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Mass spectrometric measurements of the chemical composition of residuals from small ice crystals and from supercooled droplets in free tropospheric mixed phase clouds during CLACE 3 - 6

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During the Cloud and Aerosol Characterization Experiments (CLACE 3 - 6) on the High Alpine Research Station Jungfraujoch (Switzerland, 3580 m asl) a novel Ice-CVI (Counterflow Virtual Impactor) was coupled to an Aerodyne Aerosol Mass Spectrometer (AMS), sampling and analyzing the non-refractory submicron particles. The measurements were performed in the late winter months of the years 2004, 2005, 2006, and 2007. A Q-AMS was operated in 2004 and 2005, while a HR-ToF-AMS was operated in 2006 and 2007. In 2006 and 2007, additionally a single particle laser ablation instrument (SPLAT) was employed. The combination of CVI and aerosol mass spectrometer allowed to analyze the residuals of ice nuclei as well as of supercooled cloud droplets, depending on cloud type and CVI operation mode. Within the same clouds, also interstitial aerosol was sampled and compared to the residual particles. Besides long episodes of free tropospheric aerosol, several cloud events were sampled, both within mixed-phase and pure supercooled clouds.

The results show that the submicron aerosol in the free troposphere is composed to about 95% of non-refractive material, dominated by organic and sulfate aerosol. The ice cloud residuals sampled by the CVI show negligible mass concentration in the AMS compared to SMPS data, indicating that preferably refractory particles (mainly mineral dust and black carbon) act as ice nuclei.

The HR-ToF-AMS data show a high contribution of oxygenated organic aerosol (OOA) in the free tropospheric background aerosol, confirming the assumption that photochemical aging converts primary organic aerosol emissions into oxygenated aerosol. The ice cloud residuals, in contrast, contain a small fraction of organic material that is dominated by hydrocarbon-like organic aerosol (HOA). This agrees with the finding that black carbon is also enriched in ice nuclei because HOA and black carbon have similar sources, mainly combustion processes.