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1 Increasing dust emission from ice free terrain in southeastern Greenland since
2 2000

3

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27

28 **Highlight**

29 • Annual and seasonal dust fluxes during 1960–2014 are reconstructed from
30 an ice core of the southeastern dome in Greenland (SE-Dome).

31 • The annual dust fluxes are high after 2000 primarily due to increasing dust
32 flux during autumn.

33 • Local dust emissions increased after 2000 due to a warming-induced increase
34 in snow-free land on Greenland's east coast.

35

36 **Abstract**

37 Mineral dust plays a key role in both local and global climates. At high latitudes,

38 atmospheric dust can affect ice-nuclei formation, and surface dust can reduce the

39 albedo as well as increase subsequent ice melting. As a proxy for past climate,

40 mineral dust is preserved in ice cores, but few studies have examined deposited
41 dust in ice cores during the Anthropocene, especially after 2000. We measured
42 dust concentrations in an ice core at the southeastern dome in Greenland (SE-
43 Dome), and reconstructed the annual and seasonal dust fluxes during 1960–
44 2014. We find the annual average flux during 1960–2014 to be 34.8 ± 13.5 mg
45 $\text{m}^{-2} \text{yr}^{-1}$, a value about twice that of ice cores further inland. The more recent part
46 of that period, 2000–2014, has the higher annual flux of 46.6 ± 16.2 mg $\text{m}^{-2} \text{yr}^{-1}$.
47 The annual and autumn dust fluxes highly correlate with air temperature in
48 Tasiilaq ($r = 0.61$ and 0.50 , respectively), a coastal location in southeastern
49 Greenland. Our results suggest that the local dust emissions at the coastal region
50 are increasing due to a decreasing seasonal snow-cover area arising from
51 coastal Greenland warming after 2000.

52

53 **Keyword: Greenland, dust, ice core, annual flux, seasonal flux**

54

55

56 **1 Introduction**

57 Mineral dust has global and local effects on climate (Carslaw et al., 2010;

58 Kok et al., 2018). Globally, mineral dust can directly influence the atmospheric

59 radiation budget and indirectly affect the radiation budget via the cloud
60 condensation nucleus effect (e.g., Stanelle et al., 2014). Locally, mineral dust is
61 known to affect hot, arid, and subtropical regions (e.g., Mahowald et al., 2006;
62 2014). However, it is increasingly recognized that dust produced at high latitudes
63 and cold environments may extend beyond the local source area and have
64 regional, and even global, significance (Bullard et al., 2016).

65 The motion of glaciers produces fine sediment (glacial flour) that is
66 delivered via meltwater to proglacial floodplains. Then, when the glacier retreats,
67 more land surface area with fine sediment becomes exposed to wind action,
68 meaning that local dust emissions at high latitudes are likely to increase in a
69 warming climate (Bullard, 2013; Simonsen et al., 2019). Once airborne, such
70 glacial-outwash dust has a remarkably high ice-nucleating ability under
71 conditions relevant for mixed-phase cloud formation (Tobo et al., 2019). In
72 addition, at high latitudes, the low humidity, strong winds, permafrost, and niveo-
73 aeolian processes promote dust emission and distribution of sediments. The
74 subsequent dust deposition on the surface of ice sheets and glaciers can
75 decrease ice albedo and increase subsequent glacier melting (e.g., Aoki et al.,
76 2006; Fujita, 2007; Fujita et al., 2011; Nagorski et al., 2019).

77 Ice cores drilled in high-latitude regions preserve past aerosols including
78 mineral dust (e.g., Lambert et al., 2008). The source of the mineral dust has been

79 studied mainly by using the isotopic method (Bory et al., 2003; Delmonte et al.,
80 2008). Over the long term, mineral dust contributes to the glacial–interglacial
81 cycle as a cooling factor (e.g., Seinfeld and Pandis, 1998). Dust deposited in
82 inland Greenland mostly consists of the fine grain size (<5 µm), mainly coming
83 from long-term transport from Asian source regions (Biscaye et al., 1997; A.-M.
84 Bory et al., 2002; A. J. M. Bory et al., 2003; Uno et al., 2009). On the other hand,
85 dust deposited on the Renland ice cap, a coastal region in Greenland, is
86 dominated by coarse particles (>5 µm), suggesting a dust source local to the ice
87 cap that sediments rapidly, typically within one day (Simonsen et al., 2019). Other
88 regions on the Greenland ice sheet that may have significant contributions from
89 both long-transported Asian and short-transported local sources have not been
90 examined in detail.

91 Over the local scale and short-term, the picture is less clear. Few studies
92 of deposited dust in ice cores of inland Greenland have covered the
93 Anthropocene, especially after 2000. From a lake-sediment core in west
94 Greenland, Salos et al., (2019) found that after 2006, the mean July air
95 temperature shifted 1.1 °C higher. They also found that nonlinear environmental
96 responses occurred with or shortly after the abrupt climate shift, including
97 increases in both dust and ice-sheet discharge. However, the dating uncertainty
98 of ice cores makes it difficult to compare an ice-core proxy such as dust with a

99 climatic index such as air temperature. Over the past 600 years, the warmest
100 period began after 2000. The dust in this period has been modeled and monitored
101 by satellite, but such approaches have been insufficient for understanding local-
102 scale climate systems (Bullard et al., 2016). For example, satellite detection of
103 dust is particularly challenging in the high latitudes due to darkness above the
104 polar circles in late fall and winter (Bullard et al., 2016). Thus, a record of
105 deposited dust from an ice core with accurate dating resolution would greatly help
106 us evaluate the relationship between dust and the local climate system.

107 The Greenland ice sheet has a dome in the southeastern area (SE-
108 Dome; 67.18°N, 36.37°W, 3170 m a.s.l.), located on the margin of Greenland ice
109 sheet. Despite its high altitude of over 3,000 m a.s.l., the SE-Dome is located
110 near the North Atlantic Ocean. The SE-Dome lies midway between ice-core sites
111 in inland Greenland and Renland. Thus, the SE-Dome is an ideal location to
112 evaluate contributions of both Asian and local sources of mineral dust. Also, due
113 to the extremely high accumulation rate at the SE-Dome, its ice-core record has
114 a high time resolution (Iizuka et al., 2017). Calibration between oxygen isotope
115 ($\delta^{18}\text{O}$) data from SE-Dome samples and $\delta^{18}\text{O}$ models produced a reconstruction
116 of the paleoclimate and atmospheric circulation over the last 60 years with an
117 uncertainty of ± 2 months (Furukawa et al., 2017). Thus, over this period,
118 paleoclimate reconstruction here allows one to examine the relationship between

119 dust and climate on the seasonal scale. In this study, we analyzed dust
120 concentrations in the SE-Dome ice core over this period, investigating the
121 relationship between the climate system and a high-latitude dust source, with
122 focus on the post-2000 years.

123

124 **2 Materials and Methods**

125 2.1 SE-Dome ice core and age scale

126 Dust data came from a 90.45-m depth ice core obtained at a dome site
127 on the SE-Dome (67.18°N, 36.37°W, 3170 m a.s.l., Fig. 1a). The annual mean
128 temperature at the SE-Dome is -20.9 °C, based on 20-m-deep firn-temperature
129 measurements (Iizuka et al., 2017). For a timescale, we use the SEIS2016 age
130 scale for 1960–2014, which is determined by the oxygen-isotope matching
131 method (Furukawa et al., 2017). The SEIS2016 scale has been carefully
132 evaluated with independent age markers of tritium and volcanic events, and its
133 precision is within two months (Fig. 3 in Furukawa et al., 2017). Depths marking
134 the beginning of the year are found by linear interpolation.

135

136 2.2 Microparticle (dust) concentration measurements

137 In a cold room (Institute of Low Temperature Science, Hokkaido
138 University, Japan), we divided the 90.45-m ice core into 941 sections, each of
139 close to 100 mm depth. Based on the SEIS2016 age scale, uppermost 852
140 samples cover the period from 1960 to 2014. The samples were divided using a
141 clean ceramic knife in a cold clean room (class 10000), put into clean
142 polyethylene bottles, and then were melted in the bottle at room temperature in a
143 clean room. Concentration and grain-size distributions of the microparticles,
144 hereafter dust, were measured using a Beckman Coulter Counter Multisizer 3
145 with an aperture of 30 μm (size range between 0.6 and 18 μm in diameter) in a
146 class 10000 clean room.

147 To make a measurement solution of 15 ml, we mixed 3 ml of melted ice-
148 core sample with 12 ml of a liquid dilution agent (ISOTON II, Beckman) in a 25
149 ml bottle (Accuvette ST, Beckman). To dissolve any bubbles in the melted water,
150 the solutions were kept at least 24 hours in the clean room after melting. Then,
151 to homogenize the settled (larger) particles, the solutions were gently stirred
152 using a 1000 μl pipette (Eppendorf Research) without making bubbles. For the
153 background value, we ran a blank test of the above method by using 96 samples
154 with ultra-pure water (18.2 $\text{M}\Omega\text{ cm}$). Each sample consisted of 3 ml of the super-
155 pure water and 12 ml of the diluent. The average and standard deviation of the

156 particle number and mass concentration were $1900 \pm 423 \text{ ml}^{-1}$ and $2.6 \pm 1.0 \mu\text{g}$
157 kg^{-1} , respectively. The mass concentration was calculated from dust volumes,
158 assuming a spherical approximation and a density of 2.50 g cm^{-3} .

159

160 2.3 Flux estimation

161 The annual and seasonal dust fluxes are based on the seasonal average
162 value of each dust concentration value multiplied by the seasonal accumulation
163 rate. The seasonal boundaries are March 1st, June 1st, September 1st, and
164 December 1st for spring (MAM), summer (JJA), autumn (SON), and winter (DJF).
165 The annual dust fluxes use the boundary of January 1st. These annual and
166 seasonal accumulation rates are estimated based on the SEIS2016 age scale
167 (Furukawa et al., 2017).

168 2.4 Observation of particle sizes and shapes

169 Insoluble particles were collected on a polycarbonate membrane filter
170 (Advantec 13 mm, pore size $0.4 \mu\text{m}$) following the method in Iizuka et al. (2009).
171 The filter was coated with Pt by using magnetron sputter (MSP-10, Vacuum
172 Device). Then, the particle shape was observed using a scanning electron
173 microscope (JSM-6360LV, JEOL) with an energy dispersive x-ray spectrometer
174 (JED2201, JEOL). We confirmed that almost all measured particles contained Si,

175 indicating silicate minerals (dust). To determine their shapes, we examined
176 particles in a sample from summer 1964 (depth 80.105 m) and a sample from
177 summer 2003 (24.800 m), which had the largest dust mass concentrations in
178 1960–1999 and in 2000–2014, respectively.

179 2.5 ERA-40 and ERA-Interim reanalysis data

180 To evaluate climate records in the SE-Dome ice core, we used the ERA-
181 40 (1958–2001) and ERA-Interim (1979–2014), hereafter ERA-I, reanalysis
182 datasets produced by the European Centre for Medium-Range Weather
183 Forecasts (ECMWF) (Uppala et al., 2005; Dee et al., 2011). The daily and monthly
184 mean air temperature at the SE-Dome elevation (3170 m a.s.l.) were extracted
185 from the pressure level air temperature by referring to the geopotential height,
186 both in ERA-40 and ERA-I (Sakai et al., 2015; Furukawa et al., 2017). Daily
187 precipitation was also retrieved from ERA-40 and ERA-I for the backward
188 trajectory analysis. To maintain consistency between the two precipitation
189 products for the whole period (1958–2014), the daily precipitation of ERA-40 (p_{40})
190 is calibrated with that of ERA-Interim (p_I) by a linear regression obtained for the
191 period 1979–2001 ($p_I = 1.36p_{40}$, $R^2 = 0.862$, $p < 0.001$, Iizuka et al., 2018).

192

193 2.6 Backward trajectory analysis

194 To investigate source regions of the chemical species in the ice core,
195 transport pathways of air masses are analyzed using the HYSPLIT model (hybrid
196 single-particle Lagrangian integrated trajectory), a model distributed by NOAA
197 (National Oceanographic and Atmospheric Administration) (Stein et al., 2015).
198 We followed the backward trajectory analysis as described in Iizuka et al. (2018),
199 and set four points at 10,500 m above ground level (a.g.l.) at SE-Dome for the
200 start of the three-day backward trajectory calculation.

201

202 2.7. Remotely sensed snow-free terrain

203 To investigate how variations in the distribution of snow-free areas affect
204 dust sources during 1979 to 2008, we used a 0.05° gridded daily calibrated
205 radiance and classification product derived from the Advanced Very High
206 Resolution Radiometer (AVHRR). Details of this product are in Hori et al. (2017).
207 Product flag values are defined as snow, sea ice, bare land, open ocean, and
208 cloud. To calibrate the snow-free pixels, we used the bare land flags on the
209 Greenland region defined by DiMarzio (2007). Over a month, we counted the
210 number of days a given pixel was flagged as "bare land" and the number of days
211 it was not flagged as "cloud", then took the ratio. If the ratio exceeded 0.5, it was

212 classified "snow-free" for the month. This procedure was done for each pixel of
213 the designated area for the months from April to September. The extracted snow-
214 free pixels were projected onto Lambert's azimuthal equal-area projection to
215 calculate the areal extent. The month with maximum snow-free area (July or
216 August) is defined as the "maximum month". For the analysis, the designated
217 area was that within 65–75 °N of the east coast of Greenland (Fig. 1), the most
218 likely source region of local dust based on the backward trajectory analysis
219 (maximum area of $6.5 \times 10^4 \text{ km}^2$).

220

221 **3 Results and discussion**

222 3.1 Dust concentrations

223 The average concentration of dust particle number in the SE-Dome ice
224 core over 1960–2014 is $13000 \pm 6200 \text{ ml}^{-1}$ (Table 1). (Unless otherwise noted,
225 uncertainties here are the standard deviation.) The numbers also show that the
226 concentration during 2000–2014 is $14800 \pm 6880 \text{ ml}^{-1}$, which is almost same or
227 slightly higher than that over 1960–2014. Larger particles (over $5 \mu\text{m}$) are smaller
228 in number, but show the same trend, almost same or slightly increasing from 9.08
229 $\pm 1.43 \text{ ml}^{-1}$ for 1960–2014 to $11.5 \pm 2.32 \text{ ml}^{-1}$ for 2000–2014.

230 From the profile in Fig. 2a, the dust mass concentration has an average

231 value (1960–2014) of $34.2 \pm 22.1 \mu\text{g kg}^{-1}$. The average blank error is 7.6 % in this
232 study. Table 1 shows that the average mass concentration increases after 2000
233 to $42.9 \pm 29.1 \mu\text{g kg}^{-1}$. Both are within 1σ , but the mass concentration is 25.4%
234 higher after 2000, but the number concentration is only 13.8% higher. The
235 difference is related to the relative increase of the fraction of coarser particles.

236 The dust size distribution by mass (green and purple in Fig. 3a) over
237 1960–2014 show a bimodal trend. One peak occurs around 1–2 μm , the other
238 around 15 μm , suggesting contributions from two sources. The dust size
239 distribution of Renland ice core (the RECAP Holocene) has a peak around 20 μm ,
240 suggesting high contribution from local source (Simonsen et al., 2019). Thus, the
241 large particle mode (around 15 μm) of the SE-Dome ice core implies a nearby
242 dust source. As the long-term transport from Asian source regions has particles
243 mostly less than 5 μm (Biscaye et al., 1997; A.-M. Bory et al., 2002; A. J. M. Bory
244 et al., 2003; Uno et al., 2009), we divide the two sources at 5 μm . The mass
245 concentration for dust larger than 5 μm is $10.7 \pm 10.8 \mu\text{g kg}^{-1}$, which is 31.3% of
246 the total mass concentration during 1960–2014. For the period 2000–2014, the
247 corresponding mass for dust larger than 5 μm is $12.6 \pm 12.9 \mu\text{g kg}^{-1}$, which is
248 29.4% of the total mass concentration. Thus, the size distribution by mass shows
249 a similar bimodal trend between 1960–2014 and 2000–2014, suggesting little
250 change of the source (Asian or local) contributions between the two periods. The

251 periods of dust events in summers 1964 and 2003 (red and blue in Fig. 3a) have
252 larger size distributions than those in averaged distribution (green and purple in
253 Fig. 3), suggesting high contribution from local source during the periods.

254 We examined SEM micrographs of particles from summer 1964 (depth
255 80.105 m) and summer 2003 (24.800 m). In Fig. 2a, these years have the largest
256 dust mass concentrations in 1960-1999 and in 2000–2014. These seasons did
257 not have any large volcanic eruption in Iceland. The SEM analyses show that the
258 SE-Dome ice core has a significant number of dust particles over 5 μm , especially
259 in 2003 (e.g., Fig. 3d); however, the size distributions have similar trends for
260 summer 1964 and summer 2003 (Fig. 3b). In addition, the dust-particle shapes,
261 as indicated by the aspect ratios, are similar between summer 1964 and summer
262 2003 (Fig. 3c). The similar size distributions and aspect trends suggests the same
263 contributions from each source region between the two seasons.

264

265 3.2 Annual and seasonal dust flux reconstructions

266 The annual and seasonal dust fluxes equal the product of the mass
267 concentration with their respective accumulation rate (Furukawa et al., 2017). The
268 resulting annual flux for 1960–2014 in Fig. 2a gives an average of $34.8 \pm 13.5 \text{ mg}$
269 $\text{m}^{-2} \text{yr}^{-1}$. This average is greater than the yearly values of 14–19 $\text{mg m}^{-2} \text{yr}^{-1}$ at the

270 inland ice cores of GRIP, NGRIP, and Dye3 (Bory et al., 2003), but is less than
271 the $57 \text{ mg m}^{-2} \text{ yr}^{-1}$ at Haus Tausen in the coastal region of north Greenland (Bory
272 et al., 2003) and much less than the $680 \text{ mg m}^{-2} \text{ yr}^{-1}$ at Renland on an ice cap
273 near the ice sheet (Bory et al., 2003). Over the extensive inland region of
274 Greenland, the flux values are low and nearly uniform due to the long-distant
275 transport over high-elevations to the ice sheet (Bory et al., 2003). On the other
276 hand, the high dust fluxes in Haus Tausen and Renland are mainly due to the
277 local source (Simonsen et al., 2019). The average flux value of $34.8 \text{ mg m}^{-2} \text{ yr}^{-1}$
278 of the SE-Dome ice core is about twice that of the inland ice cores. We argue
279 next that this difference indicates that some dust particles come from local
280 sources such as exposed glacial sediments, moraines, rock, soil, and sand in
281 coastal Greenland (e.g. Bullard et al., 2016).

282 The high contribution to the flux from large particles is one argument for
283 a local dust source. The average annual flux from particles larger than $5 \mu\text{m}$ is
284 $10.6 \pm 4.26 \text{ mg m}^{-2} \text{ yr}^{-1}$ (Fig. 2b, blue curve), which is 30.5% of the total flux value.
285 Due to the relatively rapid fallout of such large particles, they must be from a
286 nearby source. For particles smaller than $5 \mu\text{m}$, the average annual flux is the
287 remaining $24.2 (= 34.8 - 10.6) \text{ mg m}^{-2} \text{ yr}^{-1}$. Of this flux, some must also be from
288 the local sources. The similar method of the $5 \mu\text{m}$ threshold of dust size dividing
289 long and local transportation was done in coastal Antarctica (Baccolo et al., 2018).

290 If we assume that this remaining dust flux equals that from long distance sources
291 plus fine dust from local sources, with the former equaling that reaching the inland
292 ice cores of GRIP, NGRIP, and Dye3 ($14\text{--}19\text{ mg m}^{-2}\text{ yr}^{-1}$), then the annual flux of
293 fine particles from local sources equals $5.2\text{--}10.2\text{ mg m}^{-2}\text{ yr}^{-1}$. This amount from
294 local sources is $15.0\text{--}29.3\%$ of the total flux. Totaling both the smaller and larger
295 dust flux from the local sources, we estimate that $45.5\text{--}59.8\%$ of the dust at the
296 SE-Dome ice core is from a local source.

297 A second argument for a local source contribution comes from the recent
298 trend in annual flux. In particular, the total flux is higher after 2000 (46.6 ± 16.2
299 $\text{mg m}^{-2}\text{ yr}^{-1}$ during 2000–2014; Table 1 and Fig. 2a), but the flux from Asia is not
300 likely to have increased during this time. In particular, Liu et al., 2020 found that
301 the Asian dust intensity during 1961–2020 was high until 1980 and then
302 decreased to the present (2020). This fact suggests that the Asian dust source
303 by itself cannot explain the high flux after 2000 in the SE-Dome ice core. Given
304 that the atmospheric circulation does not change after 2000 (Fig. 1b,c), the
305 increase after 2000 must be from a local source. In addition, the annual flux of
306 the larger ($>5\text{ }\mu\text{m}$) particles also increased after 2000 to $13.3 \pm 5.33\text{ mg m}^{-2}\text{ yr}^{-1}$
307 (2000–2014; Table 1 and Fig. 2b). Thus, the contribution from local sources
308 probably increased after 2000.

309 Finally, a third argument for a local source comes from the trend in

310 seasonal flux. The seasonal fluxes during 1960–2014 are plotted in Fig. 2c. We
311 split this period into before and after 2000. Table 2 shows the averages in both
312 periods. During 1960–1999, the averages are nearly the same for all seasons
313 (7.19–8.24 mg m⁻² yr⁻¹). For 2000–2014, the highest average is in autumn at 15.7
314 ± 8.81 mg m⁻² yr⁻¹, suggesting that the high dust flux after 2000 was mainly driven
315 by the increase in autumn. A similar increase in autumn flux occurs for the large
316 particles (Fig. 2d, Table 2). In particular, the average fluxes during 1960–1999
317 are nearly the same for all seasons (2.21–2.45 mg m⁻² yr⁻¹). But for 2000–2014,
318 the highest average is in autumn at 3.72 ± 2.27 mg m⁻² yr⁻¹. However, Asian dust
319 storms tend to come in spring (e.g., Liu et al., 2020), and thus the increase in
320 autumn flux is more likely due to a local source. In the next section, we suggest
321 a cause of the increasing annual and autumn dust fluxes during 2000–2014.

322

323 3.3 Cause of the increase in annual and autumn flux after 2000

324 The distance that dust of a given size will travel will depend on the
325 fallspeed of the dust, the windspeed, and the height of the dust particles.
326 According to Tegen and Lacis (1998), dust of size 1 to 10 μm can transport in the
327 atmosphere for only about 40 hours. To determine the sources of the dust, we
328 calculated the air-mass distributions arriving at the SE-Dome site from back-

329 trajectory analyses going back three days. The air masses are separated into
330 those from 1000 and 1500 m a.g.l., and those from 10 and 500 m a.g.l. The latter
331 case of the two lower air masses can entrain dust emitted from the surface. Fig.
332 1 shows these lower air masses. The air masses mainly come from regions of
333 Greenland just north of SE-Dome, extending up to about 75°N, and include
334 regions just offshore as well as some coastal regions to the south. Both periods
335 between 1960–1999 and 2000–2014 have similar distribution patterns of air mass.
336 The similarity suggests that the increase in particle number during 2000–2014
337 was not driven by a change in atmospheric circulation, but by higher dust
338 emissions in the source regions. Given that the air-mass trajectories do not show
339 the emissions of dust, we focus on specific regions in the higher-probability areas
340 that are likely to have the greatest emissions. These regions would be the snow-
341 free areas in the coastal regions. Thus, we focus on the area marked in red in
342 Fig. 1a.

343 The area includes the cities of Tasiilaq and Itseqqortoormiit, where the
344 Danish Meteorological Institute (DMI) measures air temperature and precipitation.
345 Tasiilaq is located at a coastal region of southeastern Greenland (~190 km from
346 the SE-Dome site, Fig. 1a). Both annual dust fluxes during 1960–2014 correlate
347 more strongly with air temperature in Tasiilaq ($r = 0.61$ in Table 3, Fig. 4a) than
348 with that at the SE-Dome site (Table 3). Most coastal regions in southeastern

349 Greenland have exposed snow-free areas of rock, soil, and glacial flour, which
350 are potential dust sources. The lower air masses, which would be the main
351 atmospheric transport for larger dust, come from the eastern coast between 65
352 and 75 °N of Greenland (Fig. 1). At nearly four times the distance to Tasiilaq lies
353 Itseqqortoormiit (Fig. 1), another potential dust source because it is also on the
354 coast (Fig. 1a). But air temperature in Itseqqortoormiit has lower correlation with
355 the SE-Dome dust ($r = 0.35$) than that of Tasiilaq. This suggests that the larger
356 dust likely comes from very close to the SE-Dome site.

357 The autumn dust fluxes during 1960–2014 also have high correlations
358 with autumn air temperature in Tasiilaq ($r = 0.50$ in Table 3, Fig. 4b). Even omitting
359 the extremely high dust-flux datum from autumn 2014, the autumn average of
360 dust flux (2000–2013) is still highly correlated to the Tasiilaq air temperature ($r =$
361 0.52). The correlation suggests that a warmer autumn on the coast increases the
362 dust emission from snow-free areas exposed by a delay of seasonal snowpack.
363 The seasonal snow-free area would be an additional dust source in the coastal
364 regions, especially during autumn. Thus, high correlations between dust flux and
365 Tasiilaq air temperature, both annual and autumn, during 1960–2014 suggest that
366 the warming in the coastal regions of southeastern Greenland after 2000
367 promotes dust emission to the atmosphere.

368 The snow-free area trend by AVHRR during 1979–2008 is consistent with

369 such warming, showing a loss of seasonal snowpack in the coastal regions
370 around southeastern Greenland. The snow-free areas of the region in September
371 (Fig. 2c) show an increasing trend after 2000. The average snow-free area in
372 September is $14700 \pm 5600 \text{ km}^2$ during 1979–1999 and $21700 \pm 10800 \text{ km}^2$
373 during 2000–2008. This increase suggests that the warmer summers and
374 autumns after 2000 are causing a delay in the seasonal snowpack cover and an
375 extension of snow-free areas. These snow-free areas have high correlation with
376 the autumn dust flux ($r = 0.49$ for the maximum month, 0.60 for September in
377 Table 3; Fig. 4c). The high correlations suggest a larger snow-free area in autumn
378 increased the dust emission, leading to a higher dust flux in the SE-Dome ice
379 core.

380 On the other hand, results in Table 3 show low correlations between dust
381 flux and annual Tasiilaq precipitation ($r = 0.17$) and windspeed ($r = 0.41$). The
382 Tasiilaq precipitation is a proxy of an aridity in Tasiilaq. Dust is more likely to emit
383 from a source area under drier and windier conditions (e.g., Bullard et al., 2016).
384 The low correlations suggest that the high dust flux after 2000 is not due to a high
385 emission activity of the source area, but rather is due to extend dust emission
386 area (i.e., seasonal snow-free area).

387 We also examined the correlations between the dust flux and other
388 possible proxy variables. Results in Table 3 show low correlations ($|r| < 0.30$) with

389 the North Atlantic Oscillation (NAO), which is defined as the pressure difference
390 between the Azores High and the Icelandic Low. The low correlations with NAO,
391 as well as the Arctic Oscillation (AO, $|r| < 0.27$) are consistent with the larger dust
392 size distribution during 2000–2014 not being caused by a stronger atmospheric
393 circulation.

394 Thus, the evidence indicates that the Greenlandic warming produced a
395 larger regional dust emission via a larger seasonal snow-free area in coastal
396 Greenland. The air temperature after 2000 has been increasing throughout the
397 Arctic (Stocker et al., 2013) and specifically around southeastern Greenland
398 (Bjørk et al., 2012), consistent with a retreat of the Greenland ice sheet (Mouginot
399 et al., 2019). As mineral dust plays a key role in the climate system (Carslaw et
400 al., 2010; Kok et al., 2018), the increase in dust flux at the SE-Dome after 2000
401 is a potentially important way that high-latitude dust can affect the climate system.
402 Future studies about the dynamic of the Greenland ice sheet will need to take
403 into account the role of dust, since increased dust deposition implies an increased
404 radiative forcing on the surface of the ice sheet.

405

406 3.4 Contribution of Iceland dust emission

407 Other than the Greenland coast, the next nearest potential dust source is

408 Iceland. Iceland is also a potential local source of volcanic emissions (Groot
409 Zwaaftink et al., 2017). However, the average air-mass probability distribution
410 from Iceland is just $1.51 \pm 1.47\%$ (Figs. 1b,c), which is less than one-third that of
411 the $4.88 \pm 2.37\%$ value from 65 to 75 °N of eastern Greenland by the trajectory
412 analyses. Moreover, the fraction of air trajectories from Iceland hardly changes
413 from the earlier period of 1960–1999 ($1.53 \pm 1.47\%$) to the recent period of 2000–
414 2014 ($1.48 \pm 1.49\%$). Thus, the dust-flux increase after 2000 is not explained by
415 the contribution from Iceland.

416 Nevertheless, over shorter terms, volcanic eruptions in Iceland have
417 affected dust deposition in the SE-Dome region. For example, the extremely high
418 dust flux in the autumn of 2014 ($41.4 \text{ mg m}^{-2} \text{ yr}^{-1}$) might be due to the eruption of
419 Mt. Bardarbunga in Iceland. However, the contribution of Icelandic volcanic
420 eruptions to dust flux into the SE-Dome ice core should depend on air mass
421 trajectory. Over 1960–2014, seven eruptions in Iceland had a volcanic explosivity
422 index (VEI) exceeding 3 (Table 1). The average number and mass dust
423 concentrations of the term during these seven eruptions are $13700 \pm 8270 \text{ ml}^{-1}$
424 and $40.1 \pm 34.6 \text{ } \mu\text{g kg}^{-1}$, respectively (Table 1). These averages are higher than
425 the overall averages during 1960–2014, but are lower than those during 2000–
426 2014 (Table 1). So, some volcanic events likely contribute to dust deposition in
427 the SE-Dome region (e.g., autumn 2014); however, they cannot explain the

428 increase of dust flux in the SE-Dome ice core after 2000. Baddock et al. (2017)
429 showed similar results. They found that trajectories during Icelandic volcanic
430 seasons rarely ascend high enough to reach inland Greenland, suggesting
431 instead that Icelandic dust has more important effects on the neighboring marine
432 environment than on the cryosphere.

433

434 **4 Conclusion**

435 We measured particle-size distributions from an ice core in the
436 southeastern dome in Greenland (SE-Dome), using them to reconstruct annual
437 and seasonal dust fluxes during 1960–2014. The annual average flux over the
438 whole period 1960–2014 was $34.8 \pm 13.5 \text{ mg m}^{-2} \text{ yr}^{-1}$, a value about twice that of
439 inland ice cores. The later term of this period 2000–2014 had the higher annual
440 flux of $46.6 \pm 16.2 \text{ mg m}^{-2} \text{ yr}^{-1}$. The higher flux, together with other trends,
441 indicated that some dust in the SE-Dome region came from local sources of
442 exposed rock, soil, and sand in coastal Greenland.

443 The air-mass source locations hardly changed between the terms 1960–
444 1999 and 2000–2014, suggesting that the reason for the larger size distribution
445 during the later term was likely a higher dust production at the source. One nearby
446 coastal source area is Tasiilaq, which had air temperatures that correlated more

447 strongly to the annual and autumn dust fluxes over 1960–2014 than the air
448 temperature in SE-Dome. A probable local source is the region surrounding
449 Tasillaq, where many snow-free areas are found, especially during summer and
450 autumn. In addition, the snow-free area from 65 to 75 °N of southern and eastern
451 Greenland, possible source areas, also was highly correlated during 1979–2008
452 to the autumn dust flux, especially after 2000. The high dust flux in the SE-Dome
453 ice core after 2000 may indicate a greater influence of high-latitude dust to the
454 future climate system.

455

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665

666

667 Figure and table captions

668

669 **Figure 1.** Study area and air-mass trajectories. **(a)** Locations of southeast dome
670 (SE-Dome), Tasiilaq, and Itseqqortoormiit. Blue and red shaded regions denote
671 potential snow-free areas bordering the Greenland Ice Sheet. The red regions
672 are obtained from the air-mass trajectories in Figs 1b and 1c (east coast between
673 65 and 75 °N), and used to calculate snow-free area in September (Fig. 2c). **(b)**
674 Probability distribution (%) of an air mass arriving at the SE-Dome site from a 3-
675 day, 3-D backward-trajectory analysis, averaged over 1960–1999 for air-mass
676 starting elevations 10 and 500 m a.g.l. Color scale at bottom. **(c)** Same as (b)
677 except for period 2000–2014. Probability of the air mass is weighted by the daily
678 precipitation from combined reanalysis datasets of ERA-40 and ERA-Interim.

679

680 **Figure 2.** Trends in dust mass, dust flux, and snow-free coastal area. **(a)** Annual
681 dust mass concentration (left) and dust flux (right) in the SE-Dome ice core.
682 Dotted line shows a running average over 5 years. **(b)** Same as (a) except for
683 particles exceeding 5 μm . **(c)** At left are seasonal dust fluxes in the SE-Dome ice
684 core. Blue, green, red, and purple for spring, summer, autumn, and winter,
685 respectively, the dotted lines are running averages over 5 years. At right, snow-
686 free area in September on the east coast of Greenland within 65–75°N derived

687 from AVHRR from 1979 to 2008. (Data losses in 1980 and 1981 are due to having
688 insufficient satellite observations for that period.) **(d)** Same as (c) except for
689 particles exceeding 5 μm and without the snow-free area.

690

691 **Figure 3.** Particle distributions. **(a)** Particle mass–size distribution by the Colter
692 Counter method during 1960–2014 (purple), 2000–2014 (green), summer 1964
693 (red) and summer 2003 (blue). **(b)** Particle number–size distribution by the SEM
694 method on summer 1964 (red) and summer 2003 (blue). **(c)** Same as (b) except
695 for particles' aspect ratio. **(d)** Example of a particle larger than 5 μm from summer
696 2003.

697

698 **Figure 4.** Correlations of dust fluxes to temperature in Tasiilaq and coastal snow-
699 free area. **(a)** Annual dust flux and annual air temperature at Tasiilaq. **(b)** Autumn
700 dust flux and autumn air temperature at Tasiilaq. **(c)** Autumn dust flux and the
701 snow-free area in September during 1979–2008. Monthly air temperatures 1960–
702 2014 at Tasiilaq (65.60°N, 37.59°W) are from the Danish Meteorological Institute
703 (Cappelen, 2016).

704

705 **Table 1.** Average values and standard deviations of number and mass dust
706 concentrations during 1960–2014, 2000–2014, and seasons with large volcano

707 eruption events in Iceland. The average values and standard deviations are
708 shown in the cases of within 0.6–18 μm , and 5.0–18 μm . Seasons with 7 large
709 volcano eruption events in Iceland: 1) Bárðarbunga, autumn 2014; 2) Grímsfjall,
710 spring 2011; 3) Eyjafjallajökull, spring 2010; 4) Hekla, winter 2000; 5) Krafla,
711 autumn 1984; 6) Eldfell, winter 1973; and 7) Askja, autumn 1961.

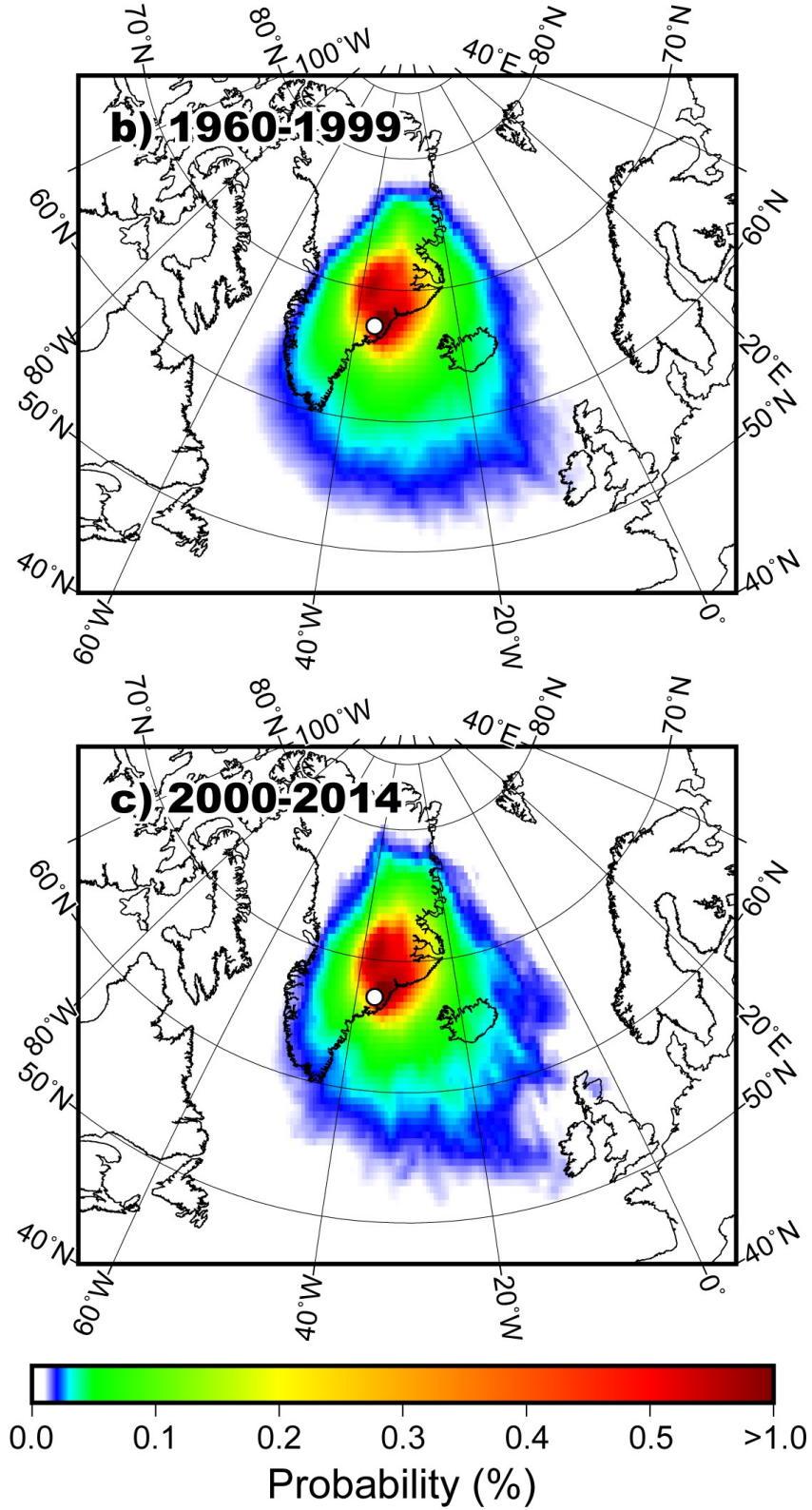
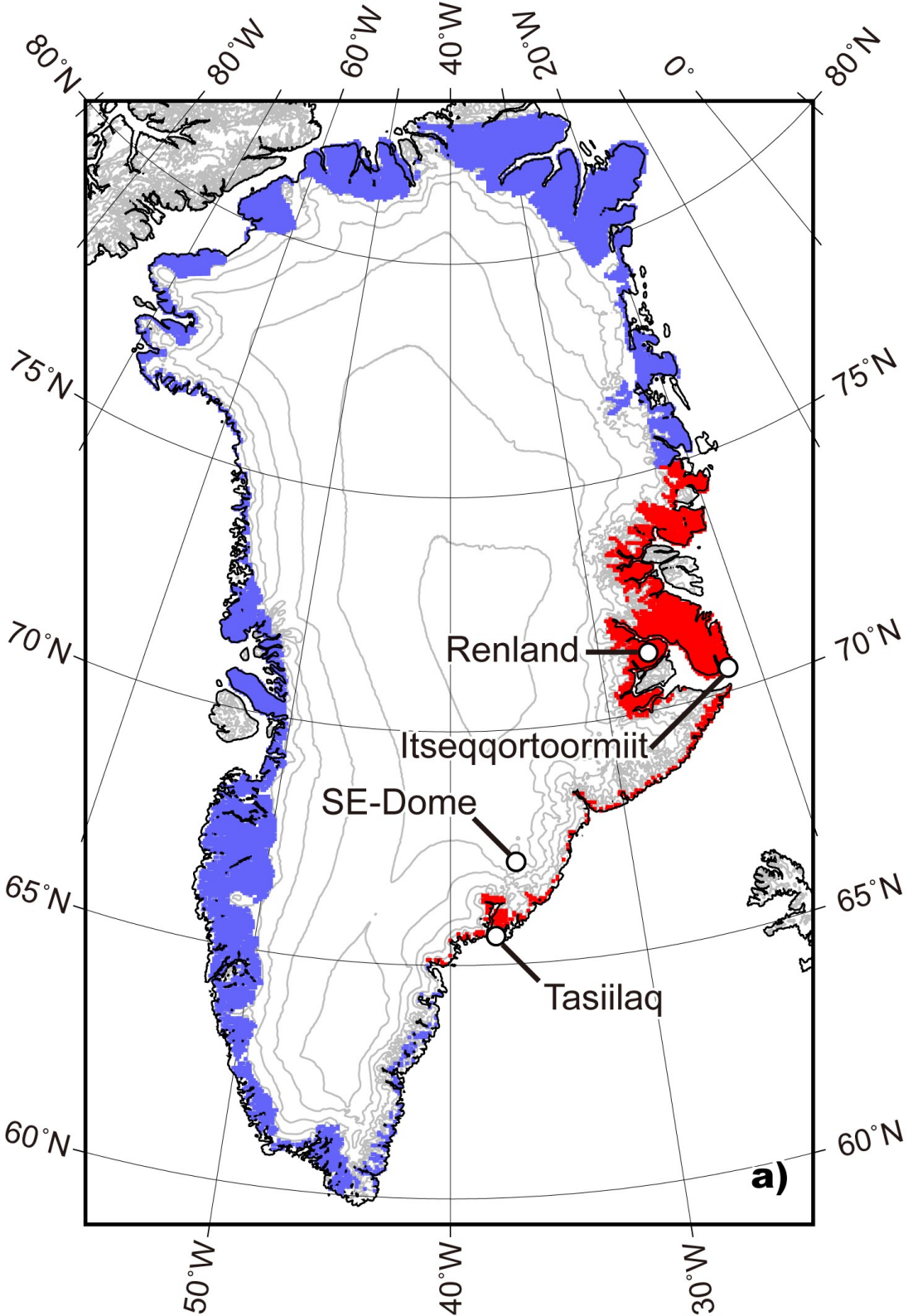
712

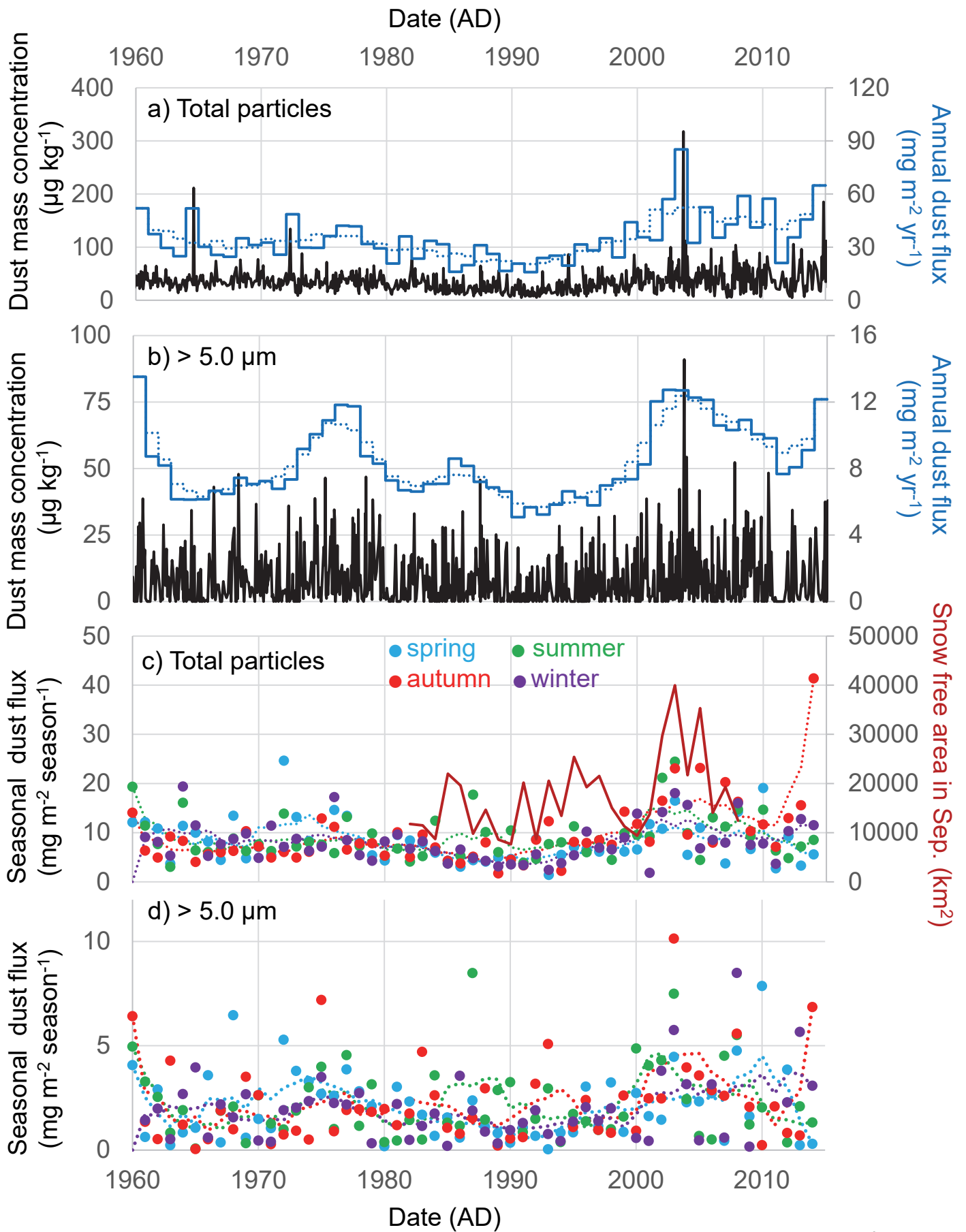
713 **Table 2.** Seasonal average dust fluxes ($\text{mg m}^2 \text{ season}^{-1}$) with standard deviations
714 during 1960–1999 and 2000–2014 in the cases of within 0.6–18 μm , and 5.0–18
715 μm .

716

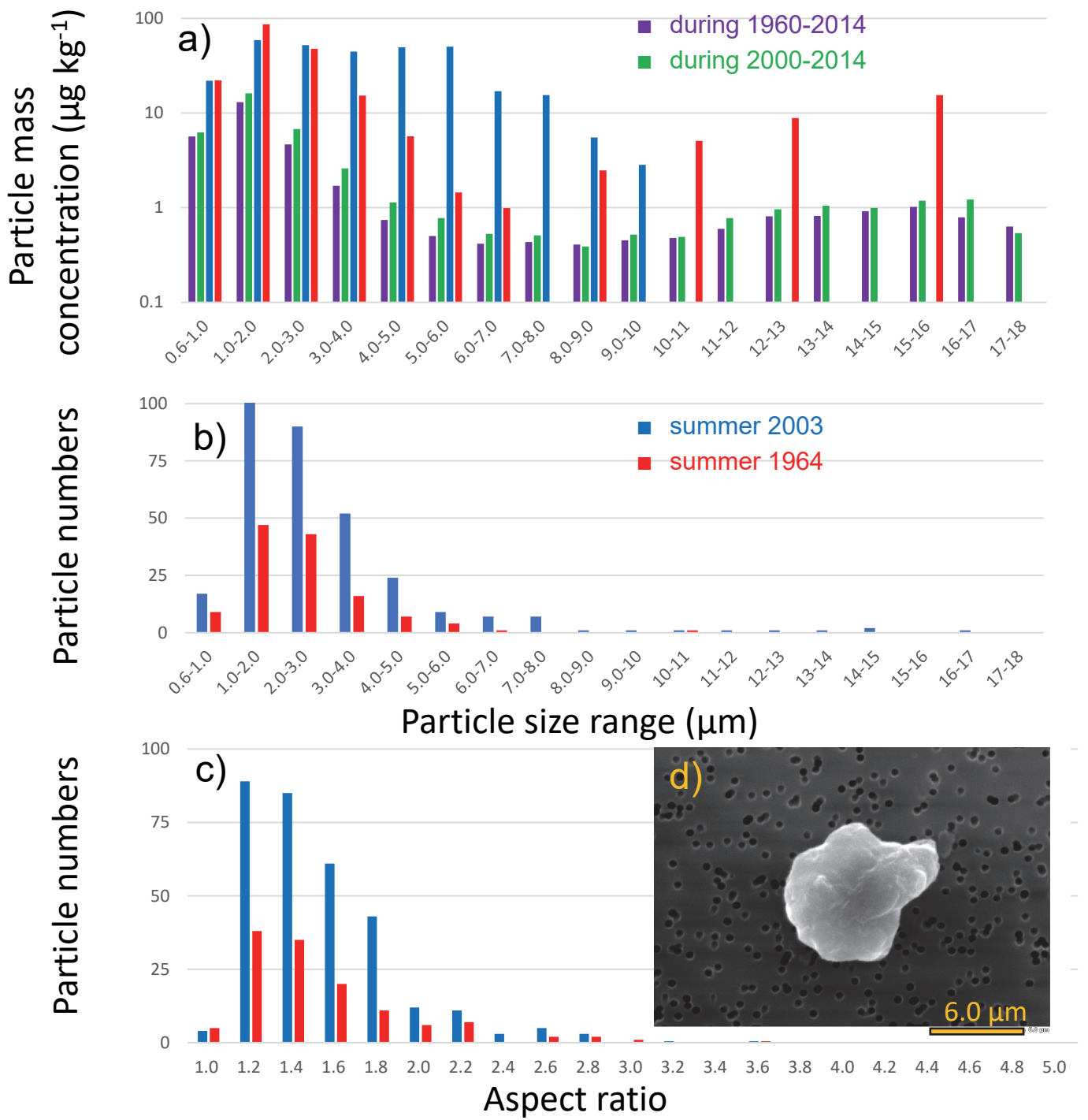
717 **Table 3.** Annual and seasonal correlation coefficients (r) between dust fluxes and
718 potentially relevant proxies during 1960–2014. Bold marks values of 0.5 or higher.
719 Gray shading marks values from plots in Fig. 4.

720

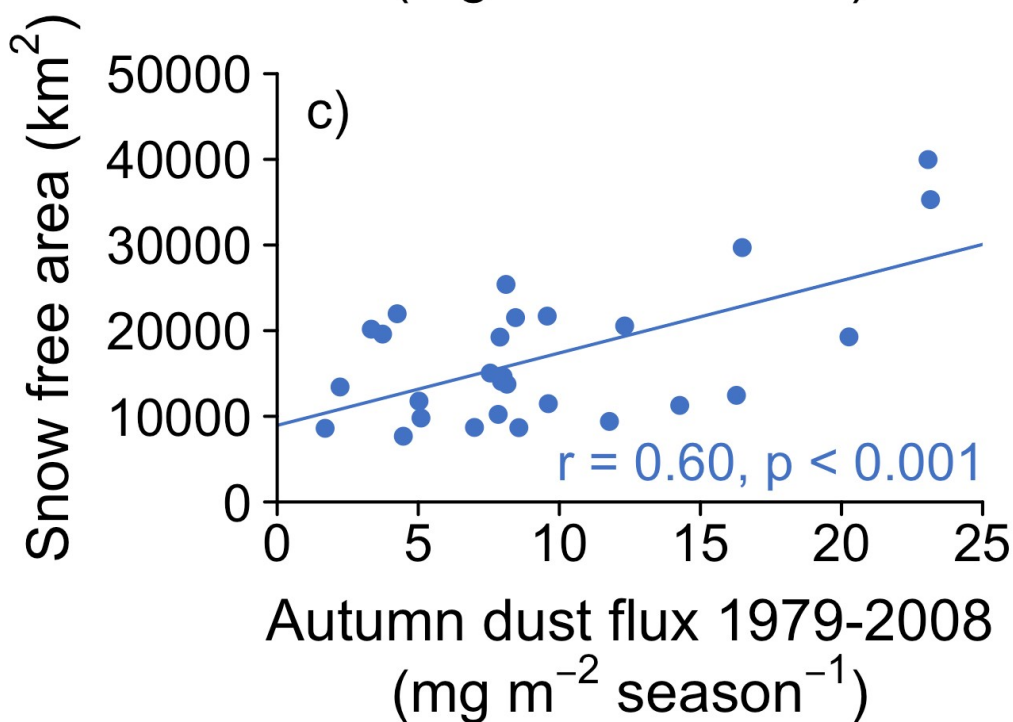
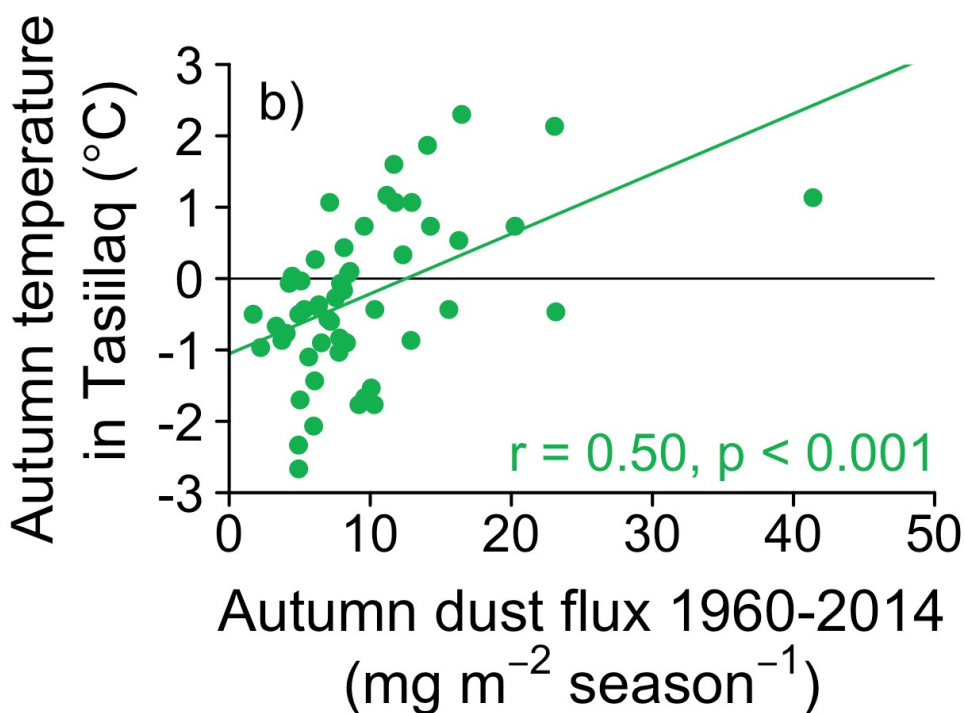
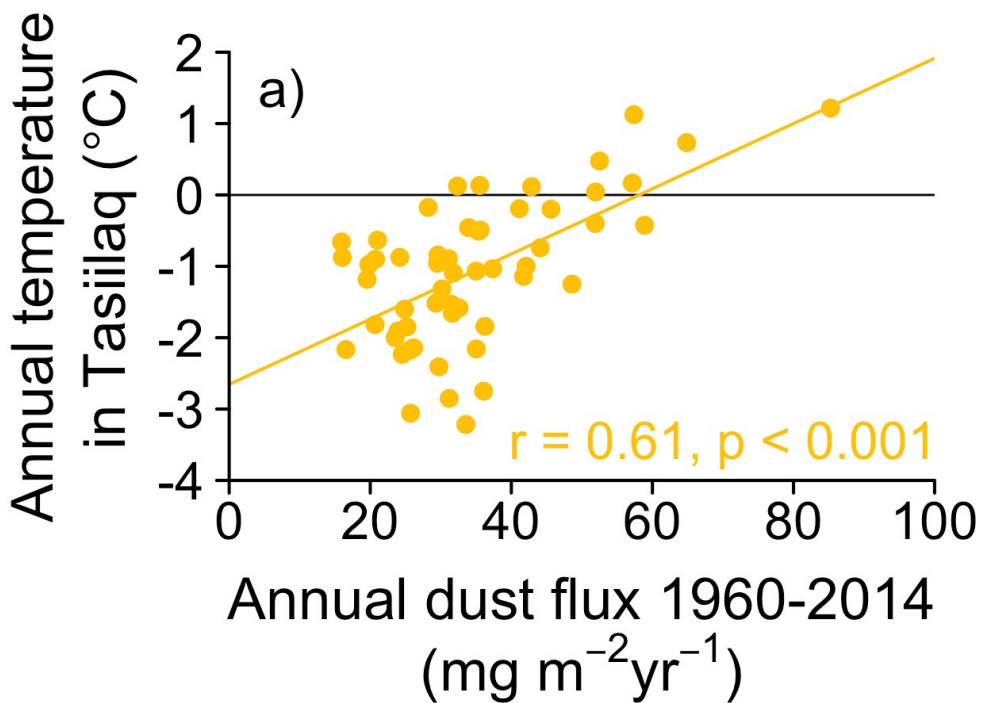




Amino et al., Fig. 2



Amino et al., Fig. 3



| Term | Particle size (μm) |
|---------------------------|---------------------------------|
| Period 1960-2014 (n =850) | 0.6 ~ 18 |
| Period 1960-2014 (n =850) | 5.0 ~ 18 |
| Period 2000-2014 (n=259) | 0.6 ~ 18 |
| Period 2000-2014 (n=259) | 5.0 ~ 18 |
| Volcanic seasons* (n=26) | 0.6 ~ 18 |
| Volcanic seasons* (n=26) | 5.0 ~ 18 |

| Number conc.(mL ⁻¹) | Mass conc. (µg kg ⁻¹) |
|---------------------------------|-----------------------------------|
| 13000±6000 | 34.2±22.1 |
| 9.08±1.43 | 10.7±10.8 |
| 14800±6880 | 42.9±29.1 |
| 11.5±2.32 | 12.6±12.9 |
| 13700±8270 | 40.1±34.6 |
| 16.4±2.82 | 14.8±14.9 |

| Period | Term | Particle size within 0.6 ~ 18 μm | Particle size within 5.0 ~ 18 μm |
|-----------|--------|--|--|
| 1960-1999 | Spring | 7.79 \pm 4.20 | 2.24 \pm 1.47 |
| | Summer | 8.24 \pm 3.74 | 2.45 \pm 1.71 |
| | Autumn | 7.19 \pm 2.96 | 2.38 \pm 1.89 |
| | Winter | 7.28 \pm 3.44 | 2.21 \pm 1.85 |
| 2000-2014 | Spring | 9.01 \pm 4.73 | 2.86 \pm 1.81 |
| | Summer | 11.2 \pm 5.46 | 3.34 \pm 2.42 |
| | Autumn | 15.7 \pm 8.51 | 3.72 \pm 2.27 |
| | Winter | 10.4 \pm 4.52 | 3.06 \pm 2.02 |

| | Ca ²⁺ | d ¹⁸ O (‰) | d-excess (‰) | NAO index | AO index | SE-Dome air temperature (°C) | Tasiilaq air temperature (°C) | Itseqqortoormiit air temperature (°C) | Tasiilaq precipitation (mm) | Itseqqortoormiit precipitation (mm) | Tasiilaq wind speed (m s ⁻¹) [1974-2014] | Snow-free area (km ²) in *maximum month [1979-2008] | Snow-free area (km ²) in September [1979-2008] |
|--------------------|---------------------|-----------------------|-----------------------|----------------------|----------------------|---------------------------------------|-------------------------------|---------------------------------------|-----------------------------|-------------------------------------|--|---|--|
| References | Iizuka et al., 2018 | Furukawa et al., 2017 | Furukawa et al., 2017 | Hurrell et al., 2003 | Higgins et al., 2001 | Uppala et al., 2005; Dee et al., 2011 | Cappelen, 2016 | Cappelen, 2016 | Cappelen, 2016 | Cappelen, 2016 | NCEI, NOAA, U. S. | This study | This study |
| annual [1960-2014] | 0.66 | 0.27 | -0.07 | -0.25 | -0.17 | 0.43 | 0.61 | 0.35 | 0.17 | 0.13 | 0.41 | 0.43 | 0.58 |
| spring [1960-2014] | 0.37 | 0.11 | -0.03 | -0.30 | -0.05 | 0.39 | 0.20 | -0.12 | 0.25 | 0.09 | -0.12 | 0.34 | 0.38 |
| summer [1960-2014] | 0.52 | 0.04 | -0.12 | -0.03 | -0.12 | 0.25 | 0.38 | 0.18 | -0.06 | -0.09 | -0.16 | 0.25 | 0.30 |
| autumn [1960-2014] | 0.83 | 0.20 | 0.00 | -0.02 | -0.13 | 0.20 | 0.50 | 0.44 | -0.09 | 0.27 | 0.38 | 0.49 | 0.60 |
| autumn [1960-2013] | 0.65 | 0.32 | -0.02 | -0.10 | -0.05 | 0.22 | 0.52 | 0.40 | -0.09 | 0.06 | 0.46 | 0.49 | 0.60 |
| winter [1960-2014] | 0.17 | 0.10 | 0.15 | -0.17 | -0.27 | 0.27 | 0.13 | 0.09 | 0.07 | 0.04 | -0.11 | 0.35 | 0.38 |

* maximum month means July or August with greatest snow-free area