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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

Anonymous Referee #2

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1. Introduction

This is an experimental attempt to determine whether the onset of homogeneous freezing of an aqueous drop is most probable in the bulk of the drop or at the liquid-vapor interface. The suggestion that the latter may be important, particularly in the atmosphere, was made in the papers 14-17, cited here. The arguments presented in those papers were largely refuted in the comment by Kay et al, which should be cited.

2. General Comment:

Unfortunately, the drop sizes used in the experiment are very large (radii of 19 and 49 microns). It was, therefore, highly unlikely an effect of surface freezing would be measurable; for such large droplets tremendous differences in the nucleation barrier would be required to make surface nucleation competitive with a volume based process. Moreover, at these drop sizes this experiment is not directly relevant to the atmosphere, where the droplets that freeze homogeneously are not larger than a few microns at most.

3. Specific Comments:

Lightning and precipitation are linked to ice formed at such high temperatures $(-20°C \leq T)$ that homogeneous freezing cannot be important.

The authors should define surface freezing; i.e., it could mean either nucleation of two-dimensional nuclei at the air/liquid interface or nucleation of three-dimensional embryos near the droplet surface. In this case they are referring to the latter.

4. Experimental:

The experimental error bars should be included in Figure 2 and the sources of uncertainty (experimental and statistical) in the parameter r_c explained.

Although there was no room for an extensive description of the apparatus, the authors should include estimates of uncertainties in temperature and humidity at the position

ACPD

4, S856–S858, 2004

of the levitated drops. How certain are they that the droplet sizes remained constant and the droplet temperatures remained uniform during the experiment?

It would be interesting to know what other experimental information might be brought to bear on the question of surface vs volume freezing. For example: What depth inside the drop does the light probe penetrate? Can it distinguish a partially frozen drop (frozen either at the surface or at its core) from a completely frozen one?

Are the results of the experiments reported here consistent with earlier laboratory results by the same authors? (Ref 12 in this paper)

The authors state their freezing rates at $T = 235.1$ K are in good agreement with published results; they should add a reference.

5. Reference

Kay, J. E., Tsemekhman, V., Larsen, B., Baker, M. and B. Swanson, Comment on evidence for surface-initiated homogeneous nucleation. Atmospheric Chemistry and Physics, 3,1439-1443, 2003.

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