Name of research institute or organization:

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

Title of project:

Investigation of the cloud droplet activation behaviour of ambient black carbon particles during CLACE (cloud and aerosol characterization experiment) 2016

Part of this programme:

BLACARAT, BACCHUS, GAW, ACTRIS, CLACE

Project leader and team:

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Project description:

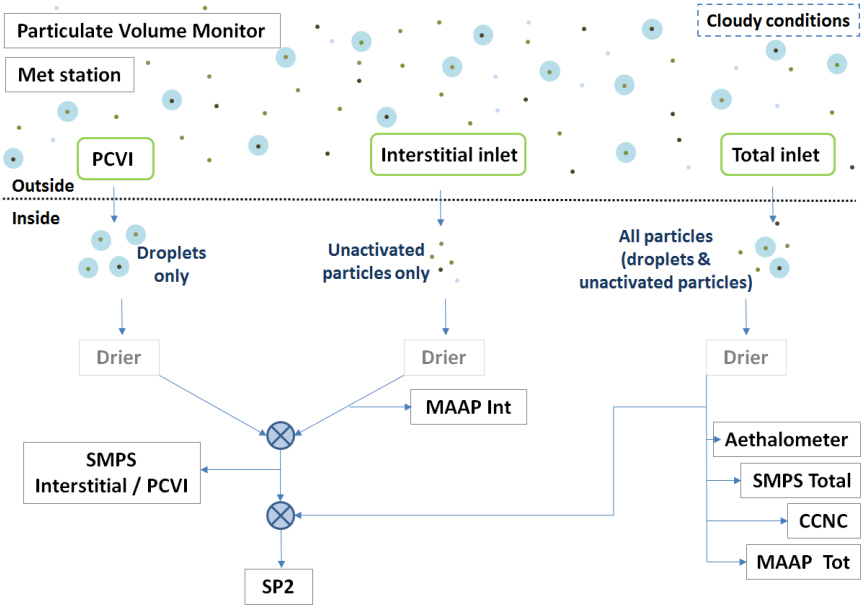
Atmospheric aerosols are small particles suspended in the air. These particles can cause various adverse effects on health and influence climate. A long-term exposure to air polluted by particles has been connected to cardiopulmonary and lung cancer mortality risk (*Pope et al., 2002*). In addition, two different effects can be distinguished concerning the aerosol's influence on atmospheric radiative transfer and, thus, on climate: aerosol scattering and absorption of short- and long-wave radiation (aerosol-radiation interactions, ari), and their activity as cloud condensation nuclei (CCN) and ice nuclei (IN) (aerosol-cloud interactions, aci). Our limited understanding of these two types of interactions makes aerosol-related uncertainties the largest among the climate forcers. Under given atmospheric conditions, cloud droplets can form on cloud condensation nuclei. An anthropogenic increase in the atmospheric CCN concentration therefore results in a modification of microphysical properties and precipitation efficiency of clouds, causing, according to the current scientific knowledge, a global cooling of the Earth's surface.

Black carbon (BC) is one of the major constituents of atmospheric aerosols, originating from combustion of fossil and biogenic fuels; most of its emissions originate from anthropogenic processes. The main difficulties in understanding and quantifying the CCN-activation process of BC come from the high heterogeneity of its sources and its short atmospheric lifetime, but also from the fact that these particles undergo various modifications during their aging. When emitted, they are generally hydrophobic upon emission, but, with aging, they can acquire a coating by the condensation of, or coagulation with, organic and inorganic material.

Conflicting results exist about the impact of black carbon on cloud formation. Accurately predicting the variations of CCN-activity from bare to thickly coated BC is a major challenge for current climate models, as it has direct implications on the life-cycle of BC: well-mixed BC-containing particles are more easily removed from the atmosphere, mostly by wet processes. This causes a decrease of the BC burden, thus reducing the time window available for aerosol-radiation interactions which influence the climate. A recent study from Lund et al. (2017) highlighted the significance of this challenge: when varying the amount of coating required for CCN-activation, they reported changes up to 25-50 % of the radiative forcing of BC via aerosol-radiation interactions compared to a baseline simulation.

Since 2000, CLACE (CLoud and Aerosol Characterization Experiment) campaigns have taken place at the Jungfraujoch in regular intervals. These comprehensive aerosol-cloud interaction studies are conducted within national and international collaborations. In this context, we performed a field campaign at Jungfraujoch during June and July 2016 (CLACE

2016) in order to investigate how the cloud droplet activation of BC particles depends on their size and mixing state and how it compares to BC-free particles. The instrumental set-up is shown in Figure 1.



PCVI:	pumped counterflow virtual impactor	> 3µm
	Interstitial inlet	< 2µm
CPC:	condensation particle counter	7 nm - 3µm
SMPS:	scanning mobility particle sizer	20 – 593 nm
SP2:	single particle soot photometer	70 – 500 nm (incand.)
CCNC:	cloud condensation nuclei-counter	0.35 – 0.75 % SS
APS :	aerosol particle sizer	0.5 – 20 µm
MAAP	multi-angle absorption photometer	>100 ng m^{-3}
	Aethalometer	0.1 - 100µg m^{-3}
PVM	particle volume monitor	0.002 - 10 g H_2O m^{-3}

Figure 1. Instrumental set-up during the CLACE 2016 campaign. The total inlet, total SMPS, total MAAP, aethalometer, and the CCNC were part of the Global Atmosphere Watch (GAW) program. "Tot" and "Int" stand for "Total" and "Interstitial", respectively.

Three different inlets were used:

-An interstitial inlet for the sampling of non-activated (interstitial) particles, with an upper cut-off of 1.8 micrometers.

-A pumped counterflow virtual impactor (PCVI, Brechtel Inc.), in order to sample cloud droplets residuals, with a lower cut-off around 2.5 micrometers.

-A total inlet, for sampling both interstitial and cloud residual particles.

Comparing size distributions of particles behind these inlets allows for the determination of size-resolved activated fractions, i.e. the number fraction of particles that activate to cloud droplets.

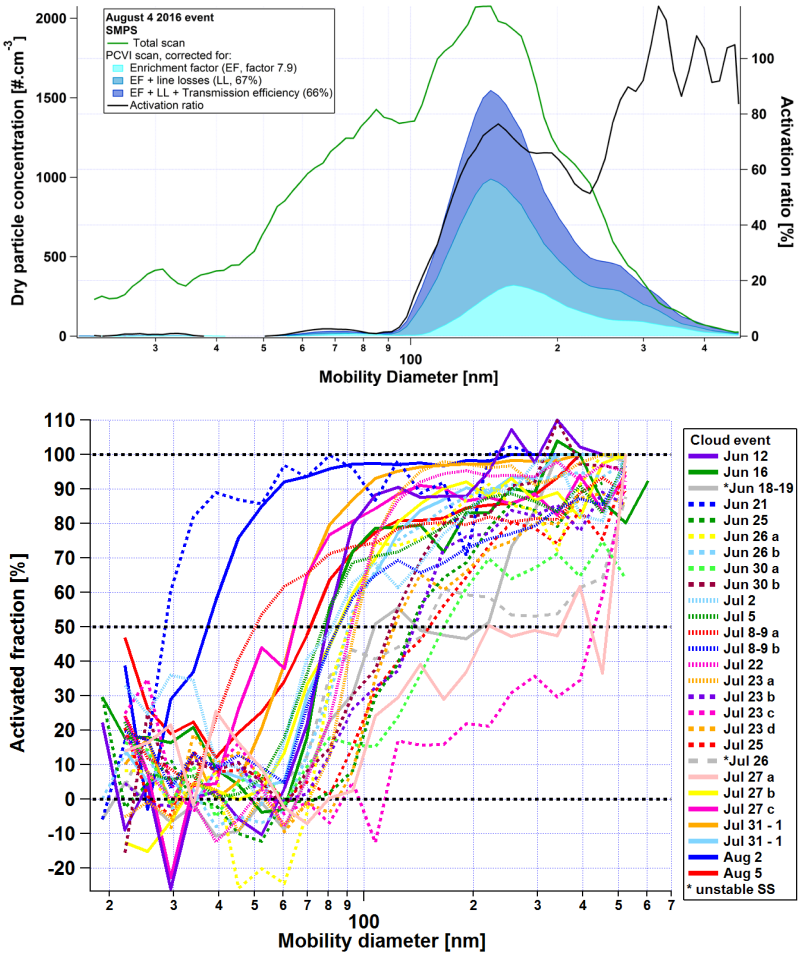


Figure 2. ***Top****: Example of the retrieval of the SMPS-derived size-resolved activated fraction using the pumped counterflow virtual impactor (PCVI) and the total inlet, after corrections for lines losses, PCVI transmission efficiency and the enrichment factor.* ***Bottom****: all SMPS-derived activated fractions using the interstitial and total inlet during the campaign.*

Very large variations of the 50% activation diameter (D_{50}, diameter at which 50 % of particles activate to droplets) were found among the cloud events during the campaign. This indicates that highly different supersaturations can occur in clouds at this altitude. This was verified by retrieving the effective peak supersaturation in clouds using a method introduced by Hammer et al. (2014). Our results confirm their findings that clouds present at the Jungfraujoch during northwest wind conditions show higher supersaturations than during southeast conditions (Figure 3).

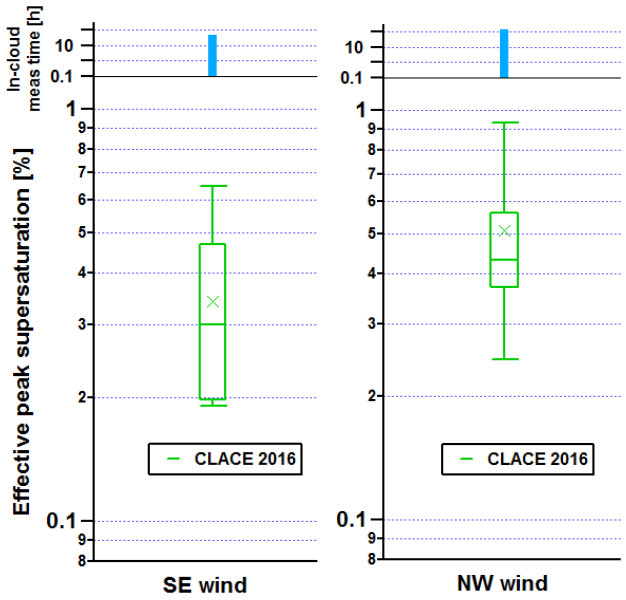


Figure 3. Boxplots summarizing the effective peak supersaturation of all cloud events during CLACE 2016, for northwest and southeast wind conditions.

It is therefore clear that cloud supersaturation has a major influence on the CCN-activation behaviour of aerosol particles. However, its influence on the activation of BC-containing particles is more complex because it needs to be isolated from the effect of mixing state, i.e. coating thickness. Cozic et al. (2007) showed, at the same site, that the scavenged mass fraction of BC is very similar to the one of the total aerosol. By comparing the scavenged (activated) fractions from the MAAP and the total SMPS, we could confirm this result (Figure 4).

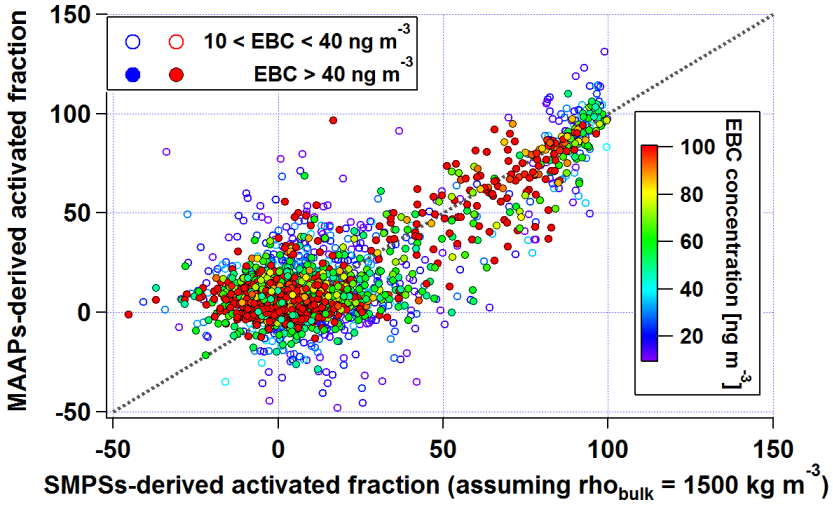


Figure 4. Comparison of the BC (y-axis) and the total aerosol (x-axis) scavenged fractions during the whole campaign.

This result suggests that BC-containing particles act as cloud condensation nuclei for clouds in the same way as BC-free particles, i.e. they form cloud droplets in a similar manner. However, conclusions from mass on number are prone to mis-interpretation. The use of a single particle soot photometer (SP2), an instrument which can measure the size of individual particles as well the refractory BC (rBC) mass, allows to discriminate the activation behaviour of rBC-containing particles as a function of their mixing state on a number basis. This is the first time that this instrument is used behind an interstitial and a PCVI inlet at Jungfraujoch, opening the door to the investigation of the influence of BC size and mixing state on cloud droplet activation and to compare it with theoretical calculations (Köhler theory). At the Jungfraujoch altitude, the dominant fraction of BC cores has substantial coatings, thus making them good CCN at BC core diameters down to 100 nm or even smaller. However, at low supersaturations the activation of BC cores with diameters smaller than 200 nm is incomplete as those cores with the thinnest coatings remain unactivated.

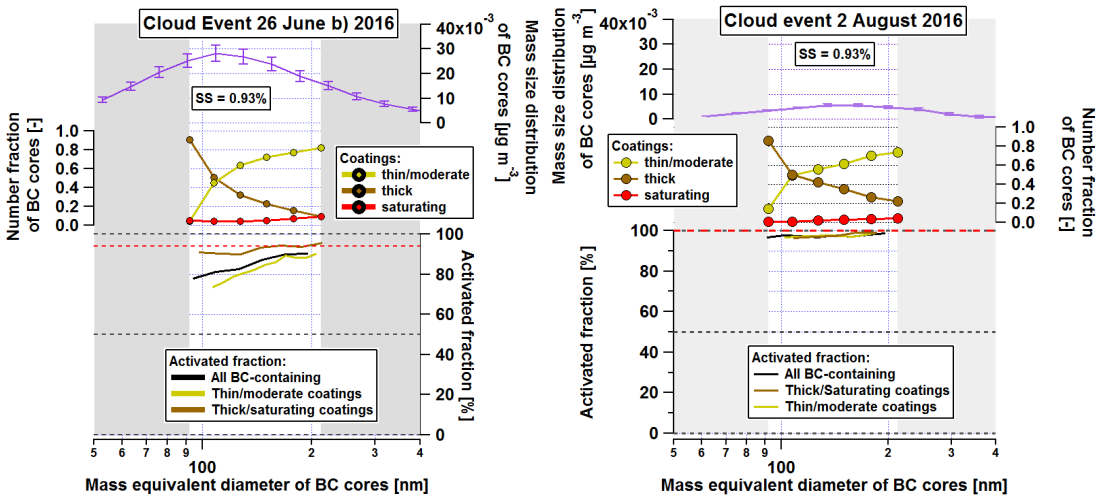


Figure 4. Mass size distribution (top panels), number fractions of coating size ranges behind the total inlet (middle panels) and activated fractions (bottom panels) of all BC-containing particles (black line), cores surrounded by thick coatings (brown line) and by thin to moderate coatings (yellow line) as a function of mass equivalent diameter of BC cores for two cloud events.

Figure 4 shows that, at cloud-relevant supersaturations, all rBC-containing particles with large rBC cores activate to cloud droplets: they are sufficiently large already without coating to activate as cloud droplets. However, the activation of small BC cores is facilitated by thick coatings compared to thin coatings. The maximum rBC core size for which coating thickness still plays a role in the CCN activation depends on the cloud supersaturation: at very high supersaturations, only very small BC cores have an activation behaviour that depends on mixing state.

References:

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