

# 1 Variability of Black Carbon mass concentration in surface snow at Svalbard

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20

## 21 Abstract

22 Black Carbon (BC) is a significant forcing agent in the Arctic, but substantial uncertainty remains  
23 to quantify its climate effects due to the complexity of the different mechanisms involved, in particular  
24 related to processes in the snowpack after deposition. In this study, we provide detailed and unique  
25 information on the evolution and variability of BC content in the upper surface snow layer during the  
26 spring period in Svalbard (Ny-Ålesund). Two different snow-sampling strategies were adopted during  
27 spring 2014 and 2015, providing the *refractory* BC (rBC) mass concentration variability on a  
28 seasonal/daily and daily/hourly time scales. The present work aims to identify which atmospheric  
29 variables could interact and modify the mass concentration of BC in the upper snowpack, the snow layer  
30 which BC particles affects the snow albedo. Atmospheric, meteorological, and snow-related physical-  
31 chemical parameters were considered in a multiple linear regression model to identify the factors that  
32 could explain the variations of BC mass concentrations during the observation period. Precipitation

33 events were the main drivers of the BC variability. Snow metamorphism and activation of local sources  
34 during the snow melting periods appeared to play a non-negligible role (wind resuspension in specific  
35 Arctic areas where coal mines were present). The statistical analysis suggests that the BC content in the  
36 snow is not directly associated to the atmospheric BC load.

## 37 **1. Introduction**

38 In the last two decades, the Arctic region has been exposed to dramatic changes in terms of  
39 atmospheric temperature rise, sea ice decrease, and increase of air mass transport from lower latitudes  
40 bringing warmer and humid air masses containing pollutants and anthropogenic derived compounds (Law  
41 and Stohl, 2007; Comiso et al., 2008; Screen and Simmonds, 2010; Eckhardt et al., 2013; Schmale et al.,  
42 2018; Maturilli et al., 2019). Long-range transport and local emissions of combustion generating aerosols  
43 like black carbon (BC) can influence the radiative budget of the Arctic atmosphere, especially the impacts  
44 of atmospheric aging on the mixing state of BC particles (Eleftheriadis et al., 2009; Bond et al., 2013;  
45 Zanatta et al., 2018). When deposited over snow, numerous aerosol species directly increase the quantity  
46 of solar radiation absorbed by the snowpack, thus favouring snow aging processes and the decrease of the  
47 snow albedo (Hansen and Nazarenko, 2004; Flanner et al., 2007; Hadley and Kirchstetter, 2012; Skiles et  
48 al., 2018; Skiles and Painter, 2019).

49 Among these light-absorbing aerosols, *black carbon* (BC) particles are the most effective in  
50 absorbing the visible and near infrared solar radiation. These primarily emitted, insoluble, refractory and  
51 carbonaceous particles originate from natural and anthropogenic sources such as open fires or diesel  
52 engine exhausts. Currently, the anthropogenic emissions are higher compared to the natural ones  
53 (Moosmüller et al., 2009; Bond et al., 2013). In 2000, the energy production sector (including fossil fuels  
54 and solid residential fuels combustion) generated approximately 59% of the total global BC emissions  
55 while the remaining came from biomass burning (Bond et al., 2013). BC particles are characterized by a  
56 mass size distribution peaking around 100-250 nm (or mass equivalent diameter), e.g. 240 nm in the  
57 Svalbard area in spring (Bond et al., 2013; Laborde et al., 2013; Zanatta et al., 2016; Motos et al., 2019).  
58 The impact of BC particles absorbing the incoming solar radiation has indeed a non-negligible role in the  
59 Arctic region, which is already threatened by a two-fold temperature increase compared to the mid-  
60 latitude areas, the so-called “Arctic Amplification” (Bond et al., 2013; Cohen et al., 2014; Serreze and  
61 Barry, 2011). BC has an atmospheric lifetime of about seven days and has been directly targeted in  
62 important international mitigation agreements (AMAP, 2015). Theoretical and experimental results  
63 showed that the cryosphere is affected both by the BC-induced warming of the atmosphere and by direct  
64 and indirect BC effects on the snow once deposited over it (Flanner, 2013),

65 Atmospheric BC measurements in the Arctic regions are still rare, despite an extraordinary effort  
66 done by the international scientific community to evaluate the sources, transport paths, concentration, and  
67 climate impact (Eleftheriadis et al., 2009; Pedersen et al., 2015; Ferrero et al., 2016; Ruppel et al., 2017;  
68 Osmont et al., 2018; Zanatta et al., 2018; Laj et al., 2020). BC mass concentrations can be directly  
69 measured by using incandescent or thermal techniques and indirectly, by absorption measurements using  
70 an appropriate mass absorption cross-section (Petzold, 2013). Various terms such as refractory black  
71 carbon (rBC) for incandescent measurements, elemental carbon (EC) using thermal techniques, or  
72 equivalent black carbon (eBC) based on optical technique are used. Forsström et al. (2009) reported  
73 measurements performed in Arctic snow in the past and new measurements of EC in snow surface using  
74 filters and a thermo-optical method. The geographical and seasonal eBC variability was investigated in  
75 the Arctic region by Doherty et al. (2010). Other BC measurement in snow samples from the Arctic  
76 region can be found in Aamaas et al. (2011), Forsström et al. (2013), Pedersen et al. (2015), Gogoi et al.  
77 (2016), Khan et al. (2017) and Mori et al. (2019). Intercomparison of different techniques agree within a  
78 factor of 2 uncertainty at Alert (Sharma et al., 2017), Ny-Ålesund, and Barrow (Sinha et al., 2017).

79 A complex combination of processes are involved in the BC particles transfer from the  
80 atmosphere to the surface snow. Via a modelling approach, Liu et al. (2011) found that approximately  
81 50% of BC's total burden in the Arctic atmosphere is removed through wet deposition-related processes.  
82 Yasunari et al. (2013) estimated the intensity of BC dry deposition on the Himalayan glaciers; they found  
83 that the surface roughness and the surface wind speed are critical parameters in order to retrieve realistic  
84 results. In a recent study, Jacobi et al. (2019) confirmed the previous estimates suggesting that  
85 approximately 60% of the BC particles are deposited on the surface snow via wet deposition in spring in  
86 the Svalbard Arctic area. Models are still not fully able to describe the actual deposition and transport  
87 processes in Svalbard, resulting in underestimating the BC concentration in the snowpack (Eckhardt, S. et  
88 al 2015, Stohl, A. et al. 2013). Although wet deposition is suggested to be the main driver of BC  
89 concentration in the snow, little is known about other environmental processes potentially affecting the  
90 BC particles concentration once deposited, i.e. physical post-depositional processes.

91 In this study we present two unique experiments performed in a clean area close to the town of  
92 Ny-Ålesund (Svalbard) at the Gruvebadet Aerosol Laboratory (78.91734 N, 11.89535 E, 40 m a.s.l.),  
93 during spring 2014 and 2015. Daily and hourly time resolution samplings were performed on the snow  
94 surface to investigate which atmospheric variables could directly or indirectly modify the BC mass  
95 concentration in the surface snow once deposited. The daily sampling lasted for approximately 85 days to  
96 assess the intra-seasonal variability covering the transition from a cold period (April) to the melting  
97 period in late June. The hourly time resolution experiment was performed to investigate the existence of  
98 potential processes affecting the BC concentration over the diurnal cycle.

## 100 **2. Experimental Methods**

### 101 **2.1 Study Area**

102 Both experiments were conducted in the proximity of the Ny-Ålesund research station (78.5526  
103 N, 11.5519 E, 25 m a.s.l.), located on the Spitzbergen Island in Svalbard archipelago. Along the west  
104 coast, Svalbard is characterized by a maritime climate with an annual average temperature of -3.9°C in  
105 Ny-Ålesund (between 1994 and 2017) (Maturilli et al., 2019). On average, the snowpack starts building  
106 up in September and melts away at the end of May (Førland et al. 2011). Ny-Ålesund has become one of  
107 the reference locations for conducting Arctic climate studies focusing on atmospheric composition and  
108 physics. Long-term monitoring of atmospheric aerosols is performed at the Gruvebadet station (Feltracco  
109 et al., 2019, 2020, 2021a, 2021b; Moroni et al., 2018; Ferrero et al., 2016; Bazzano et al., 2015; Moroni et  
110 al., 2015; Zangrando et al., 2013; Scalabrin et al., 2012, Turetta et al., 2021), and at the Zeppelin  
111 observatory (475 m a.s.l.) (Eleftheriadis et al., 2009; Tunved et al., 2013; Lupi et al., 2016, and reference  
112 therein).

113

### 114 **2.2 Snow Sampling**

115 There are no standardized methods for sampling, filtering and analytical protocols for detecting  
116 atmospheric carbon deposited in snow, even if a few protocols have been developed (Ingersoll et al.,  
117 2009; Gallet et al., 2018; Meinander et al., 2020). In the present work, two different sampling strategies  
118 were adopted regarding the thickness of the sampled layer and the temporal sampling frequency.

119 Snow samples were collected during two field campaigns: The first campaign was carried out in  
120 Spring 2014, from April 1<sup>st</sup> to June 24<sup>th</sup> for a total of 85 days, it consists of daily sampling and it is  
121 referred hereafter as the “85-days experiment”. The second campaign was conducted in Spring 2015 from  
122 April 28<sup>th</sup> to May 1<sup>st</sup>. During these three days, measurements were collected with hourly sampling. This  
123 second campaign is hereafter referred as the “3-days experiment”. Snow samples were collected about 1  
124 km North-West of Ny-Ålesund (Figure 1). The area is a dedicated clean site for aerosols and snow  
125 sampling, with no fuel engine traffic. The wind at the site is usually blowing from east to west, and rarely  
126 from North to South, minimizing the emission of the town reaching the sampling area. The main wind  
127 pattern during the experiment is presented in Figures 1 and 2. The samples for both experiments were  
128 kept frozen until the lab analyses. The samples were collected using neck nylon gloves to avoid any  
129 contamination.

130 The two experiments aim to capture the rBC mass concentration on a daily basis in the surface  
131 snow (upper 10 cm) during the seasonal change and on an hourly basis on a thinner surface snow layer  
132 (upper 3 cm) during a daily cycle. Although wet and dry deposition are the main sources of BC in the

133 Artic snow, the aim of our experiments was to evaluate if other atmospheric parameters could contribute  
134 to the snow surface rBC mass concentration variability.

135 In the 85-days experiment, the first 10 cm of surface snow were collected on a daily basis  
136 (approximately at 11.00 am, GMT+2) in the same area, using a 5 cm diameter and 10 cm long Teflon  
137 tube. The samples were collected following a straight line leaving about 15 cm between the sampling  
138 points to minimize the spatial variability. The collected snow was homogenized in a pre-cleaned plastic  
139 bag and then, without melting, 50 mL was transferred into vial (Falcon™ 50mL Conical Centrifuge  
140 Tubes) for BC, coarse mode particles number (mix of soil, mineral coarse mode and possibly coal coarse  
141 mode) concentration and electrical conductivity analyses. The 85-days experiment was designed with the  
142 aim to investigate the BC presence in the upper snow layer, where most of the snow-radiation interaction  
143 takes place and where BC particles' presence can decrease the snow albedo (Doherty et al., 2010). Snow  
144 albedos increased rapidly and asymptotically as the snow depth increased. Visible albedos reached 0.9 for  
145 a snow depth of only 5 cm (Perovich et al. 2007). Moreover, this sampling strategy allowed to evaluate  
146 the variation of BC on a seasonal basis and to capture the impacts of wind, precipitation or melting.

147 During the 3-days experiment, the first 3 cm of surface snow were collected on an hourly basis in  
148 pre-cleaned vials in a delimited area of 2 x 2 m using the same sampling tools as above (Spolaor et al.,  
149 2019). In this case the samples were collected following a straight line leaving about 5 cm between the  
150 sampling points. The aim of the 3-days experiment was to investigate the potential daily cycle of surface  
151 BC concentration; therefore, we foresaw that small variations could derive from the impact of the daily  
152 variation of short-wave radiation (SWR) and subsequent induced snow metamorphism at the surface of  
153 the snowpack, often at cm scale. To avoid dilution of the signal, we reduced the vertical sampling  
154 thickness to 3 cm to enhance our chances of observing variation in the rBC mass concentration, if such  
155 variation exists.

156 The temperature at the surface of the snowpack (at 7 cm for 85-days and at 3 cm for 3-days  
157 experiment) was always measured. The daily/hourly snow accumulation was determined by measuring  
158 the emerging part of 4 poles placed around the sampling area. The average standard deviation calculated  
159 from the four poles provides us a reasonable estimate of the variability in snow accumulation\depletion  
160 within the sampling area. The standard deviation obtained ranges from 2 to 4 cm for the entire periods,  
161 indicating a limited spatial variability.

162

## 163 **2.3 Atmospheric Optical Measurements**

### 164 **2.3.1 Aethalometer (AE-31)**

165 In this study, the equivalent BC (eBC) concentration in the Boundary Layer (around 3 m a.s.l.)  
166 was measured by an AE-31 aethalometer (Gundel et al., 1983), during the 3-day campaign. The device is

167 equipped with 7-wavelengths (370, 470, 520, 590, 660, 880, 950 nm). It determines the attenuation  
168 coefficient by using the light attenuation ratio through a sensing spot and a referenced clean spot, both on  
169 a quartz fiber filter substrate. The sampling and reference spots surface areas are  $0.5 \text{ cm}^2$ , while the  
170 volumetric flow rate is  $4 \text{ L min}^{-1}$ . The flow rate was calibrated with a TetraCal (BGI Instruments)  
171 volumetric airflow before and after the field campaign. A 5 minutes temporal resolution was used for data  
172 acquisition. However, due to the low background concentration in the Arctic, the signal/noise ratio is  
173 high, so that data were hourly averaged. The data presented in this study were processed according to  
174 Segura et al. (2014) methodology. For this purpose the multiple scattering and filter loading effect  
175 (Weingartner et al., 2003) was corrected with new values of mass absorption cross section (MAC) and  
176 multiple scattering factor ( $C=3.1$ ), reported by Zanatta et al. (2018). The MAC value was derived using  
177 observations and observationally constrained Mie calculations in spring at the Zeppelin Arctic station  
178 (Svalbard,  $78^\circ\text{N}$ ). Zanatta et al. (2018) estimated the MAC at 550 nm ( $9.8 \text{ m}^2 \text{ g}^{-1}$ ) and at 880 nm ( $6.95 \text{ m}^2$   
179  $\text{g}^{-1}$ ), which we used to estimate MAC at 520 nm ( $10.2 \text{ m}^2 \text{ g}^{-1}$ ).

180

### 181 **2.3.2 Particle Soot Absorption Photometer (PSAP)**

182 During the 85-days sampling period the aerosol absorption coefficient was also measured by  
183 means of a 3-wavelengths PSAP (this instrument was not available during the 3-days experiment period).  
184 It measures the variation of light transmission through a filter where particles are continuously deposited  
185 with constant airflow. A second filter identical to the first one remains clean and is used as a reference to  
186 take into account possible variations of the light source, i.e. a 3-color LED (blue, green and red with  
187 wavelength centred around 470, 530 and 670 nm, respectively). The correction developed by Bond et al.  
188 (1999) was applied to consider the filter loading effect. The complete eBC mass concentration time series  
189 for the 85-days experiment was retrieved using the Aethalometer (first period) and the PSAP (second  
190 period), with an overlapping period with simultaneous measurements of 5 days. For the retrieved eBC  
191 mass concentration from the two instruments to be equal during the overlapping period, the PSAP eBC  
192 was calculated dividing the absorption measurements (at 530 nm) with a MAC equal to  $7.25 \text{ m}^2 \text{ g}^{-1}$   
193 (keeping the AE31 data as reference). Daily averages were calculated from the 1-minute data to compare  
194 with the rBC daily data obtained from the snow.

195

## 196 **2.4 Surface Snow measurements**

### 197 **2.4.1 Coarse Mode Particles Number Concentration**

198 The snow samples were melted at room temperature before the on-line coarse-mode particles and  
199 conductivity measurements (the water was pumped from the vials by a 12 channels peristaltic pump,  
200 ISMATECH, type ISM942). Specifically, the number concentration of coarse mode particles in the

201 surface snow was measured with a Klotz Abakus laser sensor particle counter. This instrument optically  
202 counts the total number of particles and measures each particle's size in a liquid constantly flowing  
203 through a laser beam cavity (LDS 23/23). The measurements size range of the instrument is from 0.8 to  
204 about 80  $\mu\text{m}$  with 32 dimensional bins (Table SI 1), not overlapping with that of the SP2. Only the 32<sup>nd</sup>  
205 bin has a dimensional range above 15.5  $\mu\text{m}$ , i.e. of 80  $\mu\text{m}$ . The data were recorded by a LabView® based  
206 software obtaining a sufficient number of data points in order to have a standard deviation less than 5% of  
207 the mean value. The particles number concentration was calculated using the constant water flow value.

208

#### 209 **2.4.2 rBC Measurement – SP2**

210 The rBC mass concentration and mass size distribution were measured following the methods  
211 described in Lim et al. (2014). The snow samples were melted at room temperature prior to the analyses.  
212 The vials with the melted snow were sonicated for ten minutes at room temperature. The samples were  
213 nebulized before the injection in the Apex-Q desolvation system (APEX-Q, Elemental Scientific Inc.,  
214 Omaha, USA). The nebulization efficiency was evaluated daily by injecting Aquadag® solutions with  
215 different mass concentrations, ranging from 0.1 to 100  $\text{ng g}^{-1}$ , obtaining an average value of 61%, that  
216 was used to correct all the BC mass concentrations reported in this manuscript. More details on the  
217 method can be found in Lim et al. (2014) and in Wendl et al. (2014).

218 The SP2 data were analyzed using the IGOR based toolkit from M. Gysel (Laboratory of  
219 Atmospheric Chemistry, Paul Scherrer Institute, Switzerland). The large amount of signals derived from  
220 every single particle are elaborated achieving rBC mass and number concentrations and size distributions.

221

#### 222 **2.5 Meteorological Parameters**

223 Meteorological parameters, in addition to the atmospheric and snow ancillary measurements,  
224 were used in the statistical exercise to study the variability of rBC mass concentration in surface snow  
225 samples as a function of the atmospheric conditions. BC particles are deposited on the snowpack  
226 following a combination of wet and dry deposition. However, once deposited on/in the snowpack other  
227 processes can potentially induced a significant variability in the surface BC content. The wind direction  
228 and its velocity can modify the BC distribution in the upper snowpack due to snow-mobilization. The  
229 solar radiation and relative humidity may enhance snow sublimation and surface hoar formation thus  
230 modifying the relative BC concentration in the upper snow layer by removing or adding “water” mass to  
231 the snow surface.

232 Air temperature and relative humidity at 2 meter height have been retrieved from a meteorological station  
233 located about 800 meters north of the sampling site, using a ventilated PT-100 thermo-couple by Thies  
234 Clima and a HMT337 humicap sensor by Vaisala, respectively. Wind speed and direction at 10 meter

235 height were obtained from a Combined Wind Sensor Classic by Thies Clima (see Maturilli et al., 2013).  
236 At about 50 m distance, the radiation measurements for the Baseline Surface Radiatio Network (BSRN)  
237 provide among others the downward solar radiation detected by a Kipp&Zonen CMP22 pyranometer  
238 (Maturilli et al., 2015). Both meteorological and surface radiation measurements are available in a 1-  
239 minute time resolution via the PANGAEA data repository (Maturilli et al., 2020). The daily/hourly mean  
240 values of the meteorological parameters were used in the statistical analyses of the 85-days/3-days  
241 experiment and in Figures 2 and 3 (the physical-chemical parameters from the snow samples are punctual  
242 values).

243

## 244 **2.6 Parameters considered in the statistical analysis**

245 The snowpack evolution is primarily driven by meteorological parameters, which are responsible for  
246 adding/removing mass to the annual snowpack. Wind can affect the snow pack evolution in several ways:  
247 1) by snow redistribution, 2) favouring the ablation\sublimation, and 3) lifting particles from nearby  
248 sources and areas. Surface snow and air temperatures are two fundamental parameters required to fully  
249 understand the varying conditions of the snow pack. In our study, the temperature variables are proxies  
250 for the melting episodes and for the presence of liquid water potentially affecting the concentration of  
251 impurities. The air and snow temperatures do not have a direct effect in the rBC concertation in surface  
252 snow, but they are fundamental indicators to identify the spring warming events ( $T > 0^{\circ}\text{C}$ , called also the  
253 Rain on Snow events - ROS) that yield the snow melting. Moreover, air and snow temperature could be  
254 relevant to evaluate possible snow metamorphism and the response of the upper snowpack to the  
255 meteorological conditions. Snow and air temperatures can be used during the 3-days experiment to  
256 evaluate the daily scale frequency and be helpful to investigate the daily scale variability of rBC in the  
257 surface snow.

258 The SWR is not expected to be directly linked to the surface mass concentration of rBC, however the  
259 surface process could affect it indirectly by favouring sublimation (water mass removal), as well as hoar  
260 formation (water mass addition) during the colder parts of the day (night/early morning). The relative  
261 humidity gives an idea of the amount of water present in the atmosphere and the high RH might favour  
262 the deposition of BC suspended by the formation of water droplets through the cloud condensation nuclei.  
263 This parameter is especially significant for the selected sampling location, nearby to the shore. Indeed,  
264 relative humidity values close or higher than 90% could be associated to fog or low cloud conditions and  
265 not directly to wet or dry precipitations. The last meteorological parameter considered is the precipitation  
266 amount. This aspect is important to understand the wet deposition processes able to transfer BC particles  
267 from the atmosphere to the snow surface.



268 The additional selected parameters are 1) the atmospheric eBC mass concentration, to investigate  
269 the possible link between eBC particles present in the atmosphere and the rBC in snow surface, 2) the  
270 coarse mode particles that could have a similar transport pathways to the black carbon and gives an idea  
271 of the amount of total impurities deposition and 3) the total water conductivity, an indirect measurement  
272 of the salinity content of the snow. It is important to note that the eBC and the rBC mass concentrations  
273 are not the same physical quantities: the former is obtained from an absorption measurement assuming a  
274 constant MAC, whereas the second is obtained via a laser-induced-incandescence method with an SP2  
275 empirically calibrated with a reference material (Petzold et al., 2013). Considering the location of the  
276 sampling site (<1 km from the coastline), the contribution of the ocean emissions to the snowpack  
277 chemical composition is significant. We considered the total conductivity as an indication of sea spray  
278 deposition, and to investigate common deposition patterns and/or similarities to the behaviour of BC  
279 (although BC is not emitted from ocean surface). The conductivity was also considered to determine if  
280 there was a large sea-spray aerosol event which could, potentially affecting the SP2 measurements (see  
281 supplementary material).

282

## 283 **2.7 Statistical Analysis**

284 Multiple linear regression was carried out to evaluate the relationship between the observed  
285 surface snow rBC mass concentration and the selected set of covariates consisting of the meteorological  
286 and snow physical-chemical parameters that could have a direct effect on controlling snowpack dynamics  
287 as well on the BC concentration as discussed in Section 2.6. All the atmospheric parameters described in  
288 the previous section (wind, snow and air temperature, incoming solar radiation, relative humidity, and  
289 snow precipitation amount) were initially considered as covariates to be included in the multiple linear  
290 regression. However, wind speed and direction, as well as the atmospheric stability, expressed as vertical  
291 wind speed, were removed because preliminary statistical analyses indicate that none of them is  
292 associated with the observed variations in snow rBC mass concentrations. This does not mean that such  
293 parameters do not play a role in controlling the BC concentration, but that no statistically significant  
294 associations were found with the data collected in our study and thus these parameters were no longer  
295 considered in the statistical analyses discussed below.

296 Multiple linear regression models were fitted on the logarithm scale because the distribution of rBC  
297 concentrations in both experiments is characterized by a significant skewness. Coarse mode particles  
298 number concentrations and conductivity were also log-transformed to linearize their relationships with  
299 log(rBC). The regression model fitted on the two experiments is

300

$$\log(rBC) = \beta_0 + \beta_1 \log(dust) + \beta_2 eBC + \beta_3 temp + \beta_4 snow + \beta_5 swr + \beta_6 \log(cond) + \epsilon.$$

301  
302 In the above model, ‘dust’ indicates coarse mode particles number concentrations, ‘temp’ is the snow  
303 temperature at 7 cm depth for the 85-days experiment (daily resolution) and at 2 cm depth for the 3-days  
304 experiment (hourly resolution), ‘snow’ is a binary indicator for the presence of solid precipitation, ‘swr’ is  
305 solar incoming shortwave radiation, ‘cond’ is the conductivity and  $\varepsilon$  is a zero-mean normal error.  
306 Graphical inspection of residuals plots and normal probability plots confirmed that after the logarithm  
307 transformations, the regression models meet the assumptions of linearity, constant error variance (called  
308 *homoscedasticity* in the statistical literature) and normal errors. The statistical analyses were performed  
309 with the statistical language R (R Core Team, 2020).

310

### 311 **3. Results and Discussions**

#### 312 **3.1 Seasonal BC variability in surface snow**

##### 313 **3.1.1 Atmospheric eBC and atmospheric condition**

314 During the experimental period, the atmospheric eBC concentration ranged between from  $80 \text{ ng m}^{-3}$  to <  
315  $5 \text{ ng m}^{-3}$  (Figure 2) with an average of  $34 \pm 23 \text{ ng m}^{-3}$ . The highest concentrations were measured at the  
316 beginning of the campaign, especially from April 15<sup>th</sup> to 27<sup>th</sup>, followed by a general decreasing trend  
317 characterized by the presence of several concentration peaks (on May 8<sup>th</sup>, 17<sup>th</sup> and 24<sup>th</sup>) potentially due to  
318 Eurasian fires, as already suggested from Feltracco et al., 2020 (Figure S1). The ammonia daily  
319 concentration time series (the only available biomass burning tracer for that period in the area) measured  
320 at the Zeppelin station is plotted together with the Gruebadet atmospheric BC measurements in Figure  
321 S3. Biomass burning is a significant source of atmospheric ammonia (Andreae and Merlet, 2001), often  
322 affecting the Arctic region (Moroni et al. 2020). As shown in Figure S3, both time series have a similar  
323 behaviour at the very beginning of the campaign, from April 3<sup>rd</sup> to 8<sup>th</sup> and during the period between May  
324 7<sup>th</sup> and 21<sup>st</sup>. This suggests that the BC detected in the atmosphere could be originated from biomass  
325 burning episodes during these two time periods. During the 85-days sampling period, wind was  
326 characterized by the following median values (25<sup>th</sup> and 75<sup>th</sup> percentiles) for direction and speed:  $205^\circ$   
327 ( $152^\circ$ ,  $257^\circ$ ) and  $2.7$  ( $1.9$ ,  $3.7$ )  $\text{m s}^{-1}$ , respectively, therefore mostly coming from south-west (Figure 2).  
328 Daily air temperature at 3 m increased during the campaign from  $-15^\circ\text{C}$  to about  $+5^\circ\text{C}$  (Figure 2)  
329 following the seasonal variation of incoming solar energy: from  $100$  to  $300 \text{ W m}^{-2}$  with an average of  $185$   
330  $\pm 75 \text{ W m}^{-2}$  (Figure 2, orange line). The snow precipitation episodes are presented as daily-accumulated  
331 values (Figure 2, blue bars) ranging from zero to 12 cm.

332

##### 333 **3.1.2 Surface Snow Conditions**

334 Over the 85 days experiment, the snow rBC mass concentration varies from 0.2 to 6 ng g<sup>-1</sup> (Figure 2),  
335 with an average of 1.4 ± 1.3 ng g<sup>-1</sup>, in agreement with results available in the literature (Mori et al., 2019;  
336 Jacobi et al., 2019; Aamaas et al., 2011). An increasing trend can be observed for the rBC mass  
337 concentration in the surface snow across the sampling period. The median of the rBC mass equivalent  
338 diameter in the snow is 313 ± 35 nm (Figure 2), similar to what obtained in other studies (e.g. Schwarz et  
339 al., 2013). The rBC mass equivalent diameter show high variability, ranging from 200 to 500 nm.  
340 However, since the rBC concentrations were low, the evaluation of the geometric mean of the particles  
341 diameter for the biggest sizes, above 300 to 400 nm, has been considered as qualitative information due to  
342 the high signal noise.

343 The number of coarse mode particles (Figure 2, blue line) shows a constant concentration in the first half  
344 of the campaign (1<sup>st</sup> April - May 11<sup>th</sup> - average concentration of 3435±1824 # ml<sup>-1</sup>) whereas it increases in  
345 the second half (12<sup>th</sup> of May to 27<sup>th</sup> of June - average concentration of 7782±5683 # ml<sup>-1</sup>), especially after  
346 the 1<sup>st</sup> of June (1<sup>st</sup> of June to 27<sup>th</sup> of June - average concentration of 9352±6741 # ml<sup>-1</sup>), in concomitance  
347 with the onset of the snow melting period. The conductivity (Figure 2, green line) also shows an  
348 increasing trend at the end of the sampling campaign when snow is melting, with an overall average value  
349 of 30 ± 8 µS. The spatial variability of rBC, calculated in the same manner as proposed by Spolaor et al.  
350 (2019) for other species, was obtained from six surface snow samples collected in the four corners of the  
351 sampling area and two surface snow samples in the centre right before the beginning of the experiment.  
352 The following rBC mass concentrations were obtained: a) 3.95 ng g<sup>-1</sup>; b) 4.92 ng g<sup>-1</sup> c) 4.20 ng g<sup>-1</sup> d) 3.10  
353 ng g<sup>-1</sup> e) 3.82 ng g<sup>-1</sup> f) 3.58 ng g<sup>-1</sup>, resulting in a rBC spatial variability of 16% in the surface snow of the  
354 considered sampling area.

355

### 356 3.1.3 Statistical Results

357 The fitted multiple linear regression model for the 85-days experiment data explains the 69% of  
358 the variance of the logarithm of the snow rBC mass concentration ( $R^2 = 0.69$ ). The fitted model indicates  
359 the presence of strongly statistically significant associations of the (log transformed) snow rBC mass  
360 concentration with the coarse-mode particles number concentration ( $p < 0.001$ ) and the snow temperature  
361 ( $p < 0.001$ ). A weaker association is found with the occurrence of snow precipitations ( $p = 0.03$ ). The  
362 statistical associations of rBC mass concentration with the other covariates considered in the model are  
363 non-significant. See Table 1 for the estimated coefficients and the corresponding p-values.

364 In order to interpret the statistical results, the description of the 85-days campaign is split into two periods  
365 identified as the transition from the “cold” to the “melting” state. The first period occurred before the end  
366 of May: the rBC mass concentration often increases with snowfall episodes (April 9<sup>th</sup>/10<sup>th</sup>/11<sup>th</sup> and 17<sup>th</sup>,

367 May 17<sup>th</sup>, 22<sup>nd</sup> and 27<sup>th</sup>/28<sup>th</sup>; June 1<sup>st</sup>) as suggested by previous studies, with exceptions for April 24<sup>th</sup> and  
368 May 7<sup>th</sup>. Over the sampling period, a weakly statistically significant positive association ( $p = 0.03$ ) was  
369 found between snow rBC mass concentration in surface snow and the occurrence of snow precipitations.  
370 BC wet deposition processes are estimated to remove 50% - 60% of the total atmospheric BC burden in  
371 the Arctic (Liu et al., 2011; Jacobi et al., 2019). In our study, the wet deposition impacts could be partially  
372 masked due to the sampling frequency and the wind snow. In Kongsfjord, a strong wind is often present  
373 during the precipitation events (Figure 2). Consequently, the freshly deposited snow is frequently  
374 removed from the surface before being able to sample it. Interestingly, our observations show that, on a  
375 daily scale, the precipitation episodes are not clearly related to a decrease in the atmospheric eBC mass  
376 concentration (Figure 2). A possible explanation is that the precipitation amounts were small so that the  
377 precipitation events did not significantly alter the atmospheric BC reservoir.

378 In the second period, from the beginning of June, the atmospheric temperature increases, causing the  
379 snow-melting season's onset. At the beginning of June, the snow rBC mass concentration increases up to  
380 approximately  $5 \text{ ng g}^{-1}$ , and a simultaneous increase was detected in the coarse mode particles number  
381 concentration (peaks between June 4<sup>th</sup> and 7<sup>th</sup>). As suggested in previous studies, the surface melting  
382 process could explain the observed increase in rBC and coarse mode particles concentrations. However,  
383 we also have to consider that rBC can be dry deposited, as it has been recently suggested (up to 50-60%;  
384 Liu et al., 2011; Jacobi et al., 2019). Very few field validation data exist for estimating the amount of dry  
385 deposition at the snow surface, and this process is often used as an ancillary information since most  
386 models underestimate the BC in the Arctic snowpack compared to field measurements.

387 Our data support the hypothesis related to local sources' activation in enhancing the dry deposition  
388 impacts in an old mining town as Ny-Alesund. Especially during poor snow cover conditions, as during  
389 the snow-melting season, coarse mode particles as residuals of carbon extraction mining activities are  
390 available for wind lift\suspension (Vecchiato et al. 2018). The possible effect of local sources' activation  
391 is further supported by a recent analysis of the Brøggerbreen glacier and Ny-Ålesund annual snowpack.  
392 This analysis shows the presence of retene (an organic compound frequently used to track the presence of  
393 coal), most likely due to local sources (Vecchiato et al., 2018).

394 The simultaneous increase of rBC mass and coarse mode particle number concentrations during the  
395 second part of the experiment (e.g. visible between June 3<sup>rd</sup> and June 7<sup>th</sup>-8<sup>th</sup>) could be explained via  
396 similar post-depositional processes: snow melting and sublimation. The episodes of snow surface melting  
397 can significantly affect the snow particulate content and we hypothesize that the hydrophobicity of pure  
398 BC particles, and of several species in the coarse mode particles, might affect its physical location in the

399 snowpack (in the literature, the response of the BC particles is still debated): the hydrophobicity of the  
400 particles can cause the surface concentration to increase while losing water mass through percolation.  
401 This could lead into a positive feedback process: the increase of BC concentration can thus enhance snow  
402 sublimation (water evaporation) resulting in a further increase of BC concentration in surface snow, and  
403 so on.

404 In this study, the estimated statistical association between snow rBC mass concentration and the daily  
405 snow temperature is negative and strongly significant ( $p < 0.001$ ). During the 85-days experiment, we can  
406 distinguish two events where the temperature appeared to play a role in the BC concentration. Both of  
407 them show an increase in rBC mass concentration during melting/refreezing episodes, in agreement with  
408 other studies (Aamaas et al., 2011; Xu et al., 2006; Doherty et al., 2013; Doherty et al. 2016). The first  
409 event occurred between May 5<sup>th</sup> to May 12<sup>th</sup> and the second event after May 20<sup>th</sup>, when the proper snow  
410 melting began (Figure 2). The first event was characterized by a rapid rise of the daily air temperature  
411 (from  $-6^{\circ}\text{C}$  to  $-1^{\circ}\text{C}$ ) in concomitance to a snow precipitation event, followed by a rapid temperature  
412 decrease to  $-6^{\circ}\text{C}$ . The surface snow (10 cm) mirrored this behaviour, first rising from  $-6^{\circ}\text{C}$  to  $0^{\circ}\text{C}$ , and  
413 then cooling down to  $-6^{\circ}\text{C}$ . During this warm event, the upper snow strata underwent a melting episode  
414 with surface water percolation (although limited), making the surface BC concentration to increase. The  
415 second event started approximately on May 20<sup>th</sup> and lasted until the end of the experiment (Figure 2).  
416 During this period, the atmospheric temperature increased constantly, and the snowpack started to melt  
417 consequently. Moreover, surface BC concentration increased almost continuously from May 25<sup>th</sup> to its  
418 maximum observed on June 6<sup>th</sup>. Afterward, the upper snow rBC mass concentration tended to decrease  
419 following the rapid snowpack decline.

420

## 421 **3.2 Diurnal variation of rBC in surface snow**

### 422 **3.2.1 Surface Snow/Atmospheric Aerosol Content and Atmospheric Conditions**

423 The 3-days experiment was performed at the end of April 2015, during the Arctic spring. The  
424 samples were collected on an hourly basis over 3 days achieving a high-resolution sampling frequency.  
425 The atmospheric concentration of eBC ranged from 2 to  $50\text{ ng m}^{-3}$ , decreasing during the sampling period  
426 and not showing any particular diurnal pattern (Figure 3). The mean value of the atmospheric eBC mass  
427 concentration is  $34 \pm 23\text{ ng m}^{-3}$ , similar to the average of the 85-days experiment.

428 The surface snow rBC mass concentration undergoes to daily time scale change of surface  
429 concentration showing up to 2-fold hourly increases (Figure 3, bottom panel, smoothed dark blue line).  
430 rBC mass concentrations of approximately  $15\text{ ng g}^{-1}$  were measured in the snow samples from the  
431 beginning of the sampling to the end of the second day. Later, from the beginning of the third day until

432 the end of experiment, rBC mass concentrations show an average concentration of about  $5 \text{ ng g}^{-1}$  (Figure  
433 3). The average value over the whole sampling period is  $9 \pm 5 \text{ ng g}^{-1}$  (approximately 6 times higher than  
434 during the 85-days experiment). The rBC mass size distribution was characterized by a median value of  
435 the geometric means of about  $230 \pm 32 \text{ nm}$ , significantly lower than that which was measured during the  
436 85-days, and still in agreement with previous studies (Sinha et al., 2018; Schwarz et al., 2013). The  
437 concentrations of EC and OC measured in parallel snow samples (not of the same volume) are reported  
438 and described in Figure S4; the interpretation of the differences between the rBC and the EC  
439 measurements in snow samples was beyond this manuscript's objectives.

440 The number concentration of coarse mode particles remains stable in the first half of the  
441 experiment, until the end of April, and shows an average value over the three days of  $26642 \pm 9261 \text{ \# mL}^{-1}$   
442 <sup>3</sup>. The water conductivity shows a similar behaviour, and it is characterized by an average of  $39 \pm 9 \text{ \#S}$   
443 (30% higher than during the 85-days experiment).

444 All the measured snow impurities show two common features (see supplementary material and  
445 Figure S4): first, a decrease in the absolute values detected between 4 and 8 a.m. of April 30<sup>th</sup>, despite the  
446 absence of precipitations or any other particular meteorological episode (Figure 3); second, the impact of  
447 the snow precipitation event from approximately 4 p.m. to midnight of the April 30<sup>th</sup>, where the  
448 concentrations of aerosols in the snow slightly increased at the very beginning whereas decreasing at the  
449 end of the event. Only the BC core diameter remained above the average when the other aerosol snow  
450 content decreased (up to approximately 400 nm of mass equivalent diameter), consequently returning to  
451 the average value. The spatial variability of BC, calculated as proposed by Spolaor et al. (2019) for other  
452 species, was obtained by the analysis of 5 surface snow samples, collected in the four corners of the  
453 sampling area and one in the centre obtaining the following concentrations: a)  $10.17 \text{ ng g}^{-1}$ , b)  $10.64 \text{ ng g}^{-1}$ ,  
454 c)  $7.04 \text{ ng g}^{-1}$ , d)  $11.98 \text{ ng g}^{-1}$ , and e)  $11.91 \text{ ng g}^{-1}$ , thus resulting in a spatial variability of 19%. Clear  
455 sky conditions were observed for the duration of the sampling period except for the snowfall occurred at  
456 the end of the third day.

457

### 458 3.2.2 Statistical Results

459 The multiple linear regression model for the 3-days experiment explains the 78% of the snow  
460 rBC mass concentration variance, a percentage higher than the 85-days experiment, likely due to the more  
461 stable atmospheric conditions and the greater interaction with the atmosphere of the upper 3 cm of the  
462 snow pack compared with the depth resolution used during the seasonal experiment. Similar for the 85-  
463 days experiment we evaluate (Figure S4) the 10 days back-trajectory during the 3 days of the experiment.  
464 The result suggests that the air mass arriving in Ny-Ålesund during the experiment were mainly  
465 originated from the Arctic Ocean.

466 The fitted multiple linear regression model indicates a statistically significant association between  
467 the logarithm of the rBC mass concentration in the snow and the logarithm of the conductivity ( $p <$   
468  $0.001$ ), the logarithm of the number concentration of coarse-mode particles ( $p < 0.001$ ) and the  
469 occurrence of snow precipitations ( $p < 0.001$ ). The estimated coefficients of the covariates are reported in  
470 Table 1. In Figure 4 are displayed the 95% and 90% confidence intervals for the estimated coefficients of  
471 regression models fitted to two experiments (85-days and the 3-days). Since the covariates considered in  
472 the two experiments have quite different unit scales, Figure 4 shows the confidence intervals for the  
473 standardized covariates. The standardization simplifies the comparison among the estimated effects of the  
474 different covariates and between the two experiments, in this way allowing a visual comparison of the  
475 estimated statistical associations between the logarithm of the snow rBC mass concentration and the  
476 considered parameters.

477 The association between the logarithm of the coarse-mode particles number concentration and the  
478 logarithm of the snow rBC mass concentration is positive and strongly significant ( $p < 0.001$ ), similarly to  
479 what observed for the 85-days experiment, confirming the similar behaviour of these types of particles  
480 also in the surface snow pack (3 cm). The association between the logarithm of conductivity and the  
481 logarithm of the snow rBC mass concentration is positive and strongly significant ( $p < 0.001$ ). Snow  
482 conductivity is mostly influenced by the presence of sea salt ions (mainly coming from sea spray aerosol  
483 considering the location of the experimental site) in the snow samples. Sea spray aerosol is not considered  
484 a source of rBC and a direct effect of the sea spray emission on the rBC snow concentration is here  
485 consider negligible. However the positive association between rBC and conductivity can be explained by  
486 the fact that both sea spray aerosol and BC particles (as well dust) undergoes to similar dry deposition  
487 process (when concentration increase) favoured by the stable atmospheric condition occurred during the  
488 experiment (with the exception of the snow event during the third day) as well from similar physical  
489 removal process (concentration decrease) from the snow surface. Considering we are exploring the rBC  
490 concentration change in the upper 3 cm, we explore the possible existence of a daily cycle. The BC  
491 particles are known to be non-volatile and not photo-chemically active, therefore the decrease/increase in  
492 their concentration observed during the experiment can only be driven by physical process such as wind  
493 erosion and snow deposition. However and additional process that might drive the rBC concentration  
494 change in the upper snow pack is the condensation of water vapour on the top of the snow crystals and the  
495 formation of surface hoar as well the sublimation. The formation of surface hoar has the effect to adding  
496 “water” mass without BC particles in the snow surface causing a relative rBC dilution, while sublimation  
497 has the effect remove “water” mass causing a relative concentration increase. Surface hoar and  
498 sublimation are depending mainly by the temperature and solar radiation, two parameters that exhibits the  
499 diurnal cycle (Figure 4). From the statistical analysis no associations were found on rBC with the

500 incoming solar radiation (at hour resolution) and the snow temperature during the sampling period. These  
501 results indicate that the rBC mass concentration in the surface snow does not undergo to diurnal changes  
502 and this process are negligible in controlling the rBC snow surface concentration.

503 The occurrence of snow precipitations is negatively associated with the logarithm of the rBC  
504 mass concentration in the snow ( $p < 0.001$ ). As previously remarked, the aerosol scavenging intensity is  
505 not measurable with snow sampling strategies based on the sampling of a constant snow thickness from  
506 the surface (3 cm in this case). We tentatively explain the negative relation observed in this study with the  
507 high frequency sampling, being able to follow the evolution of the BC particles scavenged during a snow  
508 episode (from 3 to 12 p.m. of the 30<sup>th</sup> April 2015). The beginning of the precipitation episodes appeared  
509 to remove the highest amount of BC particles, leaving the atmosphere cleaner as reflected by the lower  
510 BC mass concentration revealed in subsequent samples. The snow collected at 18:00 of April 30 showed  
511 a higher amount of rBC as well as the highest coarse mode particles number concentration and  
512 conductivity. In the next few hours, from 9 to 12 p.m., the snow precipitations were depleted in terms of  
513 aerosol content and rBC mass concentration.

514

#### 515 **4. Conclusions and Future Perspectives**

516 The seasonal and daily experiments (85- and 3-days long, respectively) suggest that the rBC  
517 concentration in the upper snow layer is not only driven by a cumulative process, as it happens when the  
518 entire annual snow pack is evaluated, but it is a rather more complex process involving atmospheric,  
519 meteorological and snowpack conditions. Our results based on a multiple linear regression models  
520 suggest that the amount of BC in the surface snow is not associated to the BC atmospheric load. This  
521 finding suggests that, despite the potentially high atmospheric BC concentrations (as in the case of long-  
522 range transport of biomass burning plumes), this parameter does not seem to be the primary driver of the  
523 variations in the surface snow rBC over the experiment periods. In both experiments, the coarse mode  
524 particles are positively associated with the snow BC mass concentration, suggesting that the BC and  
525 coarse mode particles deposition undergo similar deposition and, in case, to post-depositional processes in  
526 the upper snowpack. Specifically, before the beginning of the melting season, the wet deposition episodes  
527 appeared to have major impacts, whereas the activation of common local sources favour the wind  
528 suspension from uncovered areas enhancing the intensity of dry deposition processes, might lead to an  
529 accelerated snow melting.

530 Our results also suggest that in order to explain the observed BC mass concentration variability  
531 during seasonal and diurnal time ranges other processes than wet and dry depositions should be  
532 considered. Surface melting episodes enrich the BC content in the surface layer not because of an  
533 enhanced deposition but mainly because of water mass loss. In particular, the snow mass loss is stronger



534 during the snow-melting season, where an increase in the rBC concentration could significantly alter the  
535 snow albedo and further enhance the radiative absorption, hence promoting a positive feedback. The  
536 proposed processes and the rBC concentration determined in Ny-Ålesund could be influenced by local  
537 emission in particular at the beginning and at the end of the snow season when the snowpack does not cover  
538 homogeneously the surface. However, the process described by our results could occur in other Arctic  
539 sites although with different magnitudes and impacts.

540 The remarkable diurnal and daily variability, as well as the complex interdependent mechanisms  
541 affecting the rBC mass concentration in the Arctic surface snow, makes the results of albedo-based  
542 radiative impact model of the active layer a potential source of erroneous conclusions: the impacts of long  
543 distance biomass burning episodes might be overestimated, whereas the impact of local sources and dry  
544 deposited impurities during the melting season might be underestimated. Additional empirical studies are  
545 therefore necessary in order to improve our understanding of the involved physical mechanisms and to  
546 better constrain modelling studies.

547

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563 Environments).

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566

567 **Data Availability**

568 Meteorological and surface radiation data are available at the PANGAEA database (Maturilli, 2015a;  
569 2015b; 2015c; 2016a; 2016b; 2018a; 2018b; 2018c; 2018d; 2018e). The data for precipitation amount at  
570 Ny-Ålesund can be accessed via the eKlima database of MET Norway. The BC data are available upon  
571 request.

572

573 **Author Contributions**

574 Author contributions. AS, EB, DC and MB conceived the experiments; AS, EB, DC, and LP collected the  
575 samples; MB measured the samples; KM and MMaz provided the atmospheric eBC concentrations; SC  
576 and DC provided the back-trajectories analyses; CV performed the statistical analyses with inputs from  
577 MB and AS. MB prepared the manuscript mainly with inputs from AS, J-C. G and DC (in the methods  
578 section from AS, KM, MMaz) and all co-authors contributed to the interpretation of the results as well as  
579 manuscript review and editing.

580

581 **Data repository**

582 Maturilli, Marion (2020): Basic and other measurements of radiation and continuous meteorological  
583 observations at station Ny-Ålesund (April, May 2014 and April, May, June 2015), reference list of 10  
584 datasets. Alfred Wegener Institute - Research Unit Potsdam, PANGAEA,  
585 <https://doi.pangaea.de/10.1594/PANGAEA.913988> (DOI registration in progress)

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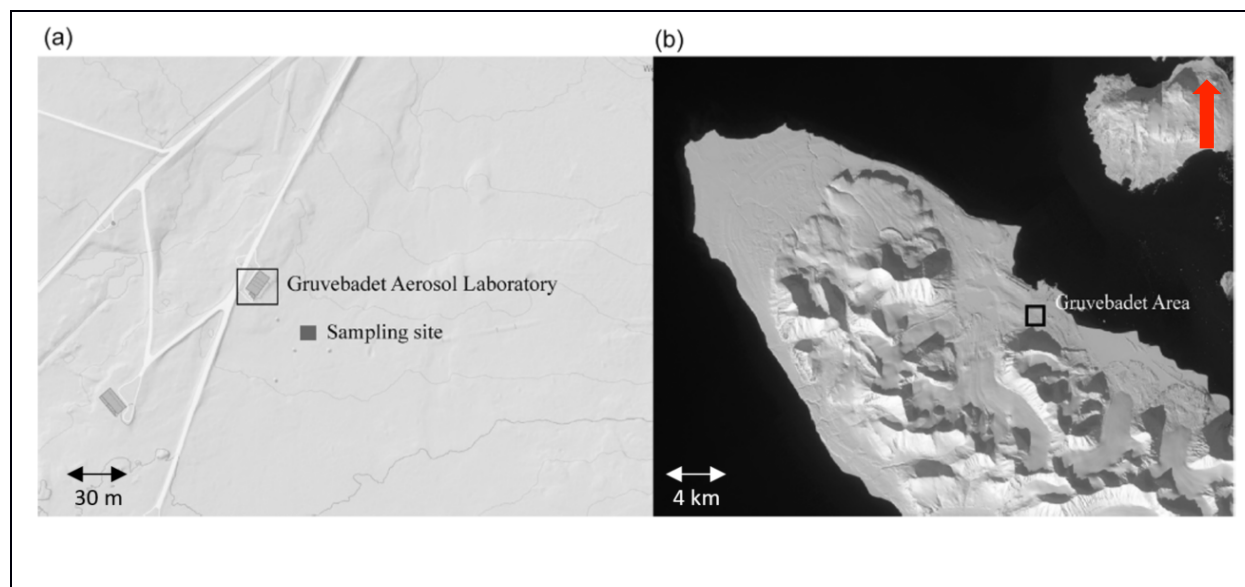
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596 **FIGURES**

597 **Figure 1.** a) Experimental sampling site location (dark grey rectangle), in proximity of the Gruvebadet  
598 Aerosol Laboratory. b) Gruvebadet area (black square), close to the Ny-Ålesund research village. From:  
599 Spolaor et al., 2019 (maps from <https://toposvalbard.npolar.no/>). The red arrow points to the North.

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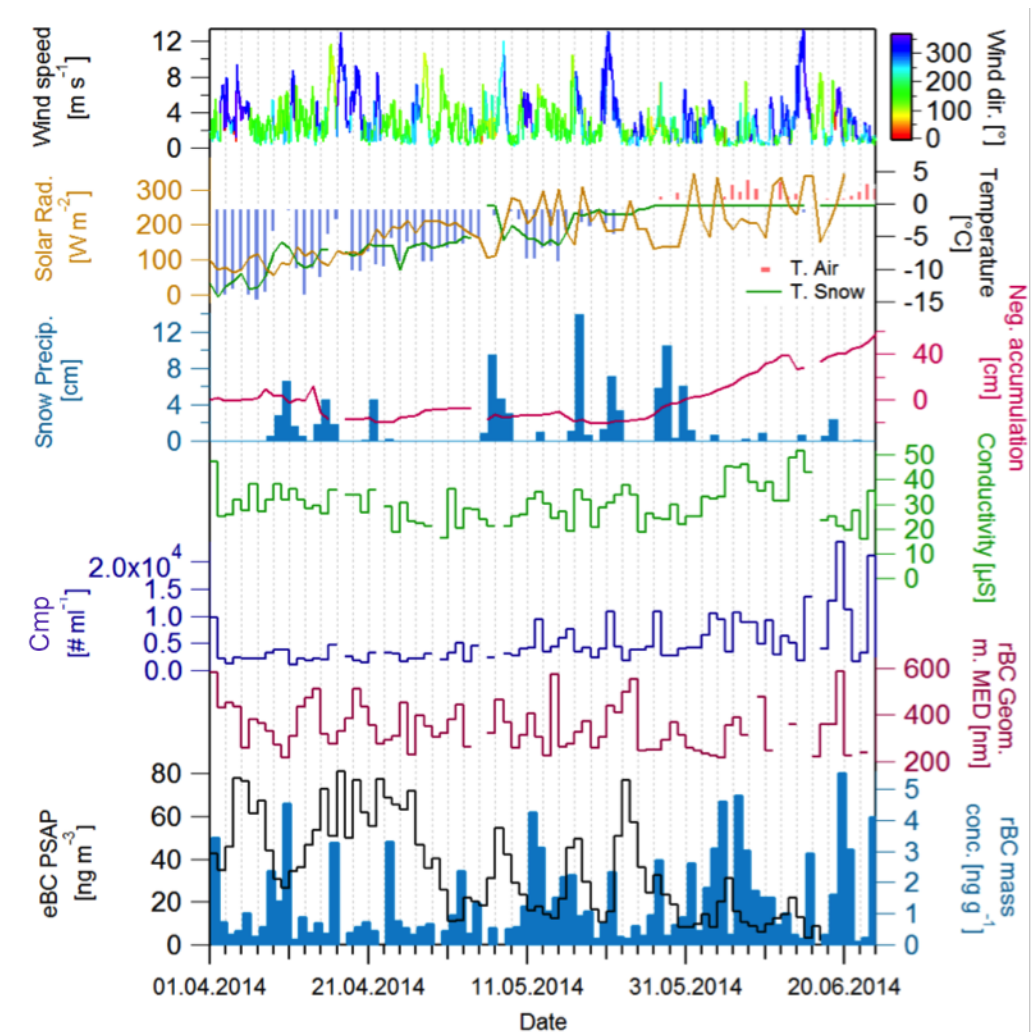
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612 **Figure 2.** The 85-days experiments daily snow samples rBC mass concentration (light blue), eBC mass  
 613 concentration in the atmosphere (black), geometric mean mass equivalent diameter (purple), number of  
 614 coarse mode particles (Cmp - blue), total conductivity (green), meteo/snow parameters used in the  
 615 statistical exercise: wind speed color coded for wind direction, solar radiation (orange line), air and  
 616 surface snow temperatures (blue bars and green line respectively), amount of fresh snow (“snow  
 617 precipitations”, light blue bars) and the snow accumulation (“Neg. accumulation”; the values where  
 618 multiplied by -1 in order to show the similar trend of the snow lost and of the air/snow temperature during  
 619 the melting period at the end of the campaign).

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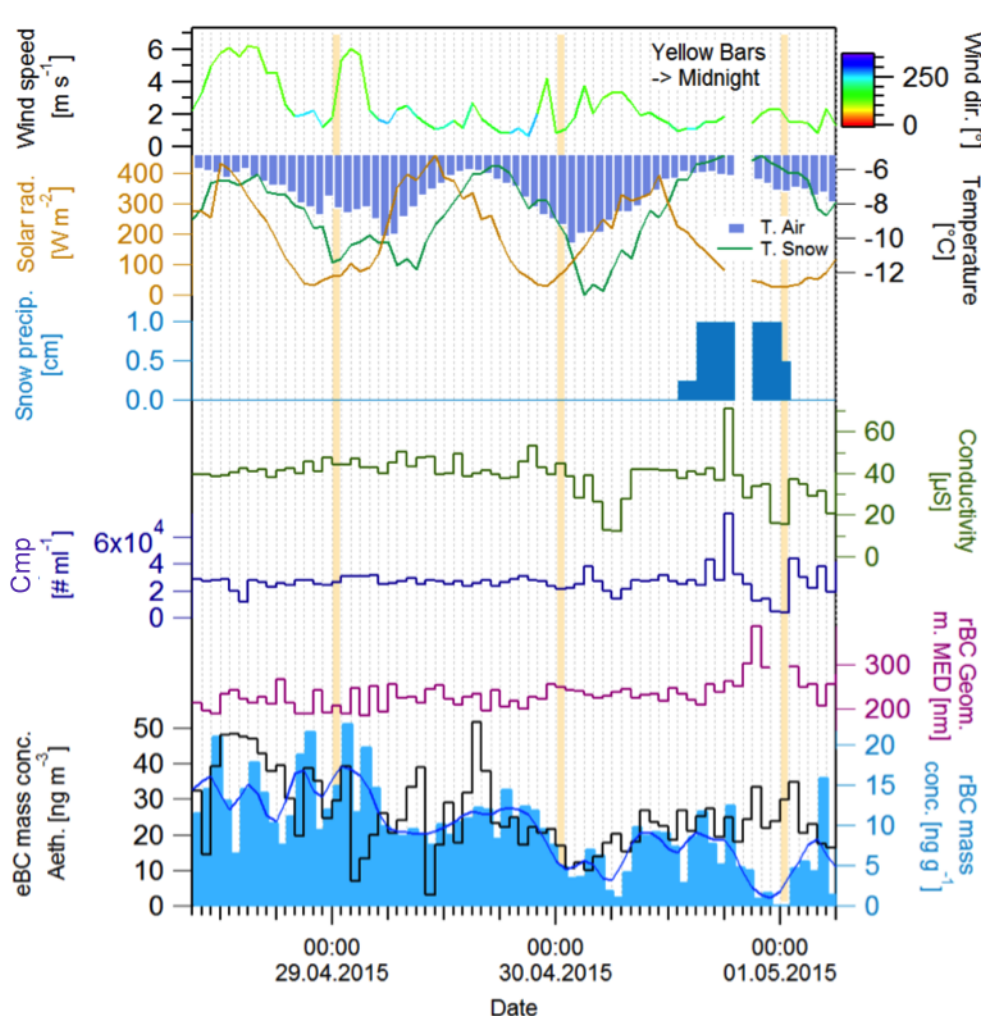


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623 **Figure 3.** The 3-days experiments snow samples hourly rBC mass concentration and smoothed line (light  
 624 blue bars), atmospheric eBC mass concentration in the atmosphere (black), geometric mean mass  
 625 equivalent diameter (purple), the number concentration of coarse mode particles (Cmp - blue) and the  
 626 total conductivity (green), meteo/snow parameters used in the statistical exercise: wind speed color coded  
 627 for wind direction, solar radiation (Orange line), Air and surface snow temperature (blue bars and green  
 628 line respectively), amount of fresh snow (“snow precipitations”, light blue bars). The yellow bars are  
 629 centered on the midnight hours for the three sampling days.

630

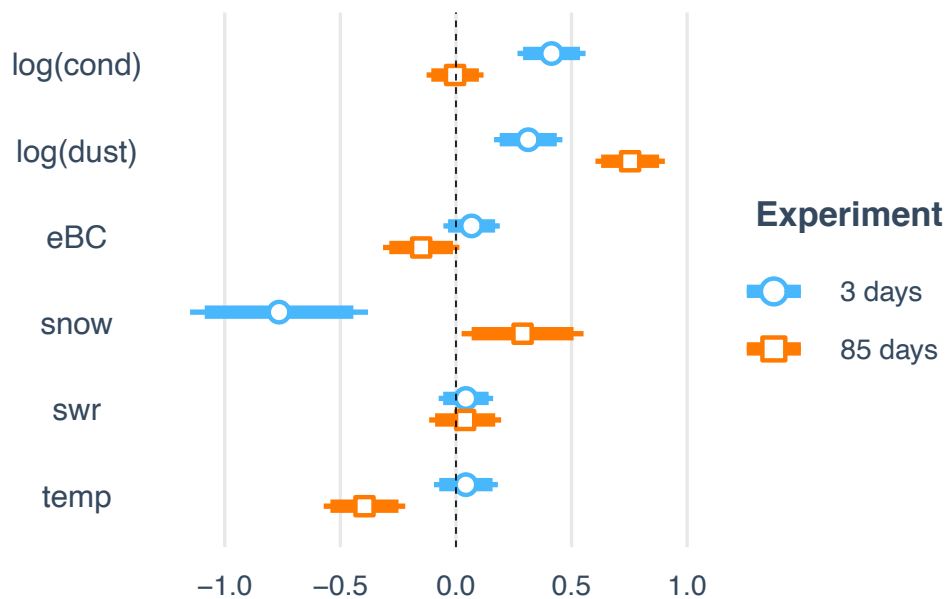


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633 **Figure 4.** Estimated coefficients of the standardized covariates of the multiple linear regression models  
 634 fitted to the 3 days and 85 days experiments. The segments correspond to 95% confidence intervals about  
 635 the corresponding estimated coefficients. The internal thicker segments correspond to 90% confidence  
 636 intervals. Intervals, that do not include the zero, correspond to statistically significant covariates. If a  
 637 confidence interval consists of positive values, then there is a significant positive association between the  
 638 corresponding covariate and the logarithm of the snow rBC mass concentration given the remaining  
 639 covariates. Vice versa, if the confidence interval consists of negative values, then the association is  
 640 negative. The abbreviations used in the plot are: “log(cond)” – logarithm of the water conductivity time  
 641 series, “log(dust)” – logarithm of the coarse mode particles number concentration time series, “eBC” –  
 642 equivalent black carbon atmospheric concentration, “snow” – presence of snow precipitation episodes,  
 643 “swr” – short wave radiation, “temp” – the snow temperature. The plot is produced with the R package (R  
 644 Core Team, 2020) jtools (Long, 2020).

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650 **TABLES**

651

652 **Table 1.** Estimated coefficients, 95% confidence intervals and the corresponding p-values for the  
 653 covariates of the multiple linear regression model fitted to the 85 days and the 3-days experiments. The  
 654 last rows of the table report the number of observations, the multiple coefficient of determination ( $R^2$ ) and  
 655 its adjusted version.

656

<i>Covariates</i>	<b>85-days</b>			<b>3-days</b>		
	<i>Estimates</i>	<i>CI</i>	<i>p</i>	<i>Estimates</i>	<i>CI</i>	<i>p</i>
(Intercept)	-6.74	-8.74 – -4.74	<b>&lt;0.001</b>	1.39	-2.30 – 5.09	0.453
Cond[log]	-0.02	-0.51 – 0.48	0.950	1.38	0.89 – 1.87	<b>&lt;0.001</b>
Dust[log]	1.29	1.03 – 1.55	<b>&lt;0.001</b>	0.74	0.39 – 1.09	<b>&lt;0.001</b>
eBC	-0.01	-0.01 – 0.00	0.074	0.00	-0.00 – 0.01	0.272
Snow[TRUE]	0.29	0.02 – 0.55	<b>0.033</b>	-0.77	-1.15 – -0.38	<b>&lt;0.001</b>
SWR	0.00	-0.00 – 0.00	0.613	0.00	-0.00 – 0.00	0.468
Temp	-0.10	-0.14 – -0.05	<b>&lt;0.001</b>	0.02	-0.04 – 0.08	0.535
Observation	72			68		
R2 /R2 adjusted	0.688/0.6590			0.779 /0.758		

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