Response letter

Dear Editor,

We have studied the valuable comments from yourself and the reviewers carefully, and made further revisions in the manuscript that address your and the reviewers' concerns. Our detailed response to the reviewers' comments follows below.

- Response to reviewer#1's comments:

We thank the reviewer for a helpful review. The reviewer's comments have guided further improvements in the logic and statement, making this work more rigorous. A detailed response follows below.

This study presents a new and efficient technique to determine the meltwater scavenging efficiency of black carbon in snow overlying sea-ice. Simply, the concentrations of BC within a melt-refreeze layer and within the overlying snow are compared, and the assumption is adopted that BC and ice have been conserved within these two layers during the melt event. Conservation within about 7% is indeed shown for a limited number of test cases, in comparison with mass measurements that were made before melt commencement. The technique is also shown to produce consistent results with those from the more rigorous repeated sampling technique employed by Doherty et al (2013). Overall, the technique described here shows promise and is attractive because of its simplicity. Several issues should be addressed prior to publication, however.

General issues:

- It is a bit frustrating to have to refer to supplementary figures and tables for a 'Brief Communication'. This may indicate that the material should instead be presented in a standard paper rather than a brief communication. Alternatively, could the supplemental figures (especially S2, which I think is important for conceptual understanding of the technique) be worked into the main body of the paper?

- Response: Thank you for your suggestion. The figures and tables in the supplementary have been merged into the main body of the revised MS.

- The abstract (and manuscript, generally) should acknowledge more clearly that the technique requires the presence of a 'melt-refreeze ice layer', or some term that is similarly precise. Although the abstract refers to sampling of an "ice layer within the snowpack", it is not clear until later what the nature of this ice layer is, and confusion arises especially because the previous sentence refers to sea ice. Furthermore, a refreeze layer will not always exist, for example when persistently warm conditions cause complete meltout of thin snow layers, and this limitation could perhaps be acknowledged more clearly.

- Response: We have clarified that the 'ice layer' here refers to the melt-refreeze ice layer that is produced from refreezing of the meltwater within the snowpack over sea ice in the revised abstract and MS.

A discussion of the limitation in this technique is included in the section of "Results and discussion", please see details in L187-206 in the revised MS.

- The applied technique also assumes that the refreezing process does not preferentially exclude BC, i.e., that the BC concentration in the ice layer will be identical to that in the melt water. Please comment on this assumption, how it could affect the utility of the technique, and any observational evidence you have that can shed light on this matter.

- Response: Thank for your advice. We have not obtained such observations during the field measurements. In theory, melting water can discharge some of impurities as it freezes, leading to BC mass concentration in the melt-refreeze ice layer be lower than that in the melt water. Therefore, ignoring this effect may result in an underestimation of MSC, theoretically.

However, suspended particles, especially those with larger surface areas, such as BC, may stay in place and freeze in the crystal lattice during the refreezing of melt water (Novotny et al., 2002). That said, the freezing process does not preferentially exclude BC. In addition, the impurities can be more effectively discharged during multiple freeze-thaw processes, while it is limited for BC particles to be expelled in the formation of melt-refreeze ice layer during one freeze event. In conclusion, BC released during the freezing process of melt water may have less impact on the MSC estimation. We further clarify this assumption in L47-50 and made a discussion about the uncertainty in the section of "Results and discussion", please see details in L212-214 in the revised MS.

Reference:

Novotny, V. and P. A. Krenkel (2002), Water Quality: Diffuse Pollution and Watershed Management, 2nd Edition, Hoboken, NJ : J. Wiley, c2003.

- Abstract, line 20: "It is concluded that MSC exhibited a regional difference in the western Arctic during the sampling period" - These differences are not very large, however (i.e., they are all substantially less than 100%, indicating inefficient scavenging of BC). Are they even statistically different from each other? If so, can you speculate on why they varied? Are there regional differences in the environment that would be expected to translate into systemic differences in the MSC? Or do these differences perhaps reflect random variability in BC properties? I suggest either downplaying these differences (because they do not appear substantial) or briefly speculating on potential sources of such differences.

- Response: Thank you for your suggestion. We analyzed the statistical significance of the differences in MSC at various locations. The Jonckheere-Terpstra test indicated that it is highly significant ($p < 0.01$) for Elson Lagoon < Chukchi Sea < Canada Basin, and the Mann-Whitney U test demonstrated that the difference from each other is moderately significant ($p < 0.1$). This analysis has been added in the revised MS (see L196-199).

The scavenging efficiency of BC is mainly determined by the particle size and the hydrophobicity, which is interfered with other impurities since BC usually occurs in the particles as an "internal mixture". These influencing factors show significant regional differences due to various sources of BC and distinguishing deposition and transport processes (AMAP, 2011; Korhonen et al., 2008; Sharma et al., 2013; Schulz et al., 2019), leading to spatial variations in MSCs, which has been confirmed by the observations at Barrow and dye-2 in Doherty et al. (2013). The sources and properties of BC aerosols at Barrow and Canadian Arctic (Alert station) are also significantly different (Sharma et al., 2006). Conway et al. (1996) found that hydrophilic BC is much more effectively scavenged with melt water than hydrophobic one. Flanner et al. (2007) further estimated that the MSC for hydrophilic BC is about 10 times that for hydrophobic one, meaning that the variations in fraction of the two in the snow with location will also have important impacts on the spatial difference in MSCs.

There is significant seasonal variation in the size distribution of BC particles in the Arctic. In contrast, few studies have been done on the spatial difference in the particle size due to lack of observations, and only a range of 160 and 220 nm has been reported in the Arctic at present (Schulz et al., 2019).

In conclusion, we speculated that the regional differences in the hydrophobicity and particle size may cause systemic differences in the MSC, which still needs further observations to confirm. We briefly discussed the potential factors those may lead to MSC spatial differences in the revised MS (See L177-179):

"The scavenging efficiency of BC is mainly determined by the particle size and the hydrophobicity, which is interfered with other impurities since BC usually occurs in the particles as an "internal mixture" in the Arctic (Doherty et al., 2013). These influencing factors show significant regional differences due to various sources of BC and distinguishing deposition and transport processes (Korhonen et al., 2008; AMAP, 2011; Sharma et al., 2013; Schulz et al., 2019), leading to spatial variations in MSCs, which has been confirmed by the observations at Barrow and dye-2 station, Greenland

(Doherty et al., 2013). Conway et al. (1996) found that the hydrophilic BC is much more efficiently scavenged with meltwater than the hydrophobic one. Flanner et al. (2007) further estimated that the MSC for hydrophilic BC is about 10 times that for hydrophobic one, meaning that the variations in fraction of the two in the snow with location will also have important impacts on the spatial difference in MSC."

Reference

AMAP (2011), The Impact of Black Carbon on Arctic Climate (2011). By: P.K. Quinn, A. Stohl, A. Arneth, T. Berntsen, J. F. Burkhart, J. Christensen, M. Flanner, K. Kupiainen, H. Lihavainen, M. Shepherd, V. Shevchenko, H. Skov, and V. Vestreng. Arctic Monitoring and Assessment Programme (AMAP), Oslo. 72 pp.

Korhonen, H., K. S. Carslaw, D. V. Spracklen, D. A. Ridley, and J. Stro¨m (2008), A global model study of processes controlling aerosol size distributions in the Arctic spring and summer, J. Geophys. Res., 113, D08211, doi:10.1029/2007JD009114.

Schulz, H., M. Zanatta, H. Bozem (2019), High Arctic aircraft measurements characterising black carbon vertical variability in spring and summer Atmos. Chem. Phys., 19, 2361–2384.

Sharma, S., E. Andrews, L. A. Barrie, J. A. Ogren, and D. Lavoue´ (2006), Variations and sources of the equivalent black carbon in the high Arctic revealed by long-term observations at Alert and Barrow: 1989–2003, J. Geophys. Res., 111, D14208, doi:10.1029/2005JD006581.

Sharma, S., M. Ishizawa, D. Chan, D. Lavoue´ , E. Andrews, K. Eleftheriadis, and S. Maksyutov (2013), 16-year simulation of Arctic black carbon: Transport, source contribution, and sensitivity analysis on deposition, J. Geophys. Res. Atmos., 118, 943–964.

- The grammar and writing in general should be proofed by a native English speaker prior to publication.

- Response: The grammar and writing have been proofed by the native English speaker.

- Response to Prof. H. Conway's comments:

We thank the reviewer for a comprehensive and helpful review. The reviewer's comments have guided further improvement in the problem statement and data interpretation. We have also reviewed the relevant literature to further support our central hypothesis and expanded the discussion of study's results. A detailed response follows below.

Others have observed and discussed the seasonal influence of black carbon in melting snow (e.g. Flanner et al, 2007; Doherty et al., 2010, 2013; Dou et al., 2017). Here the authors propose a new method to estimate meltwater scavenging of black carbon using field measurements made near Barrow, Alaska. The idea is to eliminate the need for repeat sampling at sites by collecting just one profile during the melt season, and assuming mass conservation to estimate the scavenging efficiency.

Some comments and questions:

1. Figure 1 shows locations of sites far from Barrow that are not used to support your new technique for estimating BC scavenging efficiency. I do not think that referencing these extra sites: South Korean Antarctic Ocean expeditions 2010 -18; 3rd Chinese Arctic Expedition; Hiking through the North Pole; Dye 2 etc) adds to the primary focus of your paper (i.e. meltwater scavenging of BC). I think it would be less confusing for readers if these references were not included.

- Response: Thank you for your advice. In order to make the MS more focused, the sites in Svalbard and Greenland in Fig.1 have been removed. The observational locations in the 3rd Chinese Arctic Expedition (Field executor: Tingfeng Dou, the first author) and the 1st Chinese expedition hiking through the North Pole (Field executor: Cunde Xiao, the last author) are retained because these observations provide direct evidence for the presence of ice layer within the snow pack over more extensive sea ice area, which effectively extended the application scope of this new method.

2. Not all readers will be familiar with your field measurements near Barrow, Alaska (Dou et al., 2017). It would be useful to expand on details of those measurements here. Also in the abstract it would be important to state specifically that Elson Lagoon and Chukchi Sea are in the vicinity of Barrow, Alaska.

- Response: Thank you for your comments. The description of the field measurement was included in the revised MS (See P3, L65-79):

"The field measurement involves the snow thickness, snow density and stratification. In Elson Lagoon, we measured the snow depth along a 10km line before melt onset (April 15, 2015), and determined the average snow depth in this region. A far-shore site was chosen ~12 km away from the coast where the snow depth was close to the mean value (31.6 ± 5.4 cm) of this region (Fig. 1). The snow stratification was firstly recorded, and then snow density was measured at 2.5 cm vertical resolution using SnowFork instrument. Four points were measured per time in each layer. We applied the average value of snow density to characterize the snow layer. The snow depth was recorded at ablation stakes next to the snow pit. In the Chukchi Sea, the spatial variation of snow depth is more significant as compared with the Elson Lagoon due to the presence of ice ridge. We firstly selected a relatively smooth area of sea ice, and measured the snow depth along a 200m line in the centre region of the flat ice on April 6th, 2017. The observation site was chosen at a location close to the average snow depth and the measurement procedure was the same as that applied in Elson Lagoon. Note that there was a deviation between the observation sites of 2017 and 2018 due to the interannual variation in the ice condition over the Chukchi Sea (Fig .1). In Canada Basin, we conducted the measurements of snow depth at a 100m line over floe ice due to the smaller ice size and limited operating time. Snow density was measured using Tel-Tru densitometer (Tel-Tru Manufacturing Co., Inc., Rochester, NY) with an accuracy of 1 g, and a snow shovel of

2.5-cm in thickness. The thickness of snow layer and the position of melt-refreeze ice layer were measured using a ruler."

In the abstract and MS, we clarify that Elson Lagoon and Chukchi Sea are in the vicinity of Barrow. In addition, the measurement locations in Elson Lagoon and Chukchi Sea have been shown in figure 1.

3. I do not follow your eqn. 1, which you mention comes from Flanner et al (2007). Their eqn. 3 assumes mass rate of change of BC in layer 'i' is proportional to its mass mixing ratio multiplied by a scavenging factor, which appears to be quite different to the one you are using.

Instead, it might be better to follow the method described by eqns. 2, 3 and 4 in Doherty et al., (2013), which shows how measurements of mB (average original mass per unit volume of BC before melting), and m'B (average mass per unit volume in the near surface snow can be used to calculate an average scavenging efficiency. In your case. you could use the method you propose to calculate mB = h1r· 1CB1, and calculate m'B from your measurements of CBsfc hsfc and r· sfc in the near surface snow after melt has started (from your Table 2).

- Response: Thank you for your comments. We re-checked the definitions of MSC in different literatures. It is indeed problematic to directly quote Flanner et al. (2007)'s definition in the form of their eqn. 3 that was given to facilitate the integral operation and simulation of BC in the CLM model and different to the method proposed in this study.

According to your suggestion, we calculated MB and MB' respectively by using the method proposed in this study. The BC mass per unit volume before and after the melting was respectively calculated by: MB=$\rho_1 \cdot C_{b1}$(ng/cm^3), and MB'=$\rho_2 \cdot C_{b2}$. This study mainly focused on a single melting event, so we adopt the calculation method of Eqn. (2) in Doherty et al. (2013) at n = 1:

$$MB' = MB + MB \times (1\text{-}MSC)$$

$$MSC = 2 - MB'/MB = 2 - (\rho_2 \cdot C_{b2})/(\rho_1 \cdot C_{b1})$$

Accordingly, we calculated the MSC in Elson Lagoon based on the observations in this study. Result indicates that the mean value of MSC is 0.204, which is higher than the value calculated by the method proposed in this study (0.145). It is known that Doherty et al. (2013) made two assumptions in estimating MSC of BC, leading to their estimate may represent the upper limits of MSC (section [43], P5561 in Doherty et al., 2013). Thus, the difference between the MSC values obtained using these two methods is reasonable.

The earlier studies determined MSC by comparing the BC content in snowpack before and after multiple ablation events, corresponding to a formula that is applicable to continuous sampling method. This study estimated the BC taken away from the melt water directly by measuring its content in the melt-refreeze ice layer. The new method is more suitable for estimating MSC after one ablation event. In particular, this method can be used to estimate MSC through one measurement in the case that the pre-ablation state is unknown, which is obviously different from the earlier methods. In view of this, we have retained the original Eqn. (1) and Eqn. (2) in the revised MS, but the statement for introducing this method has been modified in the revised MS (L134-136):

"By determining the burden of BC per unit area (ng BC/cm^2) in the ice layer and in the partially melted snow layer above it, the scavenging efficiency estimated using the proposed approach is given by.…."

In addition, in the revised MS, we included the results calculated by the method of Doherty et al. (2013) based on our observations, and showed the comparison to the values derived using the new method.

"With the new method, we calculated the MSC in Elson Lagoon and compared it with that estimated according to equation (2) in Doherty et al. (2013). Results indicate that the MSC (14.5%) calculated by the new method is smaller than that (20.4%) by the method of Doherty et al. (2013) based on the observations in this study. The difference in MSCs estimated by these two methods is reasonable since the latter represents the upper limits of MSC. Our estimation is close to the average value (16.2%) derived by repeated sampling (RS) introduced by Doherty et al. (2013) in the same area and is still within its best estimation [14.0%-20.0%].".

The limitations and possible uncertainties of the new method have been discussed in the last part of the MS according to the suggestions of the reviewer (Please see the response to your last comment).

4. I am confused by the data presented in Tables S1 and S2 and how they have been used to construct Figs. 2, 3, & S1. Some questions:

(i) are there 3 different sites at Elson Lagoon that were sampled in 2015 only, and another 3 different sites at Chukchi Sea that were sampled in 2017 and again in 2018?

- Response: The samples were collected at two different sites in Elson Lagoon in 2015. At one of the sites (71.3SN, 156.37W), we took samples at different dates. More details could be seen in Table 1 in the revised MS. At Chukchi Sea, the samples were gathered at two different sites selected in 2017 and 2018, respectively.

(ii) if so, are data for the first site at Elson Lake shown in Table S1: BC concentrations (ng/g) of 0.31 in the ice layer and 1.72 in the overlying snow layer measured on May 18. Does that imply melt had started by that time? On May 31, an average value of 14.9ng/g was measured in the near surface snow. What is the density and thickness of the surface layer?

- Response: Yes, snow has begun to melt on May 18, 2015. The density and thickness of the surface layer have been included in Table 1 in the revised MS. The surface melting snow in the tables refer to the surface layer of snow pack at the final stage of snow melting.

(iii) For the first site at Elson lake in Table S2: CB1=1.1, and presumably CB2 = 1.72; CBice = 0.31 and CBsfc = 14.9. What is the density and thickness of the surface layer? When were the measurements of h1, r⌧1 and CB1 made?

- Response: The density and thickness of the surface layer are indicated by ρ2 and h2 in Table 1 in the revised MS, and the measurement dates are also shown aside.

(iv) Perhaps it would be less confusing if the tables were combined with relevant data (depths, densities, concentrations for Elson Lake and Chukchi Sea) and included as a single table in the main text.

- Response: Thank you for your suggestion. All observations of thickness and density of snow cover and BC concentration, and thickness and concentration of the melt-refreeze ice layer in different regions and the corresponding measurement date are included as a single table (Table 1) in the revised MS.

5. It would be good to mention some of the limitations of the method – for example, (i) a melt-season ice layer may not form in regions of strong melt; (ii) the model does not account for influxes of BC from snowfall during the melt season; (iii) the model provides an estimate of the average seasonal scavenging efficiency but does not capture temporal variations in efficiency.

- Response: Thank you a lot for your suggestion, we have included a discussion of the limitations of the method in the section of "Result and discussion", please see details in L208-217 in the revised MS.

Brief communication: An alternative method for estimating the scavenging efficiency of black carbon by meltwater over sea ice

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Abstract. The meltwater scavenging coefficient (MSC) of black carbon (BC) is a ~~key~~ crucial parameter in snow and sea ice model, as it determines the BC enrichment in the surface layer of melting snow over sea ice and therefore modulates the BC-snow-albedo feedbacks. We present a new method for MSC estimation by sampling the melt-refreeze ice layer~~ice layer~~ that is produced from refreezing of the meltwater within snowpack~~within the snowpack~~ and its overlying snow and measuring their physical characteristics ~~in the snowpits~~ in Elson Lagoon~~-~~ northeast of Barrow, Alaska during the melting season. The bias of estimated MSC ranges from -5.4% to 7.3%, which is not exactly dependent on the degree of ablation. The average MSC value (14.5%±2.6%) calculated by this proposed method is slightly lower than that derived from the repeating sampling (RS) method in Elson Lagoon, while still is within its best estimate range. Further estimation demonstrates that the MSC in Canada Basin (23.6%±2.1%) is close to that in Greenland (23.0%±12.5%), and larger than that in Chukchi Sea (17.9%±5.0%) on the northwest of Barrow. Elson Lagoon has the lowest MSC (14.5%±2.6%) in the study areas. It is concluded that MSC exhibited a regional difference in the western Arctic during the sampling period. The method suggested in this study provides a possible approach for large-scale measurements of MSC over the sea-ice area in the Arctic~~.~~, of course, this method depends on the presence of a melt-refreeze ice layer in the observation area.

1 Introduction

BC is among the most efficient particulate species at absorbing visible light, which can reduce the surface albedo and potentially accelerate snow melting (Flanner et al., 2007; Goldenson et al., 2012; Dou et al., 2012; 2017). Previous studies suggested an annual-mean radiative forcing of 0.1–0.3 W m^{-2} over the Arctic region from BC deposition (Flanner et al., 2009; Jiao et al., 2014). However, large uncertainties still exist in the sea ice region due to lack of field measurements and poor understanding of BC enrichment by overlying snow melting.

The enrichment of BC in melting snow largely depends on MSC, as it reflects the ratio of BC concentration in the meltwater departing the snow layer to the bulk concentration in the exact layer (Flanner et al., 2007). MSC which leads to enhanced concentrations of BC in surface snow ~~has~~ is ~~been found to be~~ considerably less than 100% by very few previous studies (e.g., Conway et al., 1996; Xu et al., 2012; Doherty et al., 2013). In present snow and sea ice models (e.g., Flanner et al., 2007; Goldenson et al., 2012; Holland et al., 2012), MSC is valued as a constant of 20% and 3% for hydrophilic BC and hydrophobic BC, respectively, which were derived from the observations conducted at Snowdome (2050 m) of the mid-latitude Blue Glacier (Conway et al., 1996).

More recently, the MSC of BC was re-evaluated based on the field measurements in Elson Lagoon (Barrow, Alaska) and at Dye-2 station (Greenland) during the melting season (Doherty et al., 2013). They suggested a ~~general~~ rough range of 10% to 30% in the study area. The method adopted in previous studies requires continuous sampling for about 2–3 weeks at each site, and thus is laborious to ~~be used~~apply for large-scale measurements in the polar area. Here, as an alternative, an experimental approach for calculating MSC was proposed which may provide a new way for MSC measuring, and a further comparison between the regional differences of MSCs is presented as well.

The melt-refreeze ice layer within the snowpack results from the refreezing of meltwater that percolates into the snow~~,~~. The suspended particles, especially those with larger surface areas, such as BC, may stay in place and freeze in the crystal lattice during the refreezing of meltwater (Novotny et al., 2002). That said, the freezing process does not preferentially exclude BC. Accordingly, here we assume that the BC concentration in the ice layer is identical to that in the meltwater. ~~and thus the concentration of BC in the ice layer can represent the BC values in the meltwater departing the snow.~~ The BC concentrations in the melt-refreeze ice layer and ~~in~~ its overlying snow layer were together to determine the MSC ~~associately~~ considering the thickness and density of the two layers. We conducted ~~The~~ the field measurements and sampling ~~were conducted~~ in Elson Lagoon, the Chukchi Sea and Canada Basin during the melt season (Fig. 1). After constraining the uncertainties of this new method, the estimated MSC ~~was~~ is compared to those ~~values~~ derived from the RS method in the same area, and further the spatial variability of MSC in the western Arctic will be discussed.

2 Field measurements and sample analysis

We collected the snow samples in Elson Lagoon northeast of Barrow (Barrow expedition), in the Chukchi Sea (Barrow expedition) and in the Canada Basin (1st South Korean Arctic Ocean expedition) during the late spring and summer over the past decade (2010 to 2018). The snow physical characteristics ~~(including the snow thickness, stratification and density)~~ were also measured during the three Barrow sea ice expeditions (the year 2015, 2017 and 2018) and the 1st South Korean Arctic Ocean expedition (the year 2010). In the 3rd Chinese Arctic expedition (the year 2008), only snow physics ~~(thickness,~~

stratigraphy and density) were observed.

The field measurement involves the snow thickness, snow density and stratification. In Elson Lagoon, we measured the snow depth along a 10km line before melt onset (April 15, 2015), and determined the average snow depth in this region. A far-shore site was chosen ~12 km away from the coast where the snow depth was close to the mean value (31.6 ± 5.4 cm) of this region (Fig. 1). The snow stratification was firstly recorded, and then snow density was measured at 2.5 cm vertical resolution using SnowFork instrument. Four points were measured per time in each layer. We applied the average value of snow density to characterize the snow layer. The snow depth was recorded at ablation stakes next to the snow pit. In the Chukchi Sea, the spatial variation of snow depth is more significant as compared with the Elson Lagoon due to the presence of ice ridge. We firstly selected a relatively smooth area of sea ice, and measured the snow depth along a 200m line in the centre region of the flat ice on April 6th, 2017. The observation site was chosen at a location close to the average snow depth and the measurement procedure was the same as that applied in Elson Lagoon. Note that there was a deviation between the observation sites of 2017 and 2018 due to the interannual variation in the ice condition over the Chukchi Sea (Fig .1). In Canada Basin, we conducted the measurements of snow depth at a 100m line over floe ice due to the smaller ice size and limited operating time. Snow density was measured using Tel-Tru densitometer (Tel-Tru Manufacturing Co., Inc., Rochester, NY) with an accuracy of 1 g, and a snow shovel of 2.5-cm in thickness. The thickness of snow layer and the position of melt-refreeze ice layer were measured using a ruler.

The sample collection was performed at three stages in Elson Lagoon and the Chukchi Sea during the expeditions in 2015 and 2017. At the stage before snow-melting onset, we collected snow from 4 cm above the sea ice up to the snow surface. At the early stage of melting, the upper snow layer was firstly collected, and then the underlying ice layer was sampled separately in the same snow pit. The newly fallen snow was also collected during theonce new snowfall occurred. In order to study the spatial distribution of BC, we dug up three snow pits to sample parallelly at each site (50 meters apart from each other) and measured the physical characteristics synchronously. Observations show that the differences in BC concentrations of the three snow pits are negligible, as the standard deviation value was one order of magnitude lower than the mean concentration. We took the average BC concentration from all three pits as the BC concentration at that exact site. In At the end of the snow-melting season and when most of the snowpack had melted, we collected the top 4-cm layer of snow to analyze the BC concentration in the melted snow. In 2018, we just collected samples of melting snow in the Chukchi Sea. Table 1 shows More more details are showed in Table S1.

Sampling was performed using a pre-cleaned plastic shovel and single-used vinyl gloves. Samples were stored in polyethylene bags that had been thoroughly washed with abundant deionized ultrapure water in the laboratory prior tobefore use. In the laboratory, the snow samples were allowed to melt in ambient temperature (18–20 ºC) and immediately filtered through quartz-fibre filters (25 mm, Whatman® QM-A). The filters were stored in an insulated cabinet with blue ice and kept in low temperature avoiding any bacteria to produce and transited to the laboratory in the University of Chinese Academy of Sciences for analysis.

We used two analytical methods to measure the concentration of BC. The quartz filters were firstly dried between 60 ºC and 70 ºC and then measured using an optical transmission analytical method (Model OT-21, Magee Scientific, California USA). The OT-21 is widely used in the measurement of atmospheric BC aerosol. AThereafter that, a 1.0 cm^2 punch was cut from each filter, and was analyzed for elemental carbon (EC) using the "Thermos-optical NIOSH 5040" method (Sunset Laboratory Inc., Forest Grove, U.S., which has been applied to measure EC in Svalbard snow (Forsström et al., 2013). A comparison between EC and BC in a previous study (Dou et al., 2017) showed that the values obtained from two different methods are highly correlated (R^2 = 0.97). For consistency, we adopt BC referring to BC and EC. Five blank filters were processed following the same analytical procedure as the samples, except that they were filtered with ultrapure water. The measured BC background of the filters (0.03±0.02 ng g^{-1}) are is an order of magnitude lower than the concentration of the ice layer. The values in Table S1 and Table S2 have been corrected by excluding blank contributions.

3 Results and discussion

During two Arctic Ocean Expeditions (the year 2008 and 2010), ice layers developed in almost all snowpacks over sea ice in the measurement area, and the snow stratigraphy and thickness exhibited highly spatial variabilities. The observed thickness of ice layers ranges from ~0.3 cm to ~2.8 cm. During the field measurements in Elson Lagoon in 2015, we recorded that the ice layer came into being on May 18th and May 22th, the early stage of the sea-ice melting season. The ice layer was observed in the Chukchi Sea on May 25th–28th, 2017, and on May 30th –31th, 2018.

The ice layer results from the refrozen meltwater that percolates into cold snow along with layer-parallel capillary barriers by heat conduction into surrounding subfreezing snow (Pfeffer et al., 1998; Massom et al., 2001; Colbeck et al., 2009). It detains BC particles in the meltwater, leaving the upper snow layer. Except for the formation mechanism mentioned above, ice layers could also generate from the radiation crust or liquid precipitation re-freezing (Massom et al. 2001). However, the BC concentrations in these two types of ice layers are in the same order of magnitude as those of new or recently-fallen snow. Besides, the radiation crust usually forms on the snow surface (Colbeck et al., 2009; Dou et al., 2013). The ice layer frozen from liquid precipitation is mostly formed during winter season before the snow-melt onset (Sturm et al., 2002; Langlois et al., 2017). These two types of ice layers cannot reflect the BC scavenging with meltwater, and thus were not considered in this study.

By measuring BC in the selected melt-refreeze ice layer and its overlying snow, it can be drawnwe observed that the concentration of the ice layer is 0.42±0.08 ng g^{-1} in the measurement area, meaning suggesting that ~0.42 ng of BC particles can be carried away from the snow layer by 1 gram water. Before estimating MSC, we compared the BC concentration ~~of~~ in the ice layer ~~is also compared~~ with those of other snow layers in the measurement area at different ablation stages. The BC concentration increased from 1.32±0.20 ng g^{-1} in the new snow to 2.42±0.63 ng g^{-1} in the generally melting snow (Fig. ~~S1~~2), and the concentration in the surface layer increased up to 15.91±1.12 ng g^{-1} ~~in~~ at the end of snow ablation.

The MSC is estimated based on the observations of BC, snow density and thickness. By determining the burden of BC per area (ng BC/cm^2) in the ice layer and the average original BC mass per unit area in the unmelted snowpack, the scavenging efficiency (MSC) is given by

~~According to the definition by Flanner et al. (2007), MSC can be given by:~~

$$MSC = h_i \cdot \rho_i \cdot C_{bi} / h_1 \cdot \rho_1 \cdot C_{b1} \qquad (1)$$

where h_i (cm), ρ_i (g cm^{-3}) and C_{bi} (ng g^{-1}) are respectively the thickness, density and BC mass concentration of the ice layer (Fig. ~~S2~~3); h_1 (cm), ρ_1 (g cm^{-3}) and C_{b1}(ng g^{-1}) are the same variables but for the snow layer before the melt event (Fig. ~~S2~~3). Note that determining scavenging efficiency with this method requires measuring the above factors at a given site at least twice, before and after the melt event.

If the snow physics and BC concentration were not measured before the melt event, we would choose another method to calculate MSC. We assumed that as the surface snow melts, BC particles scavenged by meltwater are refrozen in the melt-refreeze ice layer, that is, $h_1 \cdot \rho_1 \cdot C_{b1} = h_i \cdot \rho_i \cdot C_{bi} + h_2 \cdot \rho_2 \cdot C_{b2}$, where h_2 (cm), ρ_2 (g cm^{-3}) and C_{b2} (ng g^{-1}) are respectively the thickness, snow density and BC mass concentration of the melting snow overlying the ice layer (Fig. ~~S2~~3). ~~By determining the loading of BC per unit area (ng cm^{-2}) in the ice layer and in the partially melted snow layer above it, the scavenging efficiency (MSC) is given by:~~So that

$$MSC = h_i \cdot \rho_i \cdot C_{bi} / (h_i \cdot \rho_i \cdot C_{bi} + h_2 \cdot \rho_2 \cdot C_{b2}) \qquad (2)$$

~~In fact, t~~The assumption behind the proposed new method also implies that all of the melt-water generated from the original snow column is conserved in the ice layer and its overlying snow. Thus, $h_1 \cdot \rho_1$ is also equal to $(h_i \cdot \rho_i + h_2 \cdot \rho_2)$ in the assumption.

Since the new method largely depends on the conservation of snow mass and BC content before and after the ablation event, we validate the above presumption using the observations that involve snow sampling both before and after the melt event at ~~6~~ six sites during the Barrow expeditions (Table ~~S2~~1). The average of the snow density and BC concentration of the whole layer of snow were used to represent the situation (ρ_1, C_{b1}) of the upper part (h_1) of the snow layer before ablation. Here, deviations from 100% conserved is used to measure the conservation of BC (($h_i \cdot \rho_i \cdot C_{bi} + h_2 \cdot \rho_2 \cdot C_{b2}$)/ $h_1 \cdot \rho_1 \cdot C_{b1}$-100%) and snow (($h_i \cdot \rho_i + h_2 \cdot \rho_2$)/ $h_1 \cdot \rho_1$ -100%), and to evaluate the uncertainty in the derived scavenging efficiencies. The loss of snow mass and BC content after the ablation event are both smaller than 7.0% (Fig. ~~2a~~4a), indicating that most of the meltwater and BC within it was re-frozen in the ice layer and the BC content was substantially conserved. The assumption of the proposed new method is valid in the measurement area during the sampling period.

According to Eq. (2), we estimated the MSC (MSC_2) in the measurement area and compared it with the MSC_1 calculated based on Eq. (1). The result indicates that there is a slight difference in the MSCs calculated separately by the two methods. The bias of MSC ((MSC_2-MSC_1)/MSC_1) caused by the deviation of snow and BC from 100% conserved before and after melt is small than 7.2% (Fig. ~~2b~~4b). Further analysis showed that there is no ~~obvious~~ apparent correlation between the estimated bias of MSC and the degree of snow melting (Fig. ~~2b~~4b).

With the new method, we calculated the MSC in Elson Lagoon and compared it with that estimated according to equation (2) in Doherty et al. (2013) ~~estimated by Doherty et al. (2013) in the same area~~. Results indicate that the MSC (14.5%) calculated by the new method is smaller than that (20.4%) by the method of Doherty et al. (2013) based on the observations in this study. The difference in MSCs estimated by these two methods is reasonable since the latter represents the upper limits of MSC. Our estimation is close to the average value (16.2%) derived by repeated sampling (RS) introduced by Doherty et al. (2013) in the same area and is still within its best estimation [14.0%-20.0%]. ~~The result indicates that the MSC in Elson Lagoon is 14.5%±2.6%, close to the average estimation (16.2%±8.5%) by repeated sampling (RS) introduced by Doherty et al. (2013) and is still within its best estimation [14.0%-20.0%]. Our estimation of the MSC is also broadly consistent with that adopted by Flanner et al. (2007) in their model study. They assumed that the MSC is 3% for the hydrophobic BC and 20% for hydrophilic BC, given that the total BC is a combination of the two types of BC.~~

The scavenging efficiency of BC is mainly determined by the particle size and the hydrophobicity, which is interfered with other impurities since BC usually occurs in the particles as an "internal mixture" in the Arctic (Doherty et al., 2013). These influencing factors show significant regional differences due to various sources of BC and distinguishing deposition and transport processes (Korhonen et al., 2008; AMAP, 2011; Sharma et al., 2013; Schulz et al., 2019), leading to spatial variations in MSCs, which has been confirmed by the observations at Barrow and dye-2 station, Greenland (Doherty et al., 2013). Conway et al. (1996) found that the hydrophilic BC is much more efficiently scavenged with meltwater than the hydrophobic one. Flanner et al. (2007) further estimated that the MSC for hydrophilic BC is about 10 times that for hydrophobic one, meaning that the variations in fraction of the two in the snow with location also have important impacts on the spatial difference in MSC. ~~The MSC exhibits significant spatial variability due to the different particle sizes and hydrophilicity (Flanner et al., 2007).~~ From the observations in this study (Chukchi Sea, Elson Lagoon and Canada Basin) and the results of Doherty et al. (2013) (Elson Lagoon and Dye-2, Greenland), we investigated the spatial differences of MSC in the western Arctic. The average of the MSCs in the Canada Basin (23.6%±2.1%) is basically the same as that at the Dye-2 site, Greenland (23.0%±12.5%), while is ~~larger~~ more significant than that of Chukchi Sea (17.9%±5.0%); and Elson Lagoon has the lowest MSC (14.5%±2.6%) (Fig. ~~3~~5). We further analyzed the statistical significance of the differences in MSC at various locations. The Jonckheere-Terpstra test indicated that it is highly significant ($p < 0.01$) for Elson Lagoon < Chukchi Sea < Canada Basin, and the Mann-Whitney U test demonstrated that the difference from each other is moderately significant ($p < 0.1$). The average of the MSCs in the western Arctic is 18.0%±3.8%.

This study proposes a new method for large-scale measurements of MSC over the Arctic sea ice. The estimation of MSC requires the existence of a melt-refreeze ice layer. However, the limited data from our measurements cannot support a more extensive investigation. We reviewed the snow stratigraphy records obtained during the 3rd Chinese Arctic expedition in summer 2008 and the expedition hiking through the North Pole from 88 ºN to 90 ºN in late spring 1995 (Xiao et al., 1997). The records show that the melt-refreeze ice layers were widely developing over high latitudes of the Arctic, which is also confirmed by the observations in Svalbard in late spring 2007–2009 (Eckerstorfer et al., 2011). The widely distributed melt-refreeze ice layer in the Arctic suggests broader applicability for this new method in estimating the MSC of BC in the Arctic, for example, along the cruise lines where it is not pragmatic to carry out long-term continuously sampling. Nevertheless, we need to note that a melt-season ice layer may not form in regions of intense melt, where we cannot obtain the MSC value using the proposed approach in this study.

This technique assumes that BC particles are not preferentially removed during meltwater freezing. In fact, we do not rule out that very few BC particles can still be discharged during this process. Thus, this assumption may result in an underestimation of the BC content in the melt-water, in turn leading to an underestimation of MSC. Besides, this method does not account for influxes of BC from snowfall during the melt season, which may also lead to an underestimation of MSC in the case of snowfall occurring after melt onset. The method provides an estimate of the average seasonal MSC but does not capture temporal variations in efficiency.

3 Conclusions

The MSC of BC has is been found to be much less than 100% in few previously few studies, leading to enhanced concentrations of BC in surface snow, lowering albedo and accelerating the rate of snow melting. This study proposes a new experimental approach to determine the MSC by sampling the melt-refreeze ice layer and its overlying snow in the snowpits during the melting season, assuming the complete conservation of snow and BC content before and after the ablation event. The method is different from the established methods which require repeated sampling (RS method) over an extended period. The present observations confirm that the theory adopted in the proposed method is valid in the study area and the estimation bias of the calculated MSCs is not dependent on the melting degree during the ablation.

Further estimation with the new method demonstrated that the MSC exhibits regional differences in the western Arctic. In the measurement period, the average MSC in Canada Basin is the largest, which is close to that estimated in Greenland, followed by those in the Chukchi Sea and in Elson Lagoon. The spatial difference is suggested to be considered in the future simulation of BC-in-snow over the sea ice, rather than setting MSC as a constant in the snow and sea ice model. Combined with all available observations, we estimated an average of -MSC in the western Arctic of 18.0%±3.8% ranging from 13.0% to 30.0%.

Data availability. The observations of snow thickness, snow density and BC concentrations applied in this study are available as the Supplement.

Author contribution. TD designed the experiments and performed the analyses. TD, ZD, SL, YZ, QZ, MH and CL conceived field measurements and snow sampling. All authors participated in the writing of the paper.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1: BC concentrations observed in the melt-refreeze ice layer and its overlying snow. The thickness of snow and ice layer, and snow density observed simultaneously are shown. Note that the observations before and after the ablation events in Elson Lagoon and the Chukchi Sea during Barrow expedition are shown as site1-site6. The sampling locations and dates are also shown. Refer to figure 3 for the description of variable names. 'BC (surface melting snow)' denotes BC concentration in the top 4-cm layer of melting snow in the end of melt season when most of the snowpack had melted.

Sampling area	Site	Lat (ºN)	Lon (ºW)	h_1 (cm)	ρ_1 (g/cm^3)	C_{b1} (ng/g)	Sampling date	h_i (cm)	C_{bi} (ng/g)	h_2 (cm)	ρ_2 (g/cm^3)	C_{b2} (ng/g)	Sampling date	BC(Surface melting snow, ng/g)	Sampling date	Expedition
Elson Lagoon	1	71.32	156.37	5.5	0.32	1.72	April 26, 2015	0.7	0.36	3.0	0.36	1.72	May 18, 2015	14.9	May 31, 2015	Barrow Expedition
Elson Lagoon	2	71.32	156.37	5.4	0.30	1.70	April 30, 2015	0.8	0.31	2.5	0.35	1.70	May 22, 2015	15.3	May 31, 2015	Barrow Expedition
Elson Lagoon	3	71.32	156.38	10.9	0.32	1.11	May 7, 2015	1.7	0.41	5.0	0.35	1.98	May 22, 2015	17.9	May 31, 2015	Barrow Expedition
Chukchi Sea	4	71.37	156.54	11.3	0.31	2.11	April 15, 2017	1.8	0.48	5.0	0.36	2.11	May 25, 2017	16.1	June 5, 2017	Barrow Expedition
Chukchi Sea	5	71.37	156.54	13.2	0.29	1.82	April 16, 2017	2.5	0.34	4.0	0.35	1.82	May 26, 2017	16.1	June 5, 2017	Barrow Expedition
Chukchi Sea	6	71.37	156.54	8.5	0.25	2.91	May 1, 2017	1.0	0.55	3.0	0.36	2.91	May 28, 2017	17	June 5, 2017	Barrow Expedition
Chukchi Sea	7	71.37	156.55	––	––	––	––	1.5	0.5	3.0	0.32	2.43	May 30, 2018	14.2	June 10, 2018	Barrow Expedition
Chukchi Sea	8	71.37	156.55	––	––	––	––	0.9	0.36	2.5	0.29	2.11	May 30, 2018	15.9	June 10, 2018	Barrow Expedition
Chukchi Sea	9	71.37	156.55	––	––	––	––	0.5	0.41	2.0	0.24	2.33	May 30, 2018	14.8	June 10, 2018	Barrow Expedition
Chukchi Sea	10	71.37	156.55	––	––	––	––	1.2	0.43	3.5	0.31	2.52	May 31, 2018	17.3	June 10, 2018	Barrow Expedition
Chukchi Sea	11	71.37	156.55	––	––	––	––	0.4	0.31	1.0	0.32	2.14	May 31, 2018	17.5	June 10, 2018	Barrow Expedition
Canada Basin	12	75.03	159.48	––	––	––	––	2.8	0.39	3.5	0.28	2.93	July 22, 2010	––	––	1st South Korean Arctic Expedition
Canada Basin	13	77.98	159.64	––	––	––	––	1.7	0.54	2.5	0.31	3.81	July 26, 2010	––	––	1st South Korean Arctic Expedition
Canada Basin	14	79.51	160.02	––	––	––	––	1.9	0.45	2.5	0.32	3.32	August 1, 2010	––	––	1st South Korean Arctic Expedition

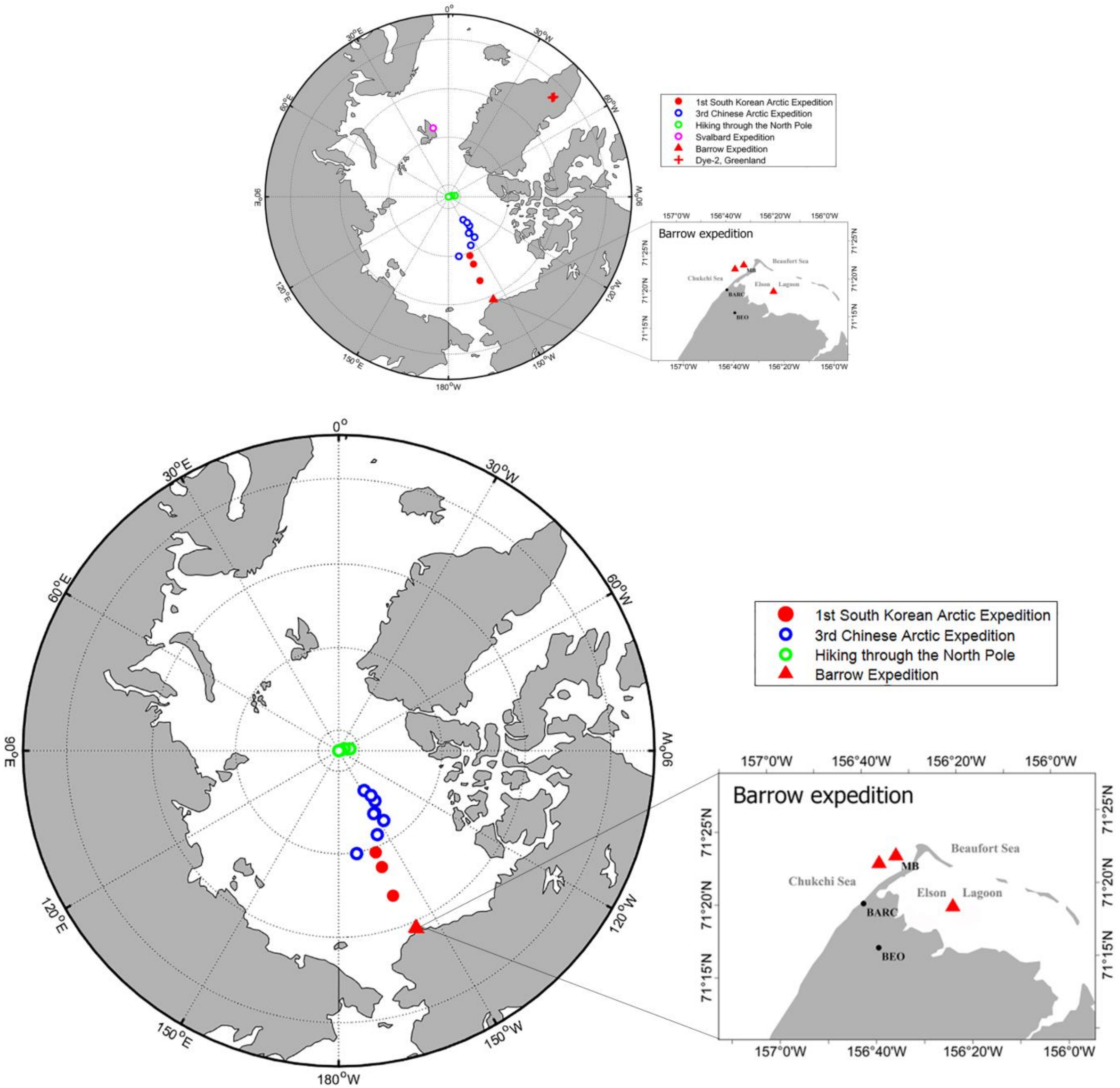
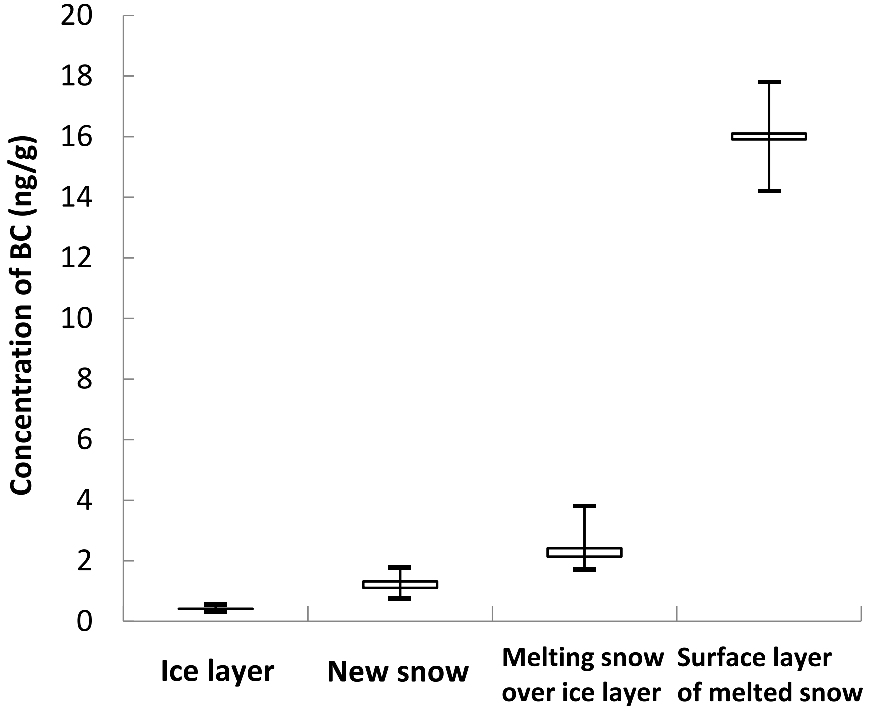


Figure 1: Locations of snow and ice layer sampling and the measurements of snow thickness and density in this study. Barrow Expeditions include the field measurements carried out in the Elson Lagoon in 2015, and in the Chukchi Sea in 2017 and 2018; the 3rd Chinese Arctic Expedition was conducted over the Canada Basin and the ~~center~~ centre region of Arctic Ocean in 2008; the 1st South Korean Arctic Expedition was conducted over the Canada Basin in 2010; the North Pole Expedition refers to the first Chinese expedition hiking through the North Pole from 88 ºN to 90 ºN in 1995 (Xiao et al., 1997)~~; the Svalbard Expedition was conducted by Eckerstorfer et al. (2011) in the field observations in 2007–2009~~. The open circle indicates the point at which the ice layer is observed. ~~Solid~~ The solid triangles and ~~squares~~ circles mark the locations for both sampling and on-site measurements. ~~Cross marks the location of Dye-2 where the MSC was estimated by Doherty et al. (2013).~~



330 Figure 2: The BC concentrations in the melt-refreeze ice layer and melting snow, and its concentrations in the new snow and the surface layer of melting snow are also shown as a comparison. New-snow samples were only collected in Elson Lagoon and the Chukchi Sea during the measurement period. The box indicates the mean (upper) and median (bottom) values of the observations, and the whiskers constrain the full extent of the observations.

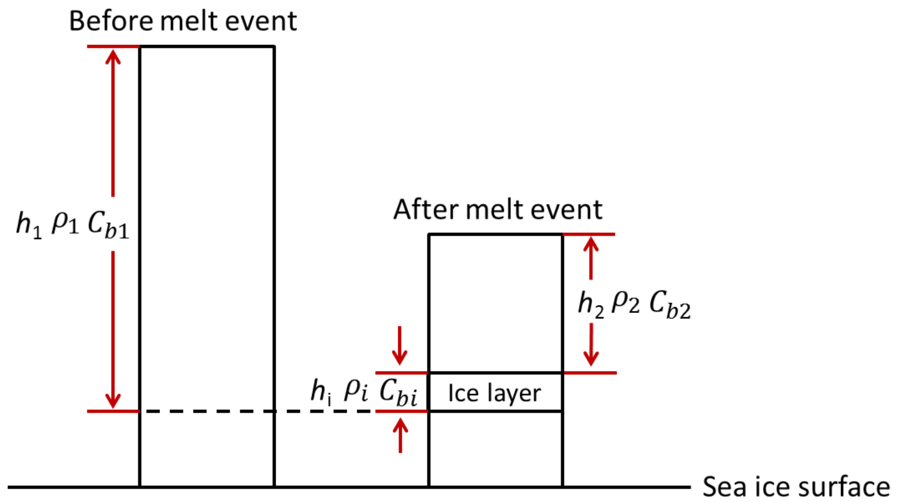


Figure 3: Conceptual sketch of snow overlying sea ice before and after the melt event. Variables relating to the snow and ice layer mentioned in Eq. (1) and Eq. (2) are shown.

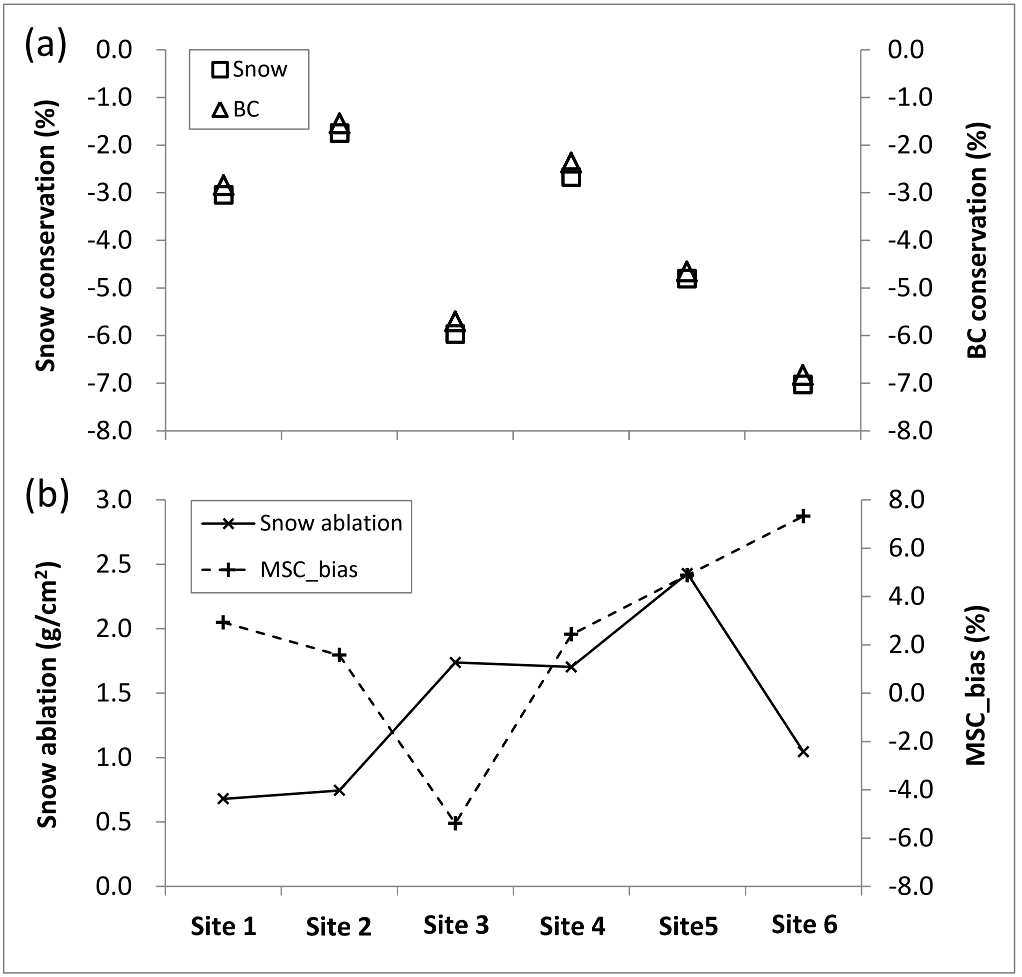


Figure ~~2~~4: Deviations from 100% conserved for snow and BC after ablation (a), snow ablation ($h_1 \cdot \rho_1 - h_2 \cdot \rho_2$) during the melt event and the bias ((MSC_2-MSC_1)/MSC_1)*100% of estimated MSC based on Eq. (2) (b). The ticks on the X-axis are matching sites given in Table ~~S2~~1.

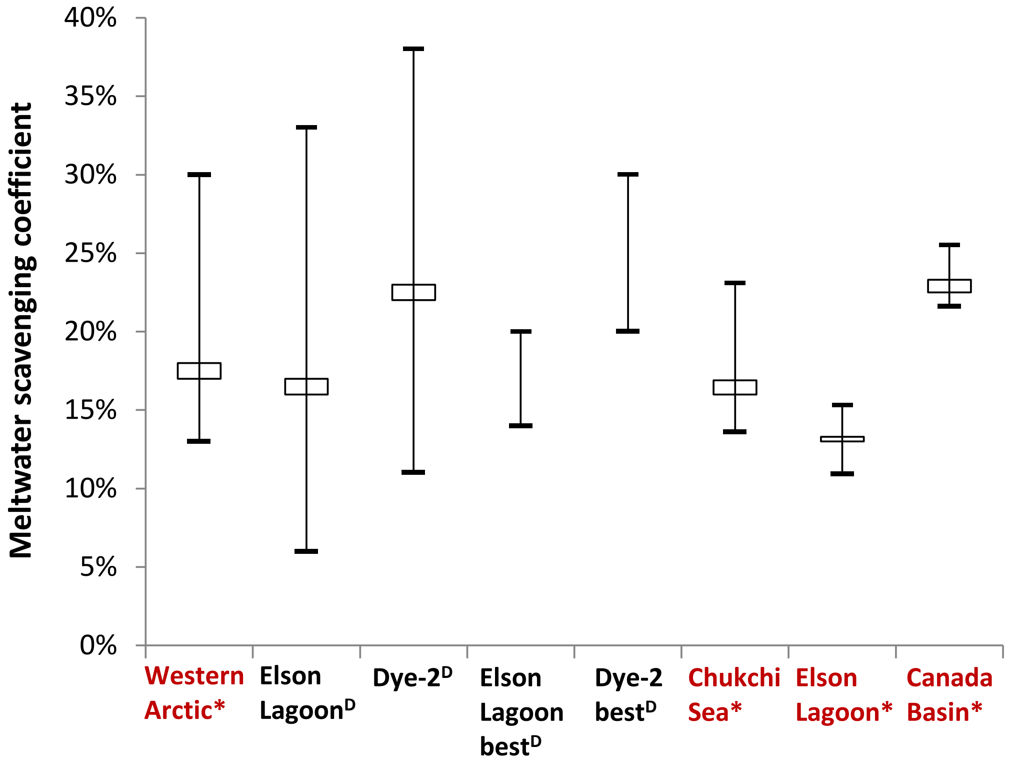


Figure 35: MSC of BC in different regions over the western Arctic. Superscript '*' indicates the results of this study (red), and '^{D}' indicates the results of Doherty et al. (2013). Elson Lagoon best^{D} and Dye-2 best^{D} indicate the best best-estimated range of MSC, respectively in Elson Lagoon and Dye-2, Greenland published in Doherty et al. (2013). The values of the western Arctic were estimated based on the observations in all measurement regions, and the best best-estimated values in Dye-2 and Elson Lagoon were employed in the estimation. The box indicates shows the mean (upper) and median (bottom) values, and the whiskers depict the extent.