Name of research institute or organization:

Laboratory of Atmospheric Chemistry, Paul Scherrer Institut

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 is believed to change the cloud droplet number concentration for a constant cloud liquid water content (LWC), and the resulting increase in cloud albedo influences the Earth's radiation budget. Cloud lifetimes and precipitation frequencies are also thought to be affected. 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 22 Global 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.

According to the recommendations of GAW's Scientific Advisory Group (SAG) for Aerosols, Regional stations should measure the optical depth, light scattering and absorption coefficient, the mass concentration and major chemical components in two size fractions. At Global stations, a larger number of measurements are envisaged. These include the Regional parameters list and in addition, the light scattering and hemispheric backscattering coefficients at various wavelengths, aerosol number concentration, cloud condensation nuclei (CCN) concentration at 0.5% supersaturation, and diffuse, global and direct solar radiation. Additional parameters, such as the aerosol size distribution, detailed size fractionated chemical composition, dependence of aerosol properties on relative humidity, CCN concentration at various supersaturations, and the vertical distribution of aerosol properties should be measured intermittently at Global stations. Data are delivered to the World Data Centre for Aerosols (WDCA, located in Ispra, Italy) using the NARSTO data format. The Institute for Tropospheric Research in Leipzig has agreed to host a World Calibration

Centre (WCC) for physical aerosol parameters, while a host for the calibration of chemical parameters still must be located.

The Jungfraujoch aerosol program is among the most complete ones worldwide. The parameters that are measured continuously are available on-line at http://www.psi.ch/gaw. As an example, Figure 1 gives the temporal evolution of the scattering coefficient, measured with a nephelometer at three wavelengths. The seasonal variation with summer maxima and winter minima are clearly visible, and are explained by the seasonal variability of thermal convection. The extraordinarily hot summer 2003 is also reflected in the highest values of the scattering coefficient since the start of the measurements, which is explained by the fact that fair weather favours vertical transport by thermal convection, which affects the magnitude of the mixing of polluted planetary boundary layer air into the free troposphere .

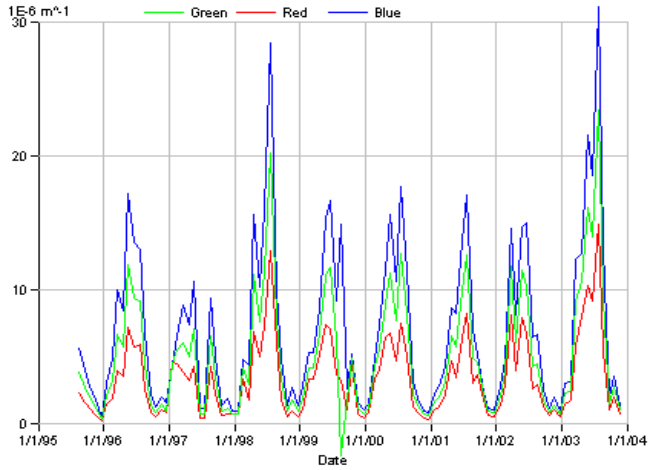

Figure 1. Screenshot from http://www.psi.ch/gaw showing the temporal evolution of the scattering coefficient measured by a three-wavelength nephelometer (at λ = 450, 550 and 700 nm).

During dedicated extensive field campaigns, important questions such as the hygroscopic growth of aerosol particles are addressed. The particles hygroscopic properties play a crucial role in air quality, acid deposition, biochemical cycles, visibility reduction, and the formation of clouds and precipitation. The hygroscopic growth factor is defined as D/D_o, where D is the particle diameter at a specified high relative humidity (typically 85 to 90%) and D_o is the reference diameter under dry conditions. At the Jungfraujoch temperatures are typically below 0°C. At these temperatures, semi-volatile compounds (such as nitrate or lower-molecular-weight organics) may be adsorbed and considerably alter aerosol hygroscopic properties. To minimize artefacts due to volatilization, aerosol hygroscopic behaviour must be measured at ambient conditions. It was therefore the goal to develop an instrument capable of measuring the hygroscopic growth at these low temperatures. This so-called low-temperature hygroscopicity tandem differential mobility analyzer (H-TDMA) selects a narrow aerosol size range from a polydisperse aerosol under dry conditions using a differential mobility analyzer (DMA). Particles are then humidified to a specified high relative humidity (RH) and the new size distribution is then measured with a second DMA at this particular RH. The new H-TDMA is capable of measuring the particles hygroscopic growth factor down to a temperature of –10°C i.e., typical of ambient temperatures at the Jungfraujoch. Such

measurements at sub-zero temperatures are thus representative of ambient conditions, as the aerosol is not heated to room temperature before analysis.

In the experiment in July 2002 the UMIST group (H. Coe, K. Bower, R. Alfarra) simultaneously operated an Aerodyne Aerosol Mass Spectrometer (AMS), which enabled the determination of the size segregated mass loadings of volatile and semi-volatile chemical components (e.g. sulphate, nitrate, ammonium and organic components).

The H-TDMA was deployed in two field experiments at the Jungfraujoch. Hygroscopic growth factors at T = -10°C were measured at D_o = 50, 100 and 250 nm. Figure 2 shows two typical examples of measured humidograms of D_o = 100 nm particles measured with the H-TDMA during winter 2000 and summer 2002. On average, higher growth factors are observed during winter than in summer. The increased hygroscopicity during winter compared to summer is explained by the different meteorological situation: during summer the station is influenced by the planetary boundary layer whereas in winter it is predominantly located in the free troposphere, suggesting that atmospheric aging processes in the free troposphere lead to a substantial increase in particle hygroscopicity. The curves in Figure 2 are characterized by a continuous increase of D/D_o with increasing RH. This behaviour is different from many pure inorganic salts which experience a clear phase transition (dry-wet), marked by a sudden increase in the growth factor. It implies that at the JFJ the particles are present as liquid droplets over a broad RH range. It is hypothesized that this behaviour is caused by the organic fraction of the mixed particles.

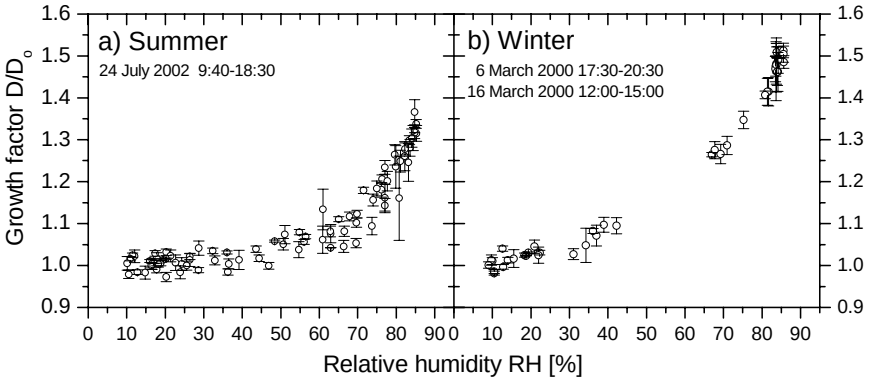

Figure 2: Typical humidograms of D_o = 100 nm particles.

During the campaigns, the H-TDMA was also operated at a constant high RH. As an example, Figure 3 shows the temporal evolution of D/D_o at RH = 85% for particles with dry diameters of D_o = 250 nm. In addition, the chemical composition of the particles as determined with aerosol mass spectrometry (AMS) and black carbon (BC) measurements was used to calculate theoretically expected growth factors. Growth factors of D/D_o (inorganics) = 1.52, D/D_o (organics) = 1.1, and D/D_o (BC) = 1.0 at RH = 85% were assumed for these compounds, and the Zdanovskii-Stokes-Robinson (ZSR) relation (Gysel et al. 2003) was used to describe the behaviour of mixed particles according to their composition. The temporal variability of hygroscopic growth factors is mainly a result of varying fractions of organic/inorganic mass. The measured growth factors and their temporal variability were well predicted with this straightforward hygroscopic model.

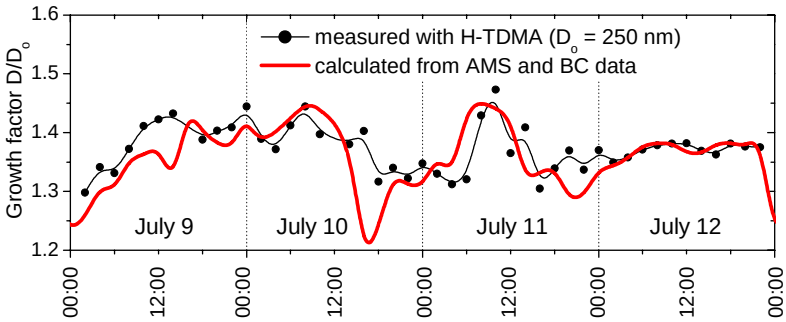

Figure 3. Temporal evolution of measured hygroscopic growth factors at RH = 85.5 ± 0.3% and growth factors calculated from the measured chemical composition.

Another activity during the year 2003 was the development of a new method for the determination of Saharan dust events and to study the occurrence of mineral aerosol at the JFJ. This work was done in collaboration with MeteoSwiss. It was found that due to the large size of mineral aerosol and to their chemical composition, the single scattering albedo ω_0 shows a deviation of the usual inverse wavelength dependence during Saharan dust event (SDE). This change in the sign of the single scattering exponent turns out to be a simple but powerful means for detecting SDE.

The single scattering albedo ω_0 denotes the proportion of the light extinction due to scattering: $\omega_0 = \sigma_{SP}/(\sigma_{SP} + \sigma_{AP})$, where σ_{SP} is the scattering coefficient and σ_{AP} the absorption coefficient. It can thus be calculated from the scattering and absorption coefficients measured at several wavelengths continuously measured within the GAW aerosol monitoring program at the JFJ since March 2001. For this analysis all the coefficients were fitted with a power law dependence on the wavelength λ (σ = constant$\cdot\lambda^{-\alpha}$). This wavelength dependence allows for the identification of characteristic features associated with SDEs (Collaud-Coen et al., 2003).

Usually the single scattering albedo decreases with increasing wavelengths so that its exponent $\alpha_{\omega 0}$ is positive. During SDEs, the ω_0 wavelength dependence is however inversed. The resulting negative $\alpha_{\omega 0}$ (Figure 4) is due to the large size of mineral aerosols, which induces a reduced wavelength dependence of the scattering coefficient. The σ_{SP} exponent, which is usually measured between 1 and 2.5, is clearly shifted below 1 during SDE. Similarly a somewhat larger wavelength dependence of the absorption coefficient (α_{AP} increases up to 1.5, while its normal value is about 1) is linked to absorbing components in the Saharan dust (such as hematite). Consequently, the resulting $\alpha_{\omega 0}$ becomes clearly negative during SDE with values usually falling between -0.1 and -0.5. Therefore, the sign of $\alpha_{\omega 0}$ allows the determination of SDEs at the JFJ with an hourly time resolution.

In 92% of all cases, SDE detected by this new method are corroborated by filter coloration, back-trajectory analysis, satellite measurements or a combination of these methods. The single scattering albedo method is however the most sensitive method to detect SDE.

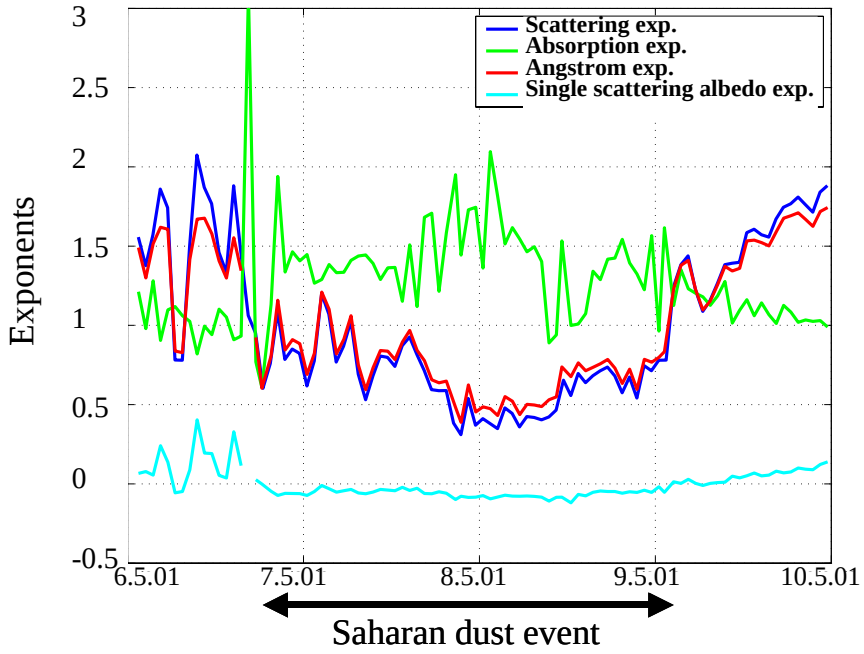

Figure 4: Exponents of the scattering, absorption and extinction coefficients, along with the single scattering albedo exponent during a SDE (6 – 10 May 2001).

According to this 22-month study, SDE occur at the JFJ about 20 times each year and last between a few hours and a week. SDE are more frequent and last longer during the March-June and the October-November period, although some events also occur during the summer (Figure 5).

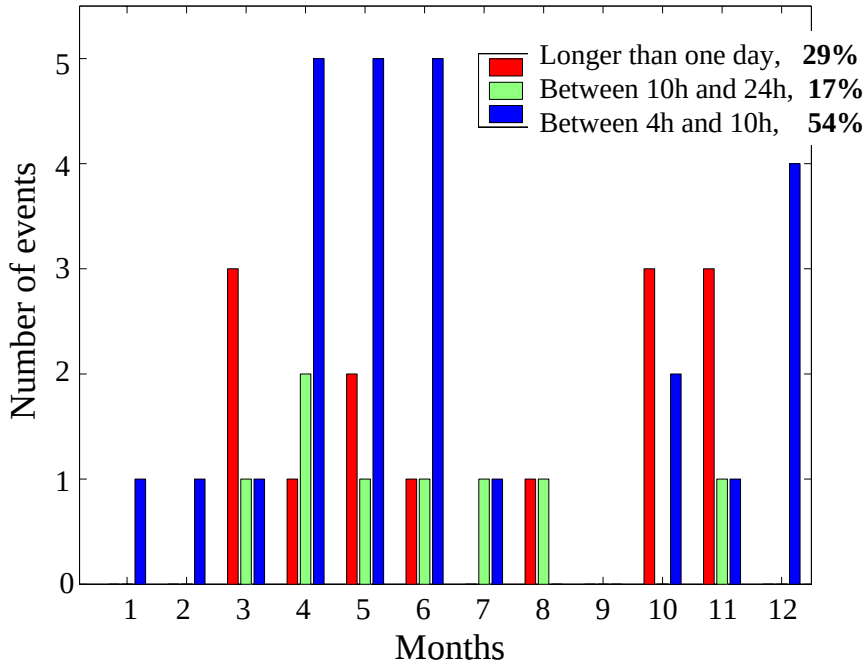

Figure 5: Climatology of the number of Saharan dust events classified by their duration for the March 2001 to December 2002 period.

Key words:

Aerosol particle, cloud condensation nuclei, direct and indirect aerosol effect, radiative forcing, hygroscopic growth

Internet data bases:

http://www.psi.ch/gaw

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Dr. H. Coe, University of Manchester Institute of Science and Technology (UMIST), Atmospheric Physics, Manchester, England

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