

Aerosol-Cloud Interactions in the Lower Free Troposphere as Measured at the High Alpine Research Station Jungfraujoch in Switzerland

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INTRODUCTION

Within the WMO Global Atmosphere Watch (GAW) program continuous measurements of aerosol parameters have been performed at the Jungfraujoch high Alpine site since 1988. This measurement site is located on an exposed mountain col in the Bernese Alps, Switzerland, at 3580 meters altitude. Throughout the year the station is within clouds about 40% of the time. In warm months, the site is influenced by injection of planetary boundary layer air into the free troposphere during sunny afternoons due to thermal convection, while in winter it is usually in the undisturbed free troposphere.

Several intensive field studies, named CLACE (Cloud and Aerosol Characterization Experiment), have been performed in both summer and winter within international collaborations. The goals of these intensive campaigns are:

- A full physical, chemical, and optical characterization of the aerosol at the Jungfraujoch in order to better quantify the direct aerosol effect.
- An investigation of the interaction of aerosol particles with clouds, for a better quantification of the aerosol indirect effect.

The cloud forming processes are studied under different meteorological conditions, with special respect to the aerosol-cloud partitioning in mixed-phase clouds. A matter of special interest is the physical and chemical characterization of ice nuclei.

Another research activity focuses on the investigation of processes responsible for the formation of new particles in the free troposphere.

METHODS

State-of-the-art instrumentation is employed to characterize the aerosol size distribution (Scanning Mobility Particle Sizer, SMPS; Optical Particle Counter, OPC). The University of Manchester and the Max Planck Institute in Mainz operated two Aerodyne Aerosol Mass Spectrometers (AMS) to measure the size segregated chemical composition. Other measured parameters were the hygroscopic properties of the particles (Hygroscopicity Tandem Differential Mobility Analyzer, H-TDMA), cloud microphysics (Particulate Volume Monitor, PVM; Forward Scattering Spectrometer Probe, FSSP; Cloud Particle Imager, CPI), and particle morphology (Environmental Scanning Electron Microscope, ESEM). An Air Ion Spectrometer (AIS) and an outdoor SMPS were also employed. The latter two instruments are especially well suited to measure nanometer sized particles (charged particles and sum of neutral and charged particles, respectively) in order to elucidate their formation mechanisms and rates.

Different inlets are used for these instruments: A heated inlet (25°C) designed to prevent ice build-up and to evaporate cloud particles at an early stage, ensuring that the cloud condensation nuclei and/or ice nuclei are also sampled. This is called the *total* inlet. An *interstitial* inlet operated with a PM2 cyclone impactor removed all cloud droplets and ice crystals from the ambient air. Within a cloud the sampled air thus represents the interstitial (or unactivated) aerosol fraction. In addition, the Institute for Tropospheric Research (IFT) operated a Counterflow Virtual Impactor (CVI, Mertes et al., 2005). The CVI was part of a new prototype sampling system (Ice-CVI) that allows for the separation of small ice particles from

large ice crystals, cloud droplets and interstitial aerosol particles. The extracted ice particles are dried airborne in the system and the remaining residual particles which correspond to the former ice nuclei were analyzed with a variety of different instruments. Differencing the response downstream of the different inlets provides insight in the fractionation of aerosol particles between the cloud phase and the interstitial phase.

RESULTS

The activated number fraction (F_N) is defined as the fraction of the total particle number ($D_p > 100$ nm) that is activated into cloud droplets (obtained from total minus interstitial). Based on more than 900 hours of in-cloud measurements from winter and summer 2004 and winters 2004 and 2005, this activated fraction has been related to several environmental factors such as liquid water content, number concentration of particles, temperature and ice mass fraction (IMF) of the cloud (Verheggen et al., 2006). Measurements show a decrease in the activated number fraction with increasing IMF from $F_N \sim 0.6$ in summer liquid phase clouds (IMF ~ 0) to $F_N < 0.2$ in mixed-phase clouds (IMF > 0.05). This is explained by the Wegener-Bergeron-Findeisen process, which describes the effect of a water vapor flux from liquid droplets to ice crystals. The lower the ambient temperature, the more liquid droplets evaporate and a higher fraction of CCN is released back to the interstitial aerosol phase.

The observed relation between number of ice crystals, particle number concentration and ice mass fraction is currently incorporated into a global climate model (Lohmann and Diehl, 2006). Model runs show that the Wegener-Bergeron-Findeisen mechanism may have a dampening effect on the aerosol indirect effect on climate.

Analyses revealed that the black carbon (BC) component of the ambient aerosol is activated into cloud droplets to the same extent as the accumulation mode aerosol (Cozic et al., 2006). The activated fraction of BC shows the same dependence on the IMF as F_N . Such behavior is not expected for freshly emitted soot particles because they are hydrophobic. The soot particles on the Jungfraujoch experienced aging processes which transformed them into an internally mixed hygroscopic aerosol.

An analysis of the size resolved mass size distributions shows that the ice residuals contain a relatively larger mass contribution from particles larger than 200 nm, suggesting that larger particles (e.g.

mineral dust) preferentially act as ice nuclei. By relating the BC mass downstream of the different inlets to their respective total mass concentration it was found that BC was also enriched in the residual particles, indicating that besides dust BC also acts as potential ice nuclei.

The formation of new particles (nucleation events) was frequently observed during CLACE. The number of particles produced is relatively small in comparison with nucleation events in the planetary boundary layer, but they typically last multiple hours, suggesting that they are regional-scale phenomena. These nucleation events will be analyzed in more detail to reveal information on their formation mechanism and on the rates of nucleation. As an example, nucleation rates of up to $1 \text{ cm}^{-3} \text{ s}^{-1}$ are estimated for one such event.

Keywords: *Cloud/Fog Aerosol Interactions, Atmospheric Nucleation, Measurement Campaigns*

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