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Does the homogeneous ice nucleation initiate at the surface or in the volume of super-cooled water droplets?

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The nucleation of ice in super-cooled water droplets affects many atmospheric processes as the initiation of precipitation and radiative transfer. Water droplets are freezing due to the formation of a critical germ initiating the freezing of the whole droplet. The common quantity to describe the creation of ice is the nucleation rate J, defined as the product of the number of critical germs and the rate at which additional molecules are incorporated into a critical germ. Nucleation of ice in a super-cooled liquid is a stochastic process and depends strongly on temperature.

Recently there was a discussion whether the germs of the new phase are formed preferentially near the surface or in the interior of the droplet. Experiments at the aerosol and cloud chamber AIDA of Forschungszentrum Karlsruhe were performed to assess this question. We produced clouds of super-cooled water droplets and deduced the ice nucleation rate J from simultaneously measurements of the number density and size distribution of liquid droplets, the number density of ice particles, and the temperature in the range between –36 and –37 °C. With different number densities of seed aerosol particles (sulphuric acid aerosol) we were able to vary the size of the nucleating water droplets between 4 μm and 9 μm diameter. The comparison of the results – by assumption of a volume dependent process - showed very good agreement both with data from literature gained from considerably larger droplets and with classical nucleation theory. The nucleation rates disagree from each other when converting them to surface-proportional values. This contradicts the hypothesis that a critical germ is formed preferentially near the surface of a super-cooled liquid droplet.