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Scavenging of black carbon in mixed phase clouds at the high alpine site Jungfraujoch

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Abstract

The scavenging of black carbon (BC) in liquid and mixed phase clouds was investigated during intensive experiments in winter 2004, summer 2004 and winter 2005 at the high alpine research station Jungfraujoch (3580 m a.s.l., Switzerland). Aerosol residuals were sampled behind two well characterized inlets; a total inlet which collected cloud particles (drops and ice particles) as well as interstitial aerosol particles; an interstitial inlet which collected only interstitial (unactivated) aerosol particles. BC concentrations were measured behind each of these inlets along with the submicrometer aerosol number size distribution, from which a volume concentration was derived. These measurements were complemented by in-situ measurements of cloud microphysical parameters. BC was found to be scavenged into the cloud phase to the same extent as the bulk aerosol, which suggests that BC was covered with soluble material through aging processes, rendering it more hygroscopic. The scavenged fraction of BC ($F_{Scav,BC}$), defined as the fraction of BC that is incorporated into cloud droplets and ice crystals, decreases with increasing cloud ice mass fraction (IMF) from $F_{Scav,BC}$=60% in liquid phase clouds to $F_{Scav,BC}$~10% in mixed-phase clouds with IMF>0.2. This is explained by the evaporation of liquid droplets in the presence of ice crystals (Wegener-Bergeron-Findeisen process), releasing BC containing cloud condensation nuclei back into the interstitial phase. In liquid clouds, the scavenged BC fraction is found to decrease with decreasing cloud liquid water content. The scavenged BC fraction is also found to decrease with increasing BC mass concentration since there is an increased competition for the available water vapour.

1 Introduction

Atmospheric aerosol particles may play a major role in climate change by absorbing and scattering solar and infrared radiation, referred to as the direct forcing and discussed in detail by e.g. Haywood and Boucher (2000). Aerosol particles can also

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indirectly influence climate by altering the microphysical properties of clouds, e.g. as reviewed by Lohmann and Feichter (2005). This indirect forcing is caused by the ability of aerosol particles to act as cloud condensation nuclei (CCN) or ice nuclei (IN). With increasing CCN number concentration, caused by e.g. human activity, more and smaller droplets are formed in warm clouds (Twomey effect). As a consequence, and under the assumption that the cloud liquid water content (LWC) stays constant, the cloud albedo and thus the reflection of solar radiation increase (Twomey, 1977). In addition, the reduction of cloud droplet size weakens precipitation development, resulting in more persistent clouds (cloud lifetime effect) (Albrecht, 1989). Although little is known about the response of warm clouds to a changing aerosol population, even less is known about the effects on and feedback mechanisms associated with glaciated or mixed-phase clouds (Lohmann and Feichter, 2005). The presence of ice crystals is important for climate, because they influence the cloud radiative properties and they lead to precipitation (Rogers and Yau, 1989; Lau and Wu, 2003). The aerosol indirect forcing has been estimated to have the highest uncertainty in assessing human impact on climate change (IPCC, 2001).

The atmospheric aerosol mainly consists of non-light absorbing components such as sulphates, nitrates and organic carbon, which scatter solar radiation and thus lead to a cooling of the climate system. Aerosol particles can also contain light absorbing material such as black carbon (BC), which exerts a positive forcing at the top of the atmosphere. This might partly offset the cooling due to the scattering (IPCC, 2001). This positive forcing can be amplified if absorption of solar radiation by BC occurs within cloud droplets or for a coated BC particle (Chylek et al., 1996). When absorption occurs in a cloud droplet, the resulting increase in temperature reduces the relative humidity and may result in the evaporation of cloud droplets (semi-indirect effect) (Lohmann and Feichter, 2005, and cited references therein). The reduced cloud cover and cloud optical depth will in turn further amplify warming of the Earth-atmosphere system.

Freshly emitted soot particles are mostly hydrophobic, i.e. they barely act as CCN (Weingartner et al., 1997). Due to atmospheric aging processes, BC is often coated

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with secondary aerosol material (such as inorganic ions) at remote locations in the atmosphere. This aged BC exhibits a larger hygroscopicity and the transfer of BC into cloud droplets via nucleation scavenging is therefore enhanced. In-cloud scavenging of BC contributes to its removal from the atmosphere and thereby affects its lifetime. It is also important for the positive radiative forcing of BC which strongly influences the uncertainties of the net cloud radiative forcing. The ability of BC to act as IN is also important to consider since it is an anthropogenic compound which may cause a glaciation indirect forcing, whereby the increased formation of ice crystals enhances precipitation, thus causing a decrease in cloud lifetime and cloud cover (Lohmann, 2002; Lohmann and Diehl, 2006). This forcing may work in the opposite direction compared to the indirect forcing of aerosols acting as CCN in liquid clouds or mixed phase clouds and thus results in a warming of the atmosphere. During our campaign, BC content of ice residuals was also measured. The ability of BC to act as IN will be discussed in a separate paper (Cozic et al., 2006a[1]).

The scavenged fraction of BC ($F_{\mathrm{Scav,BC}}$) is defined as the fraction of BC that is incorporated into cloud droplets and ice crystals. Previous studies have shown that the scavenged BC fraction increases with increasing distance from source regions, from 0.06 in heavily polluted Po Valley fog (Hallberg et al., 1992) to 0.8 in a remote area in Spitzbergen (Heintzenberg and Leck, 1994). Table 1 lists scavenged BC fractions from the literature. Some of these studies (Hitzenberger et al., 2000; Kasper-Giebl et al., 2000) calculated the scavenged fraction from the comparison between cloud impactor samples and interstitial aerosols and not as in our study with total and interstitial concentrations.

From Table 1, it is obvious that the atmospheric aging processes which transform hydrophobic, freshly emitted soot particles into hygroscopic internally mixed aerosol are important factors which increase the scavenged BC fraction. Nevertheless, the

[1]Cozic, J., Verheggen, B., Mertes, S., Petzold, A., Baltensperger, U., and Weingartner, E.: Enrichment of black carbon in ice residuals at the high alpine site Jungfraujoch, in preparation, 2006a.

slightly different results observed at Mt. Sonnblick during two different studies in the same season indicate that other parameters besides the atmospheric processing of the air mass affect the scavenged BC fraction as well. Hitzenberger et al. (2000, 2001) and Kasper-Giebl et al. (2000) found that the BC scavenged fraction increases with increasing LWC. Sellegri et al. (2003) did not find such a dependence on LWC. Hallberg et al. (1992, 1994) and Gieray et al. (1997) found that the BC scavenged fraction decreases with increasing BC total concentration. Other Studies (Hitzenberger et al., 2000, 2001; Kasper-Giebl et al., 2000) did not find such a dependence on the BC concentration.

The purpose of this study is to further increase the knowledge of the processes which determine the partitioning of BC between the interstitial particle phase (i.e. not activated particles) and the cloud phase (i.e. cloud droplets and ice crystals). The new aspect in this paper compared to the others is that measurements were carried out in liquid and mixed-phase clouds. This allowed the assessment of temperature and ice mass fraction as an additional significant factor in determining the scavenged fraction. The different sampling systems, the instrumentation and the site are first described. Then the impacts of some environmental parameters (LWC, BC total concentration, temperature, ice mass fraction) on the scavenged fraction of BC are discussed.

2 Site, sampling and analysis

Measurements were conducted during several **Cl**oud and **A**erosol **C**haracterization **E**xperiments (CLACE) at the high-alpine research station Jungfraujoch (Switzerland, 3580 m a.s.l.). Sampling was performed in March 2004 (CLACE 3), from mid July to the end of September 2004 (CLACE 3.5) and from mid February to mid March 2005 (CLACE 4). More than 3500 h of data were collected and analyzed, with approximately 1000 h of "in-cloud" measurements (37% average cloud coverage in winter and 20% in summer). A wide variety of physical and chemical parameters were measured downstream of three well characterized inlets and complemented by in-situ measurements

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of cloud microphysical parameters (see below).

Previous studies used simultaneously measured submicrometer number size distributions (total and interstitial) in order to calculate the aerosol scavenged number fraction ($F_{Scav,N}$) at the Jungfraujoch (Henning et al., 2002, 2004). The scavenged number fraction ($F_{Scav,N}$) is defined as the fraction of the total number of particles (particle diameter D>100 nm) that are incorporated into cloud droplets and ice crystals. They also found a clear dependency of $F_{Scav,N}$ on temperature. These findings have been complemented with the analysis of the $F_{Scav,N}$ dependency on cloud microphysical parameters which results are presented in a series of papers (Mertes et al., 2006[2]; Verheggen et al., 2006[3]; Weingartner et al., 2006[4]).

2.1 The Jungfraujoch station

The Jungfraujoch measurement site is located on an exposed mountain col in the Swiss Alps at 3580 m asl. It is a Global Atmosphere Watch (GAW) station where atmospheric aerosols and gases have been measured for more than a decade. The

[2]Mertes, S., Verheggen, B., Walter, S., Ebert, M., Connolly, P., Weingartner, E., Schneider, J., Bower, K. N., Inerle-Hof, M., Cozic, J., Baltensperger, U., and Heintzenberg, J.: Counterflow virtual impactor based collection of small ice particles in mixed-phase clouds for the physico-chemical characterisation of tropospheric ice nuclei: sampler description and first case study, Aerosol. Sci. Technol., submitted, 2006.

[3]Verheggen, B., Cozic, J., Weingartner, E., Bower, K., Mertes, S., Connolly, P., Flynn, M., Gallagher, M., Choularton, T., and Baltensperger, U.: Aerosol activation in mixed phase clouds at the high Alpine site Jungfraujoch, J. Geophys. Res., submitted, 2006.

[4]Weingartner, E., Verheggen, B., Bower, K. N., Mertes, S., Schneider, J., Lohmann, U., Cozic, J., Walter, S., Alfarra, M. R., Borrmann, S., Choularton, T., Coe, H., Connolly, P., Crosier, J., Curtius, J., Ebert, M., Van Ekeren, J. S., Flynn, M., Gallagher, M., Gysel, M., Henning, S., Inerle-Hof, M., Petzold, A., Sjogren, S., Weinbruch, S., and Baltensperger, U.: The Role of Ice Nuclei in Determining the Aerosol Partitioning in Natural Mixed-Phase Clouds – Implications for Climate, in preparation, 2006.

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station is regularly engulfed in clouds (30% of the time averaged over the 3 campaigns discussed in this study). Due to its location and altitude, the site is far away from significant pollution sources (only occasionally local construction work causes measurable emissions). These characteristics make the Jungfraujoch well suited to investigate continental background aerosols and clouds from a ground based platform.

All aerosol parameters measured over an extended period at the Jungfraujoch exhibit a strong seasonal cycle with a maximum in summer and a minimum in winter. This is due to the fact that in summer the site is influenced by injections of air from the more polluted planetary boundary layer caused by thermal convection during afternoons with high solar insulation. In wintertime these processes are much less frequent resulting in substantially lower aerosol concentrations (Weingartner et al., 1999; Henne et al., 2004). Therefore, the site is deemed representative of the lower free troposphere above a continental area. More information on the Jungfraujoch site and the long-term aerosol measurements are found in Baltensperger et al. (1997).

2.2 Inlets

The partitioning of aerosol particles in mixed-phase clouds was investigated by sampling air through three well characterized inlets. A heated (25°C), total aerosol inlet was used to evaporate cloud droplets and ice crystals at an early stage of the sampling process. This inlet was designed to sample cloud droplets smaller than 40 μm at wind speeds of up to 20 m/s (Weingartner et al., 1999). The total inlet sample thus consists of interstitial (unactivated) particles as well as of the residues of cloud droplets and ice crystals. This inlet is also used for the continuous GAW measurements.

The second inlet was designed to sample only interstitial aerosol by removing cloud particles from the sampled air, using an aerodynamic size discriminator ($PM_{2.5}$, Very Sharp Cut Cyclone, BGI, USA) operating at a high flow rate (~20 lpm) resulting in a cutoff of D_{50}=2 μm at ambient temperature ($T \approx -10$°C). During cloud presence, the difference in a particular instrument response between the total and interstitial inlets enables the determination of this parameter for the particles present only in the cloud

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phase. The inlet was cleaned regularly to avoid riming and clogging by snow.

The third inlet is the so-called "Ice Counterflow Virtual impactor" (Ice-CVI), which was designed to sample residual particles of small ice crystals and/or cloud droplets (Mertes et al., 2006[2]). The ability of BC to act as IN will be discussed in a separate paper (Cozic et al., 2006a[1]).

2.3 Black Carbon measurements

Two Multi-Angle Absorption Photometers (MAAP, Thermo ESM Andersen) operating at a wavelength of λ=630 nm (Petzold and Schönlinner, 2004), two Particle Soot Absorption Photometers (λ=580 nm) (PSAP, Radiance research) (Reid et al., 1998) and two 7-wavelengths Aethalometers (λ=370, 450, 570, 615, 660, 880 and 950 nm) (AE31, MAGEE Scientific) (Hansen et al., 1984) were installed to measure the particle light absorption coefficient from which the BC mass concentration was derived. The wavelengths of the individual instruments were determined in an intercomparison campaign at the IFT in Leipzig, Germany in November 2005 (T. Müller, personal communication).

One of each of the three types of BC instruments was connected to the total inlet (see Fig. 1). A second MAAP and an Aethalometer were installed downstream of the interstitial inlet and a PSAP was installed behind the Ice-CVI. The time resolution of the MAAP was set to 1 min and those of the PSAP and the Aethalometer to 5 min. The data were then averaged to a common time base of 10 min.

The principle of the PSAP and the Aethalometer is to continuously measure the transmission of a light beam with a defined wavelength through a fiber filter through which air is drawn. The primary measure of the Aethalometer and the PSAP is the light attenuation coefficient (b_{ATN}) which is defined with Beer-Lambert's law

$$I = I_o \cdot e^{-b_{ATN} \cdot x} \quad (1)$$

where I_o is the intensity of the incoming light, I the remaining light intensity after passing through the filter and x is the optical path in the filter.

For the Aethalometer, b_{ATN} is converted into an absorption coefficient, b_{abs}, which relates to particles in the airborne state:

$$b_{abs} = b_{ATN} \cdot \frac{1}{C} \quad (2)$$

C accounts for multiple scattering of light at the filter fibers and embedded particles resulting in an enhanced optical path in the filter and thus enhanced apparent absorption. The correction factor C depends on the optical properties of the deposited particles and on aerosol loading on the filter. A correction factor of C=2.14 as determined in laboratory experiments (Weingartner et al., 2003) was applied for the Aethalometer data. Equation (2) is a simplified correction which only accounts for the multiple scattering effect of the filter fibers but it does not correct for the shadowing and the scattering effects caused by the deposited particles. This simplified approach was shown to be accurate for the JFJ aerosol (Weingartner et al., 2003).

For the PSAP, data were corrected using the method described by (Bond et al., 1999):

$$b_{abs} = \left(\frac{b_{ATN}}{2 \cdot (0.5398 \cdot T_r + 0.355)} - K_1 b_s \right) / K_2 \quad (3)$$

where K_1=0.02, K_2=1.22, and T_r is the filter transmission. b_s is the light scattering coefficient at λ=550 nm which was simultaneously measured with an integrating Nephelometer (TSI 3563).

The MAAP measures simultaneously the light transmitted through and scattered back from the particle loaded filter. The light absorption coefficient b_{abs} is obtained from a radiative transfer scheme which corrects for artifacts caused by the interaction of the light with the filter material (Petzold and Schönlinner, 2004).

Black carbon is the most efficient light-absorbing aerosol species in the visible spectral range. The relationship between the aerosol absorption coefficient b_{abs} (m^{-1}) and the corresponding black carbon mass concentration BC (g/m^3) is established by the

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mass specific absorption cross section $\sigma_{abs,BC}$ (m^2/g) via the relationship

$$b_{abs} = BC \cdot \sigma_{abs,BC} \quad (4)$$

where $\sigma_{abs,BC}$ and b_{abs} are wavelength dependent.

In order to compare the absorption coefficients measured by the three different instruments at different wavelengths the b_{abs} values were extrapolated to λ=630 nm by assuming a wavelength dependence of

$$\sigma_{abs} = const \cdot \lambda^{-1} \quad (5)$$

This assumption is found to be justified for the JFJ background aerosol (Collaud Coen et al., 2004). The MAAP instrument was chosen as the reference instrument because of its improved correction algorithm and high sensitivity. The measured b_{abs} of the PSAP and the Aethalometer were extrapolated to the MAAP wavelength of 630 nm.

A thermo-optical method (OC/EC analyzer, SUNSET laboratory) was used to determine the mass specific absorption cross section (σ_{abs}) assuming that elemental carbon (EC) is equal to BC. For the PSAP, a σ_{abs} of 8.5 m^2/g was found in winter at 580 nm. For the MAAP a σ_{abs} of 7.6 m^2/g was obtained in winter and 11 m^2/g in summer at 630 nm (for the common comparison wavelength of 550 nm this would be 8.7 m^2/g in winter and 12.6 m^2/g in summer, assuming a λ^{-1} dependence; details can be found in Cozic et al. (2006b)[5].

Aerosol absorption in the visible spectrum is usually associated with carbonaceous aerosol, except during desert dust episodes when hematite (Fe_2O_3), a minor component of mineral dust, makes a small but significant contribution (Collaud Coen et al., 2004). Because of the relatively low mass specific absorption cross section of mineral dust (σ_{abs} (550 nm)~0.02 to 0.1 m^2/g (Clarke and Charlson, 1985)), the overall contribution of this component to the absorption coefficient is expected to be small during

[5]Cozic, J., Verheggen, B., Steinbacher, M., Hüglin, C., Bower, K., Crosier, J., Baltensperger, U., and Weingartner, E.: Mass closure of free Tropospheric aerosol for TSP and PM1 at the high alpine site Jungfraujoch, in preparation, 2006b.

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periods that are not influenced by Saharan dust events. Such Saharan dust episodes occur at the Jungfraujoch site on average 24 times a year (ranging from 10 to 34 per year in the last 5 years) with different duration (Collaud Coen et al., 2004), They were excluded from the following analysis by considering the wavelength dependence of the single scattering albedo as discussed by Collaud Coen et al. (2004). Another component suspected to be able to contribute to the measured absorption coefficient is light absorbing organic material. Again the low mass specific absorption cross section associated with organic compounds (e.g. for HULIS σ_{abs} (532 nm) ~0.03 m^2/g (Hoffer et al., 2005)) would require unrealistically high concentrations of these compounds to significantly contribute to the measured absorption coefficient. We therefore assume that outside Saharan dust episodes, the measured absorption coefficient is entirely due to absorption by BC.

2.4 Particle size distribution measurements

Two Scanning Mobility Particle Sizers (SMPS, TSI 3934), consisting of a DMA (TSI 3071) and CPC (TSI 3022), were used to measure the particle size distribution between 17 and 900 nm (dry) diameter, which are discussed by Verheggen et al. (2006)[3]. One SMPS was automatically switched every 6 min between the total (TOT) and interstitial inlets (INT), whereas another one was dedicated to the ICE-CVI inlet (winter 2004 and 2005 only).

During cloud-free conditions the response of the TOT and INT inlets should be identical. The INT spectrum was corrected towards the TOT spectrum by a size dependent correction factor for the small systematic difference between the two (INT was always ≤5% lower than TOT), as particle losses and/or artifacts due to a slight under-pressure were deemed more likely in the INT than in the TOT inlet.

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2.5 Cloud microphysical measurements

Cloud particles in the size range 10 to 2300 μm were imaged with a Cloud Particle Imager (CPI, SPEC Inc. 230X) (winter campaigns only). The CPI records images of individual cloud particles that pass through its sample volume. In general, the images are oversized from their true size as they are a finite distance from the object plane of the imaging optics. This means that the standard processing of the CPI data yields only qualitative size distributions. For our dataset, a semi empirical calibration (described by Connolly, 2006, and Connolly et al., 2006[6]) was used to correct the particle size and accurately define the probe sample volume. This method enables quantitative measurements of cloud particle size distributions to be made with the CPI. Another aspect that has been assumed to potentially affect microphysical aircraft probe data is the shattering of large ice crystals as they impact on the probe inlet. Since our measurements were made from a ground-based platform, droplet and/or crystal shattering due to high wind speeds (as aircraft measurements are subjected to) was minimal. Nevertheless, periods when shattering might have occurred, as evident from the measured images, were removed from further analysis.

The volume of each imaged particle, necessary to compute the ice water content (IWC), was inferred from the 2-dimensional CPI images based on the empirical size-mass relationships given by Heymsfield et al. (2004) and Mitchell (1996). Spherical particles were assumed to be predominantly liquid and were thus excluded from the IWC. A comparison of the CPI derived IWC with the condensed water content measured downstream of the Ice-CVI confirms this assumption to be valid for cloud particles up to 25 μm in diameter, which is the size region where most spherical particles reside (Mertes et al., 2006[2]).

A Forward Scattering Spectrometer Probe (FSSP-100; modified Model SPP100) was

[6] Connolly, P. J., Flynn, M. J., Ulanowski, Z., Choularton, T. W., and Gallagher, M. W.: Calibration of 2-D imaging probes using calibration beads and ice crystal analogues. Part 1: The depth-of-field, J. Atmos. Oceanic Technol., submitted, 2006.

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also employed, measuring the size distribution of cloud hydrometeors with diameters between 2 and 47 μm. Analysis of these data show that in mixed phase clouds, contamination from ice crystals is small. In any case, the response of the probe to ice is to produce a random distribution of sizes. Hence, the liquid spectrum is still clearly visible as it obeys a near lognormal spectral shape. Again, many potential problems with the inlet may be overcome by the ground based operation of the probe.

Cloud liquid water content (LWC) was measured with a Particulate Volume Monitor (PVM-100, Gerber Scientific) at 1 min time resolution. The scattered light of a laser beam is related to LWC. The instrument response was calibrated typically every cloud-free day. When the zero-reading became higher than a certain threshold value (as recommended by the manufacturer) or when the detection unit was clogged with snow, the PVM was taken inside to defrost and the lens was cleaned. The response of the PVM to the presence of ice was quantified and empirically corrected by a combined analysis of the FSSP, PVM and CPI data (Verheggen et al., 2006[3]).

These measurements allow for the evaluation of the ice mass fraction (IMF) defined as IWC/(IWC+$LWC_{corrected}$). In the following analysis, a 10-min time interval is classified as being "in-cloud" when the 15th percentile of the Cloud Water Content (CWC=IWC+$LWC_{PVM(corrected)}$) is larger than 0.02 g/m^3 and the average CWC>0.05 g/m^3. This criterion is chosen to ascertain that measurements made in cloud edges, patchy clouds, and regions influenced by entrainment are excluded from the analysis as much as possible.

3 Results and discussions

3.1 Comparison of BC measurements and characterization of the inlets

In order to validate the measurements of the BC mass concentrations obtained with the different types of instruments, the MAAP, the PSAP and the Aethalometer connected to the total inlet were intercompared. Although the Aethalometer and the

PSAP were not used for the determination of the scavenged fraction, they were included in this intercomparison in order to validate the long-term measurements of the Aethalometer performed within the GAW program. For this purpose, the measured b_{abs} of the PSAP and the Aethalometer were extrapolated (with Eqs. 2 and 3) to the MAAP wavelength of 630 nm. The comparison of the measured absorption coefficients by linear regression was performed for the two campaigns (winter 2004 and 2005) when all the instruments were operated, representing a total of 1626 h of measurements in-cloud and out-of-cloud. The correlation between the PSAP and the MAAP (see Fig. 2), for a common time base of 1 h, is very high (r^2=0.97; $b_{abs(PSAP)}$=0.99 $b_{abs(MAAP)}$+3.64×10^{-8} m^{-1}). There is also a high correlation between the Aethalometer and the MAAP, even though the slope somewhat differs from 1 (r^2=0.94; $b_{abs(Aethalometer)}$=1.32 $b_{abs(MAAP)}$+1.48×10^{-8} m^{-1}). Considering the measurement uncertainties, these graphs show a very good agreement between all three types of instrument. Nevertheless it is important to note that the Aethalometer measured approximately 30% larger absorption coefficients than the MAAP and the PSAP in March 2004 and February/March 2005. A similar result has been observed by Petzold and Schönlinner (2004) during an intercomparison between MAAP and Aethalometer, at the same site, in March/April 2003, where they found that the Aethalometer overestimates b_{abs} by 21% compared to the MAAP. The overestimation in our case may be attributed to a low value of the correction factor C (Eq. 3) applied for the JFJ conditions. Considering these results it would be necessary to use C=2.83 instead of 2.14 for the JFJ.

The evaluation of the scavenged BC fraction required an assessment of artifacts induced by the sampling procedures at each inlet. Under clear sky conditions (period for which the average cloud water content (CWC) during 10 min was below 0.01 g/m^3), the validation of the quantitative sampling of submicrometer aerosol behind the two inlets and for the instruments used for BC mass concentration determination was done by using two MAAP instruments running downstream of the total and the interstitial inlets. Figure 3 shows the comparison of two identical instruments (MAAP/MAAP)

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behind the total and the interstitial inlets. As can be seen, the comparison of BC mass concentrations from the two MAAP's shows good agreement (r^2=0.97; BC_{INT}=0.95 BC_{TOT}+1.64 ng/m^3). The BC concentrations of the interstitial inlet are slightly lower that those of the total inlet, which may be due to a slight under-pressure in the interstitial inlet caused by the 2-μm cut of the cyclone. Alternatively, the difference could be caused by the existence of some BC in coarse mode particles which were removed by the 2-μm cut of the cyclone. In order to provide the best evaluation of the BC scavenged fraction, the interstitial MAAP data "in-cloud" were divided by a factor of 0.95, to correct for this deviation, since sampling artifacts were deemed the least likely in the total inlet.

3.2 Concentrations of BC (in-cloud and out-of-cloud)

A mass closure was carried out under dry conditions by relating the BC concentration, the measured ionic chemical composition of the aerosol, its organic carbon fraction (determined with an OC/EC thermo-optical analyzer) and the submicrometer aerosol mass concentration (PM1, measured with a beta gauge, Thermo ESM Andersen FH62 I-R) on a time base of 24-h averages (Cozic et al., 2006b[5]). The average BC contribution to PM1 was found to be 2.5% in summer and 4.7% in winter.

Table 2 compares in-cloud and out-of-cloud BC concentrations measured behind the total and the interstitial inlets. As mentioned above, total and interstitial BC concentrations are very similar during out-of-cloud conditions. Higher concentrations are observed in summer than in winter for the out-of-cloud periods, which can be explained by the greater influence of boundary layer injections. During in-cloud periods the BC mass concentrations differ quite significantly between the total and the interstitial inlet: Here, the total BC concentrations are on average 1.2 and 1.8 times higher than the interstitial BC concentration for winter and summer, respectively. This is an indication that the fraction of BC containing particles in cloud droplets or ice crystals is significant (10.1 ng/m^3 in winter and 22.3 ng/m^3 in summer).

BC concentrations found at the Jungfraujoch (see Table 2) are similar to those found

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at other remote sites like the Arctic (3–174 ng/m^3), Antarctic (1.5–300 ng/m^3) and marine environments (0–500 ng/m^3) (see Krivacsy et al., 2001, and references therein).

3.3 Scavenged BC fraction

The scavenged fraction of BC ($F_{Scav,BC}$) is defined as the fraction of BC that has been incorporated into cloud droplets or ice crystals. The mass concentration of black carbon in hydrometeors is approximated by the difference between the concentrations downstream of the total and the interstitial inlets, and thus:

$$F_{Scav,BC} = \frac{BC_{cloud}}{BC_{total}} = \frac{(BC_{total} - BC_{interstitial})}{BC_{total}} \quad (6)$$

Figure 4 shows an example of the evolution of the total and interstitial BC concentrations along with the evolution of the cloud liquid water content in the case of a liquid cloud. When clouds are absent BC concentrations behind the two inlets are similar. During cloud presence, BC scavenging is observed by a decrease in the BC concentration in the interstitial phase.

The scavenged aerosol volume fraction was calculated from the measured size distributions downstream of each inlet, analogous to the scavenged BC fraction. Comparing these two provides information on the role of BC in cloud droplet activation (this paper) and heterogeneous freezing (Cozic et al., 2006a[1]).

3.4 The influence of environmental parameters on the scavenged BC fraction

A total of 1000 h of cloud periods were analyzed during winter 2004 (334 h), summer 2004 (373 h) and winter 2005 (293 h). This corresponds to an average cloud occurrence of 28%, with a larger cloud presence in winter than in summer.

The influence of the cloud liquid water content, BC mass concentration, temperature and cloud microphysics is analyzed in the next section. Several other factors were also considered, such as the wind direction, wind speed, time of the day, the wavelength

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used by the Aethalometer, but no clear influence on the scavenged BC fraction was observed for all these parameters. As no data on the updraft velocity in the cloud were available, this parameter was not evaluated in this study even though it is an important parameter in determining the available water vapour supersaturation in clouds.

In the following analysis, graphs are shown separately for low temperature (T <–5°C) and for high temperature (T >–5°C) to distinguish between (potentially) mixed-phase and liquid clouds, respectively.

3.4.1 Influence of the Liquid Water Content (LWC)

The dependence of the scavenged BC fraction on the liquid water content is shown in Fig. 5. It can be seen that $F_{Scav,BC}$ increases with increasing LWC and that it is lower for T <–5°C (b) than for T >–5°C (a), but the observed dependence is also weaker at low temperature. At cold temperatures (b) other factors limit the scavenged fraction more strongly than LWC does, as will be discussed below.

Figure 5a shows that the scavenged BC fraction increases for increasing LWC until reaching a plateau (of 60%) for high LWC (>0.4 g/m^3). At low LWC, activation of (BC containing) particles is limited by the availability of condensable water vapour. This probably relates to LWC increasing with increasing droplet number, which in turn increases with increasing aerosol number until at high concentrations there is insufficient water vapour to allow further droplet activation and hence further BC scavenging (see Sect. 3.4.2).

The increase of the scavenged BC fraction with increasing LWC has also been observed at other mountain sites such as Mt. Sonnblick (Hitzenberger et al., 2000; Kasper-Giebl et al., 2000) and Rax (Hitzenberger et al., 2001), but not at Puy de Dôme (Sellegri et al., 2003). For both the Rax and Mt. Sonnblick an increase of the scavenged fraction up to a plateau of 0.8 for a LWC around 0.3 g/m^3 was observed. This is in accordance with the results obtained here where we find a plateau for LWC around 0.25 g/m^3.

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3.4.2 Influence of the total BC mass concentration

Figure 6 shows a decrease of the scavenged fraction with increasing total BC concentration. BC mass concentrations are correlated with aerosol number concentrations, N_{total}, of the Jungfraujoch aerosol. At high concentration, for temperatures above −5°C (a), corresponding to summer data only, a larger number of available CCN may deplete the available water vapour more rapidly and decrease the super-saturation to below critical levels. As a result, the relative increase of the number of activated particles is smaller than that of the total number of particles, resulting in a lower scavenged fraction.

In winter (b), the presence of ice particles in the cloud keeps $F_{scav,BC}$ low except when BC_{total} (and thus N_{total}) is extremely low. In such clean conditions, the supersaturation is able to rise, allowing a larger fraction of aerosol particles to be activated, which results in a higher value of $F_{scav,BC}$.

In previous studies, the influence of the BC total concentration was investigated with sometimes different results. In the Po valley (Hallberg et al., 1992), at Kleiner Feldberg (Hallberg et al., 1994) and at Great Dun Fell (Gieray et al., 1997) a decrease of the scavenged fraction with increasing BC mass concentration was observed as in the present study. In contrast, no dependence appeared at Mt. Sonnblick (Hitzenberger et al., 2000; Kasper-Giebl et al., 2000) and Rax (Hitzenberger et al., 2001). The trend at T>–5°C is not very strong, which may explain the different results of the various studies.

3.4.3 Influence of temperature

The main interest of this study relates to the influence of the coexisting ice and liquid phases on the scavenged BC fraction. The influence of the ambient temperature is one of the parameters showing this effect (Fig. 7). The scavenged BC fraction decreases with decreasing temperature, from 60% in liquid clouds in summer (at T=0°C) down to <10% in mixed phase clouds in winter. This observed trend is explained with the

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Wegener-Bergeron-Findeisen process: In a mixed-phase cloud there is a flux of water vapour from the liquid phase to the ice phase due to the lower saturation vapour pressure over ice than that over liquid water. This leads to the evaporation of liquid droplets, thus releasing the aerosol material back into the interstitial phase. This in turn results in a decreased scavenged fraction, since ice nuclei (IN) (and thus ice crystals) are much less numerous than CCN. So many cloud droplets are evaporated to the benefit of the formation and growth of a few ice crystals.

At higher temperatures, $T>0^{\circ}$C, a reverse trend is observed (Fig. 7). The lowest values of the scavenged fraction can be explained by the fact that very high aerosol number concentrations ($N_{tot}>500\,cm^{-3}$) coincide with higher temperatures, which leads to a limitation of water vapour available to activate more ambient particles, as shown for the BC mass concentration in Fig. 6a.

The highest scavenged fraction (around 60%) which corresponds to a liquid cloud not influenced by high aerosol concentrations is quite high and is explained by the existence of internally mixed aerosol particles as shown in Table 1 above. Nevertheless a comparison of the scavenged fraction found at the Jungfraujoch with other studies is quite difficult since no information on the average temperature is given in the other studies.

Summarizing, the influence of LWC and BC total concentration on the BC scavenged fraction is strongest for low LWC and low BC concentration (see Figs. 5 and 6). The observed increase in $F_{Scav,BC}$ with increasing temperature is consistent with the Wegener-Bergeron-Findeisen process.

3.4.4 Influence of ice mass fraction

The scavenged BC fraction dependence on temperature was explained by the presence of ice crystals and the subsequent Wegener-Bergeron-Findeisen process. Therefore, an analysis of the influence of the ice mass fraction is presented here.

The scavenged BC fraction decreases with increasing cloud ice mass fraction (Fig. 8), as expected from the Wegener-Bergeron-Findeisen process. The best fit

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through the data of Fig. 8 is given by

$$F_{\mathrm{Scav,BC}} = 0.16 + 0.94\exp(-12.27\sqrt{\mathrm{IMF}}) \quad (7)$$

This observed trend is similar to the one observed for the bulk aerosol at low and intermediate ice mass fraction (Verheggen et al., 2006[3]) but starts to differ at high ice mass fraction where the scavenged fraction of the bulk aerosol volume, $F_{\mathrm{Scav,V}}$, decreases to near-zero.

$$F_{\mathrm{Scav,V}} = 0.02 + 0.83\exp(-6.43\sqrt{\mathrm{IMF}}) \quad (8)$$

An $F_{\mathrm{Scav,BC}}$ value of 0.05–0.1 at IMF=0.8–1 indicates that 5–10% of the BC mass concentration is found in the ice residuals. According to Eqs. (7) and (8), for an IMF=1 16% of the BC mass concentration is found in the ice residuals whereas only 2% of the aerosol mass is found in the small ice crystals. This indicates that compared to the bulk aerosol BC is enriched in the ice phase. This enrichment of BC in the ice residuals was confirmed by a detailed analysis of the fraction of BC in small ice particles (Cozic et al., 2006a[1]).

3.4.5 Relation between BC and bulk aerosol scavenging

In parallel to this study the scavenged volume and number fractions of aerosol particles were investigated (Verheggen et al., 2006[3]). The scavenged volume fraction was defined as the ratio of particle volume scavenged into cloud droplets (obtained from total minus interstitial) to the total volume of particles. The dependence of the scavenged volume and number fraction on temperature, total particle number, LWC and ice mass fraction shows similar trends as described here for BC (Verheggen et al., 2006[3]).

Figure 9 shows the comparison between the scavenged BC fraction and the scavenged volume fraction separately for $T<-5^{\circ}$C and $T>-5^{\circ}$C. The very high correlation for higher temperatures (r^2=0.99; $F_{\mathrm{Scav,BC}}$=0.94 $F_{\mathrm{Scav,V}}$) indicates that in liquid clouds BC behaves like the bulk aerosol. This is due to aged BC which exhibits a greater hygroscopicity and therefore enhances the transfer of BC into cloud droplets via nucleation

scavenging. At low temperatures, the correlation between the two scavenged fractions is slightly lower (r^2=0.81; $F_{Scav,BC}$=0.88 $F_{Scav,V}$). It has to be noted that for these values low concentrations below 40 ng/m^3 (empty circles) were not considered because they were generating a lot of scatter without changing the correlation. Including all points, the correlation was still quite high but with a lot more of scatter at low temperatures (r^2=0.245; $F_{Scav,BC}$=0.86 $F_{Scav,V}$) and no real difference at high temperatures (r^2=0.95; $F_{Scav,BC}$=0.95 $F_{Scav,V}$).

4 Summary

The influence of environmental parameters on the scavenged BC fraction in liquid and mixed phase clouds was studied. The measurements showed that the scavenged BC fraction increases with increasing liquid water content for values of LWC up to 0.13 g/m^3, and decreases with increasing BC concentration for a total BC concentration up to 35 ng/m^3. It was found that for a large temperature range (between −25°C and 0°C) the scavenged BC fraction increases with increasing temperature, up to 61% in summer. This decrease of the scavenged BC fraction with decreasing temperature can be explained by the evaporation of liquid droplets in the presence of ice crystals (Wegener-Bergeron-Findeisen process). This was confirmed by the influence of the ice mass fraction on the scavenged BC fraction. The scavenged BC fraction showed similar dependences on temperature, total particle number, LWC, and ice mass fraction, as the dependence of the scavenged aerosol volume and number fraction on the same parameters (Verheggen et al., 2006[3]). BC was found to be scavenged into the cloud phase to the same extent as the bulk aerosol due to aged BC exhibiting a larger hygroscopicity which enhances the transfer of BC into cloud droplets via nucleation scavenging. Nevertheless for high ice mass fractions BC was found to be enriched in the ice phase compared to the bulk aerosol.

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Table 1. Average scavenged BC fraction from the literature.

Sampling site	$F_{Scav,BC}$	Type of site	Papers	Conditions
Po Valley (Italy) °	0.06	Urban	Hallberg et al. (1992)	November 1989
Kleiner Feldberg (Germany) °	0.15	Rural	Hallberg et al. (1994)	
Puy de Dôme (France)	0.33	Mid altitude (1465 m)	Sellegri et al. (2003)	Feb–April 2001
Mt. Sonnblick (Austria) °	0.45	High altitude (3106 m)	Kasper-Gielb et al. (2000)	September 1995
Rax (Austria) °	0.54	Mid altitude (1644 m)	Hitzenberger et al. (2001)	March 2000–April 1999
Great Dun Fell (UK)	0.57	Rural – Coastal	Gieray et al. (1997)	April–May 1993
Jungfraujoch (Switzerland) *	0.61	High altitude (3850 m)	This work	July–August 2004
Mt. Sonnblick (Austria) °	0.74	High altitude (3106 m)	Hitzenberger et al. (2000)	September 1996 & April–May 1997
Spitzbergen (Norway)	0.80	Artic	Heintzenberg and Leck (1994)	Winter and summer (1990–1992)

* summer liquid cloud only
° scavenged fraction calculated with cloud liquid impactor

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Table 2. Average BC mass concentrations (total, interstitial) out-of-cloud and in-cloud for winter and summer, as determined with the MAAP.

[ng/m^3]	Out-of-cloud		In-cloud	
	winter	summer	winter	summer
$BC_{Total(MAAP)}$	54.2	64.9	81.4	50.3
$BC_{Interstitial(MAAP)}$	56.0	65.9	71.3	28.0

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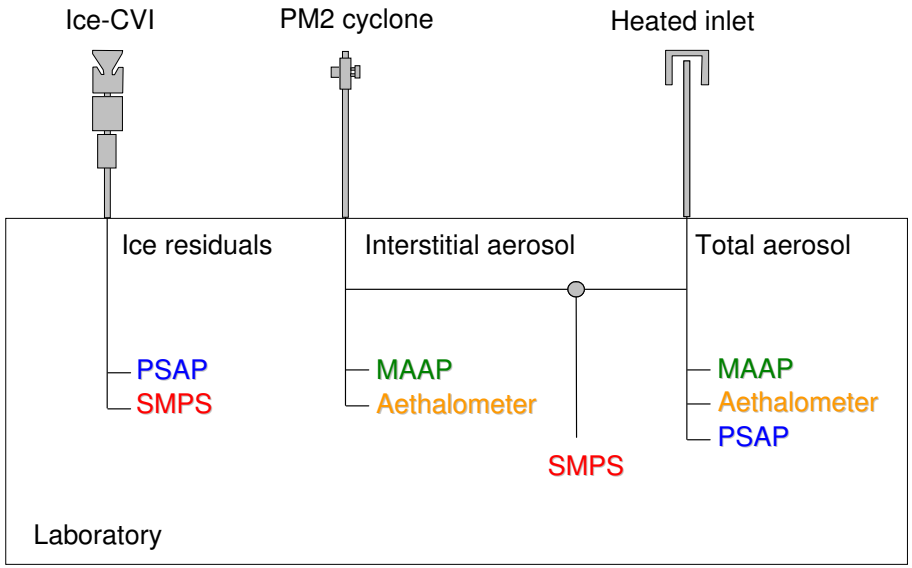

Fig. 1. Inlet systems (Total, Interstitial and Ice-CVI) and instruments employed during CLACE campaigns.

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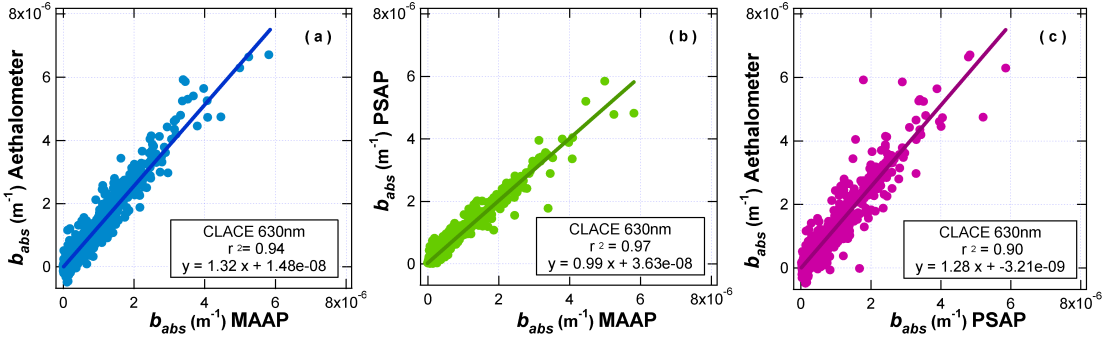

Fig. 2. Comparison of the absorption coefficients measured by the MAAP, the Aethalometer, and the PSAP) downstream of the total inlet. Symbols represent 1-h averages (total 1626 h).

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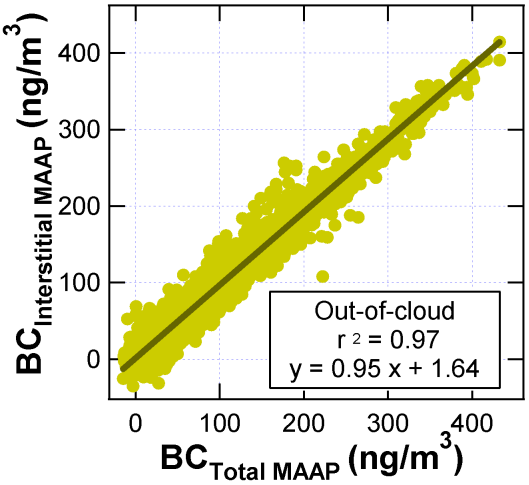

Fig. 3. Comparison of the total and interstitial inlets during clear sky conditions (CWC<0.01 g/m^3) for 1800 h of the MAAP. Symbols represent 10-min averages.

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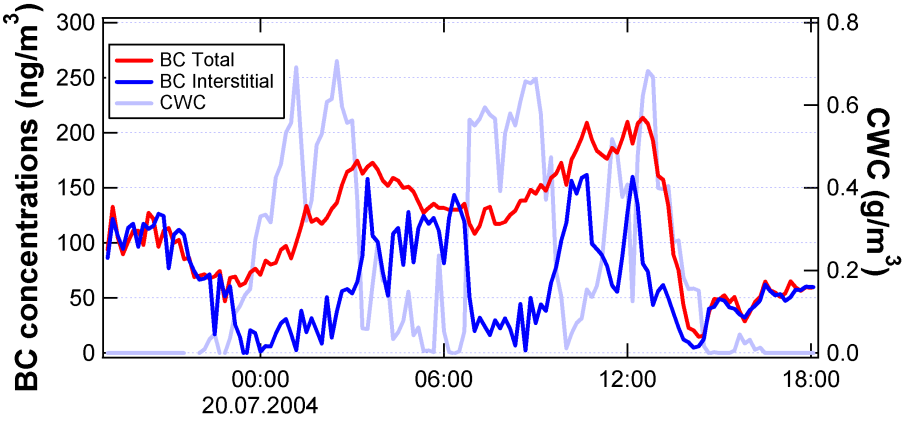

Fig. 4. Evolution of the total and the interstitial concentration of black carbon with the evolution of the cloud water content (CWC) for a liquid cloud.

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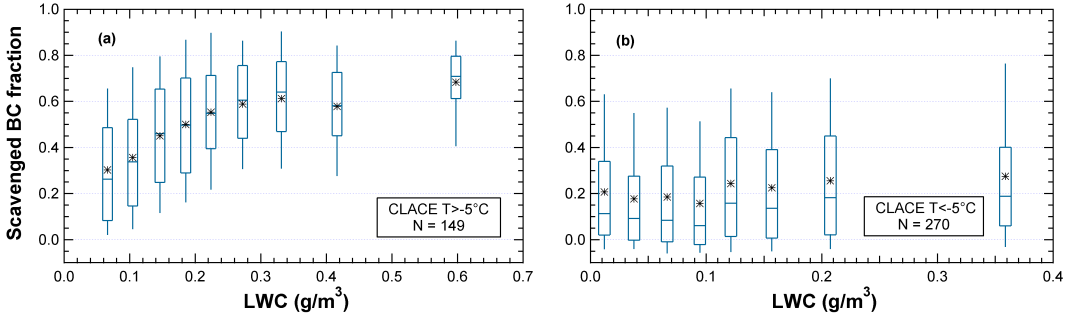

Fig. 5. Influence of the LWC on the scavenged BC fraction based on 359 h at $T > -5^{\circ}$C **(a)** and 254 h at $T < -5^{\circ}$C **(b)** of in cloud measurements. N represents the number of 10-min averages per box plot.

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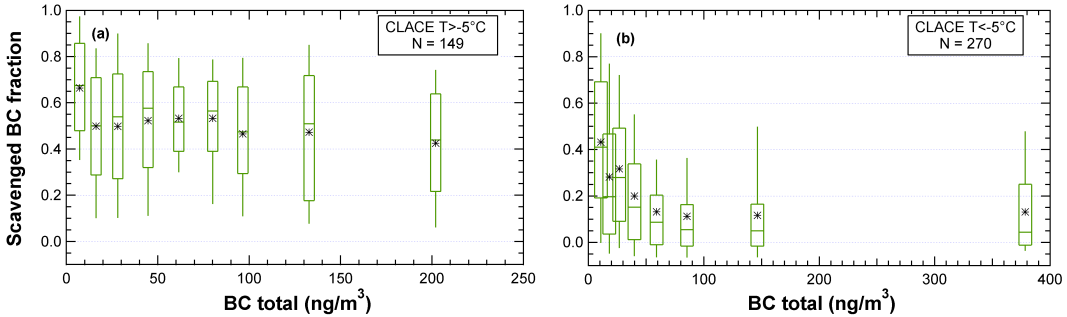

Fig. 6. Dependence of the scavenged BC fraction on the BC concentration measured downstream of the total inlet based on 359 h at $T > -5^{\circ}$C **(a)** and 254 h at $T < -5^{\circ}$C **(b)** of in cloud measurements.

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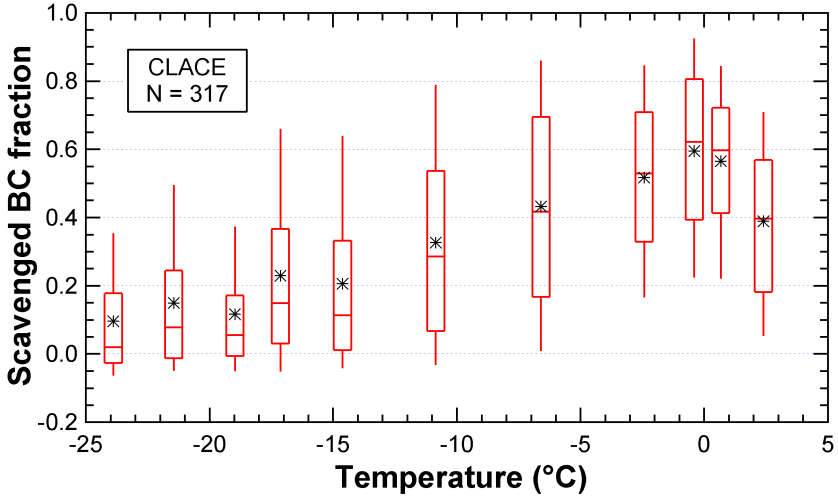

Fig. 7. Dependence of the scavenged BC fraction on temperature based on 581 h of in cloud measurements.

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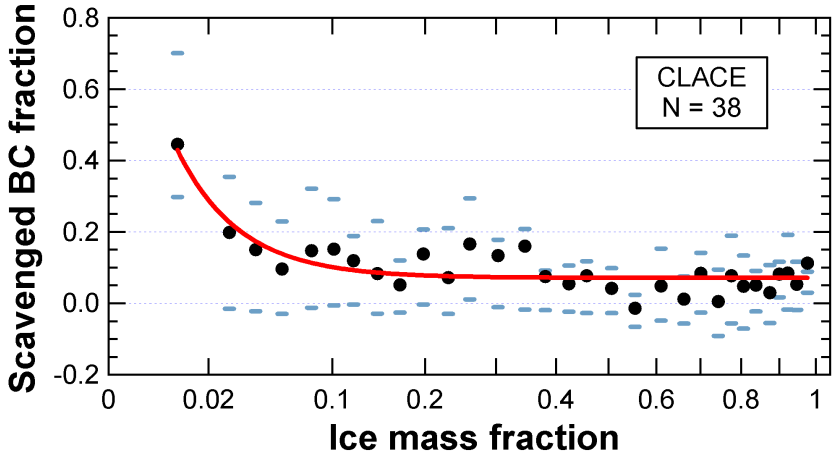

Fig. 8. Dependence of the scavenged BC fraction on the ice mass fraction based on 202 hours in winter campaigns. Circles denote the average, while horizontal lines denote the 25 and 75 percentile values. X-axis is presented on a quadratic scale for clarity.

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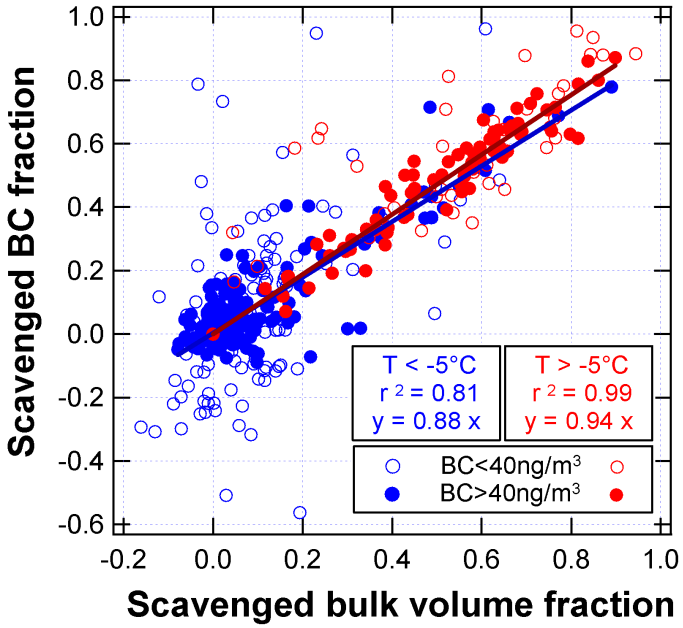

Fig. 9. Correlation between the scavenged BC fraction and the scavenged bulk volume fraction for $T < -5^{\circ}$C (blue dots) and $T > -5^{\circ}$C (red dots). Data with low concentrations (open circles; BC concentrations $<40\,\mu g/m^3$) were excluded from the correlation analysis.

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