

1 **Title:** Black carbon concentrations and modeled smoke deposition fluxes to the bare ice dark  
2 zone of the Greenland Ice Sheet

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12

13 **Abstract:**

14 Ice-albedo feedbacks in the ablation region of the Greenland Ice Sheet (GrIS) are difficult to  
15 constrain and model due in part to our limited understanding of the seasonal evolution of the  
16 bare-ice region. To help fill observational gaps, 13 surface samples were collected on the GrIS  
17 across the 2014 summer melt season from patches of snow that were visibly light, medium, and  
18 dark colored. These samples were analyzed for their refractory black carbon (rBC)  
19 concentrations and size distributions with a Single Particle Soot Photometer coupled to a  
20 characterized nebulizer. We present a size distribution of rBC in fresh snow on the GrIS, as well  
21 as from surface hoar in the bare ice dark zone of the GrIS. The size distributions from the  
22 surface hoar samples appear unimodal, and were overall smaller than the fresh snow sample,  
23 with a peak around 0.3  $\mu\text{m}$ . The fresh snow sample contained very large rBC particles that had a  
24 pronounced bimodality in peak size distributions, with peaks around 0.2  $\mu\text{m}$  and 2  $\mu\text{m}$ . rBC  
25 concentrations ranged from a minimum of 3  $\mu\text{g-rBC/L-H}_2\text{O}$  in light-colored patches at the  
26 beginning and end of the melt season, to a maximum of 32  $\mu\text{g-rBC/L-H}_2\text{O}$  in a dark patch in  
27 early August. On average, rBC concentrations were higher ( $20 \mu\text{g-rBC/L-H}_2\text{O} \pm 10 \mu\text{g-rBC/L-}$

28 H<sub>2</sub>O) in patches that were visibly dark compared to medium patches (7 μg-rBC/L-H<sub>2</sub>O ± 2 μg-  
29 rBC/L-H<sub>2</sub>O) and light patches (4 μg-rBC/L-H<sub>2</sub>O ± 1 μg-rBC/L-H<sub>2</sub>O), suggesting BC aggregation  
30 contributed to snow aging on the GrIS, and vice versa. Additionally, concentrations peaked in  
31 light and dark patches in early August, which is likely due to smoke transport from wildfires in  
32 Northern Canada and Alaska as supported by the Navy Aerosol Analysis and Prediction System  
33 (NAAPS) reanalysis model. According to model output, 26 mg/m<sup>3</sup> of biomass burning derived  
34 smoke was deposited between April 1<sup>st</sup> and August 30<sup>th</sup>, of which 85% came from wet  
35 deposition and 67% was deposited during our sample collection timeframe. The increase in rBC  
36 concentration and size distributions immediately after modelled smoke deposition fluxes suggest  
37 biomass burning smoke is a source of BC to the dark zone of the GRIS. Thus, the role of BC in  
38 the seasonal evolution of the ice-albedo feedback should continue to be investigated in the bare-  
39 ice zone of the GrIS.

40

## 41 **1. Introduction**

42 The bare ice dark zone of the southwest Greenland Ice Sheet (GrIS) is characterized by low  
43 albedo due in part to the presence of light absorbing impurities (LAIs), that create a positive ice-  
44 albedo feedback through increased surface melting, ice grain growth, and darkening (Tedesco et  
45 al., 2016). LAIs in this region are a mixture of cryoconite, ice algae (Stibal et al., 2017; Ryan et  
46 al., 2018), dust (Wientjes et al., 2011), and black carbon (BC) such as from Northern  
47 Hemisphere fires (Khan et al., 2017a), yet the relative contribution of each light absorbing  
48 particle is still uncertain. The radiative forcing of these LAIs, along with warming summer  
49 surface temperatures (Hanna et al., 2008), leads to large volumes of supra-glacial melt (Greuell,  
50 2000). Furthermore, retreat of the snowline is amplifying surface melt of the GrIS due to

51 increased bare ice exposure (Ryan et al., 2019) and the LAI-ice albedo feedbacks described  
52 above.

53 BC in and on snow and ice is known to warm the Arctic and contribute to snow and ice  
54 melting, however the magnitude of its influence is still highly uncertain e.g., (Flanner et al.,  
55 2007; Bond et al., 2013). BC concentration in air is typically operationally defined depending on  
56 the analytical technique used (Petzold et al., 2013). Many in-situ measurements of BC  
57 concentration in snow in the Arctic have been reported by the Integrating Plate and Integrating  
58 Sandwich (IS) technique, which provides analysis of light absorption of particulate impurities  
59 through spectrophotometric analysis of filter loaded with particulates collected from melted  
60 samples (e.g., Clarke and Noone, 1985; Doherty et al., 2010; Doherty et al., 2013). Doherty et al.  
61 (2010) reported a median concentration of 3 ng/g in surface snow, with higher concentration  
62 layers up to ~20ng/g in snow profiles at Dye 2. Snow samples from snowpits in the northwest  
63 sector of the GrIS were also collected in 2013 and 2014 from two traverses and analyzed for  
64 elemental/organic carbon (EC/OC). The mean concentration of the samples collected was 2.6  
65 ng/g and the mean peak was 15 ng/g. Based on these results, it was determined that EC/OC do  
66 not influence the snow albedo in the NW sector of the GrIS dry zone (Polashenski et al., 2015a).  
67 Observations of refractory black carbon (rBC) analyzed by the Single Particle Soot Photometer  
68 (SP2) have been published from snow profiles and ice cores in the accumulation region closer to  
69 the Summit research station (McConnell et al., 2007a; Keegan et al., 2014b; Lim et al., 2014).  
70 McConnell et al. (2007) presented BC concentrations from a 215-year ice-core record collected  
71 at D4 in West Central Greenland with average concentrations of 1.7ng/g in pre-industrial times,  
72 2.3ng/g over the period 1950-2002, and around 5 ng/g in the peak period of the early 1900s. The  
73 maximum monthly concentration observed was 58.8 ng/g in 1854, however, monthly

74 concentrations only exceeded 5 ng/g ~2-3 times each decade after 1950. Polashenski et al.,  
75 (2015) provides a comprehensive review of previous BC concentrations in their supplemental  
76 info, showing that the BC average ranges between 1.5 and 3 ng/g over an annual cycle, with peak  
77 deposition occurring during summer episodic events, with concentrations of 5 - 10+ ng/g only  
78 occurring a few times at a given site per decade. Similarly, rBC concentrations from the  
79 percolation zone of the GrIS have been shown to be relatively low, less than 1.5 ng/g (Lewis et  
80 al., 2021).

81 rBC measured by SP2 has been shown to provide more reliable measurements of  
82 concentration than the IS or EC/OC (from liquid and air samples, respectively) techniques  
83 because it is largely free from the interference of materials other than rBC (Kondo et al., 2011;  
84 Schwarz et al., 2012) such as pyrolyzed organic carbon artifacts (Lim et al., 2014). It also  
85 provides a lower detection limit and increased sensitivity at low concentrations (Lim et al. 2014).  
86 The SP2 coupled with a nebulizer also provides a measurement of rBC particle size distribution  
87 from liquid samples.

88 rBC particle size has been observed in some snow samples to be larger than expected  
89 from atmospheric measurements, reflecting to some degree size-dependent removal processes  
90 from the atmosphere (Schwarz et al., 2013). The rBC size distribution in snow, which at this  
91 point is constrained by direct observations not supported by detailed modeling, is a significant  
92 source of uncertainty for calculating the overall radiative forcing of BC-in-snow on the Arctic  
93 climate, as well as the global climate (e.g., Bond et al., 2013). Very few rBC size distributions in  
94 snow have been reported globally, with most measurements coming from the Arctic (Lim et al.,  
95 2014; Khan et al., 2017b; Mori et al., 2019).

96 Although, observations of BC in snow have been previously observed in the percolation zone  
97 (Dye 2) and accumulation zone (Summit Station) by the IS technique (Doherty et al., 2010, 2013)  
98 and rBC-SP2 at Summit Station (McConnell et al., 2007b; Keegan et al., 2014a; Lim et al., 2014),  
99 to the authors' knowledge, no reports of rBC concentrations with size distributions in snow and  
100 surface hoar have been reported from the GrIS, providing new insight, particularly into the  
101 dynamic bare-ice region.

102 Here we present rBC concentrations with size distributions from the bare ice region of the  
103 GrIS before and after influence by a major wildfire event, along with NAAPS modelled wet and  
104 dry deposition. Our findings suggests that rBC surface hoar concentrations in the bare ice zone  
105 reflect atmospheric conditions momentarily, before being reset, possibly by supra-glacial melt.  
106 Additionally, NAAPS model output suggest most of the biomass burning derived smoke  
107 deposition comes in the form of wet removal (i.e., removal by precipitation). These rBC  
108 concentrations and size distributions provide insight into the seasonal evolution of impurities,  
109 which are needed to constrain ice-albedo feedbacks in the bare-ice zone of the GrIS.

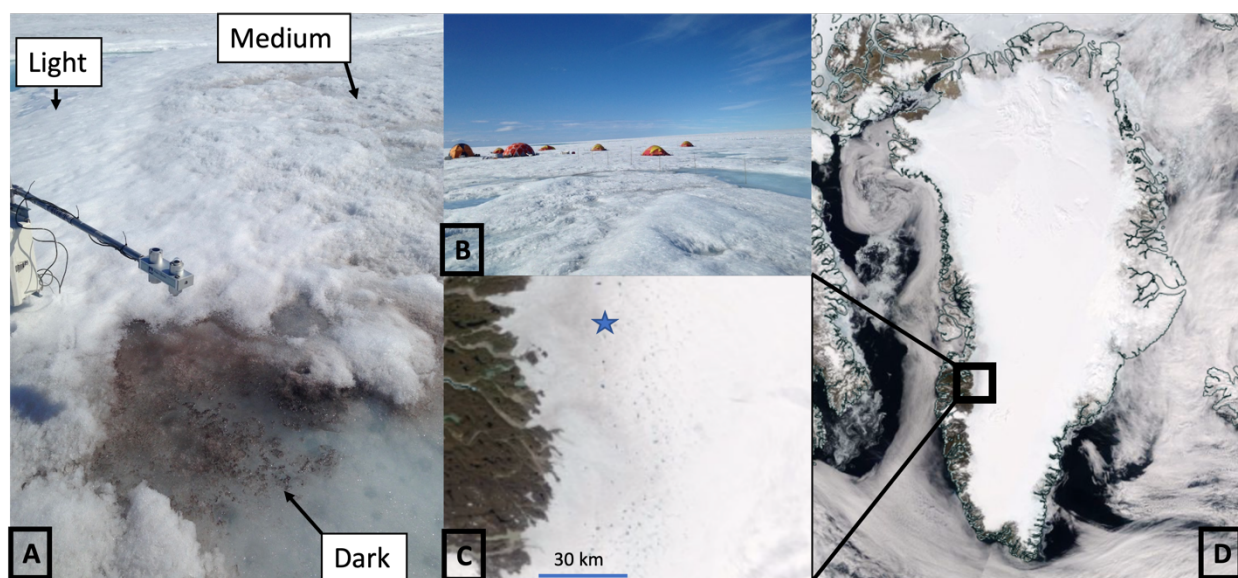
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## 111 **2. Methods**

### 112 *2.1 Site Description and Snow Sampling*

113 The field site was in the southwestern region of the GrIS near the S6 automated weather station  
114 at 67 04.779°N, 49 24.077°W, and 1011 m above sea level. More information on the study site  
115 can be found in Stibal et al. (2017). A fresh snow surface sample (2 – 3 cm), was collected just  
116 after a snow event on 2014-06-27. Three surface hoar samples (2 – 3 cm), were collected in 150  
117 mL pre-cleaned and combusted amber glass bottles four times between 2014-06-28 and 2014-08-  
118 11 across the 2014 summer melt season from visually identified light, medium, and dark patches

119 of surface hoar, for a total of 13 samples, including the fresh snow. While all sample sites could  
 120 include a mixture of ice algae, dust, black carbon (i.e, cryoconite), the dark patches especially  
 121 could represent refrozen melt that is enhanced in LAIs, including rBC. A mixture of light,  
 122 medium and dark 1 – 3 m<sup>2</sup> patches were sampled within the ~.5 km<sup>2</sup> study area to characterize  
 123 the breadth of surface types and heterogenous distribution of impurities. Samples were stored  
 124 frozen in a ‘field cooler’ dug into the ice and then transported frozen on ice to Kangerlussuaq,  
 125 and shipped on dry ice to the Denver Airport, and then transported immediately to a freezer at  
 126 the Institute of Arctic and Alpine Research (INSTAAR) at the University of Colorado – Boulder.



A and B are images collected by Dr. Alia Khan. C and D are MODIS satellite images acquired from the NASA Worldview application.

127  
 128 **Figure 1:** A) Example light, medium and dark patches of ice. B) The Dark Snow Field Camp. C)  
 129 The southwest GrIS dark zone with the field sampling location indicated by a blue star and D)  
 130 the GrIS from MODIS on July 2<sup>nd</sup>, 2014.

131  
 132 *2.2 Processing for Refractory Black Carbon*

133 The samples were transported frozen from INSTAAR to the Earth System Research Laboratory  
 134 at the National Oceanic and Atmospheric Administration where they were analyzed for rBC

135 mass mixing ratios (MMRs) by SP2 coupled to a nebulizer per the methods described in Katich  
136 et al. (2017) and Khan et al. (2018). Briefly, the samples were melted for the first time just prior  
137 to analysis with the SP2 and aerosolized with a carefully calibrated concentric pneumatic  
138 nebulizer based on a customized U5000 AT+ nebulizer (Teledyne Cetac, Inc.) which the  
139 ultrasonic piezo was replaced with a concentric pneumatic nebulizer. The SP2 was calibrated  
140 with fullerene soot (Lot# F12S011, Alfa Aesar Inc., Wood Hill, MA) with the community  
141 calibration approach (Baumgardner et al., 2012) over masses of 1 – 20 fg. Using a power law  
142 calibration dependence following Schwarz et al., [2012], the resulting linear calibration of SP2  
143 signal to rBC mass applied to mass of 80 fg was extended further to 4000 fg. The SP2 was  
144 operated with a widely staggered gain for two incandescent channels, allowing sizing of rBC  
145 mass in the range  $\sim 1 - 4000$  fg.

146 Melted snow samples were interspersed with deionized water blanks to confirm a low  
147 background, especially relative to the MMRs, indicating no appreciable contamination to  
148 concentrations and size distributions. Little size-dependence in nebulization efficiency was  
149 confirmed with concentration standards of polystyrene latex spheres (PSLs) over 220 – 1500 nm  
150 diameter, which is consistent with recent results from concentric pneumatic nebulizers (Wendl et  
151 al., 2014, Katich et al., 2017). Therefore, size dependent corrections were not necessary. During  
152 data acquisition with the SP2, its lower mass-detection limit was 1.2 fg, which corresponds to  
153 about a 110 nm volume equivalent diameter (VED) size detection limit, assuming 1.8g/cc void  
154 free density. A 510 nm diameter PSL concentration standard was sampled between melted snow  
155 analyses to track possible changes in nebulization efficiency during each day of sampling. This  
156 revealed effectively constant efficiency varying with a standard deviation less than 5%. A  
157 gravimetric mass concentration standard (Schwarz et al., 2012) was also used to evaluate

158 nebulization efficiency. The results of the PSL and gravimetric calibrations of nebulizer  
159 efficiency were consistent within uncertainties of 20% and were averaged to provide a best-  
160 estimate nebulization efficiency that was then used to produce the BC MMR values as in  
161 Schwarz et al. (2012).

162

### 163 *2.3 Global Aerosol Modeling*

164 The Navy Aerosol Analysis Prediction System (NAAPS) model is a global aerosol transport  
165 model which provides 6-hrly biomass burning smoke, anthropogenic and biogenic fine aerosols,  
166 dust, and sea salt aerosol forecasts and analyses below 100 hPa at 1/3° latitude/longitude spatial  
167 resolution and contains 42 vertical atmospheric levels. The NAAPS reanalysis (NAAPS-RA) is  
168 available 2003-current with a coarser spatial resolution (1° latitude/longitude horizontal and 25  
169 vertical levels) (Lynch et al., 2016). Total column aerosol optical thickness (AOT) is constrained  
170 through assimilation of quality-controlled satellite AOT retrievals from the Moderate Resolution  
171 Imaging Spectroradiometer (MODIS) and Multi-angle Imaging SpectroRadiometer (MISR).  
172 Near-real time satellite based thermal anomaly data enables detection of wildfires and  
173 construction of biomass burning smoke emissions (Reid et. al., 2009). Orbital corrections for  
174 MODIS-based fire detections and regional factors were applied on emissions so that the  
175 reanalysis AOT verifies well with ground-based measurements (Lynch et al., 2016). The  
176 NAAPS-RA has been applied to a broad range of science applications, and specifically the life  
177 cycle, climatology, radiative forcing, aerosol-atmosphere-ice-ocean interactions of biomass  
178 burning smoke aerosols (e.g., Reid et al., 2012; Xian et al., 2013; Markowicz et al., 2021; Ross et  
179 al., 2018; Khan et al., 2019; Carson-Marquis et al., 2021), as well as previously to corroborate  
180 wildfire smoke transport to the GrIS (Khan et al., 2017a), Arctic Canada (Ranjbar et al., 2019),



181 Svalbard (Markowicz et al., 2016; 2017), the pan-Arctic region (Xian et al., 2022a, b), the  
182 Nepalese Himalayas (Khan et al., 2020), and the Antarctic (Khan et al., 2018; Khan et al., 2019).  
183 Speciated AOT, surface aerosol concentration and deposition flux are used in this study. Here the  
184 deposition is calculated as 24-hour flux to the surface of the ice sheet in  $\text{mg}/\text{m}^2/\text{day}$ . Estimating  
185 atmospheric properties related to biomass burning is highly complex and is influenced by wide  
186 variety of factors such as the type of fuel, combustion temperature, and atmospheric  
187 conditions. Also, the chemical, optical and physical properties of biomass burning aerosols can  
188 change during atmospheric transport and dispersion. The mass ratio of rBC to total mass in  
189 biomass burning smoke particles is estimated to be 5–10% black carbon in the NAAPS-RA  
190 model based on field studies (see a summary in Reid et al., 2005) and here we chose 7% as a  
191 median value.

### 192 **3. Results and Discussion**

#### 193 *3.1 rBC Concentrations*

194 rBC concentrations in the surface hoar ranged from a minimum of  $3 \mu\text{g-rBC}/\text{L-H}_2\text{O}$  in light  
195 patches at the beginning and end of the melt season, to a peak of  $32 \mu\text{g-rBC}/\text{L-H}_2\text{O}$  in a dark  
196 patch in early August (Table 1). rBC concentrations were higher in patches that were visibly  
197 darker ( $20 \mu\text{g-rBC}/\text{L-H}_2\text{O}$ ) compared to medium patches ( $7 \mu\text{g-rBC}/\text{L-H}_2\text{O}$ ) and light patches ( $4$   
198  $\mu\text{g-rBC}/\text{L-H}_2\text{O}$ ), suggesting BC aggregates with dust and biological material on the GrIS. Light  
199 and dark patch concentrations peaked in early August. Our minimum concentrations are in the  
200 range of rBC concentrations found elsewhere on the GrIS, but our peaks are higher than  
201 previously reported concentrations from snow on the GrIS (Doherty et al., 2010; Polashenski et  
202 al., 2015; Lewis et al., 2021). Our maximum concentrations are higher than the highest  
203 concentrations observed in vertical snow with the IS (Doherty et al., 2010) and EC/OC technique

204 (Polashenski et al., 2015), but less than the highest monthly average concentration of year of  
 205 1854 reported in an ice core by McConnell et al. (2007). The concentration of rBC in the fresh  
 206 snow (3  $\mu\text{g-rBC/L-H}_2\text{O}$ ) sample was roughly the same as the light surface hoar patches on 2014-  
 207 06-28 and 2014-08-11.

208

209 **Table 1:** NAAPs Smoke Dry, Wet, and Total Deposition ( $\text{mg/m}^2/\text{day}$ ) from April 1<sup>st</sup> prior to  
 210 sample collection. Average rBC concentrations from visually light, medium, and dark patches of  
 211 surface hoar. All samples were collected at 67.07979701 degrees N and -49.40116603 degrees W  
 212 at 1005 meters above sea level in the dark zone ablation region of the SW Greenland Ice Sheet.

213 ^The fresh snow sample is a single sample.

Date	NAAPS	NAAPS	NAAPS	Average	Snow type (visual color)	rBC $\mu\text{g-rBC/L-H}_2\text{O}$
	Smoke Dry Deposition ( $\text{mg/m}^3/\text{day}$ )	Smoke Wet Deposition ( $\text{mg/m}^3/\text{day}$ )	Smoke Total Deposition ( $\text{mg/m}^2/\text{day}$ )	rBC $\mu\text{g-rBC/L-H}_2\text{O}$		
6/27/14	0.58	1.98	2.56	3.05 <sup>^</sup>	Fresh	3.05
6/28/14	0.60	6.92	7.51	8.37	Light	2.87
					Medium	9.61
					Dark	12.62
7/21/14	0.75	6.93	7.69	11.45	Light	4.21
					Medium	6.42
					Dark	23.71
8/2/14	1.51	9.44	10.95	14.15	Light	5.27
					Medium	4.71
					Dark	32.47
8/11/14	1.94	12.14	14.08	8.12	Light	2.96

Medium	8.75
Dark	12.64

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214

### 215 *3.2 rBC Size Distributions*

216 We found very large rBC are present (Figure 2A, B and C), especially in the fresh snow sample.

217 The large size distribution in fresh snow follows previous findings in the rocky mountains that

218 rBC size distributions can be larger in surface snow than expected in aerosol in the atmosphere

219 (Schwarz et al., 2013). Furthermore, the fresh event is associated with a more pronounced

220 bimodality at  $\sim 0.2 \mu\text{m}$  and  $2 \mu\text{m}$  (Figure 2A), whereas the rBC in surface hoar samples appears

221 more unimodal (Figure 2B and 2C). The average surface hoar rBC sizes, which have not been

222 previously reported in the literature, are smaller than the one fresh snow sample with a peak

223 around  $0.3 \mu\text{m}$ . This is still larger than typical modal sizes for rBC observed in the atmosphere

224 (in the range  $\sim 0.11 - 0.2 \mu\text{m}$  typically). Furthermore, no apparent patterns emerge in the size

225 distributions across the light, medium and dark patches over the duration of the season.

226 However, the surface hoar rBC size distributions likely evolve, just as the seasonal snow cover

227 evolves into bare ice and surface hoar, but we are unable to assess from this relatively small data

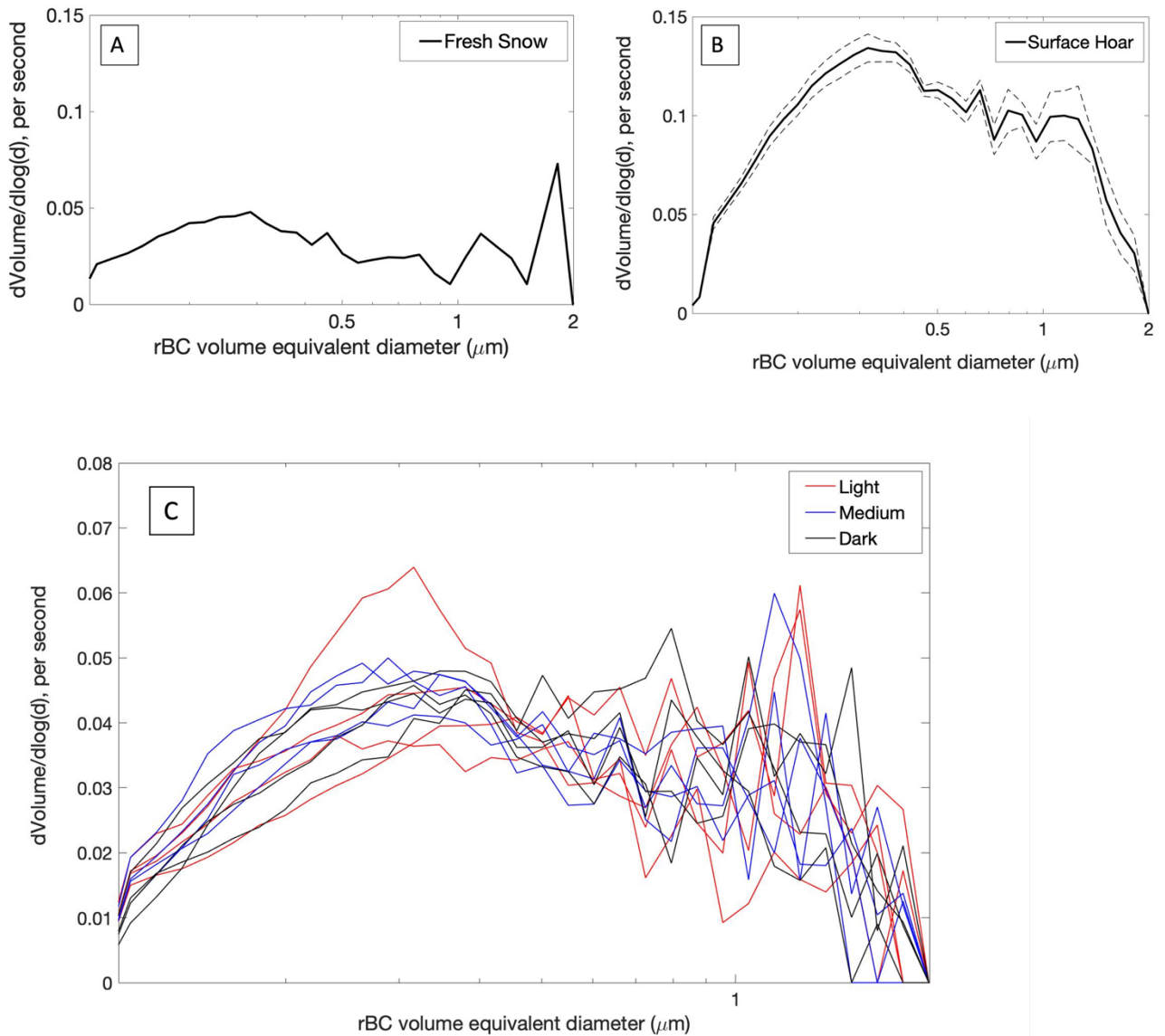
228 set. This conjecture is supported by observations that repeated freeze/thaw cycles tend to cause

229 rBC coagulation in liquid (Schwarz et al., 2013). Regardless, these initial results of rBC size

230 distributions from fresh snow and surface hoar in the bare ice region of the GrIS are important

231 for informing ice-albedo models, which are still being developed and refined for bare ice regions

232 of the ice sheet (e.g. Flanner et al., 2007).



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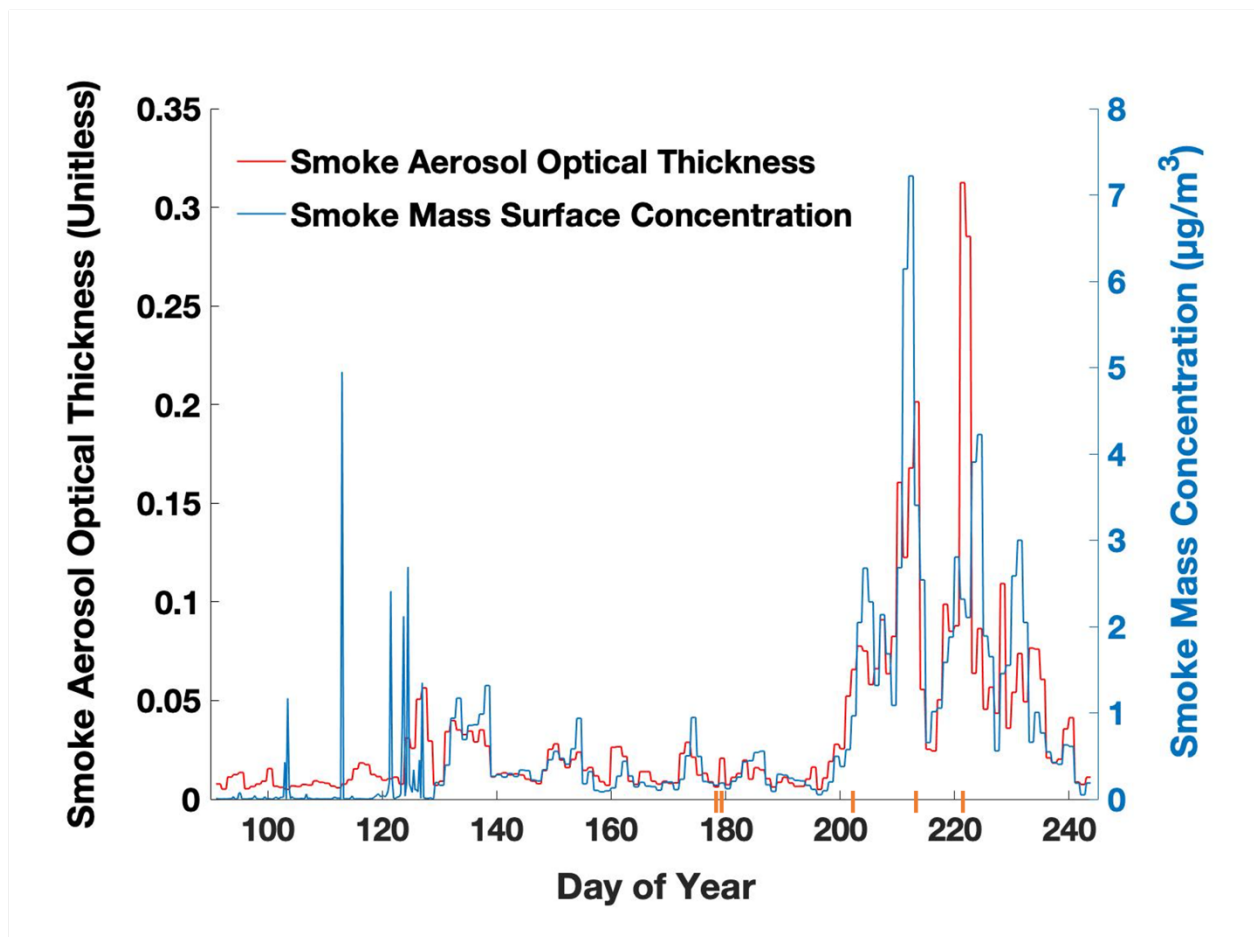
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235 **Figure 2:** A) rBC size distribution of fresh snow ( $n=1$ ), B) all surface hoar samples over the  
 236 duration of the season ( $n=12$ ) and C) the size distribution of each surface hoar sample  
 237 categorized as light, medium and dark. The dashed lines in Figure 2B represent the max and min  
 238 size distributions and the solid black line is the average.

239

240 *3.3 NAAPS Aerosol Model Comparison and Evaluation*

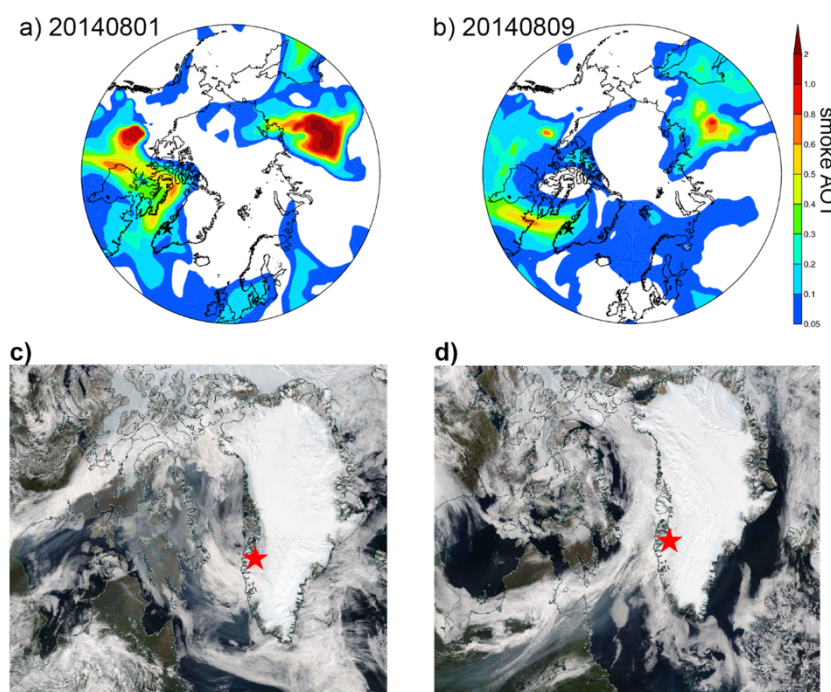
241 The ground observations were then compared to cumulative aggregates of smoke deposition  
242 fluxes modelled with the Navy Aerosol Analysis Prediction System reanalysis model. AOT  
243 derived from MODIS and modeled by NAAPS demonstrates that a large wildfire smoke event  
244 was observed just before the third sample was collected and during the time the fourth sample  
245 was collected (Figure 3). Concomitant AOT and surface concentration predictions from the  
246 NAAPS model confirms our peak concentrations are likely due to Northern Hemisphere wildfire  
247 smoke (Figure 4 A- D).  
248



249

250 **Figure 3:** Aerosol optical thickness (AOT) derived from NAAPS reanalysis

251 over the sampling season from smoke and dust. B) Smoke mass concentration ( $\mu\text{g}/\text{m}^3$ ) in the  
252 surface layer of the model (centered around 16m). The five sampling dates are marked in orange.



253  
254 **Figure 4:** Biomass burning smoke transport reaching the GrIS from the west based on NAAPS-  
255 RA daily-mean smoke AOT and MODIS TERRA true color imageries for **A and C)** Aug. 1,  
256 2014 and **B and D)** Aug. 9, 2014. The sampling location is marked with a black star in the  
257 NAAPS-RA plots (A and B), and red stars in the MODIS imageries (C and D).

258 According to NAAPS model output, the deposition flux of smoke (Table 1 and Fig. 5)  
259 onto the ice surface of the dark zone during our model study period, April 1<sup>st</sup> – August 30<sup>th</sup>, was  
260 25.6  $\text{mg}/\text{m}^2/\text{day}$  and 85% came from wet deposition. April 1<sup>st</sup> to August 30<sup>th</sup> was chosen based  
261 on the primary Northern Hemisphere wildfire season and smoke transport to the Arctic (Xian et  
262 al., 2022b). 68% of this smoke ( $17.3 \text{ mg}/\text{m}^2/\text{day}$ ) was deposited during our sample collection  
263 period from June 27<sup>th</sup> to August 11<sup>th</sup>. Prior to the first sample collected on June 27<sup>th</sup>, 10% of the  
264 total smoke flux ( $2.6 \text{ mg}/\text{m}^2/\text{day}$ ) was deposited from April 1<sup>st</sup> to June 26<sup>th</sup>. After the last sample  
265 was collected on August 11<sup>th</sup>,  $5.8 \text{ mg}/\text{m}^2/\text{day}$  of smoke was deposited between August 12<sup>th</sup> and  
266 30<sup>th</sup>.

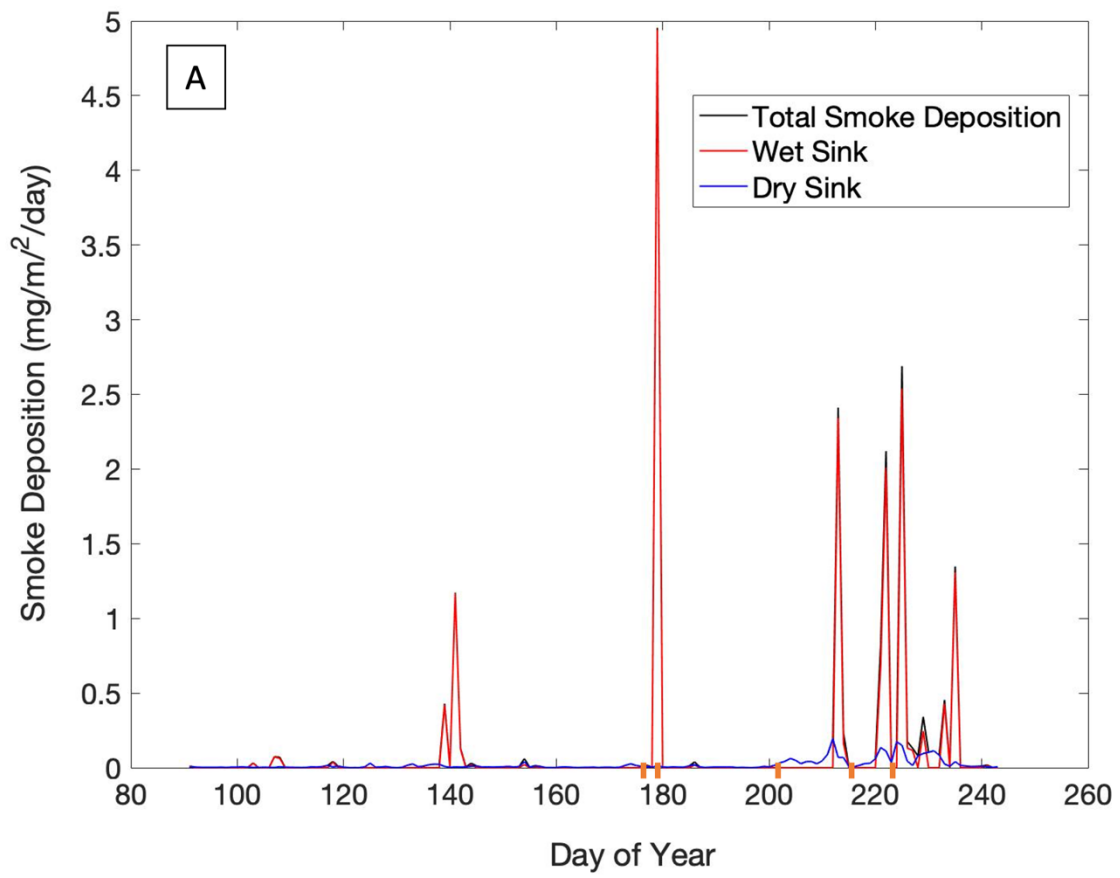
267 We evaluate the NAAPS-RA deposition flux based on the rBC concentration observed in  
268 fresh snow, which was 3  $\mu\text{g-rBC/L-H}_2\text{O}$ . The NAAPS model assumes 7% of smoke is BC. The  
269 snow event that preceded the fresh snow sample collection, had a modeled precipitation rate of  
270 10 mm/day or 10 L/m<sup>2</sup>. The modeled smoke deposition flux is 3000  $\mu\text{g/m}^2/\text{day}$  or 300  $\mu\text{g/L}$  over  
271 24 hours. At 7% BC of total smoke, that leaves us with 21  $\mu\text{g-BC/L-H}_2\text{O}$ . Therefore, the model  
272 appears to be off by roughly a factor of 7 for this one snow sample. Continued work is in  
273 progress to evaluate the model across a larger sample size of rBC ground observations across the  
274 Arctic.

275 Two case studies of interest arise in the modelled total NAAPS smoke flux when  
276 comparing wet and dry deposition. The first one is a large wet deposition flux and the second is a  
277 considerable dry deposition flux. The first wet deposition flux occurred between June 27<sup>th</sup> and  
278 28<sup>th</sup> (day of year 178 and 179), during a snow event (Fig. 5A and B). Here we see the largest  
279 increase in the total deposition flux of smoke over the study period at 5.0  $\text{mg/m}^3/\text{day}$  in just over  
280 24 hours. 99.8% of this comes from wet deposition. When we compare these model findings to  
281 the observational rBC data in the surface hoar and snow, we see the rBC concentration in fresh  
282 snow, 3  $\mu\text{g-rBC/L-H}_2\text{O}$ , is high compared to pristine fresh snow previously found in Svalbard, 1  
283  $\mu\text{g-rBC/L-H}_2\text{O}$  (Khan et al., 2017b). The average rBC concentration across the light, medium  
284 and dark patches is also relatively high for a non-human impacted site in the polar regions  
285 (Cordero et al., 2022). A previous study of black carbon in supra-glacial melt from the same  
286 GRIS site previously confirmed the dissolved BC molecular signature was indicative of wildfire  
287 smoke that likely came from Northern Canada and Alaska (Khan et al., 2017a). Between July  
288 22<sup>nd</sup> and August 2<sup>nd</sup>, the model again shows a large proportion of the total deposition flux  
289 coming from wet deposition, 77% of the 3.2  $\text{mg/m}^2/\text{day}$ . Similarly, from August 3<sup>rd</sup> to 11<sup>th</sup>, 86%

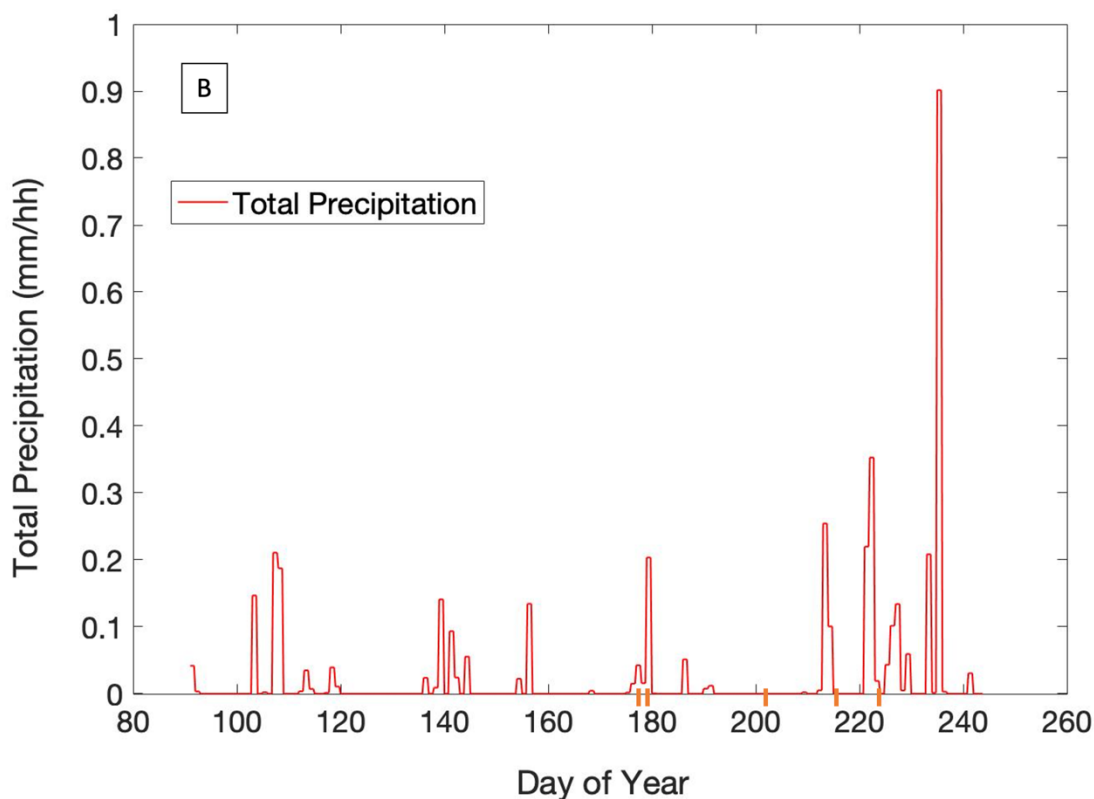
290 of the 3.1 mg/m<sup>3</sup>/day smoke deposition flux was from wet deposition (Fig. 5A). Again, this  
291 follows an increase in the total precipitation (Fig. 5B).

292         However, a dry deposition case arises on July 21<sup>st</sup>, 2014 (DOY 172). Here the NAAPS  
293 model does not produce a large total smoke deposition flux, but the rBC concentrations are still  
294 relatively high. Since the previous sampling event on June 28<sup>th</sup> (DOY 179), the model produces  
295 0.2 mg/m<sup>3</sup>/day total deposition flux, where only 16% comes from wet deposition. The majority,  
296 84%, is from dry smoke deposition. This finding is also supported by the fact that there was little  
297 precipitation during this time based on the NAAPS modeled meteorology (Fig. 5B), but it is also  
298 important to note that snow aging could also play a role in aggregation of BC particles. The  
299 decrease observed in the surface hoar rBC concentrations in the August 11<sup>th</sup> samples may  
300 suggest there was a process that removed the particles from the surface hoar, such as flushing or  
301 redistribution by supra-glacial melt, or uncontaminated fresh snow deposition which could dilute  
302 the concentrations. Further investigation into this process is warranted.





303



304

305 **Figure 5:** A) Biomass burning derived smoke deposition flux separated as wet and dry  
 306 deposition and B) total precipitation produced by the NAAPS model. The total smoke deposition  
 307 closely follows the wet deposition line. The five sampling dates are marked in orange.

308

#### 309 4 Conclusion

310 Here we present (to the author's knowledge) the first rBC size distributions from fresh snow  
 311 and surface hoar in the bare ice region of the GrIS, coupled with their concentrations. An initial  
 312 rBC size distribution in a fresh snow sample from the GrIS shows pronounced bimodality and  
 313 very large particles with the second peak almost 2  $\mu\text{m}$ . These initial rBC size distributions from  
 314 surface hoar in the bare ice dark zone of the Greenland Ice Sheet are smaller than the fresh snow,  
 315 but still much larger than observations of atmospheric rBC. There appears to be a shift in the

316 modal peak of rBC particle size in light patches over the duration of the season from  $\sim 0.3 \mu\text{m}$  to  
317  $\sim 1.4 \mu\text{m}$ , further suggesting aggregation of particles in the bare-ice region. NAAPS-AOT and  
318 surface concentration data suggest that rBC surface hoar concentrations in the bare ice zone  
319 reflect atmospheric conditions momentarily, before possibly being reset by supra-glacial melt.

320 Additionally, we demonstrate preliminary verification of BC deposition from the NAAPS-  
321 RA with *in-situ* observations. rBC measurements in dark patches from late June to early August  
322 2014 reveal an increase just after the smoke event. These elevated concentrations are closer to  
323 previously reported values in vertical snow and ice-core layers (e.g., Doherty et al., 2010 and  
324 Polashenski et al., 2015). The overall higher concentrations of rBC in visibly darker patches,  
325 where higher concentrations of ice algae were observed (Stibal et al., 2017), suggest potential bio  
326 flocculation with ice algae and mineral dust. However, NAAPS model results also indicate the  
327 increase is likely related to accumulation of episodically deposited wildfire-derived smoke. For  
328 example, the smoke event in early August, which brought smoke from the western Northern  
329 Hemisphere. Based on NAAPS deposition model and corroborated by rBC observations, wet  
330 deposition appears to be the largest source of rBC to the surface. For example, our fresh snow  
331 sample was measured at  $3 \mu\text{g-rBC/L-H}_2\text{O}$ , while the model, off by a factor of 7, produced  $21 \mu\text{g-}$   
332  $\text{rBC/L-H}_2\text{O}$ . These preliminary results suggest global aerosol models may be overestimating BC  
333 deposition; however, further investigation is warranted. These data provide utility in  
334 understanding the seasonal evolution of impurities, which are needed to constrain modeling of  
335 ice-albedo feedbacks in the bare-ice zone of the GRIS.

336

337 **Author Contributions**

338 ALK and JS analyzed the rBC samples. PX ran the NAAPS model and provided output data.  
339 ALK wrote the manuscript and PX and JS edited and contributed text. The samples were  
340 collected by ALK and the Dark Snow Project.

#### 341 **Acknowledgements**

342 The authors thank the Dark Snow Project for field support and additional sample collection,  
343 specifically, M. Stibal, J. Box and K. Cameron and N. Molotch.

344  
345 **Competing Interests.** There are no conflicts of interest.

#### 346 347 **Data Availability**

348 The rBC and NAAPS modeled deposition data are included in Table 1.

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