Name of research institute or organization:

Leibniz-Institut für Troposphärenforschung, Leipzig, Deutschland

Title of project:

Composition analysis of ice particle residuals and ice nuclei combining aerosol mass spectrometry and counterflow virtual impactor technique

Project leader and team:

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Project description:

This research activity is part of the research unit INUIT (research project 2) of the German research foundation DFG. It should contribute to a better understanding of the heterogeneous nucleation of ice particles in middle and lower tropospheric super-cooled clouds. A question closely related to the different heterogeneous ice nucleation mechanisms is the nature of the IN with respect to their chemical composition and microphysical properties. Especially the anthropogenic influence on tropospheric ice formation is hardly known as well as the atmospheric relevance between mineral dust and biogenic aerosol particles. In order to tackle these questions, INUIT co-organized a joint field campaign in real atmospheric mixed-phase clouds at the high alpine research station Jungfraujoch, called CLACE 2013. By former in-situ sampling in mixed-phase clouds at this site the role of lead in ice nucleation was found to be important. The significance of black carbon, that was observed to be enhanced in ice residuals, to serve as IN at the Jungfraujoch is still controversially discussed. In addition, the importance of primary biological particles acting as ice nuclei found to be very high could not yet be confirmed at this site.

The possibility to measure in real atmospheric mixed-phased clouds also allows the separate characterization of ice particle residuals (IPR) and IN identified by an IN counter. For the IPR there exists no direct proof that they actively contribute to ice formation although they are found in real atmospheric ice particles, whereas the particles forming ice in an IN-counter are well-defined but limited by the artificial thermodynamic conditions and heterogeneous nucleation mechanisms that can be simulated in the counter. Thus, the objectives are the physico-chemical characterization of (a) IPR within natural mixed-phase clouds (objective 1) and of (b) IN of ice particles formed in an ice nucleus counter (objective 2) at the high alpine research station Jungfraujoch in the Swiss Alps. The determination of the IPR and IN aerosol properties requires the coupling of counterflow virtual impaction (CVI) techniques with single particle mass spectrometry.

In order to investigate the scientific objectives described above, two different CVI systems were operated at JFJ: For objective 1, the physico-chemical characterization of IPR, the unique Ice-CVI inlet system was setup on the Sphinx platform, which consists of a virtual impactor (VI), a drop pre-impactor (PI) and a standard CVI, respectively. Under mixed-phased cloud conditions this sampler extracts small, freshly produced ice particles from all other solid/liquid components of the cloud by consecutively pre-segregating large ice aggregates, super-cooled drops and interstitial particles. In the PI supercooled drops freeze on the cool impaction plates whereas the small ice particles bounce off and remain in the sampling flow. Inside the Ice-CVI the ice water of the sampled ice particles is evaporated releasing the IPR for analysis. Restricting the collection to ice particle sizes between 5 and 20 µm by means of the combination of VI and standard CVI the IPR can be attributed to the original ice nuclei, because in this size range ice particles grow only by water vapor deposition. The IPR are analyzed within this proposal for number concentration, size distribution and black carbon mass by a condensation particle counter (CPC), an optical particle sizer (OPS), an ultra-high-sensitivity aerosol spectrometer (UHSAS), and a particle

soot absorption photometer (PSAP). For a more detailed chemical characterization of the IPR the single particle mass spectrometer ALABAMA (MPI Mainz) and an impactor for offline electron microscopy (TU Darmstadt) were additionally coupled to the Ice-CVI. For objective 2, the physico-chemical characterization of IN, a pumped CVI (IN-PCVI) was connected downstream the ice nucleus counter FINCH (U Frankfurt). FINCH was connected to the total aerosol inlet, heterogeneously nucleating ice particles on atmospheric aerosol under controlled conditions. The task of the IN-PCVI was to collect and separate these ice particles from non-activated particles and drops and transfer the atmospheric IN to the ALABAMA and the impactor for electron microscopy after evaporating the ice water. Moreover, the IN number concentration and size distribution was determined by CPC, APS and UHSAS.

Up to now, 24 different cloud events during the CLACE 2013 campaign were identified. Exemplarily, the number concentration time series for cloud event #12 (event indicated by the red horizontal line) measured by the CPC, OPS and UHSAS are shown in Fig.1. The small concentration of ice particle residues is significantly higher than the background counts before and after the event. Depending on the occurrence of heterogeneously nucleated ice particles, the IPR number concentration measured by the CPC (measured size range 0.01 – 30 µm) varies between 50 and 400 L^{-1}. The OPS (0.3 – 10 µm) and UHSAS (0.06 – 1 µm) see the same time course of the IPR concentration but on lower density levels due to their limited size sensitivity. Although it is expected that larger aerosol particles are the more efficient IN, the IPR number concentration is dominated by sub-micrometer particles.

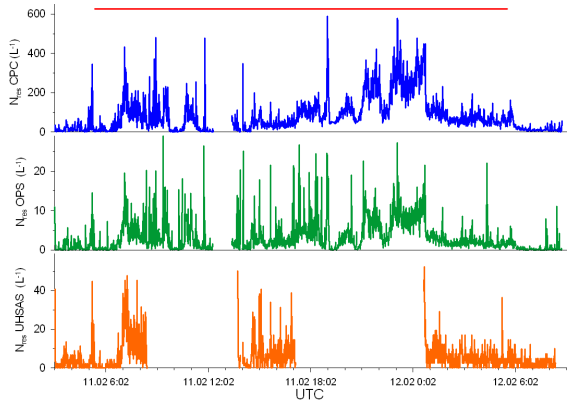

Figure 1. IPR number concentration measured during cloud event #12 with different sensors.

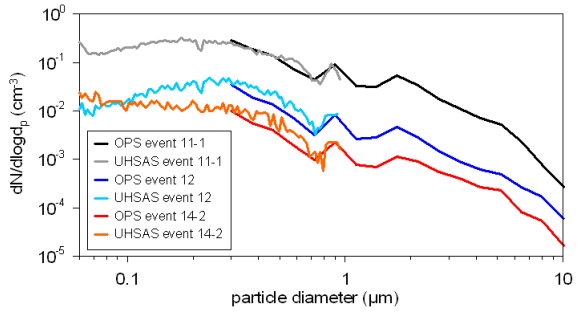

Figure 2. IPR number size distributions derived for three different cloud events.

In Fig.2 the IPR size distributions for three different cloud events derived from the UHSAS and OPS measurements are illustrated. The distributions from both instruments show a good agreement in the overlap region between 0.3 and 1 µm. Overall, the IPR number size distributions are very broad in comparison to background particles which indicates that larger

particles are enriched in the ice particles and thus are preferred to act as ice nuclei. The maxima vary but are in the size range from 0.2 to 0.3 µm.

For the validation of the particle measurements behind the IN-PCVI, the temperature and super-saturation data sets of the IN-counter were analyzed in order to determine time periods of constant conditions. Depending on the operation and thus freezing conditions inside FINCH (especially the ice supersaturation) IN number concentrations between a few and about 500 per litre were detected by the FINCH optics and a CPC behind the IN-PCVI. Especially at lower ice supersaturations the IN number concentration measured by FINCH and the IN-PCVI show a good agreement (Fig.3), indicating that the separation/collection of the ice particles nucleated inside FINCH by the IN-PCVI worked well. The ALABAMA and electron microscopy results show that mineral dust particles often mixed with organic material are the dominant IN.

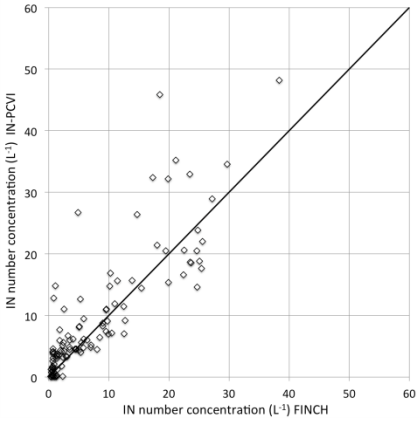

Figure 3. Scatterplot of IN number concentration measured with FINCH and IN-PCVI

The measurements of TROPOS during CLACE 2013 yield the following results so far:

- During the encountered cloud events IPR concentrations were found to be between 50 and more than 1000 L^{-1}
- The IPR number density is substantially larger than the derived IN number density
- IPR number size distributions are very broad, indicating that larger particles are preferred to act as ice nuclei
- The main IPR size modes are found to have their peaks between 0.2 and 0.3 µm.

The next step in the analysis is to determine the contribution of natural ice multiplication processes on the IPR results by means of the size resolved aerosol and ice-particle measurements (latter carried out by collaborating groups). IPR number and size need furthermore to be related to the microphysical and chemical properties of the ambient background aerosol in order to validate the assumption that the IPR have been dominantly served as atmospheric IN. In this context, the IPR results will be compared to the IN-counter based ice nuclei measurements in more detail.

Key words:

Aerosol cloud interactions, mixed-phase clouds, heterogeneous ice nucleation, ice nuclei

Internet data bases:

http://www.tropos.de/en/research/aerosol-cloud-interaction/process-studies-on-small-spatial-and-temporal-scales/aerosol-cloud-interaction/heterogenous-freezing-in-lab-and-field/ice-nulei-in-atmospheric-clouds/

Collaborating partners/networks:

Max Planck Institute for Chemistry, Mainz, Germany
University of Frankfurt, Germany
Technical University of Darmstadt, Germany
KIT Karlsruhe, Germany
Paul Scherrer Institute, Villigen, Switzerland
University of Manchester, United Kingdom

Scientific publications and public outreach 2013:

Conference papers

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