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A time dependence study of droplet freezing with immersed mineral dust particles

Tina Clauss (1), Susan Hartmann (1), Dennis Niedermeier (1), Alexei Kiselev (2), Heike Wex (1), and Frank Stratmann (1)

(1) Institute for Tropospheric Research, Permoser Str. 15, 04318 Leipzig,Germany (tina.clauss@tropos.de), (2) Karlsruhe Institute of Technology, Postfach 3640, 76021 Karlsruhe, Germany

Due to the importance of the ice phase for the radiative and microphysical properties of clouds, the freezing behaviour of aerosol particles has received an increased attention in the past years. Especially heterogeneous freezing plays a major role in atmospheric ice formation processes. There are two contrary approaches concerning the description of heterogeneous ice nucleation processes which are controversially discussed. In the first approach, the stochastic hypothesis, it is stated that the presence of an ice nucleus (IN) within a supercooled droplet increases the likelihood of the random ice nucleation but does not disturb the stochastic nature of critical cluster formation. Therefore with increasing time the likelihood of a supercooled droplet to freeze at a given temperature increases too. In contrast, the singular hypothesis describes that critical cluster formation occurs on specific sites on the IN surface at a characteristic temperature, i.e., freezing is assumed to be time-independent. To verify one of those contrary approaches immersion freezing experiments were performed in the laminar diffusion cloud chamber LACIS (Leipzig Aerosol Cloud Interaction Simulator, Hartmann et al., 2010; Stratmann et al., 2004). Size-selected monodisperse Arizona Test Dust (ATD) particles were activated to droplets which then were supercooled. The freezing of a certain fraction of these droplets was then observed for one adjusted temperature. Additionally, the residence time inside LACIS was varied between 2 s and 9 s resulting in different ice nucleation times in order to validate time dependence or independence. This experiment was performed for different freezing temperatures. For the determination of frozen and unfrozen particles downstream LACIS, a novel optical ice detecting instrument LISA (LACIS Ice Scattering Apparatus) was used. The instrument belongs to the series of Small Ice Detector 3 (SID3) instruments (Kaye, 2008), capable of capturing high-resolution two dimensional light-scattering patterns from single particles in near-forward direction with a high-speed CCD camera. Investigating the immersion freezing behavior of ATD particles for different ice nucleation times, it was found that ATD particles nucleate ice over a broad temperature range and the increase of ice nucleation time from 2 s to 9 s does not significantly change the freezing temperature distributions. These findings agree with the singular description of the ice nucleation process. But studies with the 'soccer ball' model (see Niedermeier et al., this session) showed that the variability of the surface properties (e.g., surface free energy) across the population of the very heterogeneous Arizona Test Dust particles is most plausibly responsible for the broad temperature range over which droplets freeze and for the apparent missing time dependence of heterogeneous ice nucleation.

References:

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