Name of research institute or organization:

Laboratory of Atmospheric Chemistry, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

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 25 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 2009 it has reached 15 year 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.

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; black carbon (BC) concentration
MAAP	Absorption coefficient; black carbon (BC) conc.
Filter packs	Aerosol major ionic composition (PM1 and TSP)
Betameter and HiVol[1)]	Aerosol mass, PM1 and TSP[1)]

[1)] measured by EMPA

Long-term aerosol data and trend analysis

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. As an example for the collected long-term data, Figure 1 shows the measured scattering and absorption coefficients of the last 15 years. The absorption coefficients were calculated from different instruments and were extrapolated to a wavelength of $\lambda = 700$ nm assuming a wavelength dependence of λ^{-1}. The MAAP dataset is taken as a reference and the aethalometer data are adjusted to this reference by using empirical scaling factors deduced from concurrent measurements (Weingartner et al. (2003)[1] and Collaud Coen et al. (2009)).

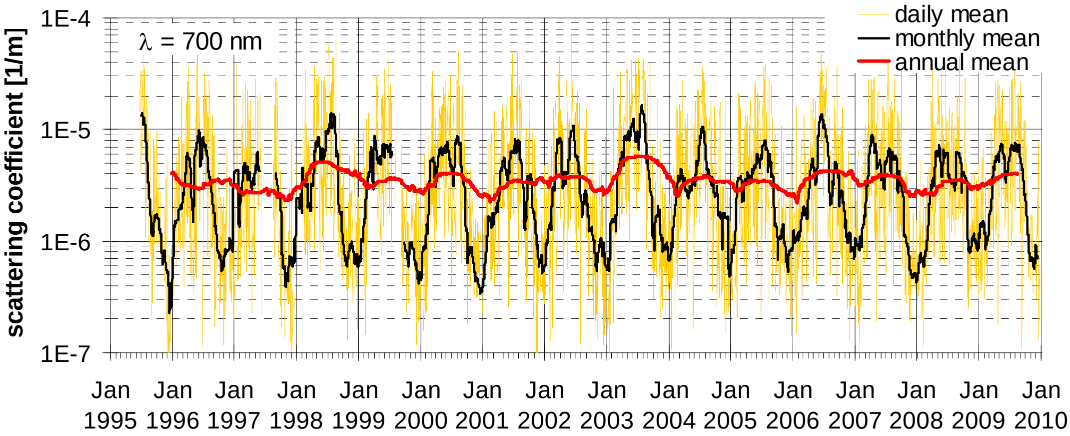
Figure 1 shows a distinct seasonality in the encountered aerosol loadings. In warm months, the site is influenced by injection of planetary boundary layer air into the free troposphere during sunny afternoons due to thermal convection, while in winter it is usually in the undisturbed free troposphere.

A thorough statistical trend analysis was performed for the measured absorption (AE31 only) and scattering coefficients with data measured before December 2005 (Collaud Coen *et al.*, 2007[2]). The summer months at the Jungfraujoch, which are strongly influenced by planetary boundary layer air masses, do not show any statistically relevant long-term trend. In contrast, a significant and distinct positive trend of 4-7% yr^{-1} was encountered for most aerosol parameters during the September to December period before 2006. A possible explanation for this increase involves a European-wide, large-scale increase of the injection of planetary boundary layer (PBL) air masses into the lower free troposphere (FT) coupled with large scale transport, or long-range transport from even more distant sources. In this sense this

[1] Weingartner, E.; Saathoff, H.; Schnaiter, M.; Streit, N.; Bitnar, B.; Baltensperger, U., Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers. *Journal of Aerosol Science* 2003, 34, (10), 1445-1463.

[2] Collaud Coen, M.; Weingartner, E.; Nyeki, S.; Cozic, J.; Henning, S.; Verheggen, B.; Gehrig, R.; Baltensperger, U., Long-term trend analysis of aerosol variables at the high alpine site Jungfraujoch. *Journal of Geophysical Research* **2007,** 112, D13213, doi: 10.1029/2006JD007995.

positive trend in this time period is interpreted as an increase of the lower FT aerosol concentration. An important future field of activity will be a careful reanalysis of trends using for the entire dataset.



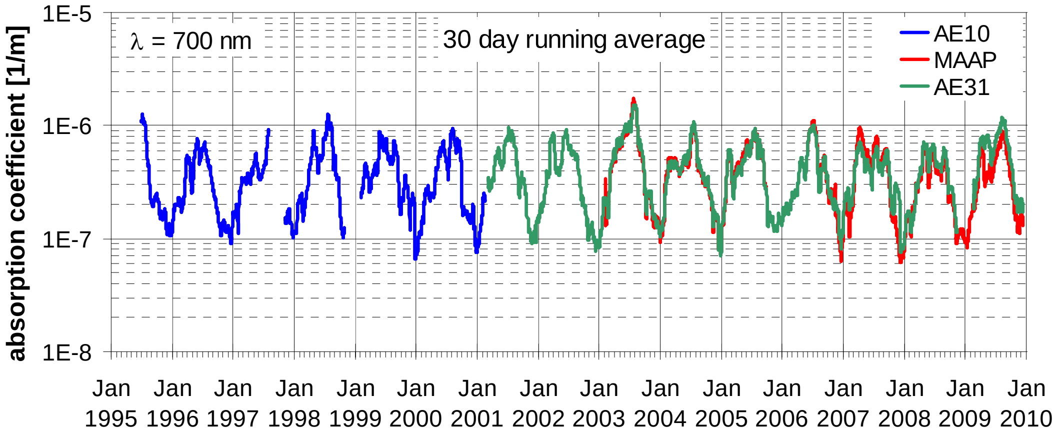


Figure 1: 15 years of measurements of the light scattering and absorption coefficients at the Jungfraujoch.

<u>On-line notification service for the presence of mineral dust at the Jungfraujoch</u>

The continuously measured wavelength dependent scattering coefficients (b_s) and absorption coefficients (b_{abs}) can be used to determine the presence of mineral dust events at the Jungfraujoch with an analysis of the wavelength dependence of the aerosol single scattering albedo, defined as $b_s/(b_s+b_{abs})$ (see Collaud Coen el al. (2004)[3] for details). This analysis is done on a regular basis at MeteoSwiss with quality controlled aerosol data from the Jungfraujoch. External groups expressed their interest to have a much faster notification service for the presence of mineral dust events. Therefore, a real-time notification service was established by a continuous analysis of the measured optical aerosol data. Computer generated notification emails are sent to interested users as soon as a possible mineral dust event is detected at the

[3] Collaud-Coen, M.; Weingartner, E.; Schaub, D.; Hueglin, C.; Corrigan, C.; Henning, S.; Schwikowski, M.; Baltensperger, U., Saharan dust events at the Jungfraujoch: detection by wavelength dependence of the single scattering albedo and first climatology analysis. *Atmospheric Chemistry and Physics* **2004,** 4, 2465-2480.

Jungfraujoch (a second email is sent as soon as the event stops). During 2009, 22 emails alerting a possible mineral dust event were sent out.

Additional measurements during 2009

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" and two EU Projects (EUSAAR and EUCAARI).

Table 2: Additional aerosol instrumentation operated in 2008-2009.

Instrument	Measured parameter	Measurement period
SMPS, OPC	Particle number size distribution, d_p = 20 - 22'500 nm	10.1.2008 - 31.12.2009
CCNC	Number concentration of cloud condensation nuclei	10.1.2008 - 15.12.2009
Sunset EC/OC	Mass concentration of organic and elemental carbon	1.4.2008 - 1.4.2009
HTDMA	Hygroscopic aerosol properties	1.5.2008 - 1.6.2009

Measurement of particle number size distributions

The permanent GAW monitoring activities include measurements of the total concentration of particles with diameters larger than 10 nm. However, the number size distribution of aerosol particles, which plays a key role for direct and indirect aerosol climate interactions, was not yet monitored on a permanent basis. Therefore, a scanning particle mobility sizer (SMPS) and an optical particle counter were installed at the JFJ in January 2008. These instruments have been fully operational since then and provide a complete size distribution form 20 nm to 22 μm.

As an example, Figure 2 shows monthly averaged size distribution parameters. Similar to most extensive aerosol parameters, the number concentration exhibits a distinct seasonal concentration change. In addition, it is found that during the colder seasons (autumn, winter and spring) the relative number fraction of small particles (d_p <50 nm) is substantially greater than in summer. This is explained by increased formation of new particles via homogenous nucleation during the colder seasons as already shown by Weingartner et al. (1999). The ratio between the CPC number concentration (dp > 7 nm) and the integrated SMPS number concentration (d_p > 20 nm) increases from 1 to 3 during wintertime, confirming the strong presence of nuclei mode particles (< 20 nm). The increased standard deviation of this ratio during wintertime also shows that the formation of new particles is occurring event-based rather than being a constant process. In contrast, the aerosol particles in summer are on average larger. This is explained by enhanced heterogeneous condensation.

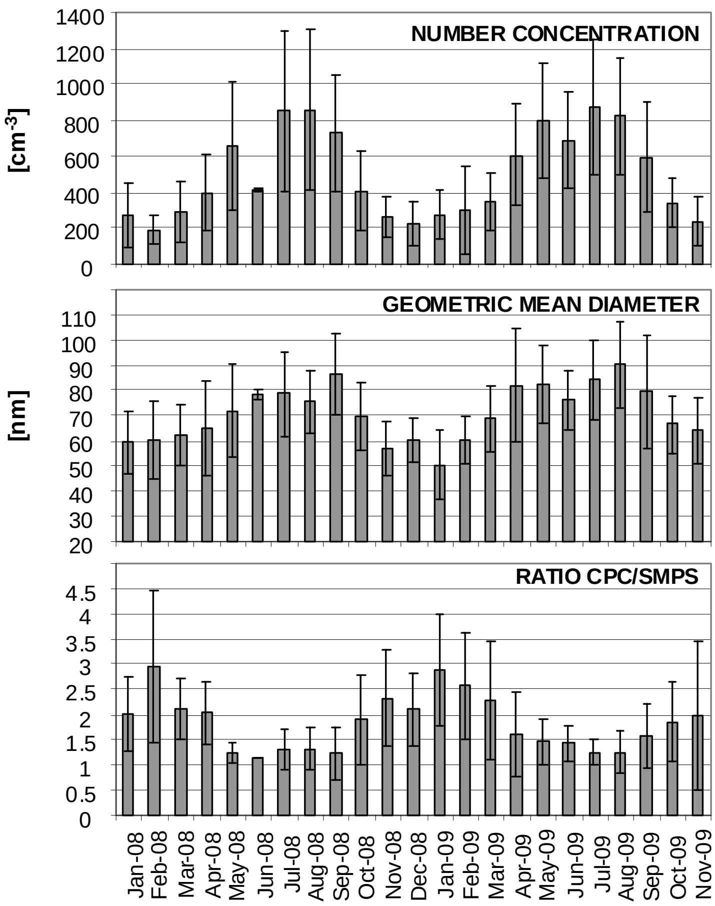


Figure 2: Monthly averaged size distribution parameters measured at the Jungfraujoch. Error bars represent the standard deviation of the monthly averages based on diurnal values. First and second panel: Parameters obtained from SMPS measurements (dp = 20 - 800 nm). Bottom panel: Ratio based on CPC measurements (TSI 3010/3772) and SMPS measurements.

Measurement of the number concentration of cloud condensation nuclei (CCN)

The cloud condensation nuclei counter (CCNC) exposes ambient aerosol particles to a defined water supersaturation (SS, in the range between SS = 0.07-1.18%) 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.

The ability of an aerosol particle to act as CCN depends on its size and chemical composition. The larger the particle, the smaller the critical SS that is needed for activation (Kelvin effect). The shape of the particle number size distribution influences hence the activation ratio at a given SS. In addition, the hygroscopicity of the particles is also important: The more soluble the particles, the more CCN active they are (Raoult effect). The variability of these two parameters is responsible for the encountered changes in the activated fraction. The monthly averages of the CCN concentration at any measured supersaturation (see Figure 3, upper panel) show the typical strong seasonal variability with a factor of 10 higher concentrations in summer than in winter. To a great extent this can be explained by the seasonally variability of the total aerosol concentration, which is caused by the injections of the planetary boundary layer into the free troposphere, occurring much more often in the summer. The additional variability can be seen in the activated fraction (i.e. the ratio of the CCN to total aerosol number concentration, Figure 3, lower panel), which correlates with the mean diameter of the size distribution. When on average the particles are smaller they activate less efficiently.

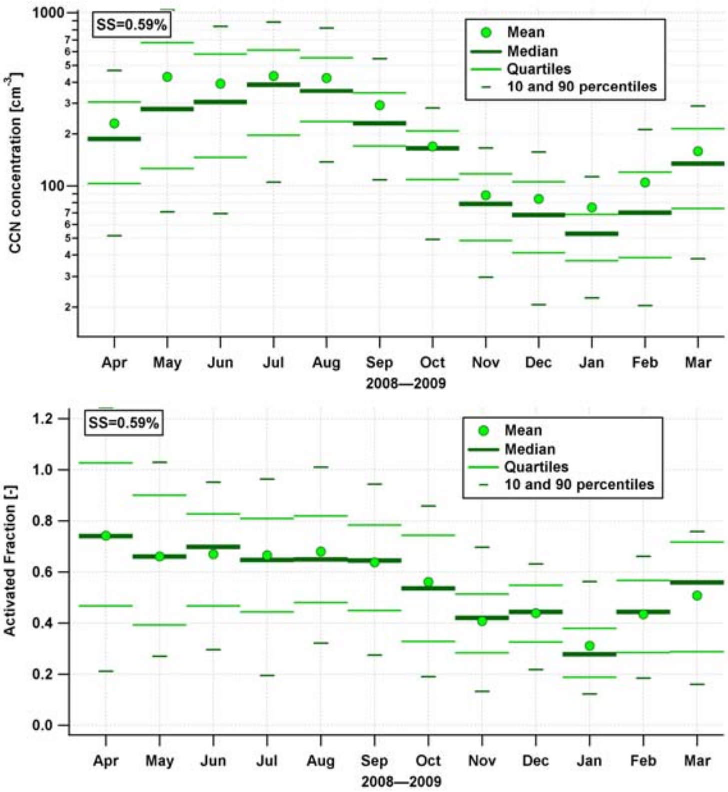


Figure 3: Seasonal variability of the CCN concentration and activated fraction at 0.59% supersaturation.

Aerosol hygroscopicity measurements

Aerosol particles can absorb significant amounts of water at high relative humidity (RH), depending on their hygroscopicity. This hygroscopic growth influences the aerosol direct effect (scattering of radiation by particles) and indirect effects (e.g. cloud-aerosol inter-actions) on climate, therefore being highly relevant for climate models. The water uptake of particles with dry diameters of 35, 50, 75, 110, 165, 265 nm was determined by a custom built HTDMA (Hygroscopicity Tandem Differential Mobility Analyzer), which measures the diameter growth factor (GF) at elevated RH relative to the dry particle diameter (D_0). In 2008 and 2009, 13 months of continuous HTDMA measurements were performed at the Jungfraujoch station.

Monthly mean GFs (GF_{mean}) at RH=90% were between 1.29 and 1.54, whereas larger particles exhibited generally larger GFs (Figure 4A). GFs depend on the chemical composition (Raoult effect) and the dry size (Kelvin effect) of a particle. Petters and Kreidenweis (2007)[4] introduced the hygroscopicity parameter κ, which captures the contribution of the Raoult term to the hygroscopic growth. Monthly mean κ values varied between 0.17 and 0.31 (Figure 4B) and were similar for all investigated D_0,

[4] Petters, M. D., and S. M. Kreidenweis (2007), Atmos. Chem. Phys., 7(8), 1961-1971.

thus implying similar chemical composition. Furthermore, the observed size dependence of GFs can largely be explained by the Kelvin effect, i.e. by the influence of particle dry size D_0. Further investigation of the growth factor probability density function (GF-PDF) revealed more detailed information about trends of GF_{mean}: Higher GF_{mean} in September 08 are due to a lower number fraction of less hygroscopic particles (Figure 4A and 4C), while lower GF_{mean} from June – August 08 are due to a shift of the dominant more hygroscopic mode to smaller GFs (Figure 4A and 4D).

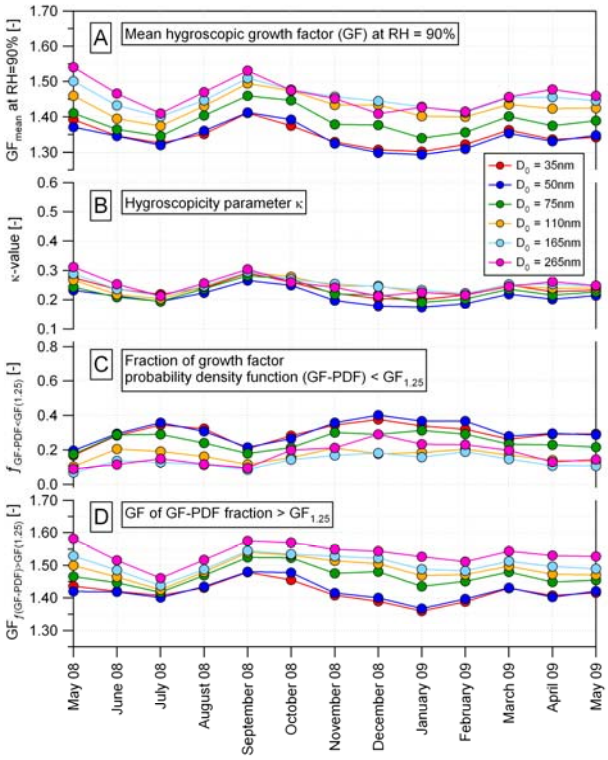


Figure 4: Annual cycles of A) the mean hygroscopic growth factor (GF_{mean}) at 90% relative humidity (RH), B) of the hygroscopicity parameter κ, C) of the fraction of the growth factor probability density function (GF-PDF) < GF1.25, and D) of the GF of the GF-PDF-fraction > GF1.25.

In conclusion, the GF of the Jungfraujoch aerosol shows little seasonal trends. This is also confirmed by the fact that the difference of GF_{mean} between May 08 and May 09 is similar in magnitude to the seasonal variability of GF_{mean}. First analyses revealed that the aerosol hygroscopicity is more influenced by the presence of planetary boundary layer (PBL) air masses during certain weather situations than by a seasonal variation. This extends findings of previous short-term studies at Jungfraujoch (Sjogren et al., 2008)[5].

[5] Sjogren, S., M. Gysel, E. Weingartner, M. R. Alfarra, J. Duplissy, J. Cozic, J. Crosier, H. Coe, and U. Baltensperger (2008), Atmos. Chem. Phys., 8(18), 5715-5729.

Key words:

Atmospheric aerosol particles, aerosol climatic effects, radiative forcing, light scattering, cloud condensation nuclei, hygroscopic growth, CCN concentration, aerosol size distribution

Internet data bases:

http://www.psi.ch/gaw
http://www.psi.ch/lac
http://aerosolforschung.web.psi.ch
http://www.meteoschweiz.admin.ch/web/en/climate/climate_international/gaw-ch.html

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Scientific publications and public outreach 2009:

Refereed journal articles

Jimenez, J. L., M. R. Canagaratna, N. M. Donahue, A. S. H. Prevot, Q. Zhang, J. H. Kroll, P. F. DeCarlo, J. D. Allan, H. Coe, N. L. Ng, A. C. Aiken, K. S. Docherty, I. M. Ulbrich, A. P. Grieshop, A. L. Robinson, J. Duplissy, J. D. Smith, K. R. Wilson, V. A. Lanz, C. Hueglin, Y. L. Sun, J. Tian, A. Laaksonen, T. Raatikainen, J. Rautiainen, P. Vaattovaara, M. Ehn, M. Kulmala, J. M. Tomlinson, D. R. Collins, M. J. Cubison, E., J. Dunlea, J. A. Huffman, T. B. Onasch, M. R. Alfarra, P. I. Williams, K. Bower, Y. Kondo, J. Schneider, F. Drewnick, S. Borrmann, S. Weimer, K.

Demerjian, D. Salcedo, L. Cottrell, R. Griffin, A. Takami, T. Miyoshi, S. Hatakeyama, A. Shimono, J. Y Sun, Y. M. Zhang, K. Dzepina, J. R. Kimmel, D. Sueper, J. T. Jayne, S. C. Herndon, A. M. Trimborn, L. R. Williams, E. C. Wood, A. M. Middlebrook, C. E. Kolb, U. Baltensperger, and D. R. Worsnop, Evolution of Organic Aerosols in the Atmosphere, Science **326** (5959), 1525. DOI: 10.1126/science.1180353, 2009.
http://www.sciencemag.org/cgi/content/abstract/326/5959/1525

Fierz-Schmidhauser, R., Zieger, P., Gysel, M., Kammermann, L., DeCarlo, P. F., Baltensperger, U., and Weingartner, E.: Measured and predicted aerosol light scattering enhancement factors at the high alpine site Jungfraujoch, Atmos. Chem. Phys. Discuss., **9**, 20063-20101, 2009.
http://www.atmos-chem-phys-discuss.net/9/20063/2009/acpd-9-20063-2009.html

Targino, A.C., Coe, H., Cozic, J., Crosier, J., Crawford, I., Bower, K., Flynn, M., Gallagher, M., Allan, J., Verheggen, B., Weingartner, E., Baltensperger, U., Choularton, T.: Influence of particle chemical composition on the phase of cold clouds at a high-alpine site in Switzerland, Journal of Geophysical Research-Atmospheres **114**, D18206, doi:10.1029/2008JD011365, 2009.
http://www.agu.org/pubs/crossref/2009/2008JD011365.shtml

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmidhauser, R., Schmid, O., and Baltensperger, U.: Minimizing light absorption measurement artifacts of the aethalometer: evaluation of five correction algorithms, Atmos. Meas. Tech. Discuss., **2**, 1725-1770, 2009.
http://www.atmos-meas-tech-discuss.net/2/1725/2009/amtd-2-1725-2009.html

Conference papers

Kammermann, L., Gysel, A., Weingartner, E. and Baltensperger, U., Hygroscopicity of Tropospheric Aerosols: One year of measurements at Jungfraujoch (3580 m asl.), 19th Annual VM Goldschmidt Conference, Davos, Geochimica et Cosmochimica Acta **73**, A618-A618, 2009.
http://www.goldschmidt2009.org/abstracts/view

Schmidhauser, R., Zieger, P., Weingartner, E., Gysel, M., DeCarlo, P.F. and Baltensperger, U., Aerosol light scattering at high relative humidity at a high alpine site (Jungfraujoch), European Aerosol Conference 2009, Karlsruhe, Abstract T047A07, 2009.
http://www.gaef.de/EAC2009/

Kammermann, L., Gysel, M., Weingartner, E. and Baltensperger, U., One-year climatology of the aerosol hygroscopicity at the High Alpine Research Station Jungfraujoch (3580 m asl.), European Aerosol Conference 2009, Karlsruhe, Abstract T170A01, 2009.
http://www.gaef.de/EAC2009/

Jurányi, Z., Gysel, M., Weingartner, E., DeCarlo, P.F., Heringa, M.F., Chirico R. and Baltensperger, U., Measuring and modeling cloud condensation nuclei concentrations at the Jungfraujoch high alpine site, European Aerosol Conference 2009, Karlsruhe, Abstract T170A04, 2009.
http://www.gaef.de/EAC2009/

Theses

Fierz, Rahel, Enhancement of the light scattering coefficient of atmospheric aerosol particles by water uptake, PhD Thesis ETH Zürich.

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