Name of research institute or organization:

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Title of project:

The Global Atmosphere Watch Aerosol Program at the Jungfraujoch.

Project leader and team:

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Project description:

Airborne aerosols affect our climate primarily by influencing the atmospheric energy budget through direct and indirect effects. Direct effects refer to the scattering and absorption of radiation and their influence on planetary albedo and the climate system. Indirect effects refer to the increase in available cloud condensation nuclei (CCN) due to an increase in anthropogenic aerosol concentration. This could lead to an increase in cloud droplet number concentration and a decrease in cloud droplet effective radius, when the cloud liquid water content (LWC) remains constant. The resulting cloud droplet spectrum could lead to reduced precipitation and increased cloud lifetime. The overall result would be an increase in cloud albedo which cools the Earth's climate. Despite the uncertainty, it is believed that in regions with high anthropogenic aerosol concentrations, aerosol forcing may be of the same magnitude, but opposite in sign to the combined effect of all greenhouse gases.

The Global Atmosphere Watch (GAW) program is an activity overseen by the World Meteorological Organization (WMO). It is the goal of GAW to ensure long-term measurements in order to detect trends and to develop an understanding of these trends. With respect to aerosols, the objective of GAW is to determine the spatio-temporal distribution of aerosol properties related to climate forcing and air quality up to multi-decadal time scales. Since the atmospheric residence time of aerosol particles is relatively short, a large number of measuring stations are needed. The GAW monitoring network consists of 23 Global (including the Jungfraujoch) and some 300 Regional stations. While Global stations are expected to measure as many of the key variables as possible, the Regional stations generally carry out a smaller set of observations.

The Jungfraujoch aerosol program is among the most complete ones worldwide. The current GAW instrumentation that is continuously run at the Jungfraujoch consists of

- CPC (TSI 3010) Particle number density (particle diameter D_p>10 nm)
- Nephelometer (TSI 3563) Scattering coefficient at various wavelengths
- Aethalometer (AE-31) Absorption coefficient at various wavelengths; black carbon (BC) concentration
- MAAP Absorption coefficient; black carbon (BC) conc.
- Filter packs Aerosol major ionic composition (PM1 and TSP)
- Betameter and HiVol Aerosol mass (PM1 and TSP)

For these measurements, ambient air is sampled via 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.

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. This causes the concentration of pollutants, including the aerosol loading, to be higher in summer than in winter (see Figure 1).

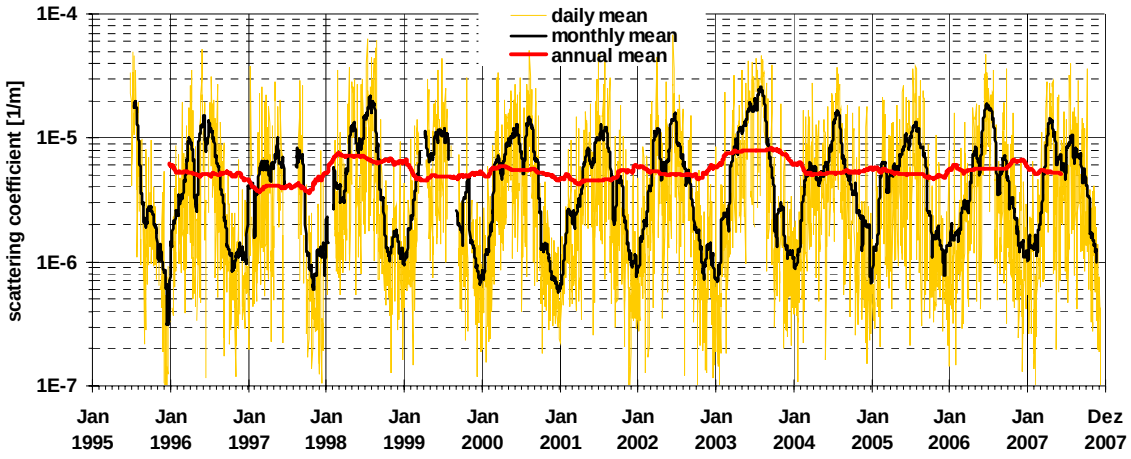

Figure 1. Long-term measurements of the light scattering coefficient (at 550 nm).

This data set was used to analyze the long-term trend of aerosol optical measurements at the Jungfraujoch (Collaud Coen *et al.*, 2007). Since the aerosol variables are approximately lognormally distributed, the seasonal Kendall test and Sen's slope estimator were applied as non-parametric methods to detect the long-term trends for each month. The yearly trend was estimated by a least-mean square fit. The most significant trend is the increase (4-7% yr^{-1}) in light scattering coefficients during the September to December period. The light absorption and backscattering coefficients and the aerosol number concentration also show a positive trend during this time of the year. This increase of the extensive aerosol properties during autumn can tentatively be related either to the general temperature increase over Europe, which lengthens the period when the influence of the PBL is preponderant and increases the injection of PBL air masses into the lower FT over large regions, or to long-range transport from areas with growing economy and increasing air pollution. The hemispheric backscattering fraction and the scattering exponent show an increase in size of small particles during most of the year, whereas the size of large particles remains constant. Generally the summer months at the Jungfraujoch, which are strongly influenced by planetary boundary layer air masses, do not show any long-term trend.

The sixth Cloud and Aerosol Characterization Experiment (CLACE-6) took place in February/March, 2007, with participation from numerous European research groups. This campaign aimed at characterizing aerosol and cloud properties with a particular focus on aerosol-cloud interactions in mixed liquid/ice phase clouds. Three different inlet systems were used the separate and further investigate the physical and chemical properties of the aerosol fractions acting as CCN, IN, or remaining in the interstitial phase in comparison to the total aerosol available. The interstitial and total inlets

operated by PSI selected the interstitial and total (interstitial plus droplet and ice residual particles) aerosol, respectively. The Institute for Tropospheric Research (IfT Leipzig) operated a novel Ice Counterflow Virtual Impactor (Ice-CVI, Mertes *et al*., 2007), which makes it possible to separate residual particles from ice crystals and/or cloud droplets from the remainder of the aerosol. Many different aerosol instruments were operated behind these inlets by the research consortium. In addition to the standard GAW equipment, PSI operated instruments to the aerosol number size distribution (Scanning Mobility Particle Sizer, SMPS; Optical Particle Counter, OPC), the enhancement of light scattering by the aerosol at enhanced relative humidity (humidified Nephelometer), hygroscopic diameter growth factors at enhanced relative humidity (hygroscopicity tandem differential mobility analyzer, HTDMA), the concentration of cloud condensation nuclei (CCN counter) and concentrations of organic and elemental carbon (Sunset Lab OC/EC analyzer).

A first set of cloud condensation nuclei (CCN) concentration measurements at different supersaturations have been conducted in February/March 2007. Parallel total measurements of the total particle concentration (CN, $D > 10$ nm) were used to calculate so-called activation ratios CN/CCN, which is the number fraction of particles having the potential to act as a CCN at a defined supersaturation. The results of these measurements are illustrated in Figure 2, indicating that the activation ratios are highly variable in time. Further parallel hygroscopicity, number size distribution and CCN measurements are planned in order to determine whether the observed variability of activation ratios is caused to a greater extent by the variability of the number size distribution or the particle hygroscopicity (composition).

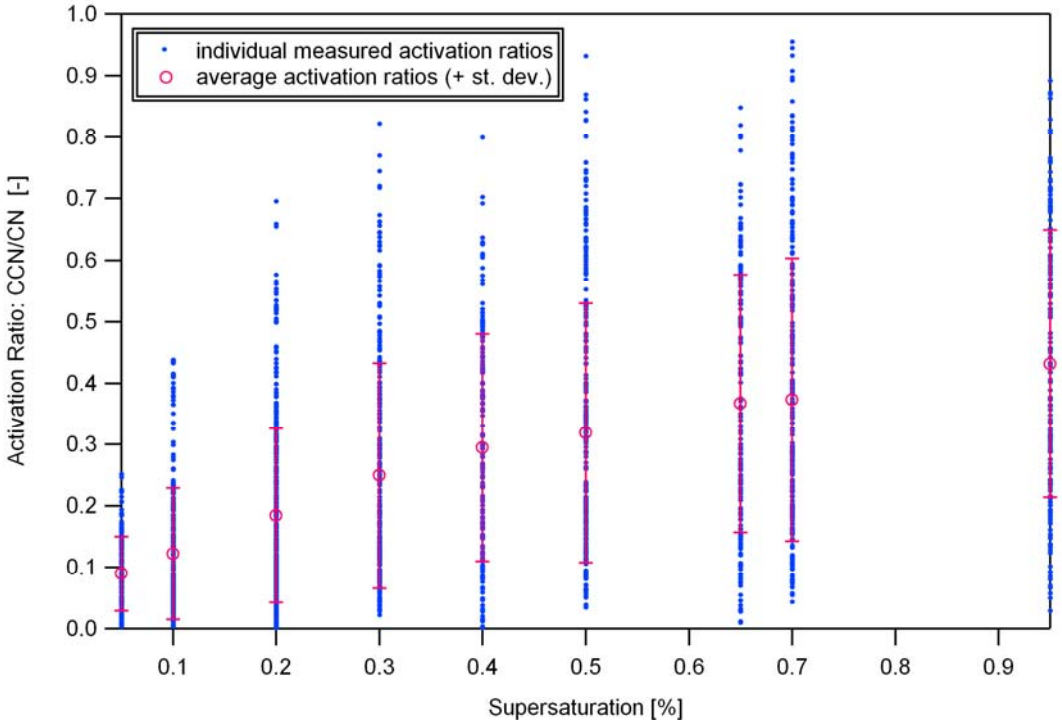

Figure 2. CCN activation ratio relative to the total number of particles with D>10 nm measured during the CLACE-6 campaign in February/March 2007.

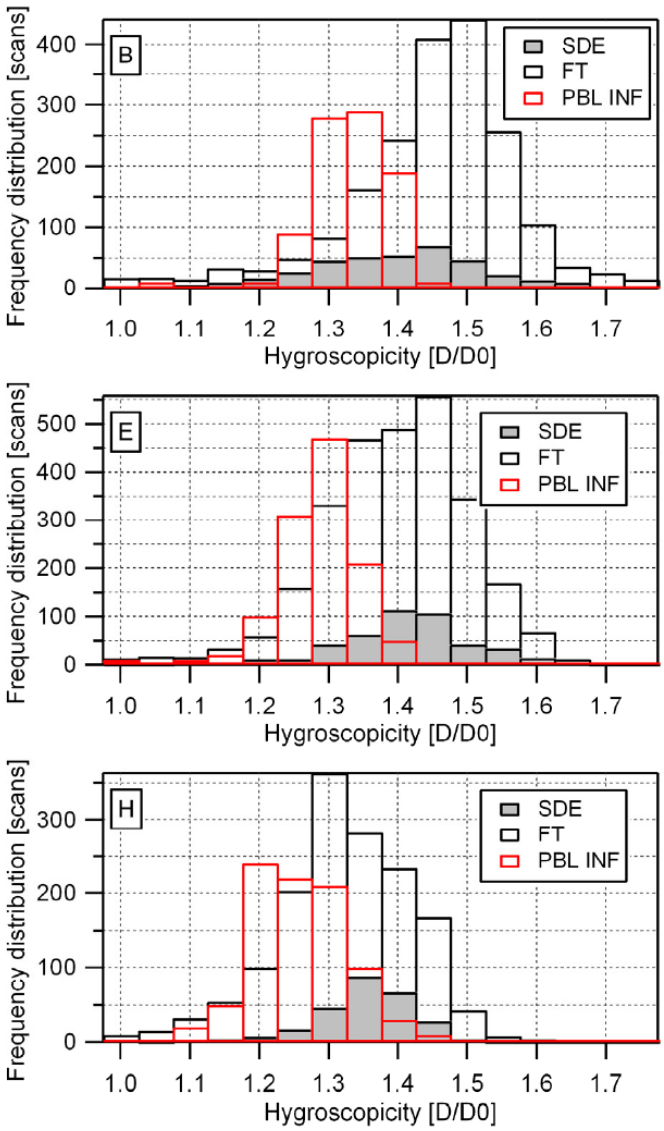

Figure 3. Histogram of mean hygroscopic growth factors at 85% relative humidity of aerosol particles with dry diameter D_0 = 250, 200, 50 nm from top to bottom (Sjogren et al., 2007).

Figure 3 shows histograms of the mean hygroscopic diameter growth factor at 85% relative humidity of aerosol particles with different dry sizes group by air mass type. Mean hygroscopic growth factors in free tropospheric (FT) air masses (black) are larger than in planetary boundary layer (PBL) influenced air masses consistently across all investigated dry sizes, thus indicating that atmospheric aging processes result in a general trend towards larger hygroscopicity. Externally mixed non-hygroscopic particles are often found at D_0 = 250 nm during Sahara dust events (SDE), thus leading to reduced mean growth factors (grey bars in top panel). This is not observed for smaller diameters, as can be seen from similar mean growth factor histograms for FT and SDE air masses (middle and bottom panel).

Another important activity in 2007 was the analysis of the complex interactions of aerosol particles with cloud hydrometeors. An important result obtained during former CLACE campaigns is that the partitioning of aerosol particles to the cloud phase is strongly dependent on the relative fraction of ice in the cloud (Verheggen *et al.*, 2007). Figure 4 shows that the scavenged volume fraction (derived from the size

distribution measurements and defined as $(V_{tot}-V_{int})/V_{tot}$ is about 60% in liquid clouds. The fraction of scavenged particles decreases with increasing cloud ice mass fraction (IMF) to reach $F_{Scav}<10\%$ in mixed-phase clouds with IMF > 0.2. This can be explained by the Wegener- Bergeron-Findeisen process, which describes the effect of a water vapour flux from liquid droplets to ice crystals. The formation of ice during the early stages of cloud development could have prevented additional particles from activating by quickly lowering the supersaturation. This is also due to the difference in vapour pressure over ice and liquid. Figure 4b shows that black carbon (BC) mass is scavenged into the cloud phase to the same extent as the total aerosol (Cozic *et al.*, 2007). Such behaviour is not expected for freshly emitted soot particles because they are hydrophobic. Most soot particles on the Jungfraujoch experienced aging processes which transformed them into an internally mixed hygroscopic aerosol. The scavenged fraction was increased in liquid cloud with increasing liquid water content (LWC) up to a plateau of 60% and decreased with increasing particle (or BC) concentration since there is an increased competition for the available water vapour.

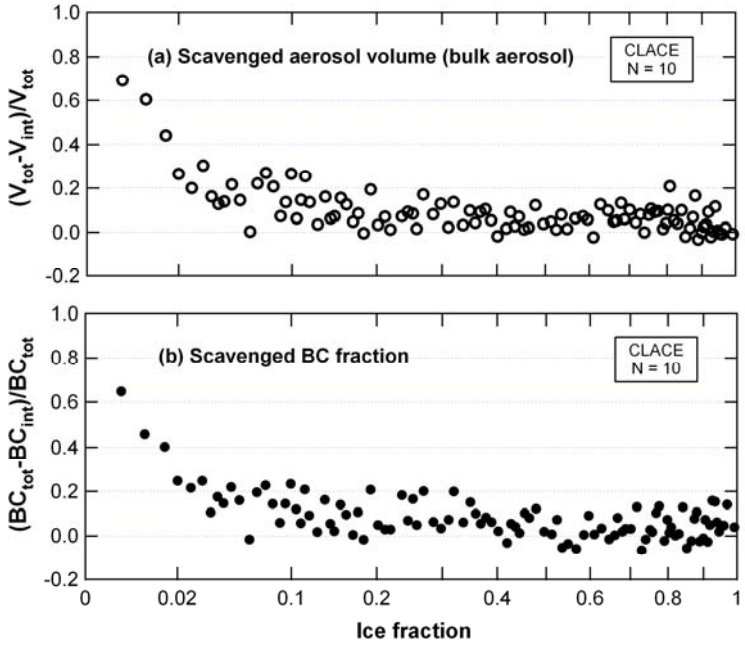

Figure 4. Scavenged fraction of aerosol volume (a) and black carbon mass (b) vs. the ice mass fraction of mixed phase clouds. Each point represents an average of 100 min of measurement.

The Ice-CVI allowed for the sampling and subsequent analysis of residual particles in small ice crystals (ice residuals). The chemical composition of ice residuals was found to be remarkably different from the total aerosol. Comparison of SMPS and AMS data confirms findings that this aerosol is composed to about 95% of non-refractory material (vaporized at 600°C) (Cozic *et al.*, 2008b). Ice residuals show a significantly different signature: Ice crystal residuals sampled by the Ice-CVI show a negligible mass concentration of non-refractory material as measured by the aerosol mass spectrometer compared to the SMPS derived mass, indicating that preferably refractory (i.e. non-volatile, such as BC or mineral dust) particles act as ice nuclei. An analysis of the size resolved mass size distributions shows that the ice residuals

experience a relatively larger mass contribution from particles larger than 300 nm, suggesting that larger particles (e.g. mineral dust) preferentially act as ice nuclei.

The BC mass fraction behind the total inlet ($BC_{tot}/(V_{tot}\cdot\rho$, assuming an aerosol density of ρ =1.5 g/cm^3)) was compared to the BC mass fraction behind the Ice-CVI ($BC_{cvi}/V_{cvi}\cdot\rho$, assuming a BC density of ρ = 2 g/cm^3). It can be observed that BC behind the total inlet represents between 3 to 10% of the total aerosol mass whereas in ice residuals it represents from 4 up to 60% of the aerosol mass. Points above the 1:1 line have a larger BC fraction in the ice residuals than in the total aerosol, while below the reverse is true. Figure 5 thus shows that most of the time, BC is enriched in the ice residuals compared to the total aerosol. On average, the BC mass fraction was 5% for the total aerosol and 27% for the ice particle residuals.

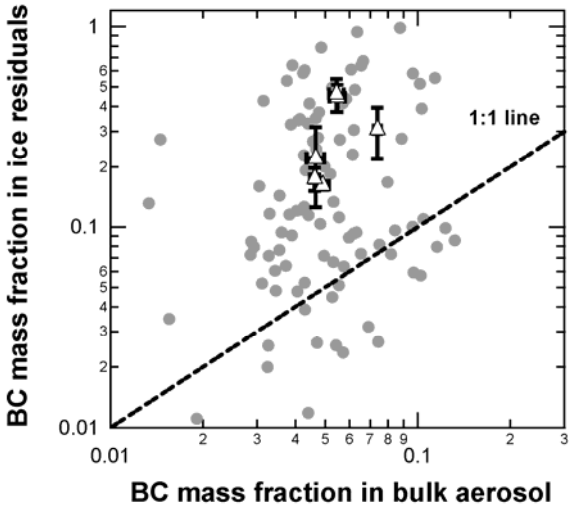

Figure 5: Comparison of the BC mass fraction in the ice residual phase with the corresponding fraction in the total aerosol phase in-cloud. Filled circles represent hourly averaged data whereas triangular points (and their errors) present averages over 6 individual cloud periods. Note that the scales are logarithmic. From Cozic et al. (2008a).

These measurements indicate that besides dust, BC (or compounds associated with BC) also acts as potential ice nuclei. If generally true, this means that in addition to an indirect effect on liquid cloud formation, there is an indirect aerosol effect via glaciation of clouds. This result is highly important for climate since BC has a predominately anthropogenic origin.

Key words:

Atmospheric aerosol particles, aerosol-cloud interactions, aerosol climatic effects, radiative forcing, cloud condensation nuclei, hygroscopic growth, light scattering

Internet data bases:

http://www.psi.ch/gaw
http://www.psi.ch/lac
http://aerosolforschung.web.psi.ch
http://www.meteoschweiz.ch/web/en/climate/global_climate_monitoring/GAW_CH_Allg/GAW-aerosol.html

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Prof. M. Kulmala, Department of Physics, University of Helsinki, Helsinki, Finland

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