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 now the Jungfraujoch, which was upgraded from a Regional to a Global station) 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).

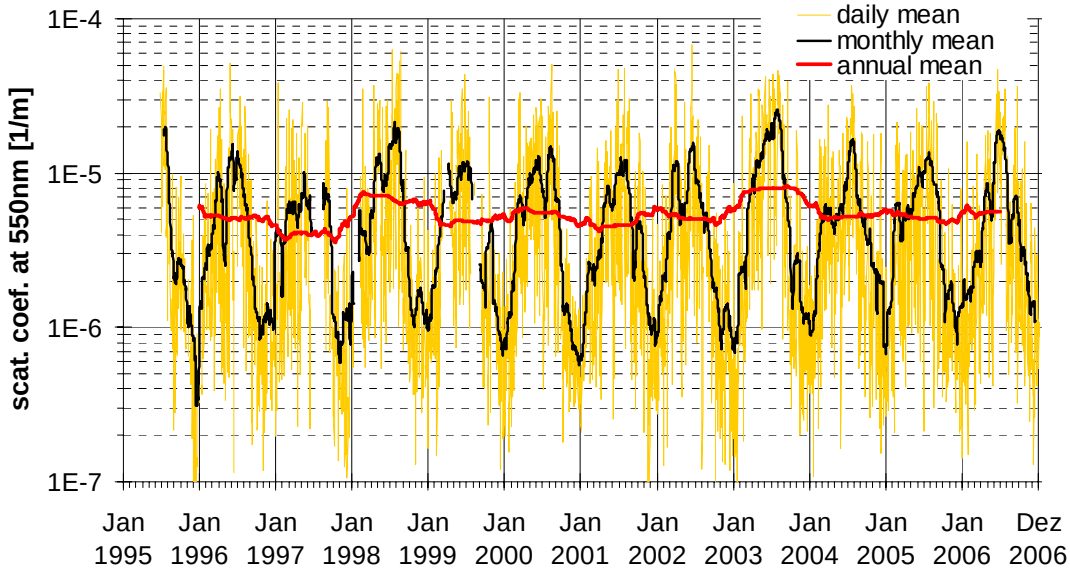

Figure 1. Long-term measurements of the light scattering coefficient (at 550 nm).

This data set was used to analyze the long-term trend of aerosol optical measurements at the Jungfraujoch (Collaud Coen *et al.*, submitted to J. Geophys. Res.). 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 fifth Cloud and Aerosol Characterization Experiment (CLACE 5) took place in February/March, 2006, with participation from different research groups. During this campaign, additional instrumentation was employed to characterize the aerosol size distribution (Scanning Mobility Particle Sizer, SMPS; Optical Particle Counter, OPC). The Max Plank Institute in Mainz (MPI) operated two Aerodyne Aerosol Mass Spectrometers (AMS) to measure the size segregated chemical composition. New in comparison with previous CLACE campaigns were the employment of an ice nucleus counter and an ATOF-MS for single particle analysis of ice nuclei by ETH Zürich.

Two additional inlets were used for these instruments: An *interstitial* inlet operated with a PM2 cyclone impactor removed all cloud particles 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*., submitted to Environ. Sci. Technol.). 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. The activated fraction 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 winter 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 of the cloud. These analyses revealed that the black carbon (BC) component of the ambient aerosol is activated into cloud droplets to the same extent as the bulk aerosol. Such a behaviour is not expected for freshly emitted soot particles because they have a hydrophobic nature. The soot particles on the Jungfraujoch experienced aging processes which transformed them into an internally mixed hygroscopic aerosol (Cozic *et al*., 2006).

During wintertime this activated fraction is generally low (below 20%), because the presence of ice crystals causes liquid droplets to evaporate, thus transforming cloud droplets back into interstitial aerosol particles, as described by the Wegener-Bergeron-Findeisen process. When the cloud exists almost exclusively of liquid droplets (i.e. ice mass fraction approaching zero), the activated fraction increases to similar values as those encountered in summer (approximately 50%) (Verheggen *et al.*, submitted to J. Geophys. Res.). This is shown in Figure 2.

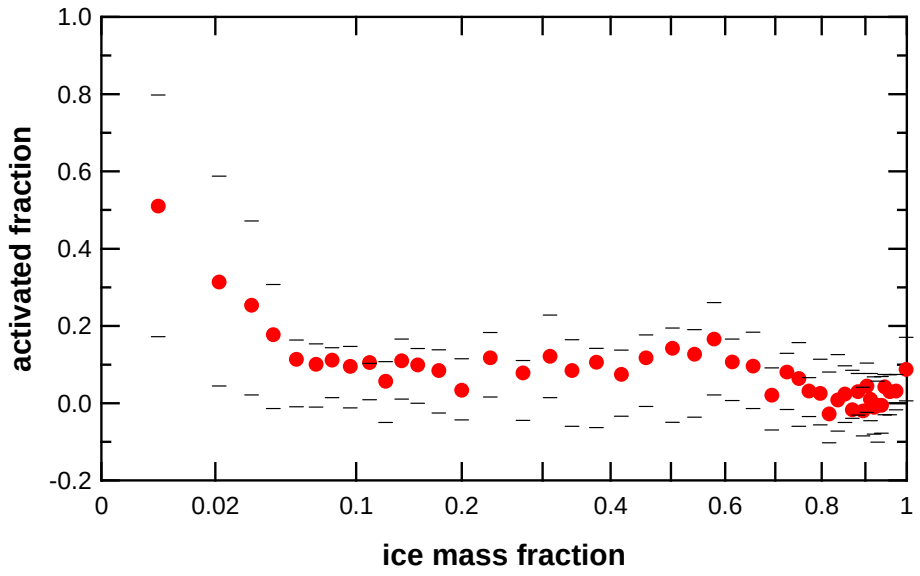

Figure 2. Activated fraction of particles as a function of ice mass fraction in the cloud. Circles denote the average, while horizontal stripes denote the 25 and 75 percentile values.

A similar analysis was performed for the partitioning of the black carbon (BC) mass concentration in mixed phase clouds. The BC mass concentration experiences a similar trend as the one observed for the bulk aerosol at low and intermediate ice mass fractions (Figure 2). A significant difference is found for the scavenged BC fraction at high ice mass fraction: The scavenged BC fraction is ~10% at ice mass fractions > 0.9 whereas the scavenged fraction of the bulk aerosol volume decreases to near-zero (see Figure 2). This indicates that the BC mass concentration is enriched in the ice phase compared to the bulk aerosol. This hypothesis is corroborated by the comparison of the BC mass fraction measured in the ice residuals with the BC mass fraction in the bulk aerosol (Figure 3). The fact that most points are located above the 1:1 line indicates that BC containing particles are enriched in the ice residuals (Cozic *et al.*, manuscript in preparation). This result is highly important for climate modeling since BC has a predominately anthropogenic origin.

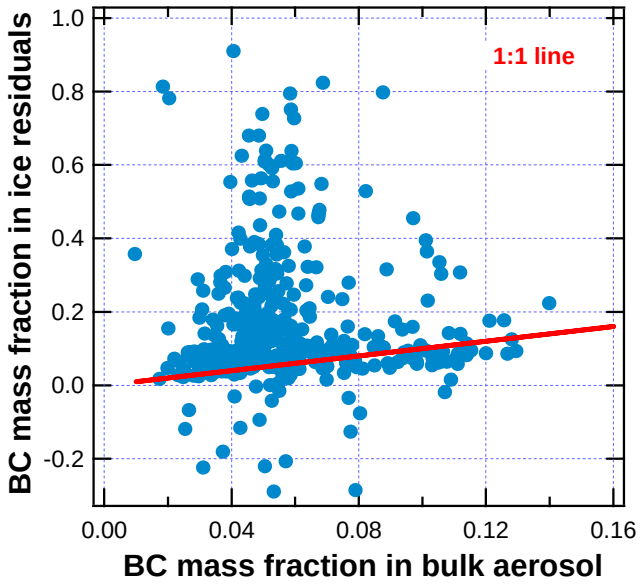

Figure 3. Simultaneously measured BC mass fraction for ice residuals and for the bulk aerosol. The BC mass fraction is defined as the ratio of the particle BC mass concentration to total aerosol mass concentration.

Incorporation of the observed relation between number of ice crystals, particle number concentration and ice mass fraction into a global climate model suggests that the Wegener-Bergeron-Findeisen mechanism may have a dampening effect on the indirect effect of aerosols on climate (Weingartner *et al*. (2006), submitted).

Key words:

Atmospheric aerosol particles, aerosol-cloud interactions, aerosol climatic effects, radiative forcing, cloud condensation nuclei, hygroscopic growth, particle nucleation

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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Scientific publications and public outreach 2006:

Refereed journal articles

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