Heterogeneous ice nucleation of soot aerosol coated with sulphuric acid and organic carbon

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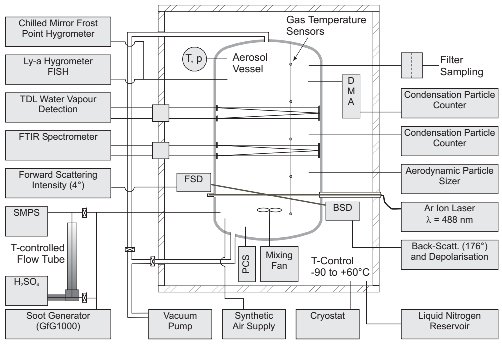
AIDA Facility for Ice Nucleation Studies



Background and Motivation

The contribution of aircraft-emitted soot aerosol particles to cirrus cloud formation may depend on their mixing state (coating) with other emitted substances, e.g. sulphuric acid and organics.

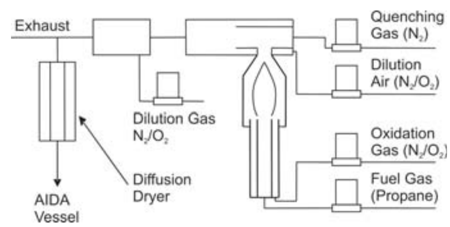
We measured the ice saturation at freezing onset, S_{IN}, for graphite spark generator soot (GSGS), sulphuric acid coated GSGS (SACS), and flame soot with organic carbon content of 16 % (CS16) and 40 % (Cs40) at temperatures between 185 K and 240 K.

Results

- GSGS was the most active soot aerosol with S_{IN} between 1.1 and 1.3, slightly above the deposition ice nucleation thresholds on mineral dust particles.
- Sulphuric acid coating of the same soot increased S_{IN} to values between 1.3 and 1.5, still below the freezing threshold of pure sulphuric acid droplets.
- FS16 nucleated ice at S_{IN} above about 1.45.
- Higher organic carbon content (FS40) markedly suppressed ice nucleation.

Combustion Aerosol Standard (CAST) Burner

Flame soot was generated with a CAST burner (see figure), mainly consisting of the flame region and a quenching and mixing gas region. The burner gas is mixed by diffusion of two concentric flows of propane and synthetic air maintained by an inner tube of 8 mm diameter and 2 mm wall thickness and a concentric outer tube of 14 mm diameter. The ratio of these flows controls the so-called elemental carbon (EC) and organic carbon (OC) contents of the resulting soot aerosol.



Method of Dynamic Expansion for AIDA Ice Nucleation Studies

Ice nucleation experiments are started at homogeneous temperature conditions, atmospheric pressure, and relative humidity close to ice saturation controlled by ice coated chamber walls.

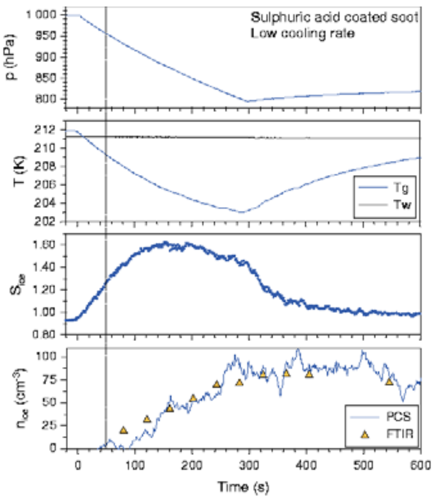
Adiabatic cooling is achieved by pumping from 1000 to 800 hPa.

Total water (gas and ice) is measured with a chilled mirror frost point hygrometer (MBW), the FISH Ly-α Hygrometer, and a photoacoustical detection system.

Ice saturation ratios are calculated from in situ tuneable diode laser absorption measurement of the water vapour pressure and the ice saturation pressure as function of the mean gas temperature.

Formation of a-spherical ice crystals is most sensitively detected by scattering intensity and depolarisation of 488 nm laser radiation at 176° scattering angle (not shown in the figure).

Number concentration and size distribution of ice crystals are measured with an optical particle counter and

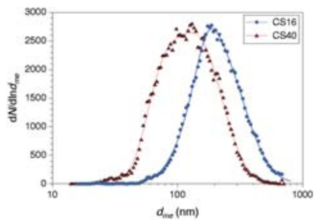


The figure shows data sets from an expansion experiment with sulphuric acid coated soot starting at a gas temperature of 212 K.

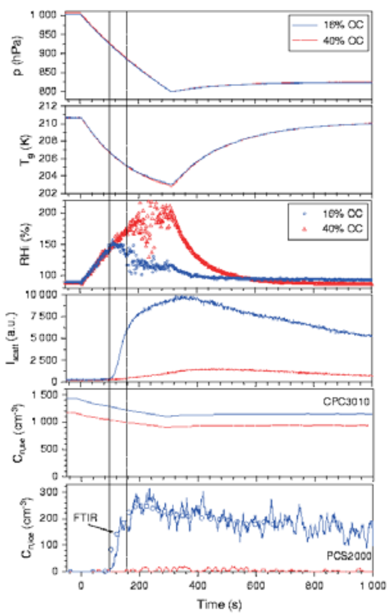
Experimental Results for Flame Soot with 16 % and 40% OC

Two expansion experiments were prepared at almost identical pressure p, gas temperature T_g, and initial relative humidity with respect to ice, RHi, but different CAST soot aerosols CS16 and CS40.

During experiment CS16, the increase of back-scattered light intensity and number concentration of particles measured with the optical particle counter clearly indicate the formation and growth of pristine ice crystals. During experiment CS40, both the number concentration and volume of ice crystals are much less compared to experiment CS16.

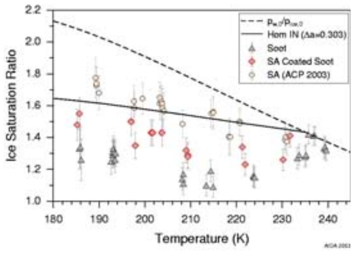


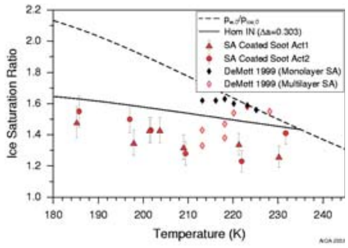
Size distributions of CS16 and CS40 soot aerosol in the AIDA chamber. Cs40 particles are smaller because the higher amount of OC causes additional shrinking of the fractal soot aggregates.



Freezing Onset of Soot and Sulphuric Acid (SA) coated Soot

Coating GSG soot particles with sulphuric acid affects both the mechanism and saturation threshold for ice nucleation. The left Figure directly compares S_{IN} measured for both GSGS (black triangles) and SACS (red-edged diamonds). Previous AIDA results for the homogenoues freezing of pure sulphuric acid solution droplets (Möhler et al., 2003) are also shown (filled circles). In the entire temperature range from 185K to 220K, the internal mixture of soot and sulphuric acid nucleates ice at S_{IN} between that of GSGS and pure sulphuric acid particles. The right Figure compares the SACS data to previous results by DeMott et al. (1999) with Degussa soot of different sulphuric acid coating.





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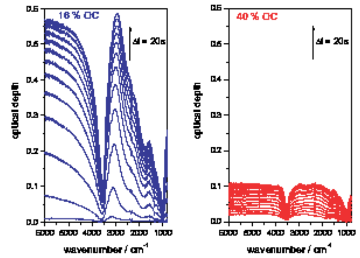
Suppression of Ice Nucleation by Organic Carbon

The formation and growth of ice crystals during experiment CS16 is also documented in series of FTIR extinction spectra measured during the expansion experiment in time intervals of 20 s. The spectra (blue lines in the Figure to the right) recorded at wave numbers between 800 and 6000 cm^{-1} show signatures of increasing volume of ice crystals as discussed in more detail by Mangold et al. (2004).

During experiment CS40, both the number concentration and volume of ice crystals are much less compared to experiment CS16 (cf. 'red' spectra). First signatures of ice formation occur at RHi between 140 and 170 %. However, RHi continues to increase to a peak above 190 % which is above liquid saturation at the respective temperature (see Figure above).



The freezing onset of CS16 soot measured at 207 K (see Figure) compares well with sulphuric acid coated spark generator soot (SACS). For comparison, AIDA expansion experiments were also performed with mineral dust aerosol samples (Arizona test dust, ATD) in the temperature range 194 to 241 K.The ATD particles nucleated ice at very low S_{IN} between 1.05 and 1.15 in all experiments.