Surface site density versus nucleation rate approaches of formulating ice formation in clouds – a comprehensive analysis based on AIDA cloud simulation experiments

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A more detailed understanding and formulation of cloud microphysical processes is still one of the major challenges for improving atmospheric cloud, weather, and climate models. These include homogeneous freezing of water and solution droplets, though this part is considered to be well understood and formulated for models (Pruppacher et al., 1997; Koop et al., 2000) as well as the various modes of heterogeneous ice nucleation (Vali, 1985) like deposition nucleation, condensation freezing, immersion freezing and contact freezing, which involve solid atmospheric aerosol particles as heterogeneous ice nuclei (IN).

The fact that only a very minor fraction of the solid atmospheric aerosol particles is active as ice nuclei, in particular at high temperatures (Pruppacher and Klett, 1997) makes it difficult to quantify and formulate the temporal and spatial distribution of ice nuclei for models. Another complication is related to the complex mixture and high variability of the atmospheric aerosol particles concerning size, chemical composition, morphology, and surface coating, which makes it difficult to formulate and calculate the IN concentration not only as a function of temperature and humidity, but also as a function of aerosol parameters.

During the previous years, an increasing number of laboratory experiments investigated ice nucleation processes for a broad variety of aerosol types (Hoose and Möhler, 2012; Murray et al., 2012). Different concepts and approaches have been developed and applied to fit the experimental data, in order to use such fit results as parameterizations of ice formation in atmospheric models. Some of them are relying on classical nucleation rate theory (Pruppacher and Klett, 1997), others describe the ice nucleation activity as function of ice-active sites on the aerosol particle surface (Connolly et al., 2009).

The nucleation rate approaches calculate the formation rate of ice directly as a function of time and particle surface area, but in most cases have to consider an empirical variation of the activation energy or freezing probability over the particle surface area at a given temperature. This is due to weaker temperature dependence of the ice formation rate in most experiments than predicted by classical nucleation rate theory with just one activation energy or contact angle (e.g. Marcolli et al., 2007).

The active site approaches consider a temperature dependent variation of the freezing probability per particles surface area and calculate the formation rate of ice particles mainly as function of the rate of temperature change (Connolly et al., 2009; Niemand et al., 2012). Up to now it appears to be unclear which approach is more appropriate to formulate and predict the abundance of ice nuclei and ice particles in the atmosphere.

Since 2003, the AIDA cloud chamber was used for comprehensive series of ice nucleation experiments with a variety of different aerosols like soot, organics, minerals or biological particles, and in wide ranges of temperature, relative humidity and cooling rate. The AIDA team has started consistent and comprehensive re-analysis of the 10 year data set to provide a uniform compilation of parameters for formulating the ice formation in atmospheric models as function of aerosol properties, temperature and humidity. The conference contribution will include an overview and discussion of the experimental data set, selected results and some basic concepts.

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