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Title of project:

The Global Atmosphere Watch Aerosol Program at the Jungfraujoch

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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 the 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 leads 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 leads to reduced precipitation and increased cloud lifetime. The overall result in the global atmosphere 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 compared 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 27 global (including the Jungfraujoch) and about 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. By the end of 2012 it has reached 18 years of continuous measurements. Table 1 shows the current GAW instrumentation that is continuously running at the Jungfraujoch. 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 inlet is called the *total* inlet.

Hourly and daily averages are calculated and the data is visualized in real-time for different time periods in the internet, see
http://aerosolforschung.web.psi.ch/onlinedata or
https://gawrtl.psi.ch.

*Table 1: Current GAW aerosol instrumentation*

| Instrument | Measured parameter |
|---|---|
| CPC (TSI 3010 or 3772) | Particle number density (particle diameter $D_p$>10 nm) |
| Nephelometer (TSI 3563) | Scattering coefficient at three wavelengths |
| Aethalometer (AE-31) | Absorption coefficient at seven wavelengths; equivalent black carbon (BC) concentration |
| MAAP | Absorption coefficient at one wavelength; equivalent black carbon (BC) concentration |
| Filter packs | Aerosol major ionic composition (PM1 and TSP) |
| Betameter and HiVol[1)] | Aerosol mass, PM1 and TSP[1)] |

[1)] measured by EMPA

Since 2008, additional aerosol parameters have been continuously measured at the Jungfraujoch (see Table 2). These measurements were conducted as part of the "GAW plus" program and three EU Projects (EUSAAR, EUCAARI, and ACTRIS).

*Table 2: Additional aerosol instrumentation operated in 2012*

| Instrument | Measured parameter | Measurement period |
|---|---|---|
| SMPS, OPC | Particle number size distribution, $D_p$ = 20 - 22'500 nm | 10.1.2008 - ongoing |
| CCNC | Number concentration of cloud condensation nuclei | 10.1.2008 - ongoing |

The number size distribution of aerosol particles plays a key role for direct and indirect aerosol climate interactions. A scanning particle mobility sizer (SMPS) and an optical particle counter (OPC) were installed at the JFJ in January 2008. These instruments have been fully operational since then and provide a complete size distribution from 20 nm to 20 μm.

The cloud condensation nuclei counter (CCNC) exposes ambient aerosol particles to a defined water supersaturation (SS, in the range between SS = 0.12-1.18%) and measures the concentration of cloud droplets that were activated at this SS. This instrument was installed in January 2008 and has been running since then. It provides valuable information on the variation, absolute value and SS dependence of the CCN concentration (Jurányi et al., 2011). Figure 1 shows the temporal variation of the measured CCN concentration from 2008 to 2012. Since December 2011 the CCNC is also part of the ACTRIS (Aerosols, Clouds, and Trace gases Research Infra Structure) network. This required a standardization of the supersaturation range and thus was changed to 0.1 to 1.0 %.

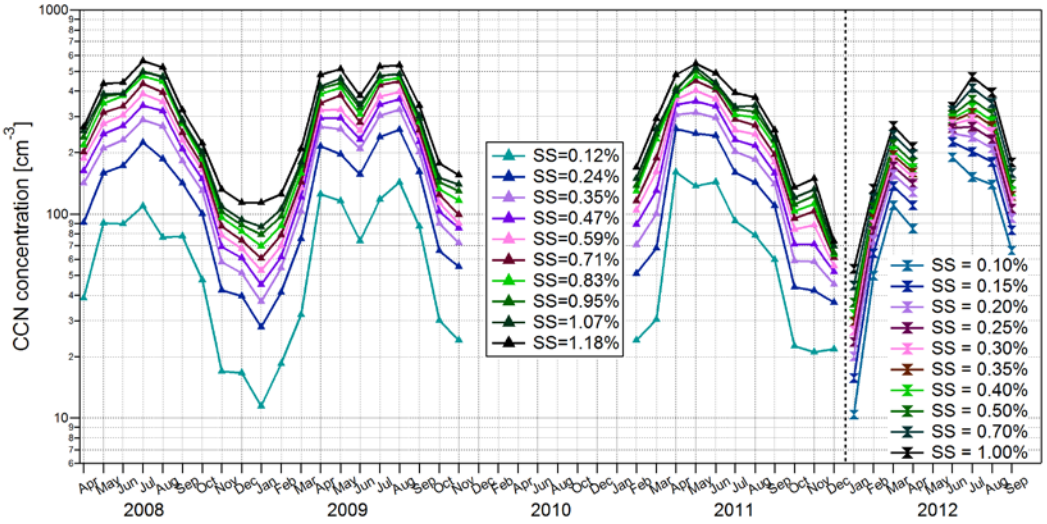


***Fig. 1:*** *Time series of CCN concentrations from April 2008 to December 2012 at 10 different supersaturations. In December 2011 the supersaturation settings were changed in order to comply with the new standards set by ACTRIS. The gap in the time series from December 2009 to January 2011 is due to the commitment of the CCNC in another campaign (MEGAPOLI in Paris) and due to a servicing of the instrument at the manufacturer. The gap in May 2012 is due to a flow problem within the CCNC.*

**<u>Angular aerosol scattering properties measured during one year at the Jungfraujoch</u>**

The angular distribution of the aerosol light scattering was measured during one year at the Jungfraujoch using a novel instrument (POLARNEPH, Aurora 4000, Ecotech Inc., Australia, see Fig. 2). This instrument measures the scattering coefficient for distinct angles using a high-precision backscatter shutter at three wavelengths. The aerosol phase function and the asymmetry parameter can be retrieved, which are important for aerosol climate forcing calculations. In a first step, the measurements at λ=450 nm were compared to the standard TSI nephelometer, which has been operated at the JFJ since 1998. Figure 2 reveals a good agreement between both instruments, which is a positive finding with respect to quality assurance issues.

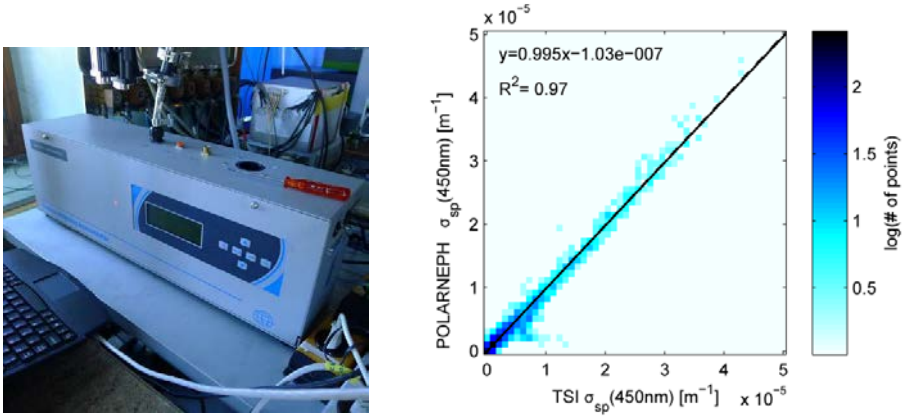


***Fig. 2:*** *Left Panel: The POLARNEPH Aurora 4000 installed at the Jungfraujoch. Right Panel: The aerosol scattering coefficient (at wavelength of 450 nm) of the POLARNEPH vs. the TSI nephelometer (GAW). The solid black line represents an orthogonal fit.*

The asymmetry parameter *g* is defined as the intensity-weighted average of the cosine of the scattering angle. It values range from +1 (all light scattered completely to the forward scattering region) and -1 (all light scattering completely backwards). It is an important factor in radiative transfer calculations which are used to estimate the radiative forcing of anthropogenic aerosols and thus also the climate effects of aerosols. While approximations exist to derive *g* from standard TSI nephelometer measurements, the POLARNEPH allows

measuring the asymmetry parameter directly. Figure 3 shows the time series of the derived asymmetry parameter at the JFJ from the TSI and the novel POLARNEPH. Clear differences between the different approximations and the direct measurement can be observed and ongoing research will clarify the reasons for theses discrepancies.

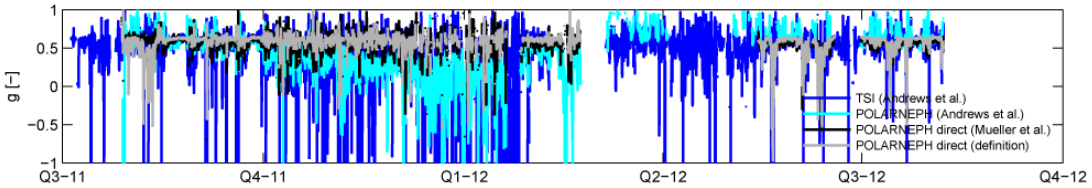


***Fig. 3:*** *The asymmetry parameter g measured at the Jungfraujoch from July 2011 to August 2012. Different algorithms and schemes have been applied to the measurements of the TSI and POLARNEPH instrument (see legend).*

It is also planned to use the recorded data for intensive closure studies using the other recorded in-situ data (size distribution, absorption and chemical data) and Mie theory and discrete dipole approximation. Furthermore, this nephelometer intercomparison is a first step in the scheduled replacement of the TSI 3563 instrument with the Ecotech Aurora 3000 instrument.

**The CLACE field campaigns: Investigation of effective peak supersaturations in liquid-phase clouds**

An important aerosol parameter for climate models is the critical supersaturation (described by Köhler theory), at which a particle forms a cloud droplet. This parameter depends on the particle's dry size and chemical composition. At ambient air conditions, the prevailing supersaturation determines the activation diameter of the aerosol particles (i.e. the diameter at which size a particle forms a cloud droplet). The highest supersaturation that a particle experiences in an ambient cloud, leading to a cloud droplet forming from that particle, is the so-called effective peak supersaturation ($SS_{peak}$).

Since 2000, several Cloud and Aerosol Characterization Experiments (CLACE) have been conducted at the Jungfraujoch. In order to determine $SS_{peak}$ in ambient clouds, the ambient activation diameter (diameter where 50% of the particles are activated to cloud droplets; $D_{50}$) was retrieved from dry particle number size distribution measurements of the total and the interstitial (non-activated) aerosol. Two different inlet systems were used to retrieve the aerosol number concentration of (1) the total aerosol including the nuclei of the hydrometeors ($N_{tot}$) and (2) the non-activated (interstitial aerosols; $N_{int}$) ones. The difference of these two numbers leads to the number of cloud condensation nuclei (CCN) that were activated at the prevailing ambient air conditions. To retrieve the ambient $D_{50}$, the activated fraction (($N_{tot}$-$N_{int}$)/$N_{tot}$ ) was calculated. The Köhler theory consists of two different laws: Raoult's law and Kelvin's law. The former is relying on the hygroscopic properties of the aerosol particles. To summarize Raoult's law with one parameter, Petters and Kreidenweis (2007) introduced the hygroscopicity parameter $\kappa$. To retrieve the prevailing ambient $\kappa$ parameter, the activation diameter was compared to those retrieved from a cloud condensation nuclei (CCN) counter measuring at various controlled supersaturations. Therewith, the effective peak supersaturation can be calculated depending on the following parameters: $D_{50}$, $\kappa$ and the temperature where the activation of an aerosol to a cloud droplet occurs.

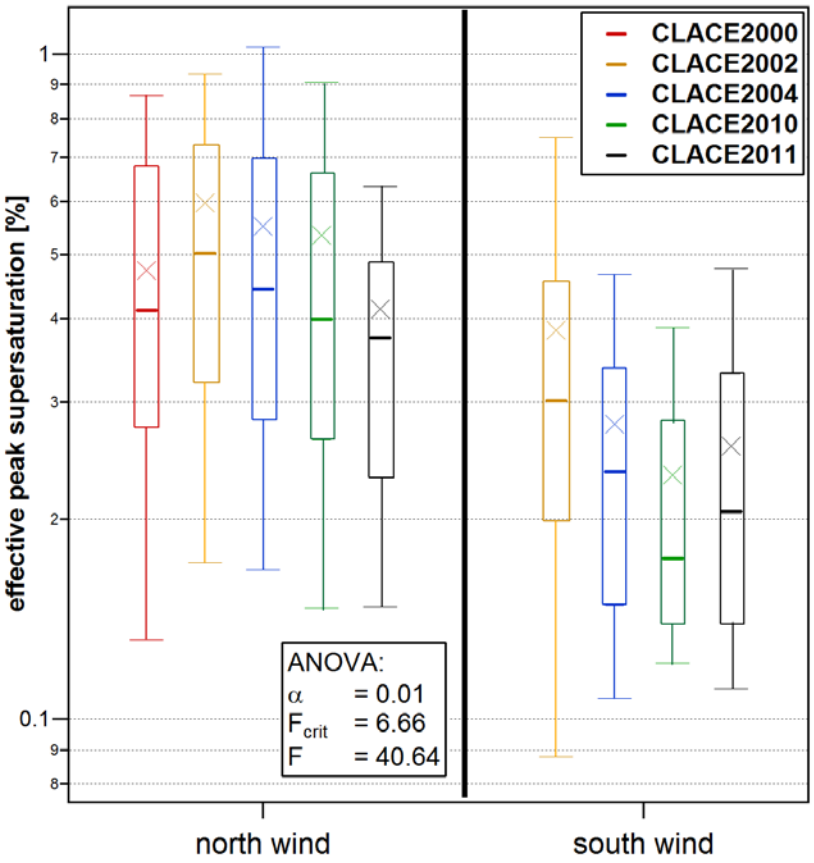


***Fig. 4:*** *Effective peak supersaturations for the two main wind fields present at the Jungfraujoch during several CLACE campaigns. North wind is classified as a horizontal wind direction between 270° to 90° and south wind in a range of 90° to 270° (Hammer et al., in prep.).*

Figure 4 shows the results of $SS_{peak}$ values for all performed CLACE campaigns during summer seasons. A range of $SS_{peak}$ values between 0.09% (10$^{th}$ percentile) and 1.62% (90$^{th}$ percentile) has been observed during several CLACE campaigns. While air masses coming from the North showed a wide range of values, $SS_{peak}$ values for air masses coming from the South were more constant at around 0.2%. This can most likely be explained by different topography from south and north of the Jungfraujoch causing different updraft velocities. While the south side of the Jungfraujoch has a rather smooth topography (Aletsch glacier) resulting in relatively low orographically induced updraft velocities, the north side is characterized by steep rock walls, with more turbulent wind conditions and high updraft velocities. The difference of $SS_{peak}$ values between the two different wind conditions is statistically significant at $\alpha$=0.01 with a variance analyses (ANOVA). Currently, the influence of particle number concentration and size distribution on the $SS_{peak}$ as a function of updraft velocity is being investigated in detail with a cloud box model.

**Aerosol decadal trends: In-situ optical measurements and number concentration at GAW, IMPROVE and ACTRIS stations**

Ground-based, in-situ measurements placed in areas away from emission sources are most suited for studying the atmospheric spatial and temporal variability of aerosol properties as well as climate relevant changes and trends in the atmospheric composition of background air masses. Indication of trends in atmospheric composition is essential, not only for our knowledge of global to regional cycling of atmospheric constituents and natural and anthropogenic changes, but also to validate past and present emission inventories, and to test validity of models at different scales.

In two recent studies (Asmi et al., 2013, Collaud Coen et al., 2013), an analysis of in-situ aerosol trends has been performed within the framework of the WMO-GAW program, using quality-controlled information provided by the NOAA-affiliated monitoring network, the EMEP and EUSAAR/ACTRIS EU-based Research Infrastructure, and the US IMPROVE network, to provide indications of long-term changes in several climate-relevant aerosol variables. Long-term (> 10 years) aerosol measurement sites in the Northern Hemisphere and Antarctica allow analyzing the trends of the aerosol number concentration, the light scattering, backscattering, and absorption coefficients as well as of the derived light scattering Ångström exponent (å) and backscatter fraction (b).

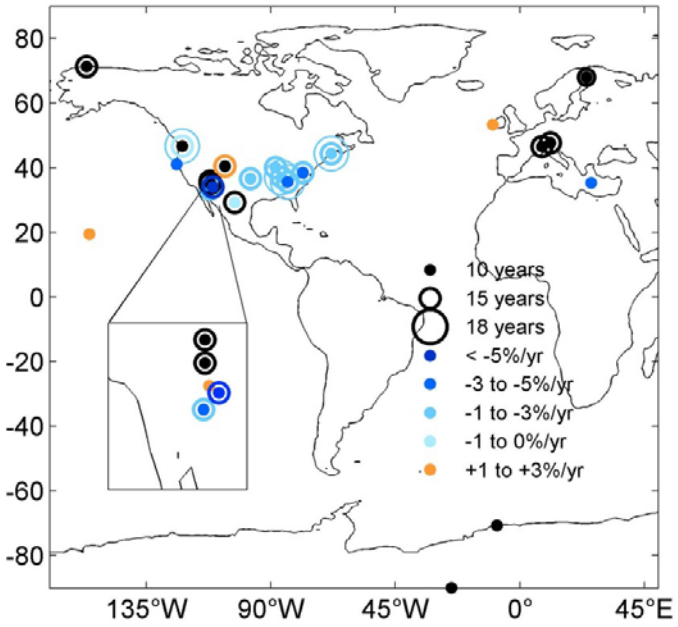


***Fig. 5****: Trends results for the scattering coefficient. Black symbols correspond to stations with no significant trends. Blue and orange symbols correspond to statistically significant negative and positive trends, respectively, the magnitude of the trends (slope) being given by the colors as stipulated in the legend. The sizes of the circles are proportional to the length of the data sets; the trend for the whole period as well as the 10 years (dots) and, if possible, 15-year trends were calculated. The largest circles denote, therefore, the trend of the longest analyzed period (from Collaud Coen et al., 2013).*

The scattering coefficient and number concentration statistically significant (s.s.) trends are predominantly negative. The trend slopes ranged between -1 and -85 %/10yr depending on the stations and the analyzing methods. Generally the absorption coefficient trends confirm the results found for the scattering coefficient. The main results are as follows:

- Over the continental US, scattering and absorption coefficients trends as well as number concentration are generally s.s. and negative. We can therefore conclude that, except for specific stations, there is a very general and robust decrease in aerosol optical parameters observed over the last 10 to 15 years in this area, detectable despite the high natural variability of the atmospheric aerosol.

- Few s.s. trends were observed for the five European sites. One Mediterranean site presents a scattering s.s. decreasing trend that can be related to regional conditions and was not confirmed by the absorption trend. In contrast, the analysis of long-term scattering measurements from two mountaintop sites (JFJ and HBP), and one site in a high latitude boreal region (PAL) revealed no s.s. trends for both the scattering and absorption coefficients for the last 10 years. For the number concentration, s.s. negative trends are found for 2 Scandinavian stations. Finally, a s.s. positive change in the scattering

coefficient is observed at the coastal Atlantic site (MHD) with only one out of the three analyzing methods, which is not observed for the absorption coefficient.

- We found no s.s. scattering trends for the Arctic or Antarctic sites. But two of the polar stations have s.s. negative absorbing aerosol trends. Number concentrations present no or negative s.s. trends for the whole measuring periods for both Arctic (13 yr) and Antarctic (37 yr), whereas positive trends are found for the last 10 yr in the Antarctic.

- The high altitude Mauna Loa (MLO) site in the middle of the Pacific Ocean has the only data set suggesting an increase of both the scattering and absorption coefficients over the last ten years. The most probable cause of the positive trends in absorption and scattering is the increased emission of pollutants in Asia being transported by high altitude winds to reach MLO. In contrast, a s.s. decreasing trend is found for the number concentration at MLO.

- No consistent, s.s. trends in b and å were observed for most of the stations. At the arctic station (BRW) and MLO s.s. negative trends in b and å were found for the 2001-2010 period. Negative b and å trends signify a relative shift towards larger particles at the lower end of the accumulation mode.

In summary, at most US continental sites, decreasing trends observed for aerosol optical properties are generally consistent with $SO_2$ and PM reductions. For continental European sites, the relation between aerosol optical properties and emissions reductions is less clear. The strong decreasing signal for $SO_2$ and, with a lower spatial homogeneity and statistical significance, for PM2.5 was not reflected in the aerosol optical properties in Europe. The European discrepancy might also be due to under-representation of continental EU PBL sites in our study. Furthermore, the difference in the timing of $SO_2$ and PM trends for the two continents is another likely explanation for the decreasing trends in aerosol number concentration and optical parameters found for most American sites compared to the lack of trends observed in Europe. The European optical property time series may not go back far enough to reflect the time period with the largest emissions reductions.

Key words:

Atmospheric aerosol particles, aerosol climatic effects, radiative forcing, light scattering, cloud condensation nuclei, hygroscopic growth, CCN concentration, aerosol size distribution, remote sensing of aerosol optical properties

Internet data bases:

http://www.psi.ch/lac
http://aerosolforschung.web.psi.ch
https://gawrtl.psi.ch
http://www.meteoschweiz.admin.ch/web/en/meteoswiss/international_affairs/GAW.html

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Scientific publications and public outreach 2012:

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http://www.eenews.net/public/Greenwire/2012/11/13/1

P. Kupiszewski, Die Mikrophysik der Wolken
Neue Zürcher Zeitung, August 8, 2012.

E. Weingartner, Hochalpine Top-Wissenschaft
Neue Zürcher Zeitung, June 12, 2012.

E. Weingartner, 75 Jahre Sphinx-Observatorium
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Tribune de Genève, May 7, 2012.

E. Weingartner, Feinstaub: Lungenschäden und Regenfälle
SDA / Schweizerische Depeschenagentur, May 4, 2012.

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E. Hammer, «Lanzi...» auf dem Jungfraujoch
Zambo, Schweizer Fernsehen SF1, December 11, 2012.
http://www.srf.ch/player/video?id=4ed8ce8a-c7c6-403f-b703-c389f9c5db60

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