

Name of research institute or organization:

**University Mainz, Environmental Geochemistry, Mineralogy**

Title of project:

Tomographic characterization of ice particles

Project leader and team:

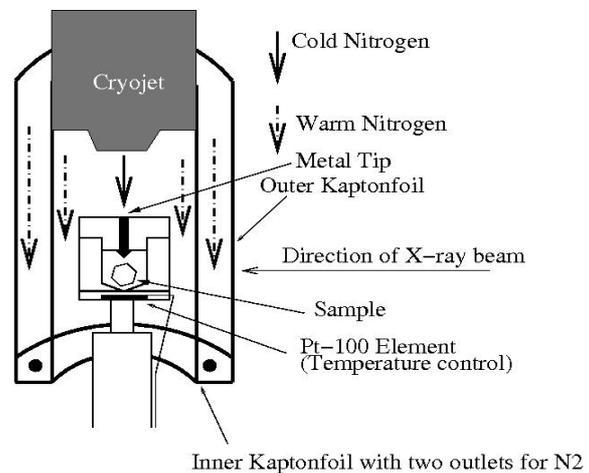
Prof. Michael Kersten, project leader

Dr. Frieder Enzmann, Dr. Thomas Huthwelker, Markus M. Miedaner

Project description:

Synchrotron based micro-tomography is a well established tool for characterizing the external and internal structure of various objects in-situ. During the last year, we developed a new setup for cryo-micro-tomography at the Swiss Light Source (SLS, Paul Scherrer Institute, Villigen, Switzerland) which allows investigating sub-millimeter particles at a spatial resolution of 1.4  $\mu\text{m}$ . Such experiments can be performed at various temperatures between 240 K and 270 K  $\pm$  1K within approximately 90 minutes. Thereby the sample is mounted inside a polyamide cup. This sample holder is cooled by directing a stream of cold nitrogen gas from a CryojetXL (Oxford Instruments) directed onto its top. Additionally, a double walled Kapton-foil cage, mounted onto the Cryojet, surrounds the sample holder to enhance cooling. Since cold surface always bear the risk of icing and condensation, all important parts of the setup are flushed with nitrogen. A detailed description of this setup and its applications is provided in the literature, while a schematic drawing is presented in Figure 1.

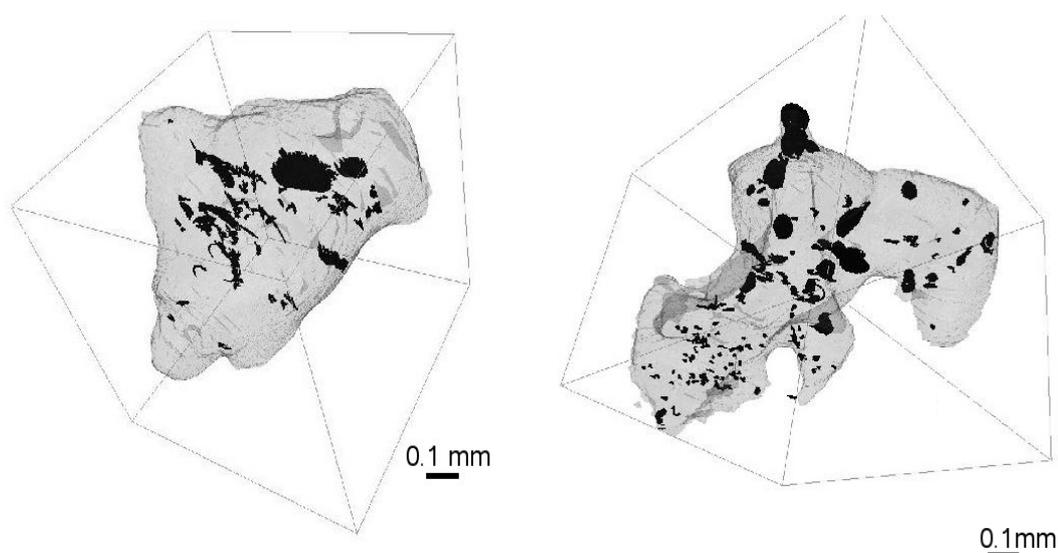
*Figure 1: Schematic drawing of the experimental setup at the SLS Material Science beam line*



Various kinds of ice particles were collected at the Jungfraujoch-Research-Station. Therefore a dewar, partly filled with liquid nitrogen, was placed directly into the precipitation. The samples obtained in such manner were stored in air-tight Zinsser vials and transported on dry ice to the SLS and subsequently stored in liquid nitrogen.

During the experimental shifts scheduled for April 2006, five of the particles collected at the Jungfraujoch were analyzed. Three of them exhibited a strictly dendritic form without any indication of riming nor inclusions detectable at a spatial resolution of 1.4  $\mu\text{m}$ . On the other hand, two particles beard air bubbles.

The voids found in them (cf. Figure 2) had an average volume of  $7017 \mu\text{m}^3$  with an average surface area of  $2837 \mu\text{m}^2$ . The sphericity was found to be 0.49. This indicates a rather elongated morphology. Indeed, Y-shaped pores were found in one of these particles. Nevertheless, all voids were homogeneously distributed all over the samples. The porosity, calculated as the ratio of volume of air divided by the total volume of the particle, was determined as 0.47% and the ratio of inner-to-outer surface ( $0.13\%$ ) was surprisingly high. We estimated the density of the collected particles as  $0.90 \text{ g/cm}^3$  by assuming that no other impurities besides air were trapped. These samples had an average external surface area of  $2.48 \cdot 10^6 \mu\text{m}^2$  while their volume was  $1.59 \cdot 10^8 \mu\text{m}^3$ .



*Figure 2: The distribution and morphology of air bubbles inside the two ice particles from the Jungfrauoch are presented in this figure. The air is colored black, while the ice appears in light gray. Note the completely different shape of both the particles and the air bubbles.*

Metamorphism has undoubtedly occurred to the samples prior to analysis. However, the following three arguments indicate that recrystallization of our samples occurred at a spatial scale that can hardly be detected with our current setup. First, the internal surface area decreases during metamorphism and therefore also the ratio of internal over external surface area. In our samples these values were an order of magnitude higher than those reported for metamorphosed snow packs. Additionally, the presence of Y-shaped voids is typical for fresh samples. As shown in a previous study, such voids transform into spherical inclusions during forced metamorphism. Finally, when atmospheric ice particles like snow recrystallize, they form rather spherical or ellipsoidal bodies. We found dendritic shapes with idiomorphic crystal facets in the majority of our samples. This would not be possible if annealing or recrystallization would have occurred to the samples on a spatial scale large than the experimental resolution.

Since nearly no metamorphism has biased the samples we continue analyzing the inclusions trapped in the particles. No other impurities besides air seem to be trapped in the samples. This may be due to the extremely low concentration and small size of all other impurities present. However, we demonstrated that even at such high

altitudes (3.5 km) ice particles beard inclusions of several micrometers in size and that a detectable degree of riming occurred to them.

Riming and inclusions effect the radiative properties of the particles. In most radiation-models, the assumption is made that the ice particles present are idiomorphic crystals or even spheres. Our samples indicate, that also non-idiomorphic or even polycrystalline particles are present. More data are nevertheless needed to evaluate the percentage of their occurrence.

Knowing the internal and external surface area of the ice particles improves the micro-physical modeling of atmospheric processes, which are often catalyzed by surfaces of ice particles. Detailed measurements allow estimating reservoir sizes and reaction capacities. Furthermore, the curvature of these surface can be extracted from the obtained data sets, which is a key-parameter in describing ice dynamics.

A better knowledge of the real density of atmospheric ice particles improves current meteorological models which assume an ideal density of 0.91 for atmospheric ice particles. As our two examples already indicate, this does not need to be the case for every particle and further particles should be scanned to elucidate the percentage of air bubble bearing particles at the Jungfrauoch.

Finally, further improvements of the spatial resolution are warranted. Obtaining tomographic data at a 10 nm scale would allow answering the question to what extend the porosity of atmospheric ice particles is open or not. Besides, a combination of X-ray tomography and X-ray fluorescence at the same scale (10 nm) would be useful to determine the chemical composition and the exact position of ice condensation nuclei and trapped impurities.

In conclusion, we have developed setup that allows determining air bubbles and other inclusions inside ice particles. Additionally, we showed that ice particles can be sampled at the Jungfrauoch-Research-Station and transported for long distances and stored for two month without detectable metamorphism. But, the obtained data sets need further extensions to provide a more rigid base for the improvement of various micro-physical and meteorological models.

Key words:

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Synchrotron, Tomography, Ice-Particles, Inclusions

Collaborating partners/networks:

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Universität Frankfurt, SFB-641, MPI-Mainz, Paul Scherrer Institut (PSI/SLS)

Scientific publications and public outreach 2006:

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### **Refereed journal articles**

Miedaner, M.M., T. Huthwelker, M. Stampanoni, F. Enzmann, M. Kersten, and M. Ammann, A new setup for synchrotron micro tomography of ice particles and their metamorphism, submitted.

### **Conference papers**

Miedaner, M.M., Huthwelker, T., Enzmann, F., Stampanoni, M., Kersten, M., and M. Amman, 2007, X-ray tomographic characterization of impurities in polycrystalline ice, Royal Chem. Soc. PCICE2006 Proceedings.

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