THE KARLSRUHE AEROSOL CHAMBER FACILITY AIDA: TECHNICAL DESCRIPTION AND FIRST RESULTS OF HOMOGENEOUS AND HETEROGENEOUS ICE NUCLEATION EXPERIMENTS

O. Möhler, A. Nink), H. Saathoff, S. Schaefers, M. Schnaiter, W Schöck, and U. Schurath*
Institute of Meteorolgy and Climate Research, Forschungszentrum Karlsruhe, Germany

Abstract
The large experimental facility AIDA of the institute of meteorology and climate research at Forschungszentrum Karlsruhe is operated and used as a cloud chamber to study processes of ice formation in tropospheric and stratospheric clouds. Like in clouds, particle freezing and growth is initiated by expansion which leads to quasi-adiabatic cooling and thus ice- and water supersaturation at constant wall temperature. Intensity and depolarisation of forward- and back-scattered laser radiation is measured, caused by particles in a small scattering volume far from the walls. The ice phase is also detected by *in situ* FTIR spectroscopy. Number size distribution of interstitial aerosol and activated ice particles is measured with an optical particle counter. Various insoluble aerosol components can be generated and added to the chamber in order to investigate their influence on ice formation processes at controlled temperatures, cooling rates, and supersaturations.

1. INTRODUCTION

Ice particle formation is an important process in the troposphere and stratosphere. It can occur either by homogeneous freezing of droplets below about –35°C [1], or be heterogeneously induced by so-called ice nuclei. E.g. it is speculated that soot particles from aircraft can act as ice nuclei [2]. A quantitative description of these processes is crucial for a better understanding of the lifetime of clouds with respect to rainout, and their optical properties. Distinction between supercooled liquid and frozen aerosol particles (cloud hydrometeors; PSC particles) is essential for the investigation of these ice nucleation processes.

Polar stratospheric clouds play a crucial role in the ozone destruction process. During recent years, various physical and chemical particle formation processes have been investigated intensively. Liquid ternary solution particles, crystalline hydrates, and PSCs mainly composed of ice or mixtures of liquid and solid particles have been detected and analysed by remote sensing [3, 4] and in situ techniques [5]. The ability of PSCs to induce chemical ozone depletion is a function of particle concentration, size, composition, and thermodynamic phase. For example, solid particles can grow bigger than liquid and thereby are thought to be responsible for denitrification of the lower stratosphere, enhancing and extending ozone depletion [6]. So far the formation processes of solid particles are not fully understood.

2. AIDA FACILITY

2.1 Experimental setup

A schematic cross section of the AIDA cloud chamber and some analytical and technical instrumentation is shown in Figure 1. This chamber (Volume = 84 m^3) is operated over a wide

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*) Now at Bayer AG, Leverkusen, Germany.

range of atmospheric conditions: -90°C < T < +60°C, r.h. under static conditions near 100 %; pressures from above 1 bar to below 1 mbar; ice and water supersaturations. This covers conditions throughout the troposphere and lower stratosphere under which water clouds, mixed clouds, cirrus clouds, and even Polar Stratospheric Clouds (PSC) are formed.

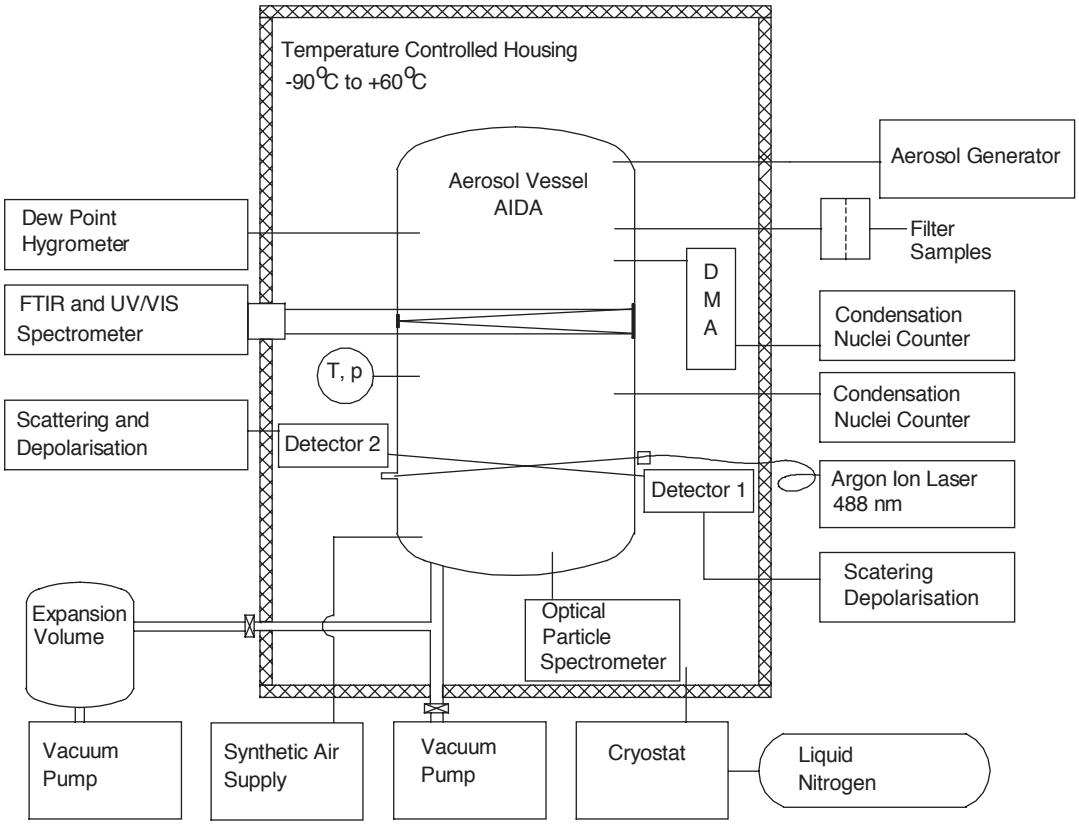

Fig. 1 AIDA main components and instrumentation available for ice activation experiments.

2.2 Ice activation experiments

Experiments investigating ice formation at supersaturated conditions typically are started at homogeneous temperature conditions, pressure between 180 hPa and 1000 hPa, and relative humidity close to ice saturation controlled by an ice covered chamber wall. Ice supersaturation is achieved by volume expansion due to controlled pumping using two large vacuum pumps at different pumping speeds. Depending on starting temperature and pumping speed, the regime between ice and water saturation is passed within a few minutes at cooling rates up to 200 K/h. Figure 2 shows an expansion started at 174 hPa and 225 K. The expansion period lasted about 8.5 min. The highest cooling rates are only achieved within the first few minutes. Hereafter, a steady state is achieved between further adiabatic cooling and heat transfer from the chamber walls remaining at constant temperature during expansion due to the high heat capacity of the 2 cm thick aluminium walls. After pumping is stopped the gas temperature increases and approaches the wall temperature on a time scale of about five minutes. Volume expansion into an evacuated vessel of 4 m^3 volume can additionally be used to sharply increase the supersaturation by up to 20 % within a few seconds. Evaporation of ice phases is forced by controlled adiabatic heating of the chamber gas due to refilling the chamber with dry synthetic air.

Water vapour is measured with three independent instruments: The FISH Lyman-a hygrometer of the ICG-1 of Forschungszentrum Jülich [7], the prototype of a novel photoacoustic water vapour sensor (PAS) developed and operated by the University of Szeged,

Hungary [8], and a commercial cooled mirror hygrometer M3 from General Eastern. All instruments are operated outside the chamber using the same heated sampling tube.

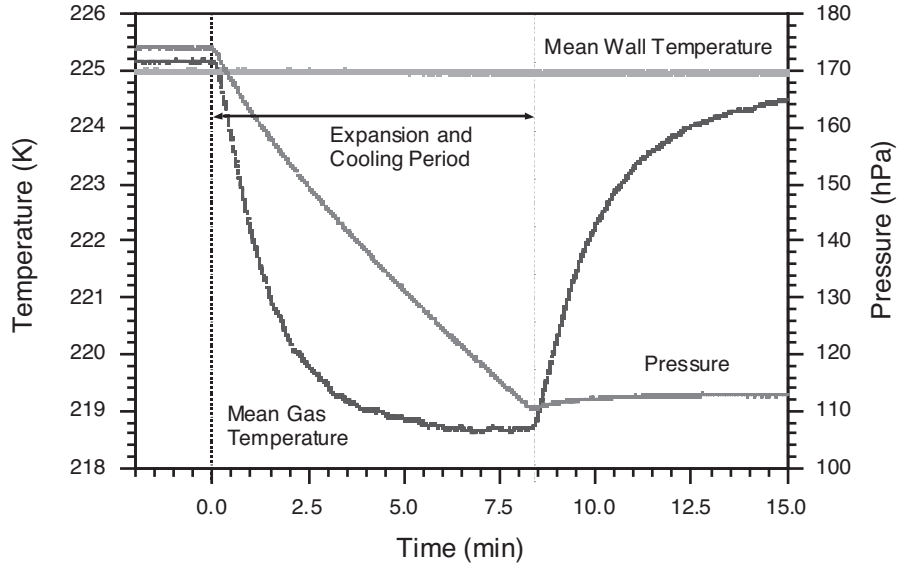

Fig. 2 Time profiles of pressure, mean wall temperature, and mean gas temperature during a typical ice activation experiment.

2.3 Detection of ice formation

An Argon-Ion laser beam (99% polarized radiation at 488 nm) is conducted into the chamber via an optical fibre which preserves the plane of polarization (Figure 3, left panel). The laser beam and the aperture of the detection optics overlap in the middle of the chamber at a distance of 2 m from the walls, providing about 2 cm^3 of scattering volume. The scattered light is split into the parallel and the perpendicular components by a Glan-Taylor prism and then detected by two independent photomultipliers (Figure 3, right panel). Detector optics are mounted at scattering angles of 176° and 4°. Photon counting is employed to achieve high sensitivity and time resolution. The laser source and the detectors can be attenuated by neutral density filters to avoid saturation, and to match the sensitivities of the forward and backward scattering detectors. This setup provides information on the volume, size, and phase of the scattering aerosol. The data set allows for a precise determination of the onset of ice formation and the formation and growth of liquid and solid aerosol particles.

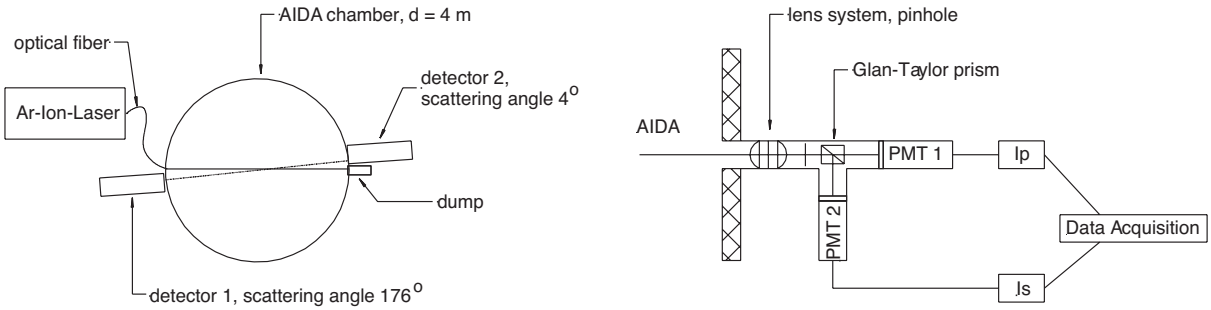

Fig. 3 Overall cross section of the laser scattering device (left) and detail of the detector for measuring backscattered light intensity and depolarisation.

Size distributions of aerosol particles before, during, and after periods of ice nucleation are measured with an optical particle spectrometer (PCS2000, Palas) operated below the aerosol chamber (c.f. Figure 1). Residence times in the cold vertical sampling tube are short enough to minimise evaporation of ice particles.

Aerosol extinction is measured by long path *in situ* FTIR spectrometry (Bruker IFS 66v, 254 m folded optical path) in the spectral range 6000 - 800 cm^{-1} with a resolution of 4.0 cm^{-1}. The FTIR measurements are made with a maximum time resolution of 40 seconds. The extinction spectra provide valuable information about size, chemical composition, and phase of the average particle volume both at equilibrium condition and periods of high cooling and heating rate.

3. HOMOGENEOUS FREEZING OF SUPERCOOLED SULPHURIC ACID PARTICLES

Binary sulphuric acid droplets with a mean diameter of about 200 nm are generated at atmospheric pressure outside the aerosol chamber. Aerosol is generated by dispersing a 20 wt% sulphuric acid solution and dried by passing through a glass tube partly filled with a 96 wt% sulphuric acid solution. The dried aerosol is passed into the chamber through a pressure reduction valve and a stainless steel tube. The size distribution of the AIDA aerosol covers the size range of stratospheric sulphuric acid background particles (c.f. Figure 4).

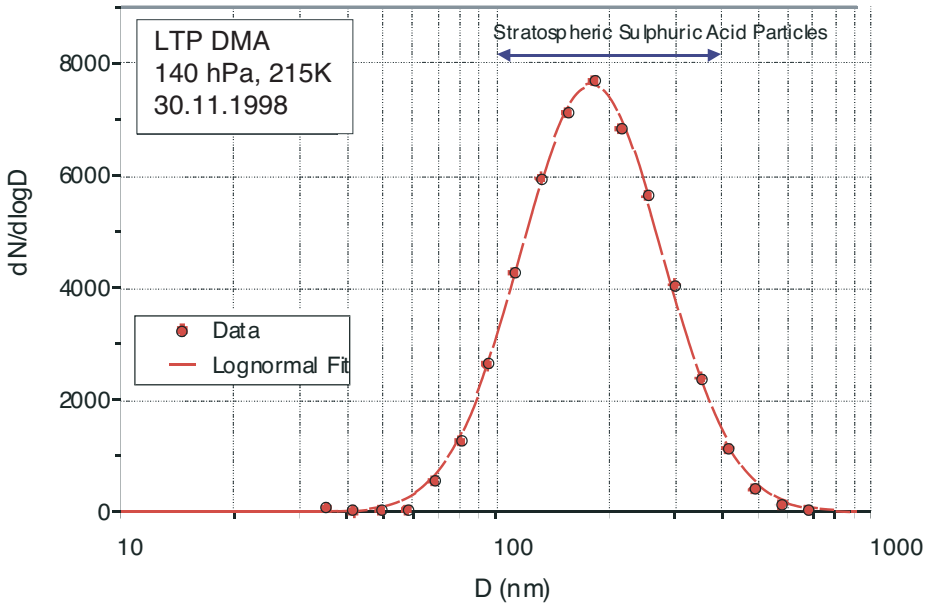

Fig. 4 Number size distribution of sulphuric acid particles measured during an AIDA PSC simulation experiment at a temperature of 215 K and a pressure of 140 hPa.

Number concentration and size distribution of sulphuric acid aerosol particles are measured with a condensation nuclei counter (CNC3010, TSI) and a differential mobility analyser in combination with a CNC3010. The CNC devices have been modified for operation at pressures from 100 hPa to atmospheric pressure. The modified DMA is operated at the same temperature as the aerosol chamber in order to avoid size change of particles by evaporation processes.

Figure 5 shows the result of a freezing experiment with sulphuric acid particles. The expansion was started at t=223 min at a pressure of 180 hPa and a temperature of 202 K. Ice formation occurred after 3 min of pumping at t=226 min, as clearly indicated by the sudden increase of the depolarisation ratio and the number of ice particles.

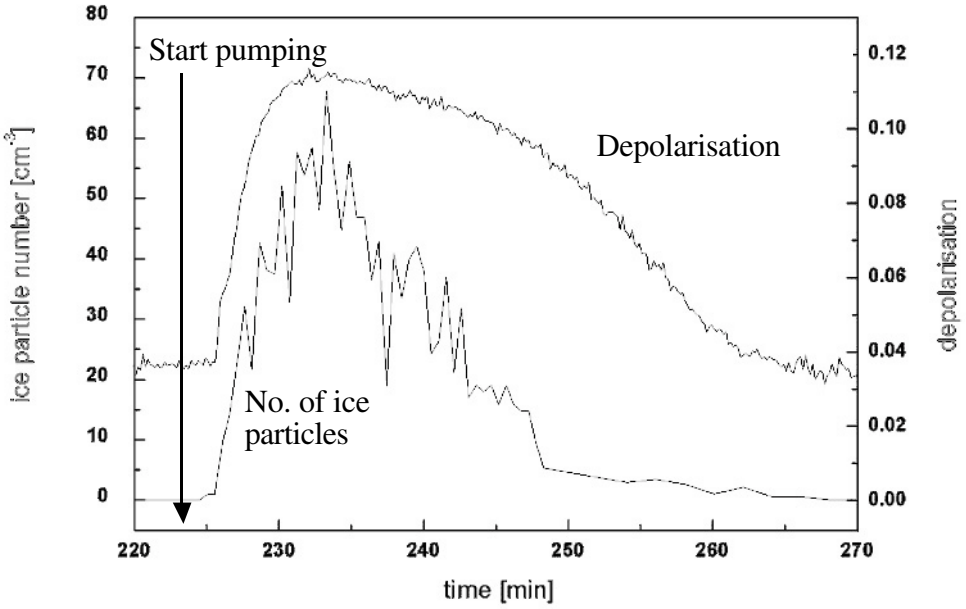

Fig. 5 Depolarisation ratio of backscattered laser light (upper curve) and number of ice particles formed during homogeneous freezing of supercooled sulphuric acid aerosol particles (lower curve).

4. HETEROGENEOUS ICE NUCLEATION OF SOOT PARTICLES

The heterogeneous ice nucleation potential of soot particles was investigated at temperatures between –62°C and –22°C. The soot aerosol was taken from a graphite spark generator (GfG1000, Palas).

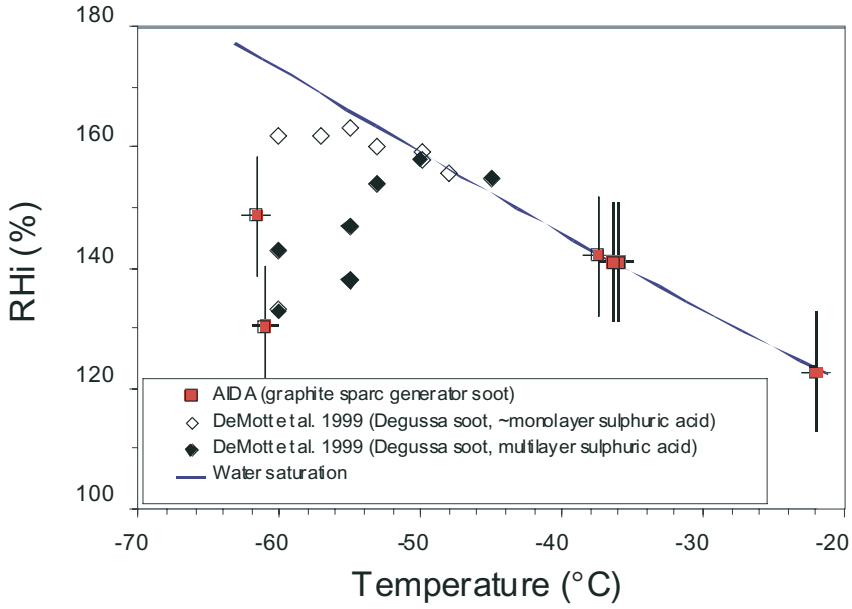

Fig. 6 Ice activation relative humidities measured for spark generator soot particles (filled squares, present study) and "Degussa" soot coated with monolayer and multilayer sulphuric acid (open and filled diamonds [2]).

The relative humidity with respect to ice, RHi, was calculated as function of measured ice frost point and mean gas temperature. Figure 6 shows RHi measured at the onset of ice formation. At higher temperatures liquid water seems to condense on the soot particles before ice activation occurs (immersion freezing). At lower temperatures ice is formed significantly below the liquid water saturation threshold. Figure 6 also depicts results from DeMott et al. [2]. Degussa soot used in that study shows significant lowering of ice onset RHi only for multilayer sulphuric acid coating. The GfG soot used in the AIDA experiments had not been coated with sulphuric acid.

5. SUMMARY AND CONCLUSION

In the AIDA experimental facility, ice activation experiments are performed by simulating cycles of ice and water super- and sub-saturations, using the method of expansion-cooling. Relative humidity can dynamically be increased in a controlled manner from ice saturation to values above water saturation within several minutes. First results on homogeneous freezing of super-cooled sulphuric acid particles and heterogeneous ice nucleation of soot prove the AIDA facility to be well suited for IN studies. One of the major advantages of AIDA ice nucleation experiments is the fact that only a minor fraction of the aerosol is lost during expansion cycles. Therefore, the same aerosol sample can be investigated in repeated activation and evaporation cycles. This opens an avenue for future experiments to study the influence of particle ageing effects (e.g. restructuring, coating) on the ice nucleation potential of relevant aerosols.

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ice formation, ice nucleation, Depolarisation, aerosol particles, wall temperature, ice particles, Particle Spectrometer, cirrus clouds, supersaturation, adiabatic cooling, volume expansion, atmospheric conditions, static conditions, expansion period, water saturation, Glan-Taylor prism, detection optics, detector, time resolution, optical particle, Argon-Ion laser beam, gas temperature, Condensation Nuclei, supersaturations, Polar Stratospheric Clouds, Dew Point Hygrometer, S. Schaefers, atmospheric pressure, Forschungszentrum Karlsruhe, formation processes, HOMOGENEOUSAND HETEROGENEOUS, AIDA, spark generator, SOOT PARTICLES, Degussa, U. Schurath Institute of Meteorolgy and Climate Research, stratospheric clouds, particle formation, sulphuric acid, optical particle counter, H. Saathoff, ice nuclei, TECHNICALDESCRIPTION, cloud chamber, M. Schnaiter, experimental facility

The Karlsruhe aerosol chamber facility AIDA: Technical description and first results of homogeneous and heterogeneous ice nucleation experiments