Scavenging of black carbon by ice crystals over the northern Pacific

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[1] Airborne measurements over the northern Pacific are evaluated to characterize properties of black carbon (BC) in cirrus crystal residuals and background aerosols in the upper troposphere. Although the mass and number concentrations of BC were 7–25 times lower in crystal residuals than in particles outside of cloud, twice as many of the residuals (31%) contained BC compared to 17% of the particles in cloud-free air. In addition the average mass equivalent diameter (MED) of BC was 10% larger in crystal residuals than in particles outside of cloud. These differences in crystal residuals compared to the background aerosols suggest that inertial scavenging is a significant mechanism for removing BC compared to removal by nucleation. The scavenging efficiency, calculated as the BC mass in condensed cloud water, ranged from 7–44 ng of carbon per gram of water, consistent with previous studies in marine stratus clouds. **Citation:** Baumgardner, D., R. Subramanian, C. Twohy, J. Stith, and G. Kok (2008), Scavenging of black carbon by ice crystals over the northern Pacific, *Geophys. Res. Lett.*, *35*, L22815, doi:10.1029/2008GL035764.

1. Background

[2] Black carbon (BC) is one of the most ubiquitous and radiatively active components of aerosol particles in the atmosphere. Even though the mass of BC in the upper troposphere is less than 10% of the mass in the boundary layer [*Schwarz et al.*, 2006], it significantly augments climate warming, as recently highlighted by *Ramanathan and Carmichael* [2008] who estimate that BC emissions are currently the second strongest contributor to global warming after carbon dioxide. One of the major problems in assessing the climate impact of BC is the large uncertainty associated with the atmospheric residence time of these particles, compounded by a paucity of measurements, especially in the upper troposphere and lower stratosphere (UT/LS).

[3] Clouds clearly play a role in removing BC particles as indicated by the BC measured in water droplets [*Twohy et al.*, 1989; *Chýlek et al.*, 1996] and ice crystals [*Ström and Ohlsson*, 1998; *Petzold et al.*, 1998; *Twohy and Poellot*, 2005; *Cozic et al.*, 2007, 2008]. There is much more BC in the UT/LS than can be explained by emissions from aircraft [*Hendricks et al.*, 2004], an indication that removal by precipitation may be less efficient than assumed. Nucleation removes only a single aerosol particle for each cloud droplet or ice crystal whereas multiple aerosols can be removed through inertial scavenging by a cloud hydrometeor. BC on ice crystal surfaces, due to inertial scavenging, will likely change the crystal optical properties differently than those inside a crystal formed by nucleation. Hence, a better understanding of the removal mechanisms of BC by clouds will improve our ability to predict the climate impacts of BC.

2. Methodology

[4] The instrument used for measuring the mass concentration of BC contained in individual particles is the Single Particle Soot Photometer (SP2), where BC is as defined by *Schwarz et al.* [2006]. Particles enter the sample cavity of the SP2 and while passing through the beam of a diode-pumped Nd:YAG laser (1.064 μm wavelength) they scatter and absorb light. Particles that contain light absorbing material at the wavelength of the laser are heated and reach a temperature at which they incandesce. The intensity of the emitted light is proportional to the mass of the light absorbing material [*Schwarz et al.*, 2006]. The SP2, as operated during the project described below, measured BC with a mass equivalent diameter (MED) between approximately 80 to 190 nm (±20 nm). (The MED is the diameter of a sphere with the measured BC mass and assumed density of 1.9 g cm^{-3}, as suggested by *Bond and Bergstrom* [2006].) The estimated uncertainty in mass concentration is ±36% [*Gao et al.*, 2008].

[5] Particles containing BC with equivalent diameters larger than 190 nm are counted but, since they saturate the detector, an equivalent diameter can not derived. Particles that saturate the incandescence detector are included in the analysis, described below as "saturated", and the mass is set to the maximum measurable value. The SP2 was mounted on the National Center for Atmospheric Research Gulfstream-V aircraft and sampled either from a forward facing inlet or from a counterflow virtual impactor (CVI) that separated cloud particles from interstitial aerosols and evaporated them using dry, heated nitrogen [*Twohy et al.*, 1997]. The condensed ice water content (CIWC) is derived continuously from the resulting water vapor measured with a tunable diode laser and the non-volatile residuals of the ice crystals were sampled with the SP2.

[6] Aerosols and crystals smaller than approximately 5 μm will not enter the CVI inlet and the concentration of larger crystals that do enter are enhanced relative to the ambient population as a result of the subisokinetic flow. The enhancement aids detection and is a factor of 30 to 50 depending on the air velocity and density and the cloud particle size. The data have been adjusted to account for this enhancement.

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Table 1a. Summary of BC Concentrations Inside and Outside of Clouds Averages and Standard Deviations[a]

Date	Type	IWC	T	Number Concentration No BC (cm^{-3})			Number Concentration BC (cm^{-3})			BC Mass Concentration ($ng\ m^{-3}$)			BC Saturated Mass Concentration ($ng\ m^{-3}$)		
				In Cloud	No Cloud	Ratio In/Out	In Cloud	No Cloud	Ratio In/Out	In Cloud	No Cloud	Ratio In/Out	In Cloud	No Cloud	Ratio In/Out
4/29	EP	**0.08**	**−31**	**0.5** 0.2	**18.3** 1.8	**0.02**	**0.3** 0.4	**4.5** 0.5	**0.07**	**3.7** 4.8	**22.8** 4.1	**0.16**	**1.0** 1.4	**3.6** 1.1	**0.29**
5/5	CPS	**0.08**	**−34**	**0.3** 0.3	**10.5** 1.5	**0.03**	**0.1** 0.1	**2.5** 0.4	**0.05**	**1.4** 0.8	**12.2** 3.0	**0.11**	**0.4** 0.2	**2.0** 0.7	**0.19**
5/5	CPS	**0.13**	**−25**	**0.5** 0.5	**35.9** 15.0	**0.01**	**0.2** 0.1	**5.4** 2.0	**0.03**	**1.5** 1.1	**36.5** 15.9	**0.04**	**0.4** 0.3	**6.3** 3	**0.07**
5/5	CPS	**0.15**	**−25**	**0.6** 0.4	**19.4** 2.3	**0.03**	**0.2** 0.1	**4.2** 0.7	**0.05**	**2.2** 1.0	**24.8** 5.0	**0.09**	**0.6** 0.3	**4.2** 1.2	**0.15**
5/14	EPS	**0.06**	**−40**	**0.2** 0.1	**28.9** 12.1	**0.01**	**0.1** .04	**3.2** 0.8	**0.04**	**0.8** 0.2	**19.1** 6.3	**0.04**	**0.2** 0.0	**3.1** 1.2	**0.06**
5/19	WPS	**0.13**	**−29**	**1.1** 1.6	**9.9** 1.6	**0.11**	**0.1 0.1**	**2.2** 0.3	**0.04**	**1.0** 0.7	**8.5** 1.9	**0.11**	**0.3** 0.2	**1.2** 0.6	**0.21**
5/22	WPSD	**0.07**	**−34**	**0.7** 0.4	**91.2** 5.1	**0.01**	**0.2** 0.1	**12.7** 1.1	**0.01**	**1.3** 0.8	**88.6** 9.3	**0.01**	**0.3** 0.2	**15.8** 2.2	**0.02**
5/22	WPSD	**0.07**	**−27**	**0.3** 0.1	**13.9** 1.2	**0.02**	**0.1** 0.1	**3.1** 0.4	**0.04**	**1.0** 0.6	**17.1** 3.2	**0.06**	**0.3** 0.1	**2.9** 0.8	**0.09**
5/22	WPSD	**0.12**	**−26**	**1.1** 0.9	**22.9** 5.0	**0.05**	**0.1** 0.1	**5.2** 0.9	**0.02**	**1.2** 0.5	**29.9** 6.2	**0.04**	**0.3** 0.1	**5.1** 1.6	**0.06**
5/23	WPDP	**0.13**	**−30**	**0.8** 0.9	**21.3** 4.0	**0.04**	**0.1** 0.1	**3.9** 0.9	**0.03**	**1.3** 1.5	**23.7** 6.0	**0.05**	**0.3** 0.4	**4.1** 1.5	**0.08**
5/23	WPDP	**0.24**	**−30**	**1.3** 2.8	**4.0** 1.4	**0.33**	**0.1** 0.1	**1.4** 0.4	**0.09**	**1.1** 1.0	**6.6** 2.8	**0.17**	**0.3** 0.2	**1.1** 0.6	**0.28**
5/24	WPDP	**0.12**	**−27**	**0.7** 0.5	**20.8** 4.5	**0.03**	**0.1** 0.1	**3.4** 0.7	**0.04**	**1.1** 0.9	**22.0** 5.6	**0.05**	**0.3** 0.2	**4.0** 1.5	**0.07**
Avg. S.D.				**0.7** 0.7	**24.7** 4.6	**0.06** 0.09	**0.1** 0.1	**4.3** 0.8	**0.04** 0.02	**1.5** 1.2	**26.0** 5.8	**0.08** 0.05	**0.4** 0.3	**4.5** 1.3	**0.13** 0.09
Correlation in to out of cloud; N = 12; P = significance level				**−0.17** **P > 0.3**	**−0.17** **P > 0.3**	**−0.17** **P > 0.3**	**0.26** **P > 0.15**	**0.26** **P > 0.15**	**0.26** P > **0.15**	**0.03** **P > 0.5**	**0.03** **P > 0.5**	**0.03** **P > 0.5**	**−0.06** **P > 0.5**	**−0.06** **P > 0.5**	**−0.06** **P > 0.5**
Correlation with IW; N = 924				**0.61** **P < 0.001**	**0.61** **P < 0.001**	**0.61** **P < 0.001**	**0.26** **P < 0.001**	**0.26** **P < 0.001**	**0.26** **P < 0.001**	**0.25** **P < 0.001**	**0.25** **P < 0.001**	**0.25** **P < 0.001**	**0.24** **P < 0.001**	**0.24** **P < 0.001**	**0.24** **P < 0.001**

[a]Averages are given in bold. EP, East Pacific; EPS, East Pacific Storm; CPS, Central Pacific Storm; WPS, West Pacific Storm; WPSD, West Pacific Storm with Dust; WPDP, West Pacific Dust and Pollution.

Table 1b. Summary of BC Derived Variables Inside and Outside of Clouds Averages and Standard Deviations[a]

Date	Seconds in Cloud	Type	IWC	T	BC Mass Equivalent Diameter (nm)			BC Number Fraction			BC Mass to Ice Water Mass $(ngC)(gIWC)^{-1}$
					In Cloud	No Cloud	Ratio In/Out	In Cloud	No Cloud	Ratio In/Out	
4/29	500	EP	**0.08**	**−31**	**140** 6	**112** 5	**1.24**	**0.38** 0.08	**0.2** 0.02	**1.89**	**43.8** 52.8
5/5	900	CPS	**0.08**	**−34**	**137** 9	**111** 7	**1.24**	**0.36** 0.06	**0.19** 0.03	**1.88**	**16.8** 6.8
5/5	500	CPS	**0.13**	**−17**	**137** 13	**126** 7	**1.09**	**0.29** 0.08	**0.13** 0.02	**2.18**	**11.8** 6.3
5/5	400	CPS	**0.15**	**−21**	**143** 6	**119** 4	**1.2**	**0.3** 0.06	**0.18** 0.02	**1.66**	**14.9** 5.4
5/14	500	EPS	**0.06**	**−40**	**117** 9	**120** 6	**0.97**	**0.34** 0.05	**0.11** 0.03	**3.1**	**12.5** 2.6
5/19	4200	WPS	**0.13**	**−29**	**135** 15	**102** 8	**1.32**	**0.12** 0.05	**0.18** 0.04	**0.65**	**6.9** 2.6
5/22	500	WPSD	**0.07**	**−34**	**121** 7	**129** 3	**0.93**	**0.23** 0.06	**0.12** 0.01	**1.9**	**17.3** 9.5
5/22	750	WPSD	**0.07**	**−27**	**130** 9	**114** 6	**1.14**	**0.26** 0.05	**0.18** 0.02	**1.42**	**13.7** 7.2
5/22	1250	WPSD	**0.12**	**−26**	**141** 6	**119** 5	**1.19**	**0.12** 0.04	**0.19** 0.02	**0.65**	**9.6** 2.9
5/23	300	WPDP	**0.13**	**−30**	**130** 13	**120** 4	**1.09**	**0.18** 0.07	**0.16** 0.02	**1.16**	**9.3** 11
5/23	700	WPDP	**0.24**	**−30**	**128** 23	**103** 14	**1.06**	**0.16** 0.09	**0.27** 0.07	**0.61**	**4.9** 3.1
5/24	100	WPDP	**0.12**	**−27**	**132** 6	**124** 5	**1.24**	**0.15** 0.05	**0.14** 0.01	**1.04**	**8.6** 5.7
Avg. S.D.					**132** 10	**116** 6	**1.1** 0.1	**0.24** 0.06	**0.17** 0.03	**1.5** .0.7	**14.1** 9.7
Correlation In to out of cloud; N = 12					**−0.21 P > 0.3**	**−0.21 P > 0.3**	**−0.21 P > 0.3**	**−0.18 P > 0.3**	**−0.18 P > 0.3**	**−0.18 P > 0.3**	**N.A.**
Correlation with IWC; N = 924					**0.21 P < 0.01**	**0.21 P < 0.01**	**0.2 P < 0.01**	**−0.27 P < 0.001**	**−0.27 P < 0.001**	**−0.27 P < 0.001**	**−0.10 P > 0.1**

[a]Averages are given in bold.

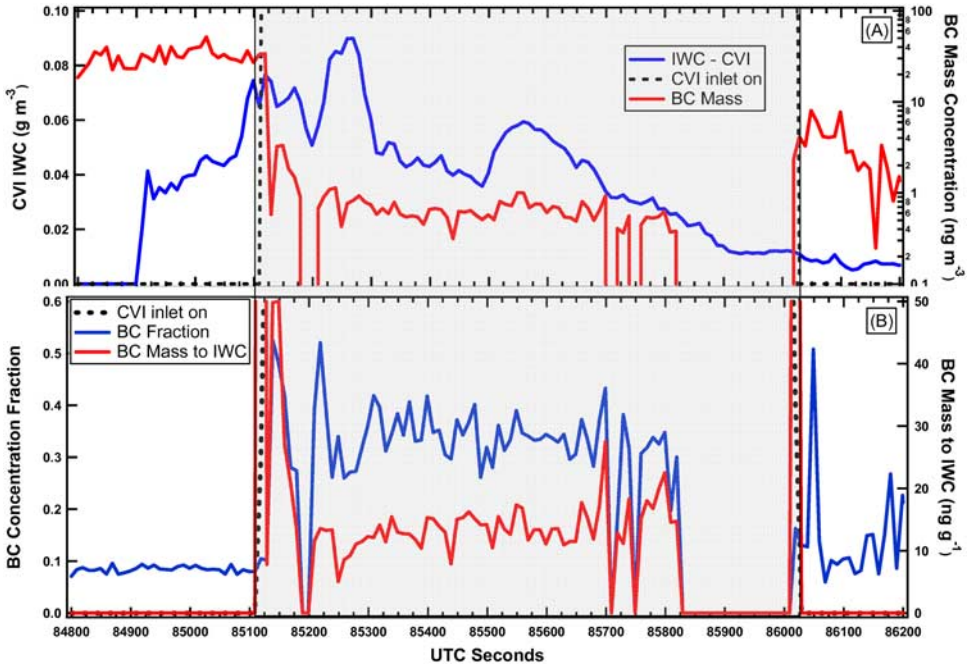

Figure 1. (a) CVI ice water and BC mass concentrations and (b) BC concentration fraction and BC mass to IWC are shown as a function of time for one of the 12 cloud passes analyzed. The shaded area shows when the SP2 is sampling through the CVI inlet. Each data point represents a ten second averaging interval.

[7] Recent studies [*Murphy et al.*, 2004] suggest that ice crystals can impact on inlets and remove material from the inlet walls that is then mistakenly classified as ambient particles. This source of uncertainty cannot be discounted in the current study but is an unlikely source of a significant fraction of the measured since clean, particle free nitrogen gas was recirculated through the inlet most of the time when not sampling in cloud, so contamination from ambient particles should be minimal and, as shown in the following analysis, the relative fraction of BC found in cloud residuals does not increase with time as would be expected if BC was accumulating in the inlet.

[8] The Pacific Dust Experiment (PacDEx) was conducted from April 29 to May 24, 2007 to evaluate the properties of aerosols and gases in air masses that originated in Asia and to study the transformation of particles as they aged and interacted with clouds (J. Stith et al., Cross-Pacific transport of Asian dust-mode particles and cloud active nuclei, submitted to *Journal of Geophysical Research*, 2008). There were 14 flights that started from Colorado, Alaska, Hawaii and Japan sampling regions all north of 20° latitude. Nine flights with cirrus encounters above eight kilometers were selected for the present study.

3. Results

[9] Twelve cirrus cloud formations were sampled with temperatures and average CIWCs from −25° to −45°C and 0.06 to 0.24 gm^{-3}, respectively (Tables 1a and 1b). The majority of the clouds were associated with cold fronts or extratropical cyclones (Stith et al., submitted manuscript, 2008). Seven of the clouds were in the West Pacific, three in the Central Pacific and two in the East Pacific. An example of trends in some the BC properties, inside and out of cloud, is illustrated by the time series in Figure 1. Each of the data points is a ten second average and corresponds to approximately 2.2 km at the aircraft research airspeed.

[10] Tables 1a and 1b summarize basic statistics of the BC properties in the ice crystal residuals and particles in cloud-free air for each of the cloud passes. Clouds are defined as air parcels with CIWC > 0.05 gm^{-3} while cloud-free regions are those within 10 km of the cloud and CIWC < 0.01 gm^{-3}. The relationship between the BC properties in cloud residuals and cloud-free particles is represented by the ratio, in/out, that follows the averages. The ensemble averages and standard deviations for the 12 cases are given at the bottom of Tables 1a and 1b. Also at the bottom of Tables 1a and 1b are correlations between the in and out of cloud properties and between the IWC and BC properties in crystal residuals.

[11] The properties of BC in crystal residuals are much more variable than in the cloud-free regions, as seen in the standard deviations about the means. The number, mass and saturated mass concentrations of BC in aerosol particles outside the clouds were approximately 25, 12 and 7 times higher, respectively, than in crystal residuals (Table 1a). In contrast, as seen in Table 1b, in 10 of the 12 clouds, the number of particles that contained BC compared to the total particles, referred to as the BC number fraction, F_n, and the MED, were larger in the crystal residuals.

[12] None of the properties were correlated between the cloud-free particles and the crystal residuals; however, all of the properties of BC in crystal residuals were significantly correlated with the CIWC.

4. Discussion

[13] BC is removed by clouds through inertial and nucleation scavenging but it is not possible to unambiguously separate these two processes with the current data set.

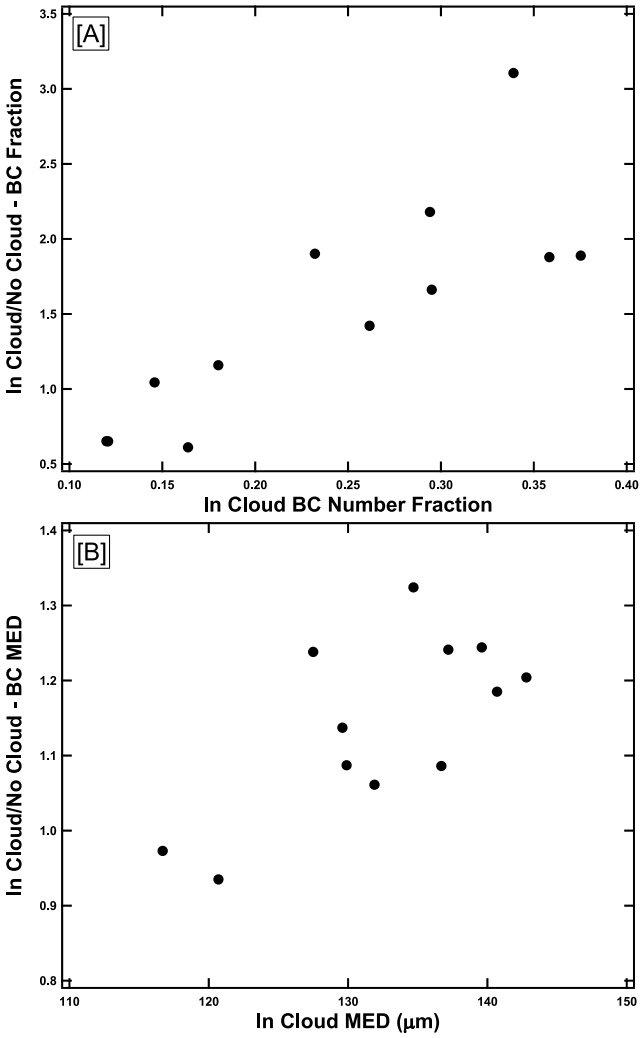

Figure 2. (a) The in-cloud to cloud-free BC number fraction is shown as a function of the average in-cloud number fraction and (b) the ratio of the BC mass equivalent diameters in and out of the cloud as a function of in-cloud MED is shown for the 12 cloud passes.

The BC on the surface of crystals, presumably captured by inertial scavenging, and BC inside a crystal formed via nucleation, will appear the same to the SP2 that measures only the residual of an evaporated crystal. A comparison of the cloud-free particle properties with those of the crystal residual does, however, provide indirect evidence of the potential importance of each of the two scavenging processes. We begin by assuming that the regions within 10 km of the cloud have the same history as the cloud itself, i.e. that the crystal residuals come from the same population as the particles in the nearby regions. Given that interstitial aerosols were not sampled we can't do a number or mass balance of particles inside and outside the clouds. We will focus instead on two prominent properties of the BC that differ between the crystal residuals and particles outside of cloud. On average, F_n and MED in the crystal residuals are 50% and 10% larger, respectively, than particles in clear air. In addition, as illustrated in Figure 2a, there is a strong, positive correlation between the average ratio of F_n (crystal residual) and F_n (cloud free) as a function of F_n (crystal residual). Given that F_n is the ratio of particles with BC to all particles, if the BC in ice is only from nucleation, when the F_n in cloud residuals increase, it must be because there are more ice nucleating BC particles in the cloud free air and the F_n out of cloud should also be increasing. If our assumption is correct, the ratio of F_n (cloud) to F_n (no-cloud) should then remain approximately constant. As seen in Figure 2a this is not the case and the increase can only occur if there is inertial scavenging adding to the BC in the crystal residuals. The same argument holds true for the observed increase in the ratio of MED (cloud) to MED (no cloud) as a function of MED(cloud) seen in Figure 2b.

[14] With respect to concentration, on average 4% of the BC by number has been removed by the cloud, 8% of the mass and 13% of the saturated mass. This implies that particles with larger MED are removed preferentially, consistent with the observations of MED on crystal residuals compared to the cloud-free particles. Preferential nucleation on larger BC particles will contribute to the observed trends; however, inertial scavenging also leads to larger MEDs when multiple BC particles are collected by a single ice crystals and then coagulate into a single larger particle when the crystal evaporates in the CVI. The much higher ratios (0.13) of saturated BC mass on crystal residuals to cloud free particles, compared to the total BC mass (0.08), is consistent with multiple BC containing particles on the ice crystals.

[15] If many of the inertially scavenged BC particles are below the detection range of the SP2 (80 nm), it will appear as if more mass is being removed than number in accordance with the observed trends.

[16] Ice crystals will also inertially scavenge particles with no BC in them so that after a period of time there will be an assortment of particles on the ice crystal surface with and without BC. When the crystal evaporates in the CVI, and they coagulate into a single particle, it will be measured by the SP2 as one that contains BC. This means that fewer crystal residuals will be classified as BC-free and the associated F_n will increase. The slightly larger MED on crystal residuals, compared to the cloud-free particles, is consistent with this hypothesis if many of the BC particles smaller than the size threshold of the SP2 are scavenged and will only increase the BC mass per crystal a small amount with an associated increase in the MED. The lack of correlations between the BC number and mass inside and out of cloud also supports this hypothesis if sub-80 nm BC particles are being collected.

[17] Some of the enhanced F_n may be the result of large ice crystals that impact at high speeds and shatter inside the CVI due to inertial stresses. In this case the BC particles originally present in a single crystal might be sampled as separate particles by the SP-2, but only if there are multiple particles in the crystal. The relatively high in-cloud number concentrations and the correlation between the BC-free residuals and IWC suggest that this occurs to some extent but these processes require further investigation.

[18] In a recent study [*Cozic et al.*, 2008] the mass of BC was estimated from the measurements of the light absorption coefficient of crystal residuals at the Jungfraujoch mountain observatory. The mass fraction of BC, computed as the mass of BC divided by the total aerosol mass, was 27% in the crystal residuals compared to 5% in the bulk

Table 2. Comparison of the Ratio of BC Mass to Condensed Water Content with Previous Studies

Water Type and Location	BC/CWC[a] Ng g^{-1}	Reference
Rainwater: Sweden	20–600	*Ogren et al.* [1983, 1984]
Rainwater: Seattle	30–400	*Ogren et al.* [1983, 1984]
Cloud water: Eastern Pacific	23–79	*Twohy et al.* [1989]
Cloud water: Nova Scotia	10–60	*Chýlek et al.* [1996]
Cirrus crystals: Northern Pacific	7–44	This study

[a]CWC is condensed water content.

aerosol. These are consistent with the values of F_n in the present study of 31% and 17%, respectively. The Jungfraujoch study also incorporated a CVI but at much lower speeds, minimizing the possible contamination by crystal impaction and removal of deposited BC on the walls of the CVI.

[19] In order to incorporate these results into global aerosol models, it is useful to express the scavenging efficiency of the ice clouds as the mass of BC with respect to mass of water. As shown in Table 1b, an average of 14 ng BC was measured per gram of water but varied from 6 to 44 ng g^{-1}. This is a conservative estimate since larger BC masses that saturate the detector are included in this estimate only as the maximum that the SP2 can detect. Previous studies [*Schwarz et al.*, 2006] have estimated that the SP2 measures approximately 50–60% of the total BC mass in the particles in the UT/LS; hence, the scavenging efficiency could be greater than twice the values estimated in this study. Table 2 shows a comparison with scavenging efficiencies reported by other investigators. The current values, although saturation limited, are consistent with the only other estimates made from aircraft measurements, those from marine stratus clouds [*Twohy et al.*, 1989; *Chýlek et al.*, 1996].

5. Summary

[20] Direct measurements of the mass of BC in upper tropospheric aerosols and cirrus crystal residuals have been made for the first time. Comparison of BC properties in the crystal residuals and cloud-free particles show that BC is scavenged by the crystals with an efficiency of as much as 44 ng of carbon per gram of ice water. The average number fraction of BC in crystal residuals was 40% greater than that of the particles in the nearby, cloud-free regions and, on average, the mass equivalent diameter of BC was 10% larger. An average of 24% of the ice crystal residuals contained BC compared to 17% in cloud-free air. The large, in-cloud variability of BC properties prevents a more rigorous proof of our hypothesis that inertial scavenging is a dominant mechanism for removal of BC by clouds. The lack of correlation between the inside and out of cloud concentrations, coupled with the differences between the number fraction and MED in the crystal residuals and the background aerosol, is consistent with the inertial scavenging process and the evidence is sufficiently compelling to warrant further study.

[21] The results presented here do not rule out nucleation scavenging and it is likely that both processes contribute to the removal of BC but at different spatial and temporal scales. The current study is being extended by analyzing the BC properties in all the clouds sampled during PacDex as they relate to the air mass origin and age.

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