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Measurements of black carbon aerosol washout ratio on Svalbard

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ABSTRACT

Simultaneous measurements of aerosol black carbon (BC) in both fresh snow and in air on Svalbard are presented. From these, washout ratios for BC are calculated and compared to sparse previous measurements of this metric in the arctic. The current ratios are significantly higher than previously found measured values. We argue that the degree of snow riming within the accretion zone can explain most of this difference. Using an analytical model of the scavenging process, BC scavenging efficiencies are estimated to lie in the range 0.25–0.5, comparable to measured values.

1. Introduction

Light-absorbing aerosol, primarily black carbon (BC), have been argued to have an important impact on the radiative balance of the arctic (Hansen and Nazarenko, 2004; Shindell and Faluvegi, 2009). In particular, the role of deposited BC in the reduction of snow albedo has received substantial attention (e.g. Clarke and Noone, 1985; Hansen and Nazarenko, 2004; Flanner et al., 2007; McConnell et al., 2007). Numerous studies have suggested that a key, and poorly understood, process in the linkage between emissions of BC and its impact on the arctic radiative balance, for both airborne and deposited BC, is the scavenging of BC by clouds and precipitation (e.g. Shindell et al., 2008; Koch et al., 2009; Garrett et al., 2010).

Few measurements of the scavenging of BC by clouds and subsequent deposition to the surface are available. Extant work has dealt mostly with scavenging in water clouds (e.g. Hitzenberger et al., 2000; Sellegri et al., 2003) and has focused primarily on nucleation scavenging. Work on mixed-phase, stratiform clouds such as those most important for arctic deposition (cf. Morrison et al., 2005; McFarquhar et al., 2007; Luo et al., 2008) are still more rare. A recent study by Cozic et al. (2007) in mixed-phase mountaintop clouds in Switzerland has suggested significant differences between the incorporation of BC into solid and liquid hydrometeors, and also suggested that the scavenging efficiencies may be influenced by the state of mixing of the BC with other aerosol constituents.

For the arctic, field data on wet scavenging and deposition are very limited indeed. The only study of which we are aware is the pioneering work by Noone and Clarke (1988), who calculated washout ratios for BC at a site in northern Sweden (Abisko). Hence, the need for further data on the BC wet deposition process in the arctic is acute, prompting us to offer this analysis of recently analysed measurements from Svalbard.

2. Venue and methodology

2.1. Venue

The data presented here are concentrations of BC in freshly fallen snow and in air, taken at two locations on the island of Spitzbergen in the Svalbard archipelago (78.917°N, 11.933°E): the Zeppelin research station (475 m MSL) and the Sverdrup research station in Ny Ålesund (8 m MSL) directly below the Zeppelin station. This venue is a favourable site for such measurements, due in part to the extensive research that has been undertaken here, which provides good context for any new measurements, and in part to its geographic position, which renders it susceptible to deposition of BC from diverse sources both inside and outside the arctic (e.g. Stohl, 2006; Eleftheriades et al., 2009). Furthermore, the altitude of the Zeppelin site is a substantial advantage in deposition analysis (as discussed later). The snow collectors were located on the roof of the Zeppelin station and on a balcony of the Sverdrup station.

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2.2. Methodology

2.2.1. BC measurements. Two sets of measurements were used in the analysis. The first set was of air concentrations of BC at the Zeppelin station. These concentrations were derived from measurements of the aerosol absorption by means of a custom built PSAP (Particle Soot Absorption Photometer) with corrections for light scattering following the procedures of Bond et al. (1999). A mass absorption cross-section (MAC) of 11 m^2 g^{-1} (at 550 nm) was used for the conversion of the absorption to BC concentration. This MAC was derived from a comparison of the absorption measurements to simultaneous measurements of BC using the thermo-optical technique (Sunset Labs OCEC analyser) and the NIOSH 5040 protocol. This conversion assumes that all measured light absorption is due to BC. The second set of measurements is that of BC concentrations in samples of freshly fallen snow at both Ny Ålesund and Zeppelin station. The BC concentrations were determined by first filtering the melted snow through a 25 mm nuclepore filter (0.4 μm pore size) and then using a multiwavelength spectrophotometer to determine the aerosol absorption on the filter as a function of wavelength. The wavelength dependence is then used to differentiate BC from other light absorbing aerosol (Grenfell et al., 2011). The value of the MAC used to convert the absorption to BC mass was 6.0 m^2 g^{-1} (at 550 nm) based on the MAC of the BC aerosol used to calibrate the spectrophotometer. This is similar to values given by Bond and Bergstrom (2006) in their review of BC for a variety of different sources (7.5 ± 1.2 m^2 g^{-1} for fresh BC).

We calculate the BC washout ratio by comparing the air and snow concentrations of BC. Hence, the discrepancy in measurement techniques between the air and snow BC is a potential source of bias and must be discussed. Several studies have suggested that the NIOSH 5040 protocol for thermo-optical measurements used here to determine the air concentrations of BC is biased low compared to optical methods such as that employed for our snow BC measurements (Reisinger et al., 2008). The Reisinger et al. (2008) study suggests that the bias may be a function of the organic carbon (brown carbon) mass present in the samples, with the bias increasing as brown carbon mass increases. Doherty et al. (2010) have estimated that ∼10–20% of the light absorption by aerosol in the snow samples is caused by non-BC constituents. If this is due to brown carbon, the results of Reisinger et al. suggest approximately a factor of two low bias in the NIOSH technique compared to optical methods. Similarly, Chow et al. (2004) compared the NIOSH approach to a thermo-optical technique using a different measurement protocol which is considered to provide a more accurate separation of BC and OC and their results also suggest the NIOSH technique is biased low by approximately a factor of two. On this basis, to ensure consistency between our snow and air values for BC/EC we have increased our atmospheric BC concentrations by a factor of 2.0 (Chow et al., 2001; Reisinger et al., 2008) when calculating washout ratios with air concentrations measured as EC. Not coincidentally, this correction would effectively be equivalent to applying a MAC to the air measurements much closer to that used for the snow measurements. Nevertheless, it is important to note that the precise magnitude of the correction to apply is as yet not certain. We feel that the factor of two we use is appropriate for our samples but is likely not universally applicable.

2.2.2. Washout ratios. The variable we use to characterize the wet scavenging of BC is the washout ratio, that is, the ratio of BC in the deposited snow to that in the air at the surface. Although this variable has a long history of usage, including in the arctic (e.g. Noone and Clarke, 1988; Davidson et al., 1989; Hewitt and Rashed, 1991; Bergin et al., 1995), it is a somewhat ambiguous metric for scavenging since it is rare that the surface air concentration is equivalent to that at the altitude at which most aerosol are incorporated into hydrometeors (cf. Davidson et al., 1993). On the other hand, it does subsume a number of individual processes such as nucleation scavenging and below cloud scavenging to give an estimate of overall scavenging. In the present analysis, we have snow measurements at both near sea level (Sverdrup Station) and at an altitude of 475 m MSL (Zeppelin Station), and air BC measurements at Zeppelin only. However, the latter measurements are likely made quite close to the riming zone of the stratiform clouds from which the deposited snow is falling at both sites and thus near the altitude where most of the BC is incorporated into the snow (cf. McFarquhar et al., 2007; Klein et al., 2009; Hoffmann et al., 2009). Indeed, airborne measurements of accumulation mode and non-volatile aerosol concentration vertical profiles at Svalbard suggest that the vertical variation in the lowest kilometre, while significant, is not great (Engvall et al., 2008). Hence, washout ratios for both Zeppelin and Ny Ålesund are likely not grossly in error.

2.2.3. Conceptual model of the removal process. Implicit in the above discussion of the washout ratio is a conceptual picture of the wet deposition process for BC. During the spring and early summer seasons, the precipitation at Svalbard, as is the case through much of the arctic, is primarily from low level, mixed-phase stratiform clouds (cf. Hobbs and Rangno, 1998; Wada and Konishi, 1998; Morrison et al., 2003; de Boer et al., 2009). In such clouds, the snow mass on the ground can arise from any of three main formation processes. These three processes are, (1) riming of snow crystals nucleated high in the cloud and falling through a liquid water zone (accretion zone), (2) aggregation of nucleated snow crystals, (3) vapour depositional growth (including the Bergeron–Findeisen process in which nucleated snow crystals grow at the expense of water drops due to their lower equilibrium vapor pressure). The three processes are illustrated in Fig 1. Of course, combinations of these three processes can occur for particular scenarios. The three processes, as shown in Fig 1, will lead to quite different washout ratios for aerosols in general and BC in particular. Essentially, the differences in overall scavenging efficiency (washout) arise due to the

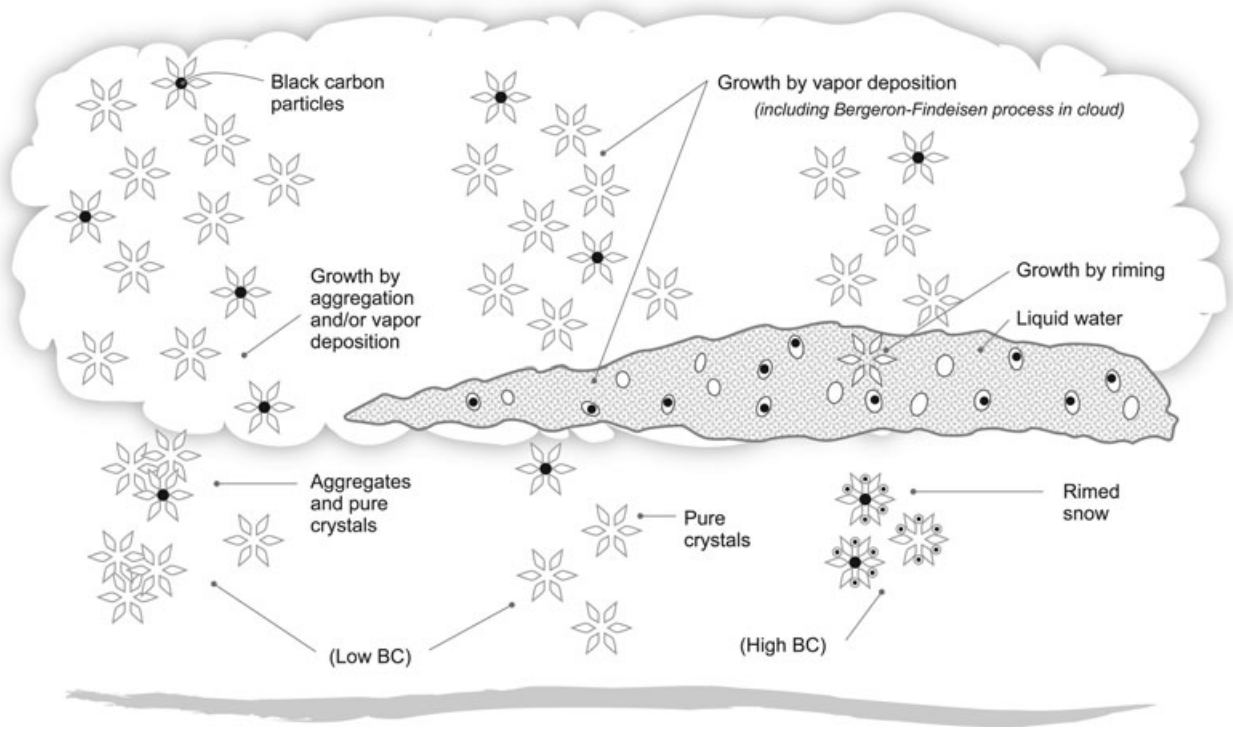

Fig. 1. Schematic diagram of the conceptual model for snow formation from arctic clouds such as those over Svalbard during our study period.

fundamental difference in the scavenging efficiency of ice and water hydrometeors.

BC mass on Svalbard resides in the accumulation mode (Heintzenberg and Covert, 1987). Most accumulation mode particles can act as CCN at modest supersaturations whereas very few such particles act as IN. Following formation/nucleation (and noting that diffusive scavenging of accumulation mode particles is definitely second order), ice particles gain sufficient mass to fall out as snow either by vapour deposition or by collection processes. In the former case there is phase transfer of water mass but not of aerosol (BC). Hence, for the snow particles, the scavenging efficiency remains low. Furthermore, for mixed phase clouds in which ice and water hydrometeors are co-located, the ice particles will grow at the expense of the water droplets until the ice particles have completely depleted the liquid water or precipitate out. This is the Bergeron–Findeisen process. Clearly, via this growth mechanism, the scavenging efficiency of BC remains low. On the other hand, growth of the ice particles to precipitation size via collection can, broadly speaking, take one of two paths. The first is usually found when only ice particles are present and the particles grow simply by collision with each other, aggregating into larger particles. This process is in principle more favourable for BC removal than pure depositional growth since each ice particle may at least possibly have a particle containing BC acting as the IN that formed it. However, it could easily have some other particle without BC acting as IN and, in any case, the ice particle concentrations are sufficiently low that little BC will be removed in contrast to the other collection pathway, riming. In riming, the ice particles collide with the relatively numerous water drops which freeze upon contact with the collecting ice particle. The collection frequency will be much higher for riming than for aggregation. During this process, both water and BC mass are incorporated into the precipitating snow with most of the BC mass deriving from the frozen cloud droplets, with their high BC scavenging efficiency. Hence, this is a relatively efficient scavenging process for BC.

In principle, below cloud scavenging of aerosol BC can also contribute to washout. However, in general, below cloud scavenging is a small contributor to removal of aerosol mass (Scott, 1978; Jacobson, 2003). Below cloud scavenging of BC mass by snow should be negligible, in accord with most recent assessments (e.g. Flossmann and Wobrock, 2010) and we neglect it in this study.

For both 'rimed or unrimed' scenarios, the incorporation of aerosol BC into the snow and its subsequent deposition can be evaluated using the analytical model of Scott (1978), originally developed for sulphate scavenging but applicable to any aerosol with appropriate adjustments to various aerosol properties such as nucleation scavenging efficiency. In this model, ice particles are nucleated in the upper, cooler portions of a stratiform cloud, with the concentration of the BC in the ice particles being determined by the relatively inefficient ice nucleation process (see below discussion). These ice particles then fall through a liquid water layer lower in the cloud, the accretion zone (as shown in

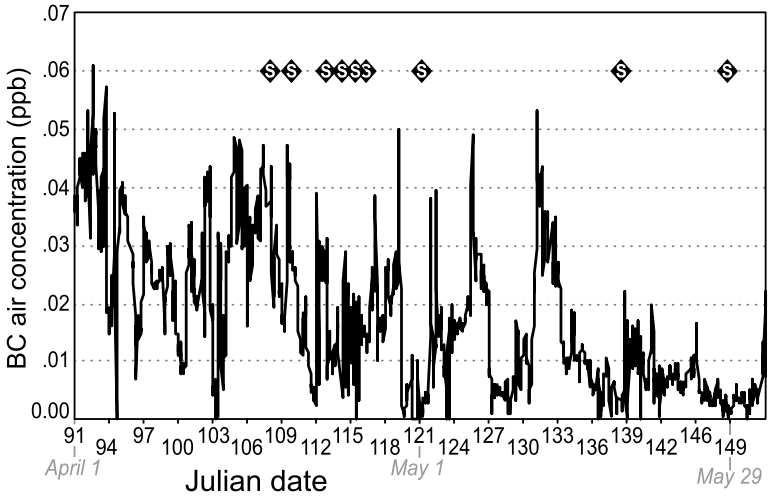

Fig. 2. Air concentrations of black carbon (BC) measured at the Zeppelin station during the study period. Periods during which fresh snow samples were obtained are indicated by 'S' along the top of the plot. The detection limit of the PSAP is a nominal 1 ppt.

Fig 1). Most of the BC is incorporated into the hydrometeors in the accretion zone, in which the scavenging of BC is primarily by nucleation scavenging, the fraction of the BC scavenged by the cloud droplets (F_{BC}) being the key parameter. While the Scott model is too simplistic and heavily parametrized to use as a prognostic model for sensitive variables such as ice particle concentration, we use it in this analysis as a diagnostic tool to explore the impact of riming on the BC washout. For this it is robust and useful.

3. Results and discussion

3.1. Measured concentrations

Temperatures at Zeppelin and Ny Ålesund ranged from –6 to –12 °C during April and from –3 to 1 °C during May. A time series of the air concentrations of BC measured at the Zeppelin research station is shown in Fig 2 for the months of April and May 2007. Concentrations ranged from ∼0 to 0.06 ppb with a mean (and associated standard error) of 0.017 ± 0.0003 ppb. These concentrations are somewhat low compared to many previous measurements at arctic surface sites such as Barrow, AK (Polissar et al., 1999), Alert, Canada (Sharma et al., 2006) and Abisko, Sweden (Noone and Clarke, 1988). However, as noted by Sharma et al. (2006), among others, there has been a downward trend in BC concentrations in the arctic, rendering comparisons of older with new data difficult. An analysis of measurements of BC at the Zeppelin station from 1998 to 2007 by Eleftheriadis et al. (2009) reports concentrations in good agreement with those found here.

Concentrations of BC in fresh snow samples (by which we mean snow from the single most recent snow event) taken at both Ny Ålesund and Zeppelin within the same time period are much more sparse (Fig 3). These concentrations can be compared with those at a variety of other arctic sites reported in Doherty et al. (2010). The range of snow concentrations measured by Doherty

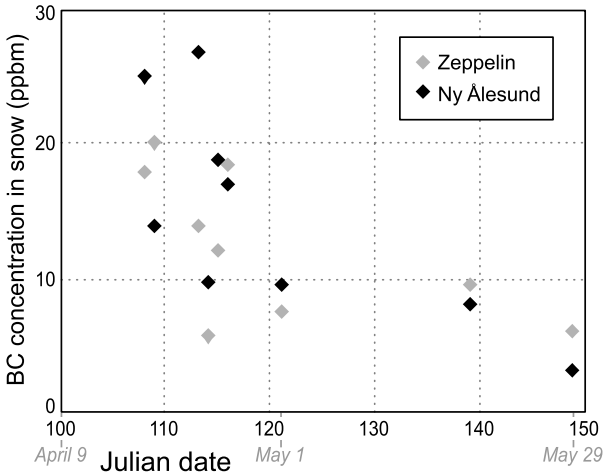

Fig. 3. Concentrations of BC in fresh snow at both the Ny Ålesund and Zeppelin locations during the study period.

et al. extended from 431 ppb (Vorkuta, Siberia) to 1ppb (several Greenland sites). However, the Svalbard values are quite similar to values reported at other comparable sites. For example, snow BC concentrations at Tromso Norway, perhaps the closest site geographically to Svalbard, were 17–19 ppb for the few samples obtained in March and April before the snow melted. Similarly, concentrations in the Canadian High Arctic ranged from 3 to 20 ppb (Doherty et al., 2010).

Calculation of the washout ratio involves a quantitative comparison of snow and air concentration. For this comparison, we average our 5 min resolution air BC concentration over each new snow event, which ranged in time from 4 to 28 h in duration. However, as discussed in section 2.2.1, the snow and air concentrations were measured by different techniques (ISSW spectrophotometer and thermo-optical, respectively, as mentioned above) and an upward correction of a factor of two has been applied to the air BC measurements to render them comparable.

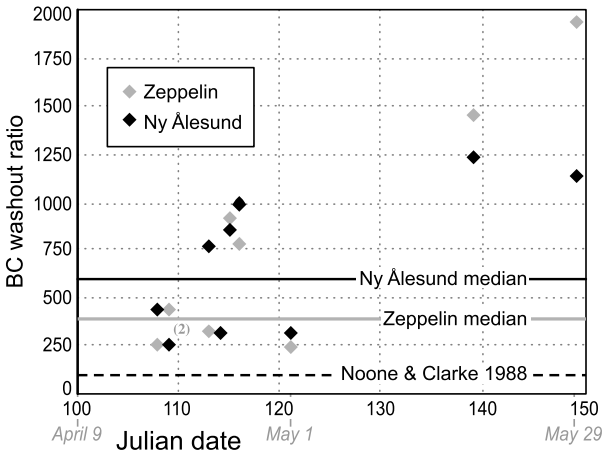

Fig. 4. Washout ratios for BC during the study period.

3.2. Washout ratios

Based on the reported concentrations of BC in the snow and air samples (modified as per the above discussion), washout ratios (BC_{snow}/BC_{air}) were calculated for seven different cases (Fig 4). Air BC concentrations were only available at Zeppelin station, so these were the air values used even for the washout ratios calculated for the Ny Ålesund snow samples, making these ratios more problematic than those for the Zeppelin site. The uncertainty in the snow BC concentrations, derived for a mean measured total absorption Ångström exponent of 1.63 (the mean over our sample set), is ±20%. This uncertainty is due largely to the uncertainties in the choices of the Ångström coefficients used for the BC and non-BC light absorbing aerosol, necessary quantities in the partitioning method used to differentiate BC from non-BC light absorption (see Doherty et al., 2010 for the details of the uncertainty calculations). The uncertainty in the corresponding air BC concentrations averaged over the corresponding snowfall periods is ±8% (standard error of the mean air BC over the fresh snow fall period). The uncertainty in the resultant washout ratios, calculated by standard Taylor series expansion is ±20%. This uncertainty does not, however, take into account the possible uncertainty in the systematic bias between the Zeppelin air concentrations and the Ny Ålesund snow concentrations (see the discussion in Section 2.2.1). There is substantial variation in the washout ratios for both sites, though, as would be expected, there is also a relatively high covariance between the two sites. Perhaps the most interesting aspect of the results is that they are systematically higher than the washout ratios previously reported by Noone and Clarke (1988) for their study site at Abisko, Sweden. These ratios ranged from ∼50 to 175. The Noone and Clarke study median (83) is shown in Fig 4 and is a factor of 4.6 lower than the median for Zeppelin and a factor of 7.2 lower than the Ny Ålesund median. Several explanations for this difference are possible. First, the air concentrations used for the Abisko calculations were true ground level concentrations in the sense they were well below cloud base, as compared to at least the Zeppelin concentrations. If BC concentrations aloft at Abisko were roughly an order of magnitude lower than those at the surface, then this alone could explain the bias. However, as the washout ratios for Ny Ålesund—and their agreement with the Zeppelin values—suggest, BC does not necessarily decrease systematically with altitude and there are no grounds for assuming it does here. Alternatively, it is possible that the discrepancy is due in part to actual differences in the scavenging efficiency (fractional removal) of the air BC by hydrometeors, dominated by the nucleation scavenging by water drops, in the accretion zone (F_{BC}) at the two sites. Values of the scavenging efficiency of BC have been found to differ substantially, ranging from quite low values in polluted environments (e.g. 0.06 for the Po valley as per Hallberg et al., 1992) to values similar to those for sulphate and other soluble accumulation mode aerosol in more remote locales (e.g. 0.80 at Svalbard itself as per Heintzenberg and Leck, 1994). This variance is presumably due to the transition from fresh, largely hydrophobic BC to aged BC coated by various soluble species. Finally, for mixed-phase clouds, it is also possible that differences in the nature of the precipitation formation process at the two sites, as per the discussion in Section 2.2.3, affect the washout ratio. Cozic et al. (2007) present data from the Jungfraujoch (Switzerland) that suggest that an active Bergeron–Findeisen process can significantly reduce the effective scavenging by essentially evaporating off cloud drops containing previously scavenged BC particles and returning them to the gas phase. Snow particles subsequently deposited on the ground, grown largely by vapour diffusion, would thus have appreciably less BC in them than would snow particles whose mass was mostly due to riming by cloud drops. An active Bergeron–Findeisen process, or simply growth through vapor diffusion in general, with little or no riming at Abisko as compared to Svalbard might thus explain the observed difference in washout ratios. We find this more plausible than fresh, hydrophobic BC at so remote a site as Abisko and aged, hydrophilic aerosol at Svalbard leading to differences in the washout ratios. Certainly it is possible that both differential aging as well as the vertical gradient issue discussed above could contribute to the observed difference in scavenging between the two sites but there is no real evidence for either. We prefer to first consider the more plausible difference in precipitation formation processes.

Support for differing snow formation processes at the two sites is in fact present. The snow sampled at Abisko was observed to show no evidence of riming, consisting almost exclusively of pristine crystals (K. Noone, personal communication, 2010). The falling snow at Svalbard, on the other hand, was commonly though not always reported as showing evidence of rime. Furthermore, we also know that the stratiform clouds over Svalbard just prior to our sampling period were mixed phase (Campbell and Shiobara, 2008) and that such clouds do lead to rimed snow at Svalbard in the spring (Wada and Igarashi, 1998).

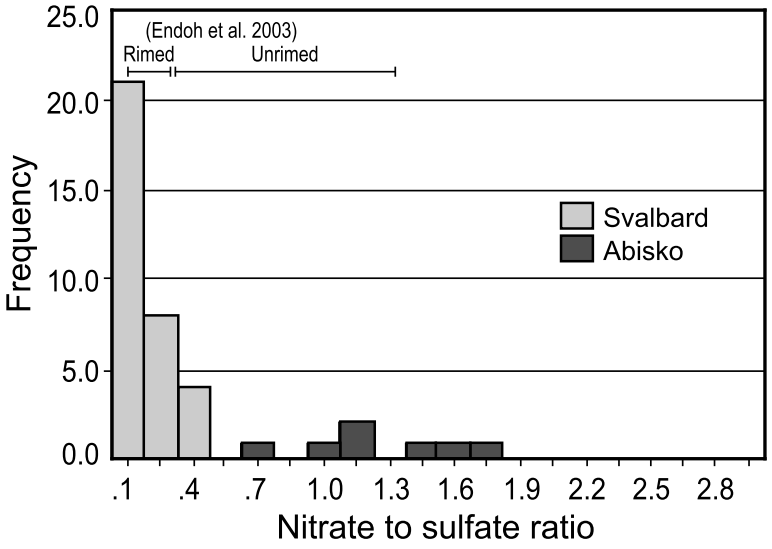

Fig. 5. Histograms of the nitrate to sulphate (NSS) ratio in the snow sampled at both Svalbard and Abisko. For reference, the ranges for both rimed and unrimed snow sampled at Svalbard by Endoh et al. (2003) are also shown.

Perhaps most convincingly, the snow chemistry itself suggests riming at Svalbard. It has been known for some time that nitrate is preferentially scavenged by snow flakes, as compared to sulphate and various other predominantly condensed phase species (cf. Raynor and Hayes, 1983), and that this leads to a marked dichotomy in the nitrate to sulphate ratio in rimed as opposed to unrimed snow (Takahashi et al., 1996). This phenomenon has in fact been observed at Svalbard (Endoh et al., 2003). Histograms of the snow nitrate-to-sulphate ratio at both Svalbard and Abisko are shown in Fig 5. There is a striking difference in the nitrate-to-sulphate ratios at the two locations; the two frequency distributions do not overlap at all. The much lower Svalbard ratios are clearly indicative of more riming. Indeed, they are comparable to those found in rimed snow on Svalbard by Endoh et al., as can be seen in Fig 5. It is also important to note that, when the snow at Svalbard was not rimed, the nitrate-to-sulphate ratios reported by Endoh et al. (see Fig 5) are essentially the same as those found at Abisko. This eliminates the possibility that the observed difference between our Svalbard ratios and those for Abisko are simply due to more nitrate being present in the air at Abisko, an unlikely but conceivable scenario.

It is also of interest to compare the values of the washout ratio measured at Svalbard with those incorporated into the large numeric models used to assess the impact of BC on the arctic climate (e.g. Jacobson, 2004; Flanner et al., 2007). The model used by Flanner et al., predicts a washout ratio for BC at Svalbard during April 2007 of ∼1400 (data provided by Mark Flanner, personal communication). The measured washout ratios for April range from ∼ 250 to 1000 and for May from ∼ 250 to 2000. The mean washout ratio for April (combining Zeppelin and Ny Ålesund data) is 554 ± 81 while that for May is 1057 ± 270. Hence, the model washout ratio is somewhat high by comparison. However, given that the model prediction is the mean for a grid square far larger than Svalbard and given the large observed variability in the ratio, the disagreement is not severe. Furthermore, it must be remembered that a correction factor of 2 was applied to the air concentrations. As per the discussion in Section 2.2.1, there is some uncertainty in this value and a somewhat reduced correction would render the modelled and measured ratios still more comparable.

3.3. Washout ratio as a function of precipitation rate and of BC scavenging efficiency

The variance in the washout ratios shown in Fig 4 is substantial. This is not surprising. The washout ratio for BC, or any other aerosol constituent, as per the above discussion, is a function of a number of different variables. Two critical controlling variables are the precipitation rate and the BC scavenging efficiency. It is quite plausible that much of the variability in the washout ratios shown in Fig 4, including the difference between the results reported here and those of Noone and Clarke (1988) is due to differences in these variables. To explore this, we make use of the model of Scott (1978), which postulates that the precipitation mass formation process is primarily due to accretion (or collection for warm clouds) of cloud droplets in an accretion/accumulation zone when larger hydrometeors (either ice or water) fall through it. This model is in accord with the studies of arctic clouds discussed in Section 2.2.3. Indeed, our measured precipitation rates (∼0.1 to 2 mm h^{-1}) are quite similar to those from mid-latitude systems for which the Scott model was developed (e.g. Herzegh and Hobbs, 1980) and in fact the case study to which Scott (1978) initially compared his model had a precipitation rate of 0.25 mm h^{-1}, the precipitationconsisting of rimed crystals as was the case for Svalbard. The washout ratio for the Scott model is given by the following equation (eq. 19 of

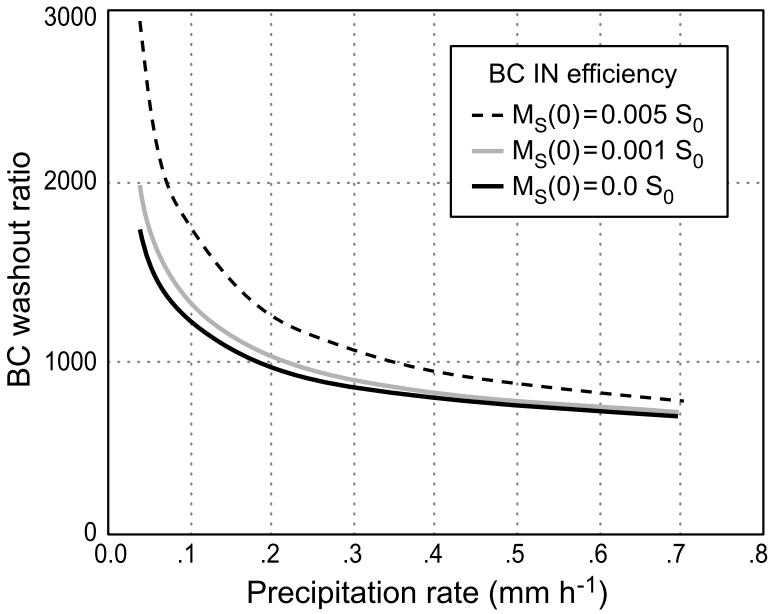

Fig. 6. Washout ratio as a function of precipitation rate and BC IN efficiency for an assumed BC scavenging fraction (F_{BC}) of 1.0. The curves are generated by the model of Scott (1978) for three assumed values of $M_s(0)$.

Scott, 1978).

$$\mathrm{WR(BC)} = \frac{14000 M_s(0)}{S_0 R^{0.88}} + \frac{1000 F_{BC}(1 - 0.0441 R^{-0.88})}{(1.56 + 0.44 \ln R)}, \quad (1)$$

where WR(BC) is the BC washout ratio, $M_s(0)$ is the concentration of BC in hydrometeors (per unit volume of air) at the top of the accretion/accumulation zone, F_{BC} is the fraction of air BC scavenged by hydrometeors in the accretion/accumulation zone, S_0 is the air concentration of BC, and R is the precipitation rate in mm h^{-1} (water equivalent in the case of snow). Here, the first term in Eq. 1 accounts for incorporation of BC into the ice crystals as IN at the top of the accretion zone and the second term accounts for incorporation in the cloud droplets that are added to the falling ice crystals (or raindrops for warm processes) via riming (or collection).

The least well-defined parameter in eq. (1) is undoubtedly $M_s(0)$, the concentration of the scavenged species at the top of the accretion zone. Scott developed the parametrization expressed in eq. (1) for aerosol sulphate. He derived a value of 0.1 S_0 for warm clouds based on the growth of the hydrometeors falling into the accumulation zone (for warm clouds) via condensation and autoconversion. For mixed-phase clouds in which the collecting hydrometeors are ice, he set $M_s(0) = 0$, assuming, quite reasonably, that sulphate particles would be very poor ice nuclei (IN) and hence that the sulphate concentration would be negligible compared to that due to collection in the accretion zone itself. However, BC is known to act, at least at times, as effective IN (DeMott et al., 1999) and we thus must examine possible values in more detail. The number of aerosol particles that act as IN is quite small, the fraction being at times literally one in a million (10^{-6}, Pruppacher and Klett, 1980). For arctic IN, Rogers et al. (2001) report a median value for the fraction of all particles that act as IN of 2×10^{-5} within a range extending from 0 to 2×10^{-2}. However, accumulation mode (and larger) particles are normally preferentially more active as IN than particles as a whole (cf. Richardson et al., 2007) and this is where most of the BC mass in the Svalbard area appears to reside (Heintzenberg and Covert, 1987). As alluded to above, within this larger size range BC are relatively more effective as IN than are most other particle types (DeMott et al., 1999). Hence, one would expect an IN active fraction for BC particles substantially in excess of the Rogers et al. value. We have made an ad hock selection of $M_s(0) = 0.001 S_0$ in part on this basis and in part because a plot of washout ratios as a function of precipitation rate generated by eq. (1), with an assumed value of $F_{BC} = 1.0$, reveals that the ratio is not a strong function of $M_s(0)$ even for low precipitation rates so long as $M_s(0)$ does not exceed 0.001 S_0 (see Fig 6). This relative insensitivity to $M_s(0)$ arises because most of the BC is incorporated into the hydrometeors during the riming process (second term in eq. 1) if riming does take place, and this is not a function of $M_s(0)$.

Washout ratios as a function of precipitation rate and scavenging efficiency are given in Fig 7 for $M_s(0) = 0.001\ S_0$. Superposed on this plot are the measured values of the washout ratio at Zeppelin station for those cases for which the precipitation rate could be calculated. (These rates were calculated from the change in snow depth in the collector divided by the time period during which snow fell as noted by an observer at the site.) We used only the Zeppelin values since the Ny Ålesund values, as discussed above, are less certain. Two main points can be inferred from Fig 7. First, from the model generated curves, for low precipitation rates, the washout is a much stronger function of the precipitation rate than of the scavenging efficiency while at higher rates the converse is true. Second, the positions

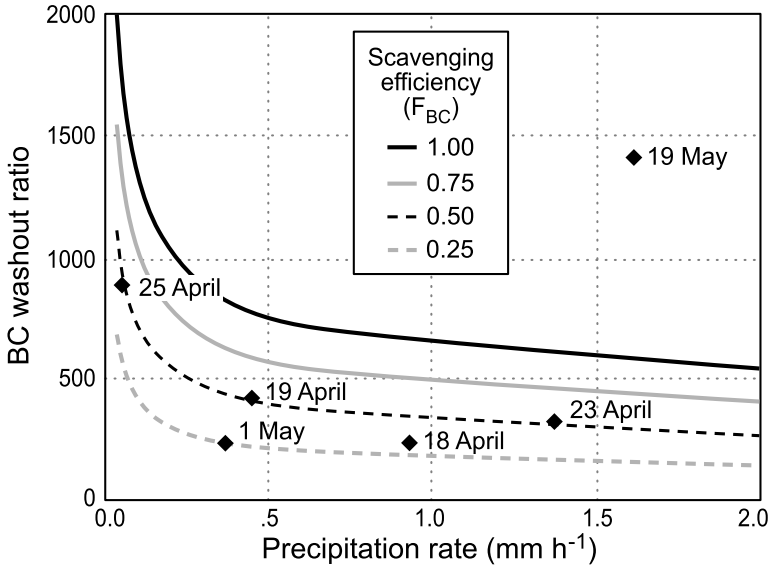

Fig. 7. Washout ratios for BC as a function of precipitation rate and scavenging efficiency for a BC IN efficiency of 0.001 [$M_s(0) = 0.001$]. The curves are generated with the model of Scott (1978). The six measurement points, all for Zeppelin station, are labelled by the sample dates (note that all dates are in 2007).

of the measured ratios for five of the six cases on the curves are consistent with the Scott model if the scavenging efficiency is allowed to vary from ~0.25 to 0.5. This range in scavenging efficiency is similar to that reported by Cozic et al. (2007) for their measurements on the Jungfraujoch and to similar measurements by Hitzenberger et al. (2000) on Mt. Sonnblick but is somewhat lower than the value of 0.8 reported some years ago by Heintzenberg and Leck (1994) for Svalbard itself.

The sixth measured washout ratio shown in Fig 7, that for May 19 (Julian date of 139), is well above any of the curves generated from Eq. 1. One distinguishing characteristic of this day was the relatively warm air temperature. Indeed, the temperature at Ny Ålesund was actually above freezing, the only above-zero day observed during the study. We speculate that on this day the precipitation process involved liquid as well as ice hydrometeors falling through the accretion zone, though of course all the hydrometeors would eventually freeze. For such a scenario, a value of $M_s(0)$ closer to that suggested by Scott for warm rain processes might be in order. If we use a value of $M_s(0) = 0.1\ S_0$ (Scott's warm rain value) coupled with a value of $F_{BC} = 0.5$, eq. (1) predicts a washout ratio of 1200 for the May 19 case, in reasonable agreement with the observed value of 1400.

Finally, the Scott model provides support for interpreting the marked difference between the washout ratios observed here and those reported by Noone and Clarke for Abisko in terms of differences in the precipitation process between the two sites. The lack of riming in the Abisko samples suggests that the second term of eq. (1), which accounts for the contribution to BC washout by the riming process, should be set to zero for the Abisko scenario. If we do this for our Svalbard cases, leaving everything else the same [i.e., $M_s(0) = 0.001S_0$], the median predicted washout ratio for the five cases in Fig. 7 that overlay the Scott curves would be 33, lower than but much closer to the median Abisko value of 83 than the measured Svalbard ratios.

4. Conclusions

The values of the BC washout ratio reported for Svalbard in spring of 2007 substantially enhance the meagre database for this parameter in the arctic. These measurements show substantially higher washout ratios than earlier reported values at Abisko, Sweden. However, the current values are not inconsistent with the Abisko data when interpreted with the aid of the Scott analytical model, which predicts the greater than factor-of-three difference in washout ratios for precipitation generated primarily through riming versus pure-ice nucleation processes. This model also qualitatively explains much of the variance in the Svalbard washout ratios in terms of measured variations in the precipitation rate and realistic variations in the BC scavenging efficiency. It highlights the importance of accurately representing not only precipitation rates and BC concentrations but also BC scavenging efficiencies and, critically, precipitation formation processes in models trying to represent BC deposition to surface snow.

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