Atmospheric Chemistry and Physics Discussions

Interactive
Comment

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

General comments, mentioned by both referees:

We thank both referees for pointing us to the instructive comment on the Tabazadeh papers by Kay et al. We have include that reference in the revised manuscript.

We agree with both referees that the droplet size used in our experiments is rather large compared to the droplets of interest with regard to homogeneous freezing in the atmosphere. Unfortunately we therefore are only able to give an upper limit (4 μm) for the droplet radius below which surface nucleation might be important. Nevertheless we

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believe that our experiments are relevant, as the analysis of Tabazadeh et al. (refs. 14-17) is based largely on experimental work which was performed just in the same size range as our work. In future work we might be able to investigate somewhat smaller droplet sizes, but with our experimental approach we are limited to droplets significantly larger than about 5 μm which will at most limit r_c to somewhere around 500 nm.

Specific Comments of Referee 2:

We did not meant to say 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. Nevertheless we have removed the reference to thunderstorms in the introduction.

We indeed have considered the formation of three-dimensional nuclei in the surface region in our analysis, we have clarified that point explicitly in the revised manuscript.

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 the counting of droplets, so no error bar can be attributed here. The x-axis 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). That the symbols seem to scatter 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 is given in Table 1.

Great care was taken to keep the temperature uniform throughout the experiment. In our type of experiment the determination of the absolute temperature is not so important as the relative temperature changes when switching from large to small droplets and vice versa. The relative temperature fluctuations in our experiment were about 20 mK (single sigma). At the low temperatures of our experiment the droplets did hardly

evaporate at all after the initial thermalization process. From the light scattering probe we estimate that thereafter 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 and that the initial stage is a very rapid process which takes only about 100 μs for a r=40 μm droplet. When the germ becomes detectable its size is already in the order of a few μm 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 (Kramer et al. 1999), (Stockel et al. Footnote 1) and the STO data by Taborek (Taborek 1985). We have specified these references in the final manuscript.

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