Name of research institute or organization:

Laboratory of Atmospheric Chemistry, Paul Scherrer Institute

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 by PSI consists of

- CPC (TSI 3010) Particle number density (particle diameter D_p>10 nm)
- Nephelometer (TSI 3563) Scattering coefficient at various wavelengths
- Aetalometer (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).

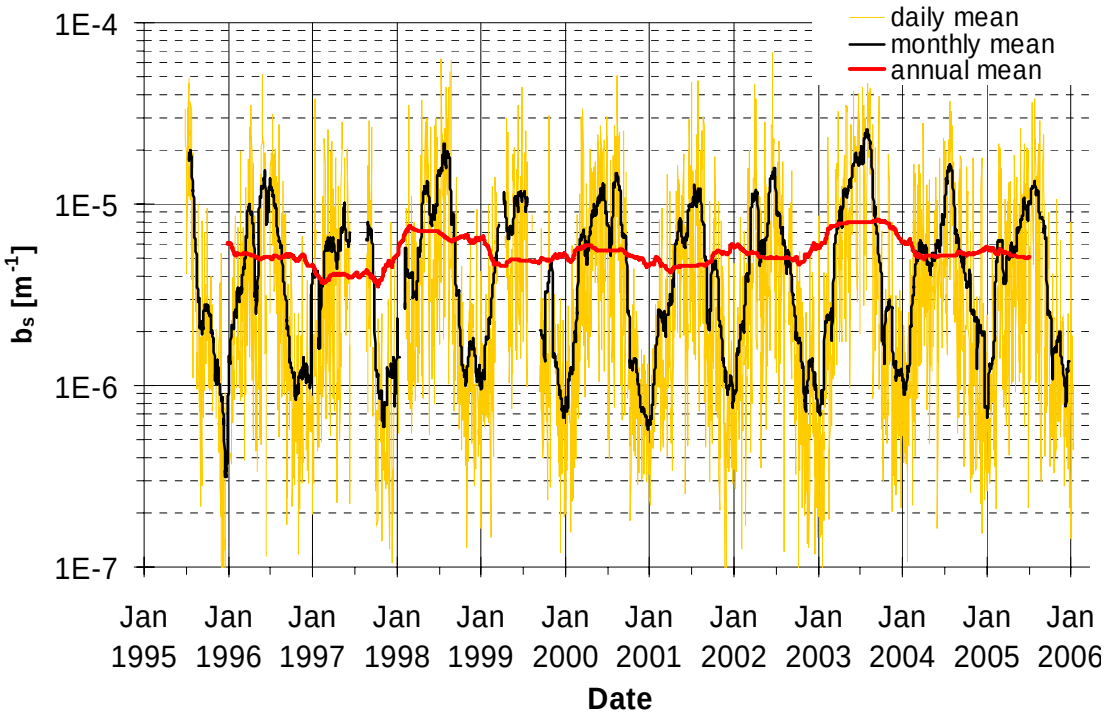

Figure 1. Long-term measurements of the light scattering coefficient bs (at 550 nm).

The data set was used to determine long-term trends for each month and for meteorological seasons (Collaud Coen et al., in preparation). The most significant trend is the increase (2-4% per year) of the aerosol light scattering coefficients at 450, 550 and 700 nm. This autumn increase can be explained by a greater background aerosol load, which relates to long-range transport of air masses, and can be compared to the similar increase of background ozone concentration in autumn and winter at high elevation sites (Ordòñez et al., in preparation). In general, the summer months, which are strongly influenced by the PBL, do not show any significant long-term trend. It seems therefore that the measured decrease of anthropogenic aerosol emissions in Europe is compensated by other effects on a larger scale.

The fourth Cloud and Aerosol Characterization Experiment (CLACE 4) took place from February 15 to March 15, 2005, with participation from ten 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 University of Manchester (UMIST) and the Max Plank Institute in Mainz (MPI) operated two Aerodyne Aerosol Mass Spectrometers (AMS) to measure the size segregated chemical composition. Other measured parameters were the particles hygroscopic properties (Hygroscopicity Tandem

Differential Mobility Analyzer, H-TDMA), cloud microphysics (Particulate Volume Monitor, PVM; Forward Scattering Spectrometer Probe, FSSP; Cloud Particle Imager, CPI), and particle morphology (Environmental Scanning Electron Microscope, ESEM).

New in comparison with previous CLACE campaigns was the contribution of more chemical analyses (several GC/MS techniques, TEM), and the operation of an Air Ion Spectrometer (AIS) and of an outdoor SMPS. The latter two instruments are especially well suited to measure very small particles (charged particles and sum of neutral and charged particles, respectively) in order to elucidate their formation mechanisms and rates. In addition, a nano-SMPS was being operated for 10 days, on loan from TSI Inc., Germany.

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). 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 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, manuscript in preparation).

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%) (Weingartner et al., Verheggen et al., manuscripts in preparation). This is shown in Figure 2.

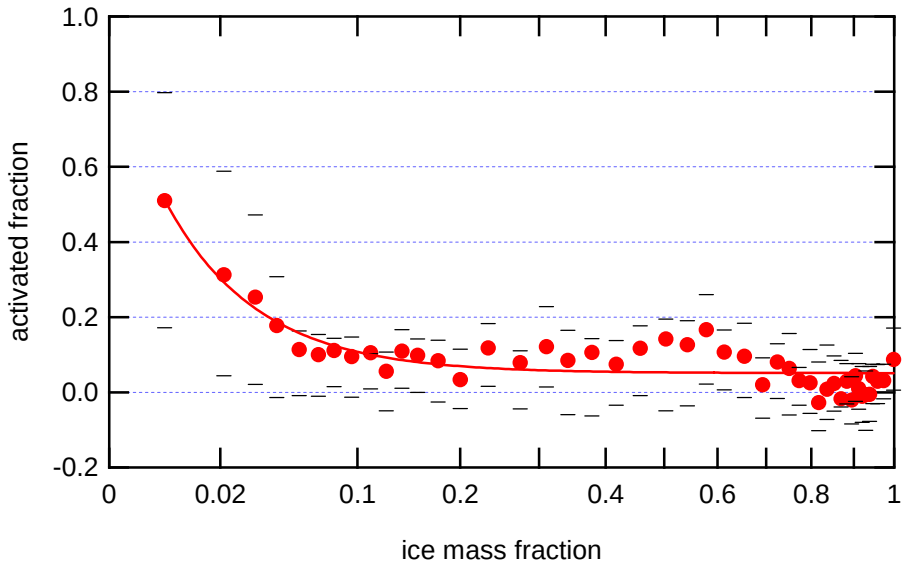

Figure 2. Activated fraction of particles as a function of ice mass fraction in the cloud. Circles denote the average, while horizontal stripes denote the 25 and 75 percentile values.

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., manuscript in preparation).

Nucleation events were frequently observed during CLACE 4. The number of particles produced is relatively small in comparison with nucleation events in the Planetary Boundary Layer, but they typically last multiple hours, suggesting that they are regional-scale phenomena. These nucleation events will be analyzed in more detail to reveal information on their formation mechanism and on the rates of nucleation. As an example, nucleation rates of up to 1 cm^{-3} s^{-1} are estimated for one such event.

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

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