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Ice nucleation rates of ammonium sulphate and sulphuric acid aerosol droplets at upper tropospheric temperatures: Experiments in the aerosol chamber AIDA

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High altitude cirrus clouds influence the radiative balance of the earth both by scattering incoming sunlight and by absorbing infrared radiation emitted by the earth. Important properties of these clouds are determined by the ice nucleation mechanisms. Ice crystals are formed either by homogeneous freezing of pure liquid solution droplets, by heterogeneous freezing of droplets promoted by solid inclusions, or via direct deposition of water vapour on suitable dry particles.

It is well known from field campaigns that sulphate aerosols including sulphuric acid and ammonium sulphate are abundant throughout the upper troposphere. The aerosol and cloud chamber AIDA of Forschungszentrum Karlsruhe was used to investigate ice formation in these aerosols. The instrumentation of the chamber is presented by Wagner et al. in a separate contribution. The chamber was used to expose sulphuric acid and ammonium sulphate aerosols (as the limiting cases of atmospheric sulphates) to realistic conditions with regard to temperature, cooling rate, and ice super-saturation. Sulphuric acid aerosol neither converts to dry particles at low relative humidities, nor does it form solid hydrates at the temperatures covered by this study. Therefore, ice formation in pure sulphuric acid particles is expected to occur by homogeneous freezing only, as described by the water activity model of Koop et al. This contrasts with supersaturated ammonium sulphate particles which may or may not effloresce to form dry crystals before passing the water activity threshold of homogeneous freezing. Whether ammonium sulphate aerosol exists in the form of liquid super-cooled droplets, or has partially been converted to dry solid particles is not always clear. This

is important because a small fraction of dry ammonium sulphate particles may serve as ice nuclei for deposition nucleation well below the homogeneous freezing relative humidity. It has even been suggested that the water activity model of Koop et al. does not apply to the homogeneous freezing of super-cooled liquid sulphuric acid particles.

In our cloud chamber AIDA we performed ice nucleation experiments with initially liquid as well as initially solid particles in the temperature regime from -50˚C to -70˚C to assess these issues.