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 leaders 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 the SAG, Regional stations should measure the optical depth, light scattering coefficient, the mass concentration and major chemical components in two size fractions. Those stations wanting to add aerosol number concentrations for health effects are advised to do so. 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, the light absorption coefficient, 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.

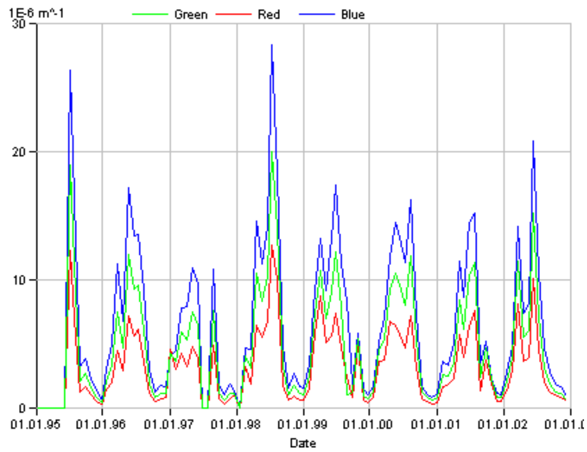

Figure 1. Screenshot from http://www.psi.ch/gaw showing the temporal evolution of the scattering coefficient measured by a three-wavelength nephelometer (at λ = 450, 550 and 700 nm).

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. 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 behavior 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. A detailed description of the so-called low-temperature hygroscopicity tandem differential mobility analyzer (H-TDMA) is found in Weingartner et al. (2002) and Gysel et al. (2002). Briefly, this instrument 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.

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 humidograms measured on two different days during the winter campaign (Weingartner et al., 2002). The acquisition of the spectra in Figure 3 over the range 20% < RH < 80% took only 3 hours on each of the two days.

It is interesting to note that the humidograms are characterized by a continuous increase of D/D_o as a function of RH - no distinct deliquescence behavior is observed during these increasing RH scans which means that the particles were always present in a liquid state in the H-TDMA. It is well known that multi-component aerosols may exhibit hysteresis behavior different from that of pure salts, i.e. the deliquescence relative humidity DRH of mixed salts is always lower than the DRH of the individual salts in the particle. A RH of <10% in the first DMA was obviously not low enough to dry the particles completely. The solid line in Figure 2 is an empirical model. This model fits the data well and can be used to extrapolate the hygroscopic growth to RH = 90% yielding D/D_o = 1.55, 1.62, and 1.67 for D_o = 50, 100, and 250 nm, respectively. The lower growth factor at D_o = 50 nm is mainly due to the Kelvin effect.

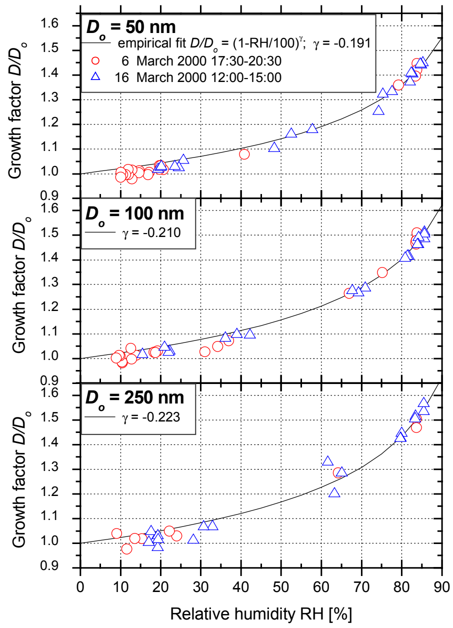

Figure 2. Humidograms of D_o = 50, 100 and 250 nm particles measured at T = -10°C during increasing RH scans at the high-alpine site Jungfraujoch. The greater scatter for the D_o = 250 nm particles is due to very low number concentrations. The data points were acquired with a temporal resolution of 5 min.

Figure 3 shows the hygroscopic growth distribution for three different aerosol types, i.e., for an urban aerosol (Milan), as well as for the Jungfraujoch during the winter and the summer campaign. The Milan aerosol exhibits a bimodal distribution of the growth curve, with a less- and more-hygroscopic mode (see, e.g. Baltensperger et al., 2002). In contrast, the Jungfraujoch is characterized by a narrow monomodal growth distribution. This implies that the particles in the observed size range were to a large extent internally mixed with regards to their hygroscopic and chemical properties. It can also be seen that the winter aerosol at the Jungfraujoch presents the highest hygroscopicity, followed by the summer Jungfraujoch aerosol, and the Milan aerosol, which is close to the aerosol sources. This indicates that atmospheric aging processes lead to a substantial increase in particle hygroscopicity.

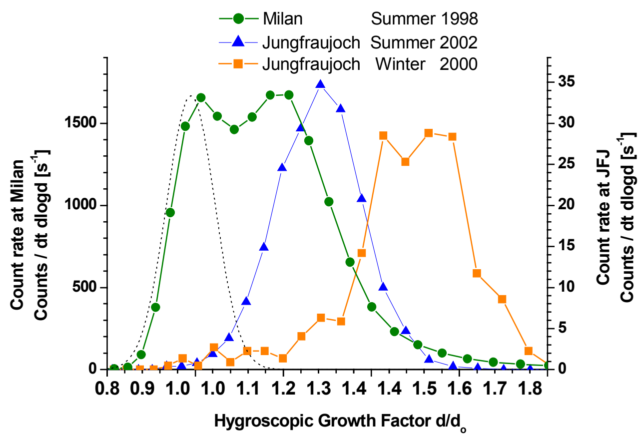

Figure 3. Typical examples of measured hygroscopic growth distribution at RH = 85% in Milan and at the Jungfraujoch. The dotted line is the distribution of the originally dry, monodisperse aerosol particles with a modal diameter of D_o = 100 nm (data from Weingartner et al. (2002) and Baltensperger et al. (2002)).

The measured hygroscopic properties are also important for the continuously measured GAW aerosol parameters. Since these in situ measurements are performed by sampling through an inlet tube into the laboratory at room temperature (typically 25°C) and thus dry conditions (RH < 10%), the measured aerosol properties may differ from the properties determined at ambient conditions (with temperatures typically < 0°C and RH > 40%). This bias was quantified in Nessler et al. (2003): As an example, Figure 4 shows number size distributions measured at indoor and outdoor conditions together with the corresponding values obtained by correcting the indoor values according to the outdoor relative humidity and resulting hygroscopic growth factors (Figure 2). It is seen that this correction is quite significant for enhanced RH.

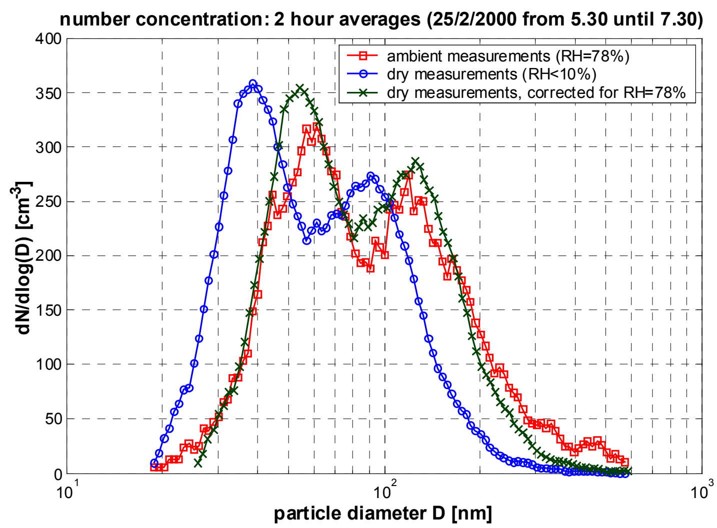

Figure 4. Measured and corrected particle number size distributions at the Jungfraujoch.

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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