dimensional nuclei in the surface region is considered in our analysis but we will clarify that point explicitly in a final manuscript.

Experimental

It is difficult to include error bars in Fig. 2 as the values on the ordinate are calculated as the logarithm of the ratio of unfrozen droplets to the total droplet number. These integer numbers are obtained from droplet counting, so no error bar can be attributed here. The x- error bars regarding the time measurement would be much smaller than the symbol size. (The accuracy of the measurement of the freezing time is given in the text). The scatter of the data points around the theoretical linear decline has to be attributed to the statistical nature of homogeneous freezing. The magnitude of this scatter should decrease as the number of droplets under investigation is increased. The error limits for the determination of the nucleation rate, which is largely due to the limited number of droplets under investigation, are given in Table 1.

Great care was taken to keep the temperature uniform throughout the experiment. In the type of experiment presented here the determination of the absolute temperature less important than the relative temperature changes especially when switching from

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large to small droplets and vice versa. The temperature fluctuations in our experiment were about 20 mK (single sigma). After the initial thermalization process the droplets do hardly evaporate at all at the low temperatures of our experiment . From the light scattering probe we estimate that their radius changes less than about 50 nm/s. Given the short time until freezing occurs, evaporation of the droplets is no concern in our experiments. We will specify these numbers in the final manuscript.

In an independent set of measurements we have been able to record the light scattering transient during freezing of water droplets with high temporal resolution. From these type of measurements we infer that freezing of droplets is a two stage process, the initial stage being a very rapid process which takes only about 100 μ s for a r=40 μ m droplet. When the nucleus becomes detectable its size is already in the order of a few μ m and then its origin (surface or bulk) cannot be determined. (The notion that the laser light probes only a small region under the surface is only true if the laser wavelength hits a so called morphology dependent resonance, then the penetration depth is a few wavelength of light). We believe that a discussion of these issues goes beyond the scope of our article.

Our results are in excellent agreement with earlier measurements by us (Krämer et al. 1999), (Stöckel et al., footnote 1) and the STO data by Taborek (Taborek 1985).

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 3077, 2004.

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