The Cloud and Aerosol Characterization Experiment (CLACE) at the Jungfraujoch

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Introduction

The observation of cloud and aerosol particles has been conducted at the JFJ since 1988 and has established the site to be suitable for the long-term monitoring of the FT background aerosol [1,2]. It forms the joint European Global Atmosphere Watch (GAW) baseline station.

With the participation of 8 German, Hungarian, and Swiss institutes, the intensive field campaign CLACE (Cloud and Aerosol Characterization Experiment) took place during February/March 2000 at the high alpine research station Jungfraujoch (JFJ, 3580 m asl, Switzerland). The main goals of CLACE were to examine cloud formation processes under different meteorological conditions, to characterize organic aerosol compounds, to compare particle size distributions measured under different conditions and to determine hygroscopic properties of aerosols.

Clouds have an effect on the earth's energy budget by reflecting the solar radiation back to space. This property is known as the indirect effect of the atmospheric aerosol and has rarely been quantified yet [3]. It is therefore of special interest which particles become activated under which conditions. With the increase of the aerosol concentration caused by human activity, more particles are possibly available for activation. Under the assumption that the amount of water vapor stays constant, more and smaller water droplets are formed than without anthropogenic influence. This will cause an increase of the cloud albedo and thus of the reflection of solar radiation [4].

Experimental

During CLACE, with the instrumentation and expertise of the contributing scientists, the following aerosol parameters were measured simultaneously: number and surface area concentrations, aerosol and cloud droplet number size distributions, aerosol volatility, chemical composition (major ions and organic compounds), particle morphology (scanning electron microscopy and transmission electron microscopy), single particle composition (time of flight mass spectrometry), spectrally resolved light scattering and absorption, water soluble fraction, and hygroscopic growth.

Ambient aerosol was sampled by different inlet systems: A heated inlet, as described in [5], was used to measure the total aerosol. In addition, the interstitial aerosol was sampled by a cascade impactor, which was designed to remove cloud droplets.

Total and interstitial particle size distributions were measured with three different Scanning Mobility Particle Sizers (SMPS, for the diameter range D = 0.02 to 0.8 μm).

One SMPS was operated in the laboratory at approx. 25°C and sampled the aerosol alternately from both inlets. Another SMPS system measured the interstitial aerosol, which passed through a thermodesorber system [6]. A third SMPS system was located outdoors and measured the size distribution at ambient relative humidity (RH) and temperature conditions.

Results

Figure 1 shows a typical example of total and interstitial aerosol size distributions measured at room temperature for a cloud event as well as for a cloudless period. In Fig. 1 (a) the cloud droplet size distribution as measured with a forward scattering spectrometer probe (FSSP) is also given.

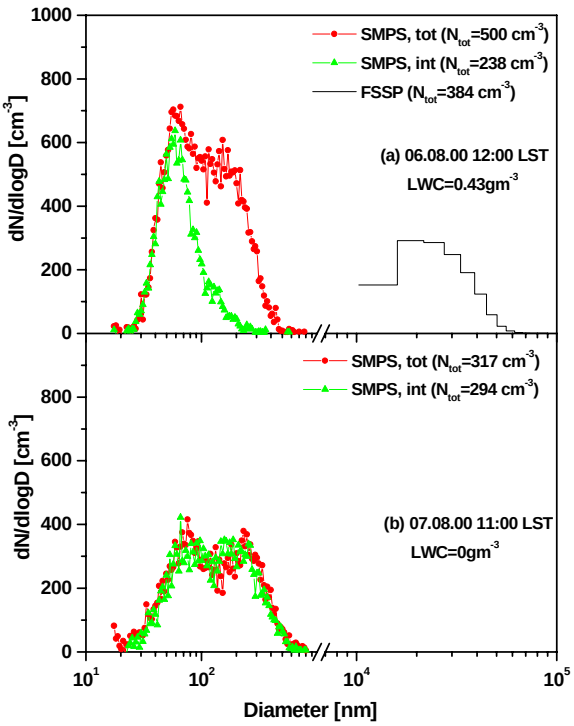

Fig. 1: Averaged size distribution (1-hour averages) of the total (up triangles) and interstitial (circles) aerosol during a cloud event (a) and without cloud (b).

Aitken (20 nm < *D* < 100 nm) and accumulation mode (100 nm < *D* < 800 nm) particles were observed in both cases, with mean diameters for the cloud event of 65 and 150 nm, respectively, and 90 and 200 nm, respectively for the cloudless period. During the cloud, the accumulation mode aerosol was scavenged completely, while almost the whole Aitken mode remained. This can also be seen in the number concentrations, with 500 cm^{-3} for the total and 238 cm^{-3} for the interstitial aerosol. The total droplet concentration of 384 cm^{-3} calculated from the FSSP data shows a reasonable agreement with the one calculated from the difference of the particle concentrations between total and interstitial aerosol (~260 cm^{-3}). An analysis of the whole dataset showed that in dense clouds (liquid water content, LWC > 0.3 g m^{-3}), particles with *D*> 100 nm were activated to cloud droplets.

The capability of an aerosol particle to act as a cloud condensation nucleus depends on its dry size as well as on its hygroscopic properties. Aerosol particles usually contain a substantial amount of water as soon as the relative humidity (RH) has once reached the deliquescence point of an individual compound. Due to the hysteresis phenomenon, the particles stay in a metastable state well below the deliquescence point. As an example, the water content of pure ammonium sulfate is about 25% at RH=50%. Thus, the atmospheric aerosol typically contains a high fraction of water at elevated RH, which significantly influences the particle properties such as its ability to scatter light.

During the campaign, the hygroscopic growth of the particles was measured by means of two hygroscopicity tandem differential mobility analyzers (HTDMA). One instrument performed these measurements at temperatures below 0°C [7].

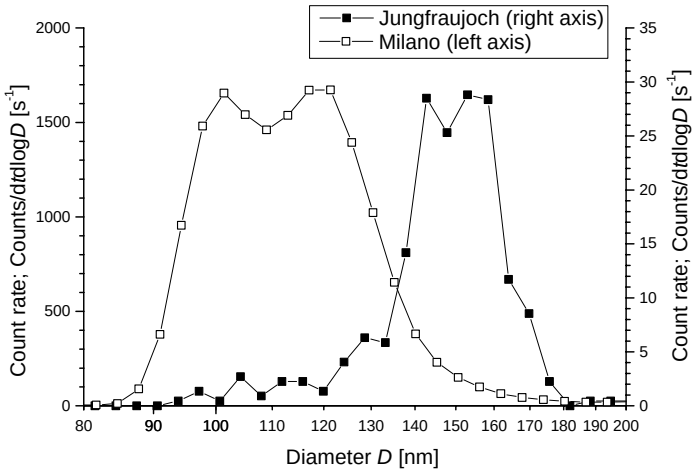

Fig. 2: Size distributions of monodisperse aerosol particles with a (dry) diameter D_0 = 100 nm after exposure to a relative humidity of 90%. (Milano data from [8]).

Figure 2 compares HTDMA data from an area close to major particle sources (Milano, Italy) to the situation on the Jungfraujoch located in the lower free troposphere. The increase of the growth factor is clearly seen, which is due to the deposition of water soluble material on the particles by a number of processes (e.g., coagulation with water soluble particles, (photo)-chemical degradation of the aerosol particle surface, gas to particle conversion (heterogeneous nucleation), adsorption and reaction of gaseous molecules, or cloud processing).

Together with the hygroscopic growth, there are also other factors, which may influence the number size distribution of aerosol particles. Figure 3 shows the 24-h evolution of the particle number concentrations at the Jungfraujoch measured with different systems. On these particular days the station was in clouds until approx. 5 a.m. of Feb. 21. The ambient temperature ranged between –16 and –20°C. The number concentration of the interstitial aerosol measured indoors tracks well with the total aerosol measured via the other inlet by the same system as well as with the total particle number concentration measured by a Condensation Nucleus Counter, CNC. In contrast, the variation of the interstitial aerosol measured outdoors shows two additional peaks that are significantly higher than the level of the interstitial aerosol measured indoors. These two phenomena range over 3-4 hours before the particle concentration drops back to the level of the other variations. During these periods, the outdoor SMPS recorded particle number size distributions with nearly identical modes, but with a particle concentration higher by a factor 3 in the range 20 - 100 nm and by a factor 2 in the range 100 - 400 nm, respectively. Obviously, a significant fraction of the particles is composed of material with a high volatility.

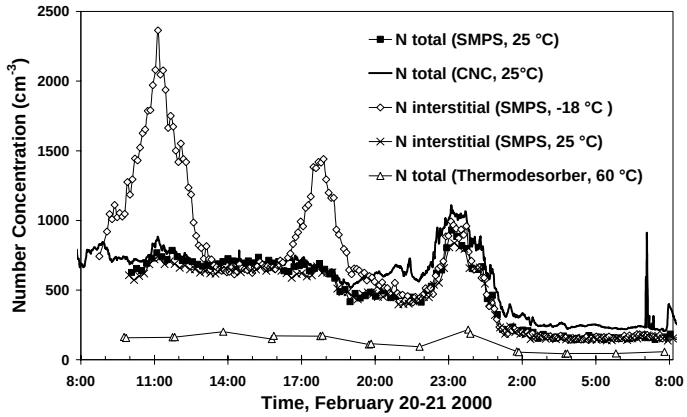

Fig. 3: 24-h variation of the particle number concentration measured by different instruments and inlet systems.

Further evaluation of the data with incorporation of the chemical analysis will show to what extent organic compounds are responsible for these phenomena.

Acknowledgements

We thank the International Foundation Jungfraujoch and Gornergrat and the staff of the high-alpine research station Jungfraujoch for their support. This work was funded by MeteoSchweiz under the auspices of the Global Atmosphere Watch (GAW) Programme of the World Meteorological Organization.

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