New Homogeneous Ice Nucleation Results from Measurements at a Large Atmospheric Simulation Chamber

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Phase transitions in clouds such as droplet formation by the condensation of water vapour or droplet freezing are in most cases heterogeneous processes triggered by aerosols. On the other hand, homogeneous ice nucleation without the participation of aerosols is an important process in deep convective systems, where almost pure water droplets are supercooled until homogeneous freezing occurs at temperatures below about 238 K.

A well cited theory which describes homogeneous ice nucleation was published by Pruppacher and Klett (1997). It is called classical nucleation theory and includes a number of uncertain parameters like the interface tension between the supercooled liquid and the ice phase or the activation energy. Therefore it is difficult to exactly formulate classical nucleation theory for a certain nucleating system like supercooled water drops. What is done instead is to fit classical nucleation theory formulations to experimental data, which also involve errors and uncertainties.

Here we present a reanalysis of droplet freezing experiments at the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud chamber at KIT, Karlsruhe, Germany, which follows experimental methods described by Benz *et al* (2005) who already published first homogeneous ice nucleation rates in supercooled water droplets derived from measurements at the AIDA facility.

At the large aerosol and cloud chamber AIDA the interaction between aerosol particles and clouds is being investigated. The facility operates under atmospherically relevant conditions within a wide range of temperature, pressure and humidity, whereby investigations of both tropospheric mixed-phase clouds and cirrus clouds can be realised. By controlled adiabatic expansions the ascent of an air parcel can be simulated. In order to investigate homogeneous freezing, sulphuric acid seed aerosol is typically injected into the chamber (Figure 1). By starting the expansion the gas temperature decreases and therefore the relative humidity rises whereby water condenses at the sulphuric acid droplets. They then grow until they are almost pure water drops. First ice particles appear at lower temperatures by homogeneous freezing. The ice particles then rapidly grow in size at the expense of the remaining droplets (Wegener-Bergeron-Findeisen process) with the result that at the end of the experiment only ice particles are left inside the chamber.

During these expansions, the aerosols, droplets and ice particles are monitored by a number of instruments. For the calculation of homogeneous ice nucleation rates the measurements of an optical particle counter (welas, PALAS GmbH) are used. The

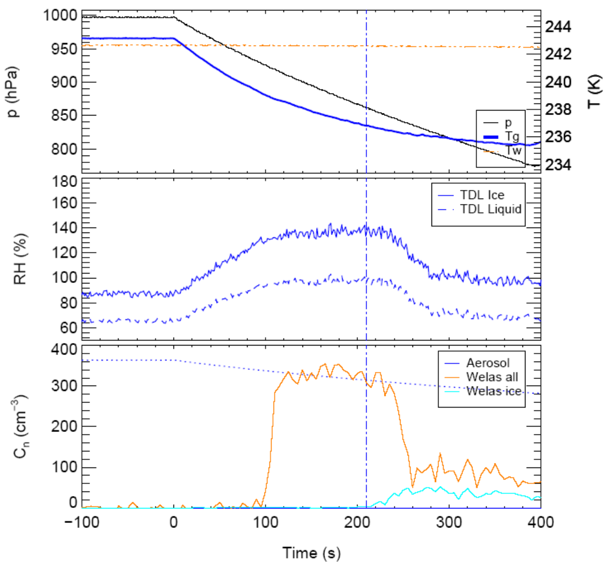

Figure 1. Data time series of a typical AIDA homogeneous droplet freezing experiment.

instrument detects the size of the resulting droplets and the number concentration of droplets and ice particles during the expansion. With these values homogeneous ice nucleation rates in supercooled water droplets can be calculated.

On the base of the new and improved data set we hope to achieve a more accurate fit of classical nucleation theory parameters. In addition, these new homogeneous ice nucleation results from the AIDA cloud chamber facility will be compared with homogeneous ice nucleation rates derived from other experiments like the cold stage where a droplet ensemble can be examined (Di Natale, 2012) or the Paul trap where single droplets can be studied by levitating them in an electrodynamic balance (Duft and Leisner, 2004; Rzesanke *et al*, 2012).

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