Name of research institute or organization:

Max Planck Institute for Chemistry, Mainz
Particle Chemistry Department

Title of project:

Mass spectrometric analysis of residuals from small ice particles and from supercooled cloud droplets during the Cloud and Aerosol Characterization Experiments (CLACE)

Project leader and team:

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Project description:

The identification of ice nuclei is crucial for the understanding of heterogeneous ice nucleation in supercooled clouds, which is the main initiation process of precipitation in middle latitudes. Until today it is not well understood which chemical components (e.g. sulphuric acid, ammonium, nitrate, various organic substances, mineral dust, sea salt, soot, or other materials) contained inside or on the surface of aerosol particles enable a particle to act as an ice nucleus (IN). While water soluble compounds are expected to favour the formation of liquid cloud droplets, insoluble materials like mineral components may favour the formation of ice particles.

During the 3rd and 4th Cloud and Aerosol Characterization Experiments (CLACE-3, CLACE-4) in February/March 2004 and 2005, mass spectrometric measurements of residuals of supercooled cloud droplets and small ice particles were performed at the High Alpine Research Station Jungfraujoch. An Aerodyne Quadrupole Aerosol Mass Spectrometer (Q-AMS) was used to measure chemically resolved mass concentrations and size distributions of various non-refractory aerosol components (sulphate, nitrate, ammonium, organics) in the size range of 20 – 1500 nm.

A novel sampling system for freshly formed ice particles (ICE-CVI, Institute for Tropospheric Research, Leipzig) was coupled to the Q-AMS. By pre-segregation of other mixed-phase cloud constituents and evaporation of the ice water fraction, the residual particles, which are expected to be the original IN, were made available for analysis with the Q-AMS. Depending on cloud type and ICE-CVI operation mode, the combination of Q-AMS and ICE-CVI allowed the analysis of residuals of ice particles as well as of supercooled cloud droplets. Alternatively, the interstitial and out-of-cloud aerosol was sampled and compared to the residual particles.

Figure 1 gives the time series of mass concentrations of ammonium, nitrate and sulphate, measured for interstitial aerosol (during cloud events) and out-of-cloud aerosol. The concentrations during the cloud free time periods vary significantly and are relatively low, as expected for free tropospheric aerosol.

Filter samples taken by the Paul Scherrer Institute, Villigen, allowed an additional chemical characterization of the aerosol. Figure 2 shows a comparison between sulphate mass concentrations derived from the filter samples and from the Q-AMS. The data agree well within the uncertainties.

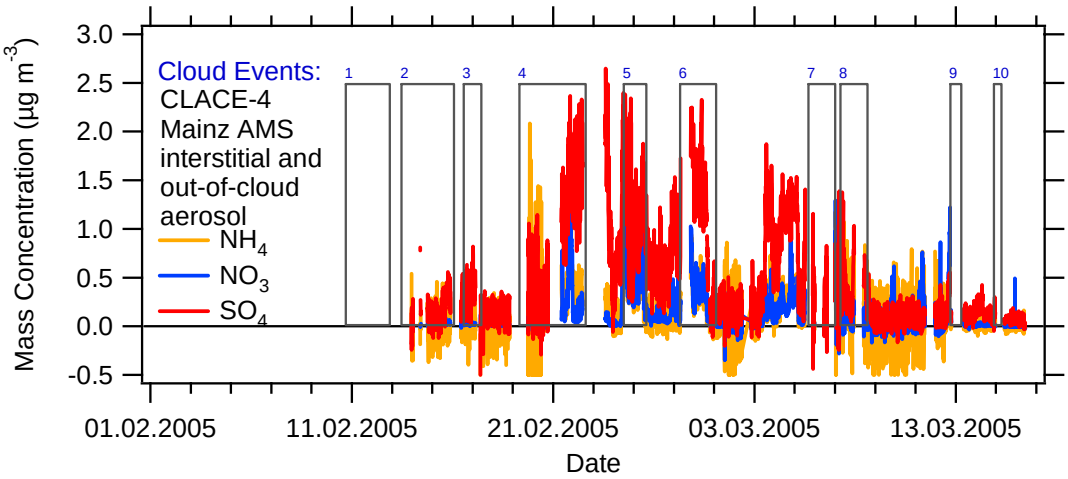

Figure 1: Time series of Q-AMS mass concentrations or interstitial and out-of-cloud aerosol

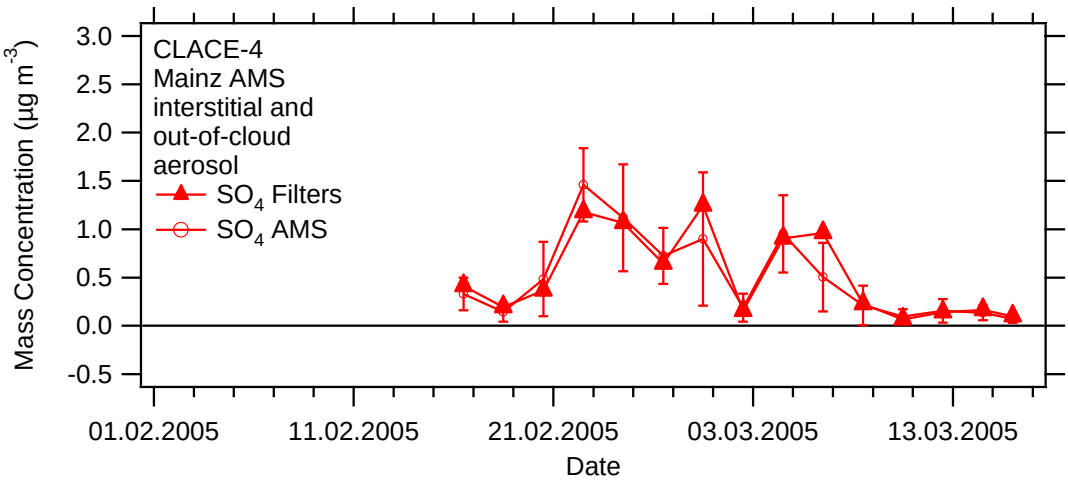

Figure 2: Comparison of SO_4 mass concentrations measured with the Q-AMS and filters

Integrated mass concentrations for the transmission range of the AMS inlet system (vacuum aerodynamic diameter < 1000 nm) from both AMS and SMPS are given in Figure 3. The SMPS data were converted from mobility into vacuum aerodynamic diameter using densities inferred from comparison of the measured diameters (mobility and vacuum aerodynamic diameter), assuming spherical particles. For the shown time periods the densities were found to be 1.5 g cm^{-3} for both interstitial and out-of-cloud aerosol. For the ice residuals, a density of 2.0 g cm^{-3} was chosen.

The unknown density of the ice residuals was varied between 1.5 and 2.5 g cm^{-3}, indicated by the error bar added to the SMPS value. The comparison of SMPS and AMS data indicates that the out-of-cloud aerosol was composed to about 80% of non-refractory material. The interstitial aerosol was found to contain a larger fraction of refractory compounds than the out-of-cloud aerosol. The ice cloud residuals sampled by the CVI show very low mass concentrations detected by the AMS, although the total mass below 1 µm, as inferred from the SMPS, was well above the AMS detection limit. This implies that a large fraction of the ice residual mass (≈ 86%) could not be detected by the AMS. Since the amount of black carbon (not shown) does not account for this difference, this finding implies that preferably refractory particles like mineral dust act as ice nuclei.

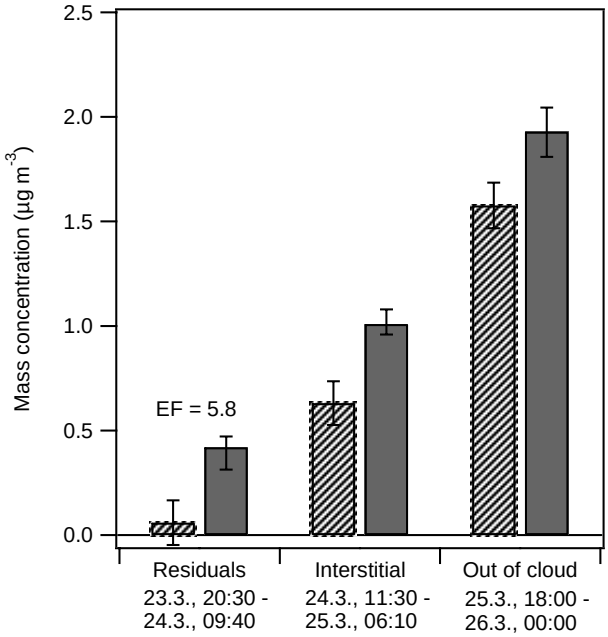

Figure 3: Mass concentrations measured by the AMS in comparison with mass concentrations inferred from the SMPS for ice cloud residuals, interstitial and out-of-cloud aerosol particles (CLACE-3). Note that the residuals are enriched by a factor of 5.8 compared to the other data.

Results from measurements with a similar setup using additionally two single particle laser ablation mass spectrometers during a follow-up experiment (CLACE-5) in February/March 2006 will help to gain further insight into the chemical composition of ice cloud residuals.

Key words:

Aerosol mass spectrometry, Cloud-Aerosol interactions

Collaborating partners/networks:

E. Weingartner et al., Laboratory of Atmospheric Chemistry, Paul Scherrer Institut, Villigen, Switzerland
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K. Bower et al., School of Earth, Atmospheric and Environmental Sciences, University of Manchester, UK
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J. M. Balzani-Lööv et al., ETH, Zürich, Switzerland
G. Legreid et al., EMPA, Dübendorf, Switzerland

Scientific publications and public outreach 2005:

Conference papers/contributions

S. Walter et al., Mass spectrometric analysis of residuals from small ice particles and from supercooled cloud droplets during CLACE-3 and CLACE-4, oral presentation at the European Aerosol Conference, Ghent, Belgium, 2005

E. Weingartner et al., An overview of the Cloud and Aerosol Characterization Experiments (CLACE) conducted at the high alpine research station Jungfraujoch in Switzerland, oral presentation at the European Aerosol Conference, Ghent, Belgium, 2005

S. Mertes et al., Sampling and physico-chemical characterisation of ice nuclei in mixed phase clouds at the high alpine research station Jungfraujoch (3580 m asl) during CLACE, oral presentation at the European Aerosol Conference, Ghent, Belgium, 2005

B. Verheggen et al., Nucleation and activation of aerosol particles during CLACE campaigns (Jungfraujoch, 3580 metres a.s.l., Switzerland), oral presentation at the European Aerosol Conference, Ghent, Belgium, 2005

M. Ebert et al., Identification of the ice forming fraction of the atmospheric aerosol in mixed-phase clouds by environmental scanning electron microscopy, poster presentation at the European Aerosol Conference, Ghent, Belgium, 2005

J. Crosier et al., Comparing winter and summer submicron aerosol chemical composition and size distributions at the Jungfraujoch, poster presentation at the European Aerosol Conference, Ghent, Belgium, 2005

E. Weingartner et al., An overview of the Cloud and Aerosol Characterization Experiments (CLACE) conducted at a high alpine site in the free troposphere, solicited oral presentation at the EGU General Assembly, Vienna, Austria, 2005

U. Baltensperger et al., Aerosol hygroscopic growth closure by simultaneous measurement of hygroscopic growth and chemical composition at the high-Alpine station Jungfraujoch, solicited oral presentation at the EGU General Assembly, Vienna, Austria, 2005

J. Cozic et al., Aerosol - cloud interaction: highlights from the Cloud and Aerosol Characterization Experiments (CLACE) conducted at the high alpine research station Jungfraujoch in Switzerland, oral presentation at the 1st ACCENT Symposium, Urbino, Italy, 2005

S. Walter et al., Mass spectrometric analysis of ice and supercooled cloud residuals during CLACE-3, poster presentation at the EGU General Assembly, Vienna, Austria, 2005

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