

1 **Replies to Referee Comments (RC1, RC2) and Short Comment (SC1)**

2 We thank both referees No.1 and No.2 for their detailed and insightful comments which helped to
3 further improve the manuscript. Below we address all referee comments as well as the contributed
4 short comment (in italics). Revised text, keyed to the ACPD online version, is shown in blue, and
5 is included in the final manuscript we are submitting to ACP. Updated figures and tables as well as
6 material for a supplement are attached at the end of our reply.

7
8 **RC1:** *Source assignment of proxies is a basic prerequisite for interpreting climate archives in terms*
9 *of past climate as well as climate change. Concerning polar ice cores, ionic impurities originate pri-*
10 *marily from aerosol deposition. Amongst them, interpretation of sea salt aerosol deposition archived*
11 *in ice cores is especially challenging and contro- versial because the contribution of two different and*
12 *competing sources - viz. open water versus sea ice - is up for debate. In addition it became apparent*
13 *that sea salt aerosol production over ice-covered oceans may contribute significantly to the global sea*
14 *salt aerosol budget. The manuscript at hand addresses this pivotal subject and provides thorough*
15 *and direct observational evidence of sea salt aerosol production from blowing snow above sea ice.*
16 *The important conclusions drawn are based on comprehensive state of the art ship-borne aerosol*
17 *and snow measurements during winter / early springtime in the Weddell Sea region. Although the*
18 *main conclusions are primarily restricted to the chosen site, there are certainly strong implications for*
19 *climate research in the Southern Ocean realm and climate related interpretation of sea salt profiles*
20 *from ice cores in general. The authors have accomplished a clear, well-organised and concise paper.*
21 *The methodology is sound and assumptions are identified clearly and conscientiously. From my point*
22 *of view, all parts, including figures, are essential. The manuscript certainly addresses the scientific*
23 *scope of ACP and I recommend a final publication after some minor revisions I specified below.*

24 **Reply:** We thank the reviewer for the positive assessment of this work.

25
26 **RC1:** *1. Abstract, page 1, line 13 and Conclusions, page 24, line19: The authors state that bromine*
27 *enrichment was typical at 29 m height, but from Chapter 3.4.2 and Fig. 15, bromine depletion is evi-*
28 *dent. Please clarify.*

29 **Reply:** In both cases it should read "depleted", which is now corrected.

30
31 **RC1:** *2. Chapter 2.3, Aerosol chemical composition: Could you assess the impact of pollution on*
32 *chemical aerosol composition? Was the bulk aerosol sampling contamination controlled?*

33 **Reply:** The setup of this study did not include any contamination control of the bulk aerosol filter
34 sampling such as pump control based on wind speed and direction. As described aerosol num-
35 ber concentrations at the crows nest showed significant spikes, when air came from the direction
36 of the ship stack, whereas no evidence of pollution was detected in aerosol number concentrations
37 observed on the sea ice. We clarified text in section 2.2 including also the fraction of CLASP data
38 filtered out:

39 *Raw aerosol number concentrations at the crow's nest showed significant spikes, when air came from*
40 *the direction of the ship's engine stack, whereas no evidence of pollution was detected in the obser-*
41 *ventions on the sea ice. Pollution spikes were effectively filtered out prior to averaging by excluding all*
42 *data when relative wind direction was in the 135–225° sector encompassing the ship's engine stack.*
43 *A total of 21% of the available 1-second data was removed from the crow's nest data.*

44 We now include an assessment of the potential impact of pollution on bulk aerosol chemistry from
45 filters and added the following text to section 2.3:

46 *In order to assess the impact of potential pollution on bulk aerosol chemistry from filters collected at*
47 *the crow's nest we calculated for each filter sample the fraction of the total filter run time during which*
48 *relative wind direction was within the 135–225° sector encompassing the ship's engine stack. Con-*
49 *sidering all filters sampled from June to September 2013 (N=141) the fraction of total filter run time*
50 *with winds from the polluted sector was on average 9.5%. Polluted time fraction and atmospheric*
51 *concentrations of Na⁺, Cl⁻, SO₄²⁻ and Br⁻ did not show any correlation (R²<0.05), suggesting that*
52 *the impact of pollution on the respective ion concentrations is small. A weak, but significant negative*
53 *correlation was found between polluted time fraction and depletion factors DF_{S₄O₂⁻}* (R²=0.19, p<0.01)

54 and DF_{Br^-} ($R^2=0.13$, $p<0.01$) suggesting that enrichment in sulphate (and bromide) may be more
55 likely during polluted conditions. The bulk aerosol chemistry observations on the sea ice showed
56 no evidence of pollution. Thus, in the case of sulfate we cannot rule out that some of the sulfate
57 enrichment in atmospheric aerosol observed at the crow's nest may be due to ship exhaust rather
58 than presence of mirabilite. It follows that estimates of sea ice contributions to total SSA derived from
59 depletion factors discussed in section 3.4.4 have to be considered as lower bounds of true values.

60
61 **RC1:** 3. Chapter 3.2: Impact of snow precipitation on blowing/drifting snow: Did you access the
62 regular weather reports from the ships meteorological office in this case?

63 **Reply:** The available 3-hourly weather reports from the ship provided only limited information, but
64 confirmed for the case of precipitation shown in Fig.10 (3-4 July 2013) overcast skies, variable visibil-
65 ity from 0.5 (fog) to 10 km, and occasional ice needles. As stated in the text we inferred occurrence of
66 precipitation qualitatively from direct observation supported by webcam images, if usable, and pres-
67 ence of clouds (p9-114). The still images of a webcam installed at the crows nest (p8-116) allowed to
68 see at times, including 4 July, large airborne snow crystals during night time in the beam of the ships
69 search lights. We added a sentence in section 3.3.2 to further clarify.

70 For the early morning of 4 July 2013 webcam images from the crow's nest confirmed the presence of
71 large airborne snow crystals visible during darkness in the beam of the ship's search lights, whereas
72 the ship's 3-hourly weather report noted the presence of airborne ice needles.

73
74 **RC1:** 4. Pages 12/13 and Fig.7: Regarding the salinity (S_p) of blowing snow, corresponding S_p -
75 values of the uppermost surface snow layer are decisive. Did you take samples from surface snow;
76 say <1 cm deepness below surface? Figure 7: The reader cannot get an idea about the salinity of
77 the surface snow layer from this graph. It would be informative as well to specify the total depth of
78 the snow layer shown here, not just the snow height above sea ice.

79 **Reply:** Typically snow pit profiles were measured at 2 cm depth resolution (see methods section 2.4);
80 except at ice station S6 some profiles include a surface snow sample from a layer of ~0.5-1.0 cm
81 thickness. As discussed, S_p in blowing snow is consistent with the local surface snow measurements
82 (section 3.2.2). In Figure 7 depth information is readily available since data points at the top of each
83 profile, i.e. with the largest snow height above ice, represent the surface snow layer. We updated
84 Figure 7 and caption accordingly (Fig. 1).

85
86 **RC1:** 5. Chapter 3.3.2, Snow particle size distribution: Is it possible to rate the impact of the ships
87 profile on the local wind field and eventually on the measured snow particle size distribution?

88 **Reply:** We did not attempt to quantify the distortion of the local wind field by the ship and its impact
89 on measured snow particles, and had therefore included a respective caveat in the method descrip-
90 tion (p4 line 30): "It should be borne in mind that the distortion of flow caused by the ship may mean
91 both that speed at 39 m is not representative of flow in the far field at that height, and further, the tur-
92 bulent field strength, which governs the gradient of the logarithmic profile, may be a residual from a
93 different, likely lower, height. Thus, we suggest care when interpreting the data, and estimate that the
94 conversion from particle counts to number density be seen as an estimate suitable for comparison,
95 rather than quantitative with a well behaved uncertainty."

96
97 **RC1:** 6. Chapter 3.4.1, page 19, lines 28-30: As for Antarctic winter, acid induced Cl^- loss is rather
98 extraordinary because production of acidic sulphur compounds usually cease at the end of summer
99 / fall. Are there any indications for alternative HNO_3 induced Cl^- loss in your data?

100 **Reply:** We agree. Sea salt reaction with atmospheric HNO_3 is a plausible alternative chloride loss
101 process in winter; e.g. at Halley on the nearby Antarctic coast observations in winter show low but
102 non-zero levels of atmospheric HNO_3 of 1-2 pptv (Jones et al., 2011). Unfortunately, no usable filter
103 data of aerosol nitrate are available from this study to test the suggested process due to a very high
104 lab procedure blanc. However, we include the point in section 3.4.1 as follows.

105 Snow on sea ice follows closely the theoretical mirabilite fractionation line, whereas aerosol shows
106 large scatter and a tendency to apparent Na^+ enrichment with respect to Cl^- of up to 20 %, equivalent

107 to Cl^- depletion with respect to Na^+ of 17% (Fig. 14). Dechlorination of sea salt aerosol observed
108 in Antarctica has a maximum in spring/summer, when gaseous acidic species (nitric, sulfuric and
109 methanesulfonic acid) are available to replace chloride on sea-salt aerosol (Wagenbach et al., 1998;
110 Rankin and Wolff, 2003; Legrand et al., 2017). Acidic sulphur species are close to zero during winter
111 in coastal Antarctica e.g. at Neumayer (Weller et al., 2011), whereas nitric acid is low but non-zero,
112 e.g. 1-2 pptv at Halley (Jones et al., 2011). Thus nitric acid induced Cl^- loss from sea salt is a plausible
113 explanation for the observed Cl^- depletion either in airborne SSA or as a sampling artefact from
114 sea salt already accumulated on the filter surface as suggested previously (Wagenbach et al., 1998;
115 Legrand et al., 2017). Unfortunately no usable filter data of aerosol nitrate are available from this
116 study to further test the association between nitrate and sea salt due to a very high lab procedure
117 blank.

118
119 **RC1:** 7. Chapter 3.4.2, Chemical fractionation of Br^- , lines 28-33 and Fig. 15: There is strong
120 bromine depletion during polar night in July when global radiation was about zero (Fig. 15b). This
121 peculiarity deserves some discussion.

122 **Reply:** We did mention (p21-line16) that bromide escape from aerosol was detected previously year-
123 round at DDU in coastal Antarctica, including during winter months, except in June (Legrand et al.,
124 2016). As suggested, we expand the discussion in section 3.4.2:

125 Contrary to expectation bromide depletion of aerosol was significant even during winter darkness
126 from mid June to mid July (Fig. 15b), whereas previous observations at DDU showed a similar trend
127 but less bromide depletion and none in June (Legrand et al., 2016). At DDU DF_{Br^-} in bulk aerosol
128 increased gradually from a minimum in June (0.04), intermediate values in July to Sep (0.22-0.39) to
129 a maximum in October (0.42) (Legrand et al., 2016). Light conditions are unlikely a cause of differ-
130 ences in bromide depletion, since DDU is located at a similar latitude ($66^\circ 40'S$) as the area covered
131 by this study. However, one of a number of processes identified leading to bromide loss from snow
132 or aerosol involves HOBr oxidation of bromide, which leads to its autocatalytic release (Abbatt et al.,
133 2012). The early laboratory study by Oum et al. (1998) has shown that the required HOBr can be
134 chemically produced in darkness through the reaction of ozone with bromide. Another study during
135 the ANT-XXIX/6 expedition reports significant bromoform (CH_3Br) production in sea ice during winter
136 darkness (Abrahamsson et al., 2018), which requires HOBr (and organic matter) as precursors, and
137 therefore indicates that bromine loss processes were active in the sea ice in the absence of sunlight.
138 It therefore appears plausible that the same reactions may have caused significant bromide depletion
139 observed here in sea salt aerosol, provided the aerosol pH was low enough.

140
141 **RC1:** 8. Figure 5 and page 12, lines 8-9: By the way: During late afternoon of the 11 July, there
142 is an outstanding Na^+ peak associated with corresponding sulphate depletion, while the wind speed
143 seemed just close to the threshold value (well below 10 m/s throughout the whole day). Any ideas?

144 **Reply:** From midnight to the early morning of 11 July 2013 wind speed was indeed at or slightly
145 above the snow drift threshold ($= 7.1 \text{ m/s}$) (Fig.5a) suggesting that drifting snow near the surface was
146 present and after sublimation contributed to the observed sodium peak. The increase in SSA num-
147 ber densities and atmospheric sodium occurred in the afternoon a few hours after wind speed had
148 dropped again below the threshold, consistent with a similar phasing observed during the blowing
149 snow event on 14-16 July and discussed in section 3.2.2. To better illustrate episodes of snow drift
150 we include in Figures 4-6 of the revised manuscript a horizontal line marking the estimated threshold
151 wind speed U_t .

152

153 **RC2:** *Frey et al present an observational study of sea salt aerosol (SSA) production from blowing*
154 *snow above sea ice, through measurements during winter 2013 in the Weddell Sea, Antarctica.*
155 *Since the modelling hypothesis presented by Yang et al (2008, GRL), the mechanism of SSA produc-*
156 *tion from blowing snow has been implemented in numerous modelling studies, unfortunately without*
157 *observational evidence of the mechanism itself. This work provides a detailed study of the pro-*
158 *posed mechanism through measurements of size distributions and inorganic chemical composition*
159 *of aerosols and blowing snow, and comparisons to modelled parameters of blowing snow SSA pro-*
160 *duction. Given the prevalence of the use of the blowing snow SSA production parameterisation, this*
161 *is a very valuable study.*

162 **Reply:** We thank the reviewer for the positive assessment.

163
164 **RC2:** *My comments mainly focus on clarification of the manuscript and assessment of statistical*
165 *significance throughout. Given the significant length and many figures and tables, the authors are*
166 *encouraged to consider moving some material to a supplementary information file if appropriate.*

167 **Reply:** We consider tables and figures all essential, in agreement with reviewer 1. However, addi-
168 tional material as suggested is now included and presented in a supplement (see below).

169
170 **RC2:** *One overarching and major comment that needs to be addressed throughout the manuscript*
171 *is for uncertainties (or standard deviations) to be listed with average values. This is important for as-*
172 *sessing data variability, as well as for assessment of statistical significance. Indeed, statistical tests*
173 *of significance should be applied to inform whether 'trends' and 'differences' are indeed statistically*
174 *significant, which would greatly strengthen the findings presented in the manuscript. This is impor-*
175 *tant because trends sometimes seem to be overstated in the text when compared to large scatter*
176 *shown in the figures. Routine statements of statistical significance would significantly strengthen the*
177 *conclusions throughout.*

178 **Reply:** We added standard deviations for all averages reported in the tables (Tables 1, 2) and include
179 in the final manuscript significance of trends and differences, where appropriate. As an example be-
180 low updated text in section 3.4.4.

181 *During storms median atmospheric sea salt concentrations from both estimates showed increases*
182 *above background values (Fig. 17a) that were statistically significant based on the Wilcoxon rank-*
183 *sum test ($p < 0.01$).*

184
185 **RC2:** *I highly recommend reorganising the manuscript to improve readability. Section 3.2 relies sig-*
186 *nificantly on depletion factors. Therefore, I recommend reorganising to move Sections 3.4.1-3.4.3 to*
187 *be before Section 3.2. Also, the current Section 3.4.3 would be best after Section 3.3.*

188 **Reply:** We are considering this suggestion for the final manuscript.

189
190 **RC2:** *Major Comments: Page 1, Line 21 & Page 25, Lines 14-15: These sentences state generally*
191 *that 'similar processes take place in the Arctic', yet no supporting discussion is provided. Since the*
192 *current work focuses on the specific conditions of the Antarctic work and no data are provided to*
193 *evaluate this statement, these sentences should be removed.*

194 **Reply:** Agreed. We removed the sentence referring to the Arctic from abstract and conclusions, and
195 added text in the conclusions as follows:

196 *Similar in situ measurements are needed to corroborate the importance of sea salt aerosol produc-*
197 *tion from blowing snow also in the Arctic to validate atmospheric and ice core models (e.g. Rhodes*
198 *et al., 2017; Huang and Jaeglé, 2017).*

199
200 **RC2:** *Page 1, Lines 2-3 and Page 3, Lines 5-7: The statement 'validating a model hypothesis to*
201 *account for winter time SSA maxima in polar regions not explained otherwise' generalises beyond*
202 *the Antarctic, which is not appropriate, and it also not consider other factors, such as lower bound-*
203 *ary layer height and lead-based SSA production. This statement should be rephrased to focus on*
204 *validating wintertime SSA production from blowing snow (which is excellent), as a comprehensive*
205 *discussion of wintertime SSA maxima causes in both the Arctic and Antarctic is not presented in*

206 *this work. Further, the work of Huang and Jaegle (2017) did not consider the observed influence of*
207 *lead-based SSA production in the Arctic (May et al. 2016, JGR). I suggest focusing on the Antarctic,*
208 *as this is the strength of this work.*

209 **Reply:** We agree and rephrased in abstract (1.) and introduction (2.) accordingly.

210 1. Two consecutive cruises in the Weddell Sea, Antarctica, in winter 2013 provided the first direct
211 observations of sea salt aerosol (SSA) production from blowing snow above sea ice, thereby validat-
212 ing a model hypothesis to account for winter time SSA maxima in the Antarctic. 2. Indeed, model
213 agreement with SSA winter maxima observed at a number of locations in the polar regions is much
214 improved when a SSA source from blowing snow based on the parameterisation of (Yang et al., 2008)
215 is included in the model (Huang and Jaeglé, 2017; Yang et al., 2019).

216
217 **RC2:** *Figure 1; Page 3, Lines 30-33; Page 13, Lines 22-23: Please provide a legend for sea ice con-*
218 *centration. It appears that stations S2, S3, and S9 were in areas of reduced sea ice concentration.*
219 *While there is significant evidence for blowing snow SSA production based on chemical analyses, a*
220 *discussion of the distance to open leads, in addition to open water (Page 3, Line 32), needs to be*
221 *included, since there is measurement evidence of wind-dependent lead-based SSA production (e.g.,*
222 *Nilsson et al. 2001, JGR).*

223 **Reply:** A legend is now included in Fig. 1 and we added the following text in section 2 (1.) and in the
224 discussion p13 - after line26 (2.).

225 1. Sea ice concentrations in mid July 2013 derived from Nimbus-7 satellite microwave radiometer
226 measurements (Comiso, 2018) show areas with 85-95 % ice cover near ice stations S2-3 and S7-9
227 indicating that open leads may be present (Figure 1). 2. Open leads, which may have been present
228 in areas of reduced sea ice concentration e.g. near ice stations S2-3 and S7-9 (Figure 1) are another
229 potential wind-dependent source of SSA from open water as observed in the Arctic (Nilsson et al.,
230 2001; May et al., 2016), albeit with a much smaller flux contribution per surface area compared to the
231 open ocean due to reduced fetch and low fraction of surface coverage (<15%).

232
233 **RC2:** *Page 7, Lines 3-5: Please clarify whether these time periods of ship exhaust influence were*
234 *also removed from the aerosol size distribution data, as they should be.*

235 **Reply:** Ship exhaust influence on measurements of aerosol size and concentration was removed by
236 using a wind-sector filter (section 2.2). We clarified text in section 2.2 including also the fraction of
237 CLASP data filtered out as follows (see corresponding reply to RC1):

238 Raw aerosol number concentrations at the crow's nest showed significant spikes, when air came from
239 the direction of the ship's engine stack, whereas no evidence of pollution was detected in the obser-
240 vations on the sea ice. Pollution spikes were effectively filtered out prior to averaging by excluding all
241 data when relative wind direction was in the 135–225° sector encompassing the ship's engine stack.
242 A total of 21% of the available 1-second data was removed from the crow's nest data.

243
244 **RC2:** *Page 7, Line 9 and Table 3: LODs are normally defined as 3*sigma, rather than 2*sigma. What*
245 *is the authors justification here? Also, LODs should be reported with one significant figure (too many*
246 *shown in Table 3, which can be misleading).*

247 **Reply:** Here we follow Wagenbach et al. (1998) who employed a similar aerosol filter method and de-
248 fined the mean detection limits as 2*sigma. Two figures for LOD were reported in Table 3 to account
249 for increased LOD at the shorter run times of filters deployed on the sea ice. However, we removed
250 that line to report only one figure for LOD and clarified the footnote of Table 3 as follows.

251 ^cbased on crow's nest mean air sample STP-volume (6.4 m³); mean air sample STP-volume for filters
252 deployed on the sea ice was 3.3 m³ increasing respective LODs by a factor 1.6

253
254 **RC2:** *Tables 4-5: Data below the LOD should be labeled as such, as exact values below LOQs are*
255 *not meaningful.*

256 **Reply:** Agreed. Snow concentrations (Tab. 2) were typically 2 orders of magnitude above the LOD
257 of ~2 ng g⁻¹, whereas some aerosol concentrations (Tab. 1) were below the estimated LOD. In the
258 final manuscript a corresponding footnote is added to those values in Table 4:

259 °below the estimated LOD (see Table 3)

260

261 **RC2:** Page 8, Lines 3-5: *Instead of reporting depletion factors, I highly encourage the authors to*
262 *consider reporting 'enrichment factors' (e.g. Krvanek et al. 2012, Atmos. Environ.), which are more*
263 *intuitive to understand in my opinion (i.e. enrichments are >1, depletion corresponds to <1).*

264 **Reply:** Deviations from bulk sea water ion ratios are reported in the literature in both ways, either
265 as enrichment or as depletion factors (e.g. Sander et al., 2003). Since the focus here is on depletion
266 processes we choose to report depletion rather than enrichment factors, also to be consistent with
267 some of the previous related work (Yang et al., 2008). To help interpretation we added a sentence to
268 section 2.4:

269 For example, $DF_x = -1.5$ or 150% enrichment means the respective ion concentration is 2.5 times
270 that in reference sea water.

271

272 **RC2:** Page 8, Lines 8-11: *I am quite concerned that data were selectively removed from the datasets*
273 *presented. I can understand if certain samples are not used for externally identified reasons, but if,*
274 *for example, sulfate concentration is removed for a given sample, I'm concerned about continuing*
275 *to use other ions from that sample, as appears to have been done based on the numbers shown in*
276 *Tables 4 and 5. I worry that the presented datasets are skewed based on the removal of these data*
277 *points. What fraction of the time did ship emissions impact the dataset? It needs to be clarified what*
278 *fraction of the data were removed. This data treatment is very important for later statements about*
279 *the distribution of depletion factors (e.g., statements on Page 10, Lines 7-9).*

280 **Reply:** No snow data were removed whereas the fraction of aerosol filter data removed was relatively
281 small, and is now mentioned in the revised text. Filter samples suspected of contamination based
282 on anomalous sulfate enrichment (total of 6 samples) are not anymore used in the statistics. The
283 pollution impact on filter chemistry is now discussed (see reply to RC1 above). Bromide depletion
284 factors below a threshold of -7 are considered outliers and removed. The corresponding statistics in
285 Table 4 are updated. Follow up statements are not affected by any of these changes.

286 A total of 6 (= 6% of all crow's nest samples) $DF_{SO_4^{2-}}$ values were below that of pure mirabilite (=
287 -7.3) and are attributed either to sulfate contamination from the ship's engine emissions discussed
288 below or measurement error. We therefore removed all ion concentrations of the corresponding filter
289 samples from the dataset. DF_{Br^-} only below -7 were considered outliers due to measurement error
290 and removed: a total of 4 (= 3% of all samples) from the crow's nest data, and a total of 6 (= 14% of
291 all samples) from the sea ice data.

292

293 **RC2:** Page 9, Lines 28-30; Page 10, Lines 1-3: *Please reference where these data are presented,*
294 *or please add them as supplementary information.*

295 **Reply:** Agreed. In a supplement we include now a Figure S1 (Fig. 2) with an overview of the available
296 observations during ANT-XXIX/7, and Table S1 and Table S2 (Table 3, 4) with the statistics of particle
297 concentration and size. The text has been amended as follows:

298 1. At 29 m mean total number densities N_{46-478} were $8.7 \times 10^3 \text{ m}^{-3}$ during ANT-XXIX/6 and very similar
299 $7.2 \times 10^3 \text{ m}^{-3}$ during ANT-XXIX/7 (Table S1, Figure S1c).

300 2. At 29 m mean total number densities $N_{0.4-12}$ were $2.1 \times 10^6 \text{ m}^{-3}$ during ANT-XXIX/6 (Table S2, Fig-
301 ure 2d). $N_{0.4-12}$ mean values at 2.0 and 0.2 m during ice stations were 1.4×10^6 and $1.7 \times 10^6 \text{ m}^{-3}$,
302 respectively, about the same as the number densities observed during the same time at 29 m (Ta-
303 ble S2). The median aerosol particle diameters \bar{d}_p at the measurement heights 0.2, 2.0m and 29 m
304 ranged between 0.60 and 0.66 μm (Table S2) showing dominance of sub-micron sized particles in
305 atmospheric aerosol below the instrument particle size cut-off (>11 μm).

306 3. Median $DF_{SO_4^{2-}}$ values at 29 m were very similar during ANT-XXIX/6 (=0.34) and ANT-XXIX/7
307 (=0.30), but larger near the sea ice surface (=0.49), suggesting throughout a significant contribution
308 to the total SSA burden from a fractionated sea ice source (Table 4, Figure S1e).

309

310 **RC2:** Section 3.4.2 and associated text in Conclusions: *The authors should be mindful that only*
311 *aerosol and snow bromine were measured and that no measurements of reactive bromine are pre-*

312 *sented. Therefore, the strength of the implications for reactive bromine production should be weak-*
313 *ened to account for this uncertainty and other factors that contribution to reactive bromine production*
314 *and abundance.*

315 **Reply:** Indeed, we do not infer any details on speciation of reactive bromine chemistry. We added a
316 sentence in section 3.4.2 (1.) and amended a sentence in conclusions (2.):

317 1. Detailed measurements of participating bromine species in air, snow and aerosol are needed to
318 further understand relevant processes and constrain the mass budget.

319 2. It is found that SSA produced by blowing snow is depleted in bromide suggesting it is a source of
320 reactive bromine to the atmosphere, which then can contribute to ozone depletion events.

321
322 **RC2:** *Page 21, Lines 9-10: Depletion factors examine the degree of depletion, but they do not provide*
323 *information on the mass present. Therefore, the data here cannot assess contribution to the fraction*
324 *of net bromine release, as currently presented, especially without reactive bromine measurements.*

325 **Reply:** Agreed, we don't discuss detail of the bromine mass budget. We amended the correspond-
326 ing sentence in section 3.4.2, conclusions and abstract (1.), as well added a note (2.) (see reply to
327 previous comment):

328 1. On average snow on sea ice and blowing snow showed no or small depletion of bromide relative
329 to sodium with respect to sea water, whereas aerosol at 29 m was depleted suggesting that signifi-
330 cant bromine loss takes place in the aerosol phase between 2 and 29 m above the sea ice surface.

331 2. Detailed measurements of participating bromine species in air, snow and aerosol are needed to
332 further understand relevant processes and constrain the mass budget.

333
334 **RC2:** *Page 19, Lines 22-25: This analysis is only valid if you assume there is no precipitation of*
335 *NaCl.2H₂O. Please verify that based on temperature, and perhaps take out the very low temperature*
336 *points.*

337 **Reply:** A complete model of freezing seawater is beyond the scope of this study. Thus we acknowl-
338 edge that precipitation of NaCl.2H₂O introduces some uncertainty to this analysis by adding the
339 sentence below.

340 Further Na⁺ depletion may arise from the precipitation of hydrohalite (NaCl·2 H₂O) once ambient tem-
341 perature drops below the threshold of -22.9 °C (e.g. Butler et al., 2016), which occurred here during
342 some periods of time (Fig.2b). In the analysis below however we consider only the precipitation of
343 mirabilite.

344
345 **RC2:** *Page 19, Lines 127-28: Does this also mean that the aerosols collected were a mixture of sea*
346 *salt emitted from the ocean and sublimation of blowing snow?*

347 **Reply:** Mixing with a pool of non-fractionated sea salt aerosol from the open ocean ($DF_{Na^+}, DF_{SO_4^{2-}}=0$)
348 would move data points towards the origin in Figure 14, but would not explain apparent Na⁺ enrich-
349 ment or Cl⁻ loss in aerosol at a given SO₄²⁻ depletion. We believe a plausible explanation for the
350 deviation of aerosol observations from the mirabilite precipitation model is HNO₃ induced Cl⁻ loss
351 from sea salt either in airborne SSA or as an artefact on filters, as stated in the reply to reviewer 1.
352 Below we repeat the amended text.

353 Snow on sea ice follows closely the theoretical mirabilite fractionation line, whereas aerosol shows
354 large scatter and a tendency to apparent Na⁺ enrichment with respect to Cl⁻ of up to 20 %, equivalent
355 to Cl⁻ depletion with respect to Na⁺ of 17 % (Fig. 14). Dechlorination of sea salt aerosol observed
356 in Antarctica has a maximum in spring/summer, when gaseous acidic species (nitric, sulfuric and
357 methanesulfonic acid) are available to replace chloride on sea-salt aerosol (Wagenbach et al., 1998;
358 Rankin and Wolff, 2003; Legrand et al., 2017). Acidic sulphur species are close to zero during winter
359 in coastal Antarctica e.g. at Neumayer (Weller et al., 2011), whereas nitric acid is low but non-zero,
360 e.g. 1-2 pptv at Halley (Jones et al., 2011). Thus nitric acid induced Cl⁻ loss from sea salt is a plausi-
361 ble explanation for the observed Cl⁻ depletion either in airborne SSA or as a sampling artefact from
362 sea salt already accumulated on the filter surface as suggested previously (Wagenbach et al., 1998;
363 Legrand et al., 2017). Unfortunately no usable filter data of aerosol nitrate are available from this
364 study to further test the association between nitrate and sea salt due to a very high lab procedure

365 blank.

366

367 **RC2:** Page 22, Lines 32-33: A conversion factor is used to calculate [SSA] based on Na⁺ and using
368 seawater composition, but this seems to undermine and not take into account the sulfate-depletion
369 observed.

370 **Reply:** It does not. The impact of the depletion due to mirabilite precipitation on our calculation is
371 indeed very small, and is therefore neglected. We added the text below to clarify:

372 As shown in section 3.4.1 depletion of SO₄²⁻ due to the precipitation of mirabilite decreases Na⁺ by up
373 to 12%. Reduction in both ions decreases the mass fraction of Na⁺ in the depleted sea salt aerosol
374 by a maximum of ~0.7% compared to reference seawater. Thus, by not considering the depletion
375 effect conversion factor and calculated SSA mass are underestimated by up to ~0.7%, which is neg-
376 ligible given all other uncertainties.

377

378 **RC2:** Page 21, Lines 30-31 and elsewhere: Is this U_{10m} and the associated data in Fig 16 an aver-
379 age, or threshold? It isn't clear how the data were binned. Please clarify calm and stormy conditions.
380 Does calm represents U_{10m}<5 m/s? How about stormy?

381 **Reply:** We used a relatively narrow wind speed range for calm and windy conditions. We amended
382 this to include more data, particularly for the open ocean case when only a few days of measurements
383 were available. Aerosol data are now selected based on a wind speed threshold: calm conditions
384 when U_{10m} <4 m s⁻¹ and windy conditions when U_{10m} >9 m s⁻¹. We updated Figure 16, including also
385 the standard deviation of the mean, to show statistical significance of differences in size distributions,
386 as suggested further below (Fig. 3). And the text in section 3.4.3 is clarified as follows:

387 Average aerosol number density and volume distributions observed in the Weddell sea show that
388 during calm conditions (U_{10m}<4 m s⁻¹) concentrations across most of the size spectrum were smaller
389 above sea ice than above the open ocean (Fig. 16a). Depending on particle size the variability was
390 relatively large as illustrated by the standard deviation of the mean values (Fig. 16a). Thus differ-
391 ences in mean size distributions were statistically significant only for d_p <2 μm in the case of aerosol
392 number density, and d_p 1-8 μm in the case of aerosol volume distributions (Fig. 16b). The wind speed
393 threshold chosen for calm conditions is well below the mean snowdrift threshold wind speed U_t
394 of 7.1 m s⁻¹ observed during this study and within the range when breaking of waves commences (3-
395 4 m s⁻¹; O'Dowd et al., 1997). ... During stormy conditions (U_{10m}>9 m s⁻¹) average aerosol number
396 densities above sea ice increased significantly for particle diameters d_p<2 μm, reaching at the lower
397 end of the size spectrum levels similar to those observed above the open ocean (Fig. 16a). Average
398 aerosol volume concentrations above sea ice also showed an increase during storms, significant for
399 particle sizes d_p 0.8 to 9 μm (Fig. 16b).

400

401 **RC2:** Page 22, Lines 16-19: It seems "not all water is lost" could represent a large uncertainty of blow-
402 ing snow sublimation. This is important for reactions that depend on the surface area of aerosols.
403 It could be highlighted in the abstract or conclusion. Also, please justify how to get 10⁻³ μm. Using
404 snow salinity of 0.06 psu from Table 5, median snow particle of 100 μm from Table 6, yields d(dry) of
405 1 μm.

406 **Reply:** We agree the degree to which water ice is lost on particles during sublimation has implica-
407 tions for heterogeneous chemistry, something future experiments will need to address; text below has
408 been added to the conclusions.

409 The degree of water ice loss from particles has implications for particle surface area and heteroge-
410 neous chemistry, which future experiments will need to address.

411 **Reply:** We disagree regarding the calculation of d_{dry}: to convert S_p from psu (equivalent to g of
412 dissolved salt per kg of sea water as defined in Section 2.4) into units of kg per kg in order to be
413 consistent with units of density (kg of salt per m³ of salt) requires division by one thousand as the
414 equation states (Page 22, Line 16), correctly yielding d_{dry} of ~1 nm.

415

416 **RC2:** Page 23, Lines 1-3: Please show this comparison and data in a supplementary file.

417 **Reply:** We included Figure S3 in the supplement to show the comparison (Fig. 4), and amended the

418 sentence as follows:

419 The sea salt mass estimates show that most filter-based values have a low bias compared to median
420 sea salt concentrations derived from $N_{0.4-12}$ during filter sampling intervals (Fig. S3), on average of
421 $\sim 26\%$. The bias shows also a weak but significant positive correlation wind speed ($R=0.4$, $p<0.01$)
422 (Fig. S3). A low bias of the filter samples especially during high wind speeds is expected because
423 the smaller cut-off diameter ($<6\ \mu\text{m}$) compared to the optical particle counter ($>11\ \mu\text{m}$) limits capture
424 of coarse sea salt aerosol, where much of the particle mass is located (Fig. 16b).

425
426 **RC2:** *Page 25, Lines 5-10: This is not a new finding and has been presented in other work. There-*
427 *fore, either these sentences should be removed here or other work should be referenced to further*
428 *support these findings.*

429 **Reply:** Presenting the links between snow salinity, differences in sea ice age and SSA source
430 strength of blowing snow together with direct observations is of course new. However, we refer-
431 ence relevant work on sea ice and snow on sea ice as follows:

432 - at a given salt migration distance from the sea ice surface it is total snowpack depth, that determines
433 the salinity probability distribution of snow on sea ice consistent with previous studies (Domine et al.,
434 2004; Massom et al., 2001). FYI can therefore be distinguished from MYI based on snow salinity, be-
435 cause snow on FYI is in general more shallow than on MYI. Secondary factors potentially increasing
436 the difference in salinity between FYI and MYI and identified previously (e.g. Massom et al., 2001)
437 are more frequent flooding of FYI with seawater due to negative freeboard and MYI desalination due
438 to brine drainage.

439
440 **RC2:** *Data Availability: Since the current work is expect to be very valuable for informing future mod-*
441 *elling work and other studies, I highly encourage the authors to put these data in a public archive.*

442 **Reply:** All data from this study used are stored in the UK Polar Data Centre. The DOI is provided in
443 the final manuscript.

444 All data are stored in the UK Polar Data Centre, Natural Environment Research Council, UK Research
445 and Innovation (<https://doi.org/10.5285/853dd176-bc7a-48d4-a6be-33bcc0f17eeb>, Frey et al., 2019).

446
447 **RC2:** *Figure 7: Please add a legend to give meaning to the colors presented. Also, it is stated*
448 *throughout the manuscript that the surface snow is typically significantly sulfate depleted (justifying*
449 *the sea ice source for sulfate-depleted aerosol), but here the surface is more often near 0. Please*
450 *clarify.*

451 **Reply:** Figure 7 now includes a legend (Fig. 1). There is significant spatial heterogeneity in the
452 sampled local snowpack profiles, whereas blowing snow integrates over a wider area of sea ice. We
453 clarified the discussion of the snow pit observations (Page 13 - Lines 9-14) as follows:

454 $DF_{SO_4^{2-}}$ profiles exhibited large scatter: except at one location surface-near snow showed no or small
455 depletion, whereas most profiles showed significant depletion in deeper layers within 5-10 cm of the
456 sea ice surface (Fig. 7c). ... However, the $DF_{SO_4^{2-}}$ values of blowing snow were at the top end of the
457 range observed only in the deeper and more saline local snowpack (Fig. 6c). A plausible explanation
458 for this observation during the storm on 15 July is that blowing snow integrates snow contributions
459 from a wider area. And given the spatial heterogeneity of local snowpack thickness and composition
460 blowing snow contributions must have dominated from areas where fractionated snow was at or near
461 the surface such as seen in one of the profiles sampled on 12 July (Fig. 7c).

462
463 **RC2:** *The highly relevant work of Giordano et al. (2018, ACP) 'The importance of blowing snow*
464 *to halogen-containing aerosol in coastal Antarctica: influence of source region versus wind speed'*
465 *should be considered in this manuscript.*

466 **Reply:** We agree and correct the oversight by referring to this work in the introduction:

467 A recent observational study in the Ross Sea sector of coastal Antarctica also shows a significant
468 association between increased SSA and high wind speed suggesting a link to blowing snow above
469 sea ice as a source (Giordano et al., 2018).

470

471 **RC2:** *Minor/Technical Comments: Throughout the manuscript, watch for 'paragraphs' that are only*
472 *1-2 sentences, as this disrupts the flow and limits discussion. Consider reorganization to prevent this.*
473 **Reply:** We reorganised, where appropriate.

474
475 **RC2:** *Page 1, Line 9: Please state the size of the sulphate-depleted aerosol.*
476 **Reply:** Done.
477 Similar depletion in bulk aerosol observed in the 1-6 μm range suggests that most sea salt originated
478 from snow on sea ice and not the open ocean or leads, e.g. on average $\sim 93\%$ during the 8 June and
479 12 August 2013 period.

480
481 **RC2:** *Page 1, Line 13: Based on the data presented later, 'enriched' is likely a typo and should be*
482 *'depleted' here with respect to aerosol at 29 m.*
483 **Reply:** This is now corrected (see reply to reviewer 1 above).

484
485 **RC2:** *Page 2, Line 20: Provide a reference to a SSA review here.*
486 **Reply:** The reference below is now included.
487 de Leeuw et al. (2011)

488
489 **RC2:** *Page 4, Lines 27-28: I think it is dividing kappa instead of multiplying. Please check. Also,*
490 *please provide the value for the von Karman constant in parentheses.*
491 **Reply:** Corrected as follows:
492 To do this a logarithmic wind profile $U(z)$ is assumed given by $U(z) = u_*/\kappa \ln(z/z_0)$ (e.g. Li and Pomeroy,
493 1997), with measurement height z , the von Karman constant κ ($= 0.4$), friction velocity u_* and the
494 surface roughness length of momentum z_0 set to 5.6×10^{-5} m as measured very consistently above
495 snow at Halley (King and Anderson, 1994).

496
497 **RC2:** *Page 5, Lines 12-14: Please provide a greater description of the inlet. Also, please clarify*
498 *whether the data presented were corrected for these particle loss estimates ('we adopt' is confusing*
499 *phrasing).*
500 **Reply:** Clarified as follows.
501 Particle losses to inlet walls are minimised by using a short and straight inlet tube of 0.3 m length sim-
502 ilar to the original configuration (Hill et al., 2008, Figure 9). We assume as an upper limit of particle
503 losses those estimated previously for a similar inlet configuration (Norris et al., 2012), which amount
504 to 43% at $d_p = 11.32 \mu\text{m}$, 19% at $d_p = 6.06 \mu\text{m}$ and 0.1% at $d_p = 0.44 \mu\text{m}$, respectively.

505
506 **RC2:** *Page 5, Lines 22 and 27: Please clarify the size range of aerosol collected.*
507 **Reply:** Clarified as follows.
508 Filters were estimated to collect aerosol in the diameter range $\sim 0.3 \mu\text{m}$ to less than $6 \mu\text{m}$. The lower
509 end of the range is based on previous measurements of collection efficiencies of PTFE filters as a
510 function of particle size (Soo et al., 2016), whereas the upper end is based on the estimated cut-off
511 diameter described below.

512
513 **RC2:** *Page 9, Line 8: I assume the authors are discussion temperature in degrees Celsius, but this*
514 *needs to be stated.*
515 **Reply:** Added.
516 Near-zero or positive ambient temperatures T_a in degrees Celsius ...

517
518 **RC2:** *Page 9, Line 14: Where is the timing of the snowfall presented/shown?*
519 **Reply:** Only the timing of airborne snow particles is shown. Occurrence of precipitation is based on
520 3-hourly ship's weather reports and occasional webcam images. We rephrased accordingly.
521 Winter storms occurred frequently with wind speeds ranging between 10 and 20 m/s, occasionally
522 exceeding 20 m/s, and coincided often with snowfall based on the ship's 3-hourly weather report,
523 occasional webcam images and presence of clouds (data not shown).

524

525 **RC2:** Page 9, Line 22-23: Please provide a reference that connects the friction velocity with the
526 boundary layer conditions. Also, reference where these data are shown, or add to a SI.

527 **Reply:** We included Jacobson (2005) and Nishimura and Nemoto (2005) as references, as well as
528 a figure (Fig. 5) in the supplement showing the correlation between friction velocity u_* and horizontal
529 wind speed U .

530

531 **RC2:** Page 10, Line 16: Please clarify 'two 7-10 day long periods'. I'd suggest wording such as 'two
532 periods, one lasting 7 days and another 10 days', or similar.

533 **Reply:** Added as follows.

534 Two periods, one lasting 7 days and another 10 days, were chosen based on data coverage to dis-
535 cuss key features of observed blowing snow and associated SSA increases.

536

537 **RC2:** Page 11, Lines 3-4: Please provide concentrations in parentheses for context.

538 **Reply:** Clarified as follows.

539 Near the surface spectral number densities $N_{0.4-12}$ for particles with $d_p < 2\mu\text{m}$ during the storm on 24
540 June remained with 10^5 m^{-3} below those seen at 29 m (10^6 m^{-3}) likely due to scavenging of aerosol
541 by snow particles (Fig. 4d-e).

542

543 **RC2:** Page 13, Lines 16-17: The direct comparison of $N_{0.4-12}$ to $d_p < 2\mu\text{m}$ here is confusing since
544 these are different size ranges.

545 **Reply:** Clarified as follows.

546 Aerosol size spectra show that number densities of particles with size $d_p < 2\mu\text{m}$ increased during in-
547 dividual storms by 2-3 orders of magnitude above background levels.

548

549 **RC2:** Page 14, Line 15: Please define SWE (snow water equivalent?) and the 'saltation layer' (what
550 height?).

551 **Reply:** Amended as follows.

552 1. (mm day^{-1} snow water equivalent) 2. The saltation layer is a layer just above the snow surface
553 usually several centimetres thick (e.g. Déry and Yau, 1999).

554

555 **RC2:** Page 15, Line 3: What does '(0.001)' correspond to here? Please clarify.

556 **Reply:** Amended as follows.

557 ... when snow drift density μ right above the snow surface exceeds a critical value μ_c ($= 0.005\text{ kg}$
558 m^{-3}). For comparison a lower value of μ_c ($= 0.001\text{ kg m}^{-3}$) is also considered.

559

560 **RC2:** Page 15, Lines 6 and 11: Please clarify that U_t and u^*_t are calculated, not observed.

561 **Reply:** U_t and u^*_t are not calculated. Windspeed and snow particle number densities are both mea-
562 sured quantities; thus drift threshold wind speed is an observed quantity based on the combination
563 of two measurements (symbols in Figure 8) as opposed to modelled values (Eq 4). Similar for friction
564 velocity u^* . We added a sentence to clarify.

565 The observed threshold wind speed U_t and friction velocity u^*_t are the respective measurements at
566 the onset of drifting or blowing snow.

567

568 **RC2:** Page 15, Line 15: Please show how u^*_t values were calculated.

569 **Reply:** u^*_t is not calculated. See reply above.

570

571 **RC2:** Page 15, Line 32: Please define what you mean by 'minor' here. Please quantify.

572 **Reply:** We did not run the model but the model bias in absolute values will cancel out because ratios
573 are used (see Eq.2). Clarified as follows.

574 The model bias in q_{bsalt} is expected to cancel out in estimates of bulk sublimation rate Q_s (Eq. 2) and
575 therefore also of SSA production Q_{SSA} (Eq. 1) because the calculation uses not absolute values but
576 ratios of actual q_{bsalt} and its maximum q_{b0} .

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RC2: *Page 17, Line 13: Please delete 'have' typo.*

Reply: done

RC2: *Page 17, Line 3: Didn't mean d_p increase?*

Reply: Decrease is correct. Expected is a decrease of d_p with height above the surface snow particle source in the absence of snowfall due to gravitational settling.

RC2: *Page 19, Line 32: Do you mean 0.1204 here?*

Reply: This has been corrected.

RC2: *Page 20, Lines 8-10: The wording 'well established' should be removed, as the Yang et al papers are models based on a hypothesis rather than measurement based and this associated uncertainty should be noted.*

Reply: Agreed and clarified as follows.

Modelling studies suggest that sea salt may be an important source of atmospheric bromine species in the mid to high southern latitudes, and that SSA from blowing snow releases bromine (Yang et al., 2008, 2010) driving ozone depletion events observed during or after snow storms (Jones et al., 2009).

RC2: *Page 20, Line 27: Data in Table 5 are presented in $\mu\text{g g}^{-1}$. Please fix or clarify.*

Reply: Corrected as follows.

Median bromide concentrations in snow ranged between 0.07 and $0.18 \mu\text{g g}^{-1}$ (Table 5).

RC2: *Page 21, Line 11: Change "due a" to "due to a".*

Reply: Corrected.

RC2: *Page 21, Line 14: No data are presented examining the acidity of the surface snowpack.*

Reply: Agreed, pH of aerosol and snow was not measured. We therefore removed reference to acidity.

The bromine release from SSA produced by blowing snow may be more efficient because it has a large fraction of sub-micron sized particles (see section 3.4.3), and resides at the well ventilated top of the blowing snow layer.

RC2: *Page 23, Line 22: Delete extra "the".*

Reply: Corrected.

RC2: *Page 23, Lines 29-30: Remove "always" and replace with "often" to more appropriately reflect the data shown.*

Reply: Agreed and amended.

RC2: *Page 25, Line 27: "LL & MM"?*

Reply: Mentors who prefer to remain anonymous

RC2: *Figure 16: The variations in these distributions (e.g. standard deviations) should be shown.*

Reply: Agreed, we updated Figure 16 including the standard deviation of the mean values, and corrected the caption (Fig. 3).

RC2: *Figure 17: This figure is difficult to understand currently.*

Reply: We updated Figure 17 and clarified the caption (Fig. 6).

627 **SC1:** *This manuscript describes an interesting set of measurements and detailed analysis confirm-*
628 *ing the blowing snow as a significant source for sea salt aerosol in the vicinity of sea ice in coastal*
629 *Antarctica. We agree that this is an important result with significant implications for polar tropospheric*
630 *aerosol loadings and heterogeneous halogen chemistry. However, it would be helpful to both the au-*
631 *thors and readers of this article to refer to prior work also published in ACP showing similar results*
632 *from measurements taken on sea ice in the Ross Sea. Giordano et al., 2018 also clearly identifies*
633 *blowing snow on sea ice as a significant source of chlorine rich sea salt aerosol from online Aerosol*
634 *Mass Spectrometer measurements of aerosol composition, optical measurements of blowing snow*
635 *and interstitial aerosol concentrations and offline measurements of surface and blowing snow com-*
636 *position. The consistency between the results from observations using different techniques and on*
637 *opposite sides of the Antarctic continent further indicates the importance of this mechanism to the*
638 *overall Antarctic aerosol budget.*

639 *Lars Kalnajs and Peter DeCarlo*

640
641 *Reference: Giordano, M. R., Kalnajs, L. E., Goetz, J. D., Avery, A. M., Katz, E., May, N. W., Leemon,*
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643 *containing aerosol in coastal Antarctica: influence of source region versus wind speed, Atmos. Chem.*
644 *Phys., 18, 16689-16711, <https://doi.org/10.5194/acp-18-16689-2018>, 201*
645

646 **Reply:** We agree and apologise for the oversight of this interesting study (see also reply to RC2
647 above). We now refer to this work in the introduction:

648 [A recent observational study in the Ross Sea sector of coastal Antarctica also shows a significant](#)
649 [association between increased SSA and high wind speed suggesting a link to blowing snow above](#)
650 [sea ice as a source \(Giordano et al., 2018\).](#)

651

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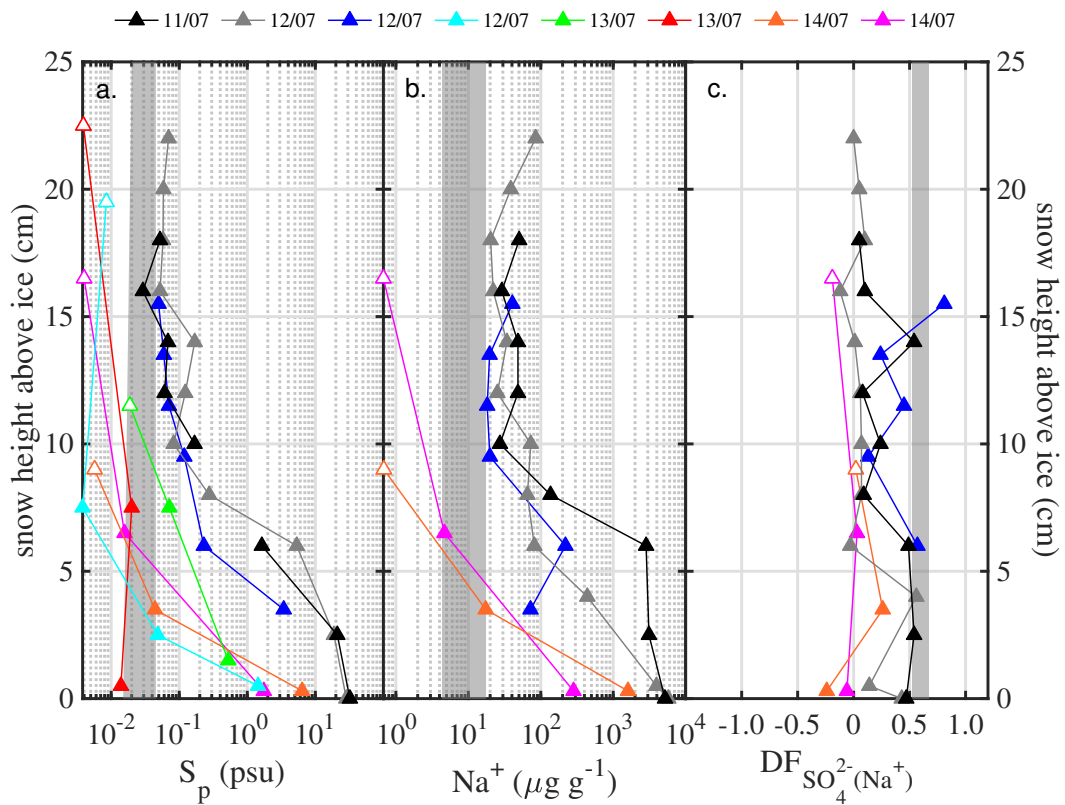


Figure 1: manuscript Figure 7 - Vertical snowpack profiles sampled at various locations on the ice floe of ice station S6 during the 11–14 July 2013 period (color indicates day of sampling): (a) salinity S_p , (b) Na^+ concentrations and (c) sulfate depletion factor $\text{DF}_{\text{SO}_4^{2-}}(\text{Na}^+)$ with respect to Na^+ as a function of snow height above the sea ice surface. Symbols illustrate averages for snow layers of 2 cm thickness, except those with white face color indicating 0.5-1.0 cm layer thickness. Data points at the top of each profile represent the surface snow layer, thus adding half the snow layer thickness to snow height yields total snowpack depth. Shaded areas illustrate the range of the respective parameter measured in blowing snow on 15 July 2013 (Fig. 6e-f).

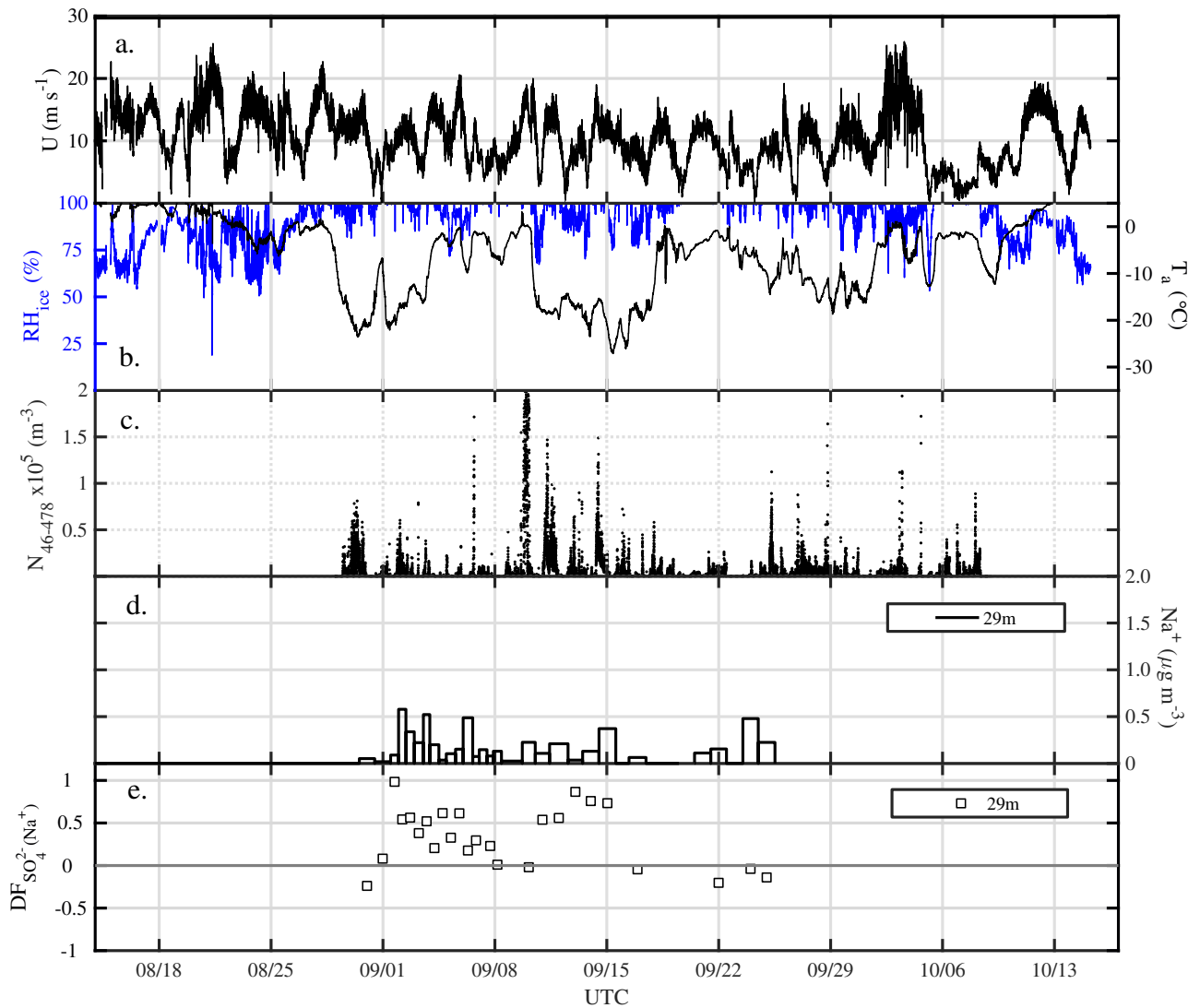


Figure 2: manuscript Figure S1 - Overview of atmospheric observations in the Weddell Sea from 14 August to 17 October 2013 (ANT-XXIX/7): (a) horizontal wind speed U at 39 m. (b) ambient temperature T_a and relative humidity with respect to ice RH_{ice} at 29 m. (c) total number densities N_{46-478} of airborne snow particles at 29 m. (d) aerosol Na^+ concentrations and (e) sulphate depletion factor $DF_{SO_4^{2-}}$, both at 29 m.

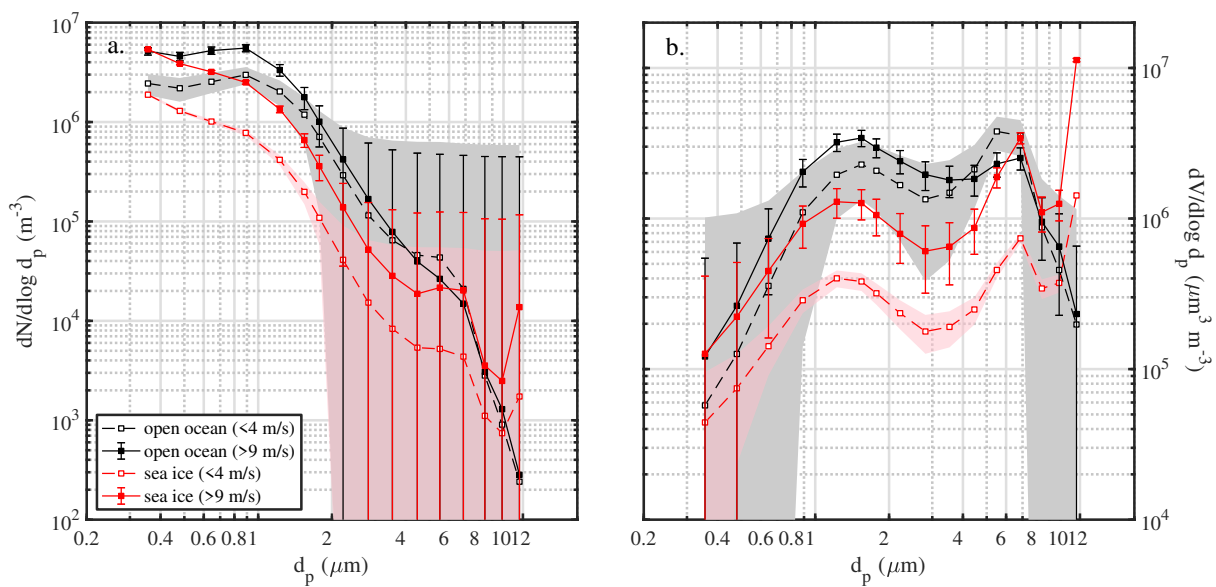


Figure 3: manuscript Figure 16 - Comparison of mean number distributions (panel a) and volume distributions (panel b) of aerosol above the open ocean (13 to 16 June 2013) and sea ice in the Weddell Sea (18 June to 21 July 2013) during calm ($U_{10m} < 4 \text{ m s}^{-1}$) and windy ($U_{10m} > 9 \text{ m s}^{-1}$) conditions. Shaded areas and error bars show the standard deviation of the mean during calm and windy conditions, respectively. Data included are observations from 29 m above the sea surface at ambient RH .

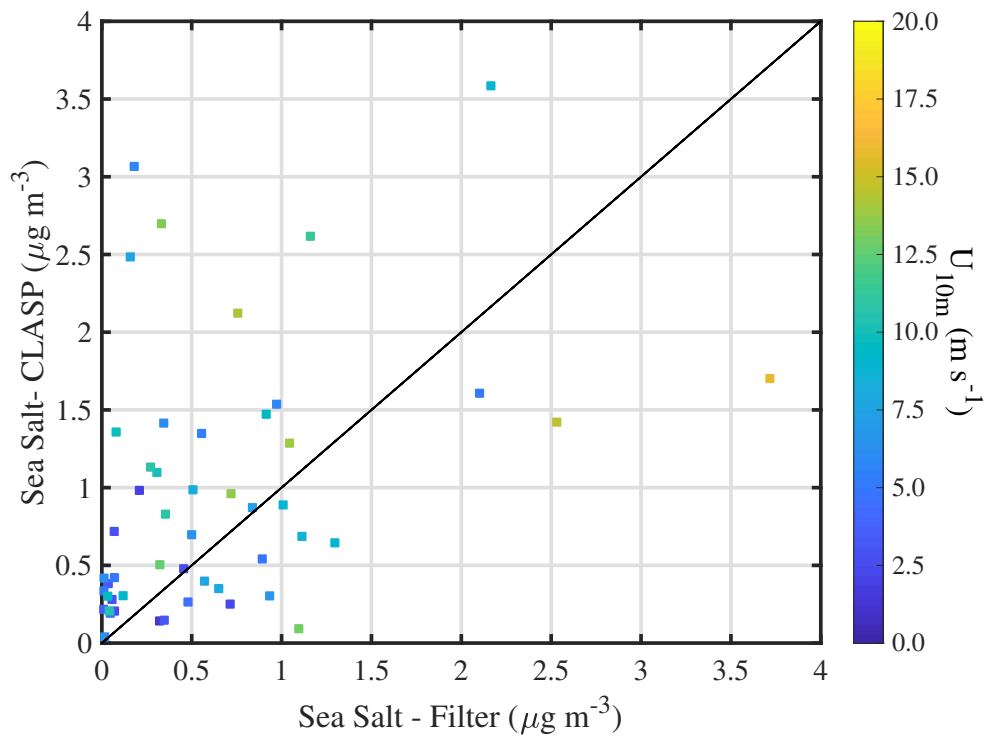


Figure 4: manuscript Figure S3 - Comparison of atmospheric sea salt concentrations during the 8 June to 26 July 2013 period derived from filter measurements and from median number densities $N_{0.4-12}$ measured with the CLASP during filter sampling intervals. Data included are observations from 29 m above the sea surface. Symbols are color coded based on wind speed U_{10m} .

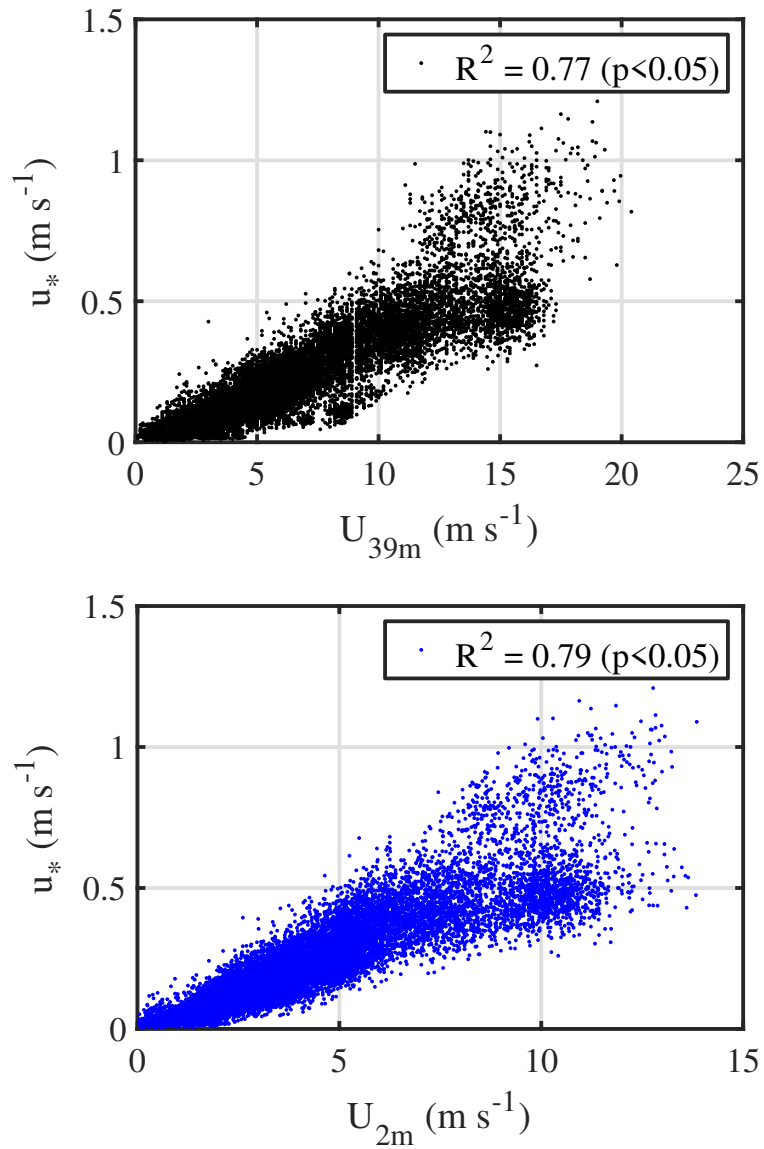


Figure 5: manuscript Figure S2 - Comparison between friction velocity u_* and horizontal wind speed U at 2 and 39 m above the sea ice surface. The legends shows respective coefficients of determination of the linear regression. Note that U_{2m} has been derived from the 3-D wind measurements of the sonic anemometer.

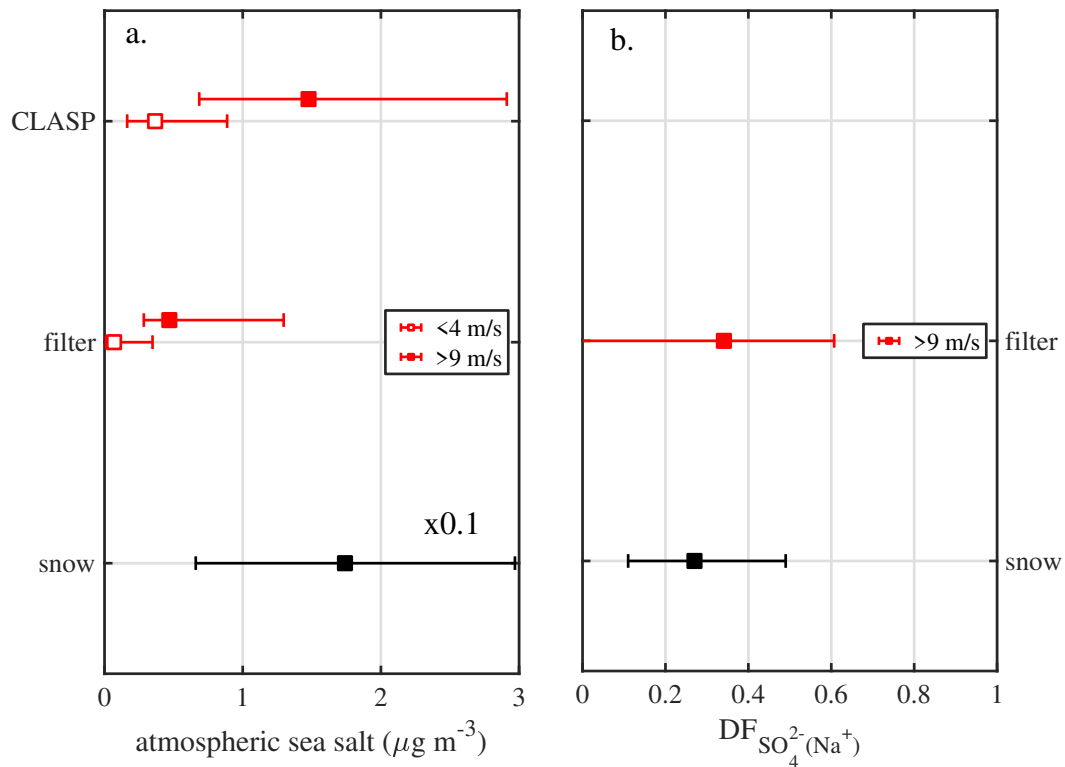


Figure 6: manuscript Figure 17 - The partitioning of sea salt between atmosphere and snow, and sulfate depletion above first year sea ice from 18 June to 21 July 2013. Panel (a) shows median atmospheric sea salt concentrations during calm ($U_{10m} < 4 \text{ m s}^{-1}$) and windy ($U_{10m} > 9 \text{ m s}^{-1}$) conditions derived from aerosol filter measurements (filter) and spectral particle number densities $N_{0.4-12}$ (CLASP) (see text). For comparison, a potential atmospheric concentration is calculated assuming that all sea salt observed in the top 0.1 mm of snow on sea ice was released by sublimation and mixed into a 100 m thick boundary layer (for better comparison multiplied here by 0.1). Panel (b) shows for the same time period median sulfate depletion factors $DF_{\text{SO}_4^{2-}}$ (with respect to Na^+) in in surface snow and in aerosol during windy ($U_{10m} > 9 \text{ m s}^{-1}$) conditions. Symbols and errorbars represent median and lower and upper quartiles, respectively.

Table 1: manuscript Table 4 - Descriptive statistics of the aerosol chemistry during ANT-XXIX/6 (ANT6) and ANT-XXIX/7 (ANT7) with mean and median values weighted by the filter sampling interval. Ion and sea salt concentrations are in units of ng m^{-3} . See section 2.4 for definition of depletion factors DF .

Parameter	ANT6			ANT7			ANT7		
	at 2 m mean $\pm\sigma$	median	N^a	at 29 m mean $\pm\sigma$	median	N^a	at 29 m mean $\pm\sigma$	median	N^a
sea-salt ^b	707 \pm 1500	336	43	1253 \pm 2319	639	106	559 \pm 486	425	28
Na ⁺	217 \pm 460	103	43	384 \pm 711	196	106	171 \pm 149	130	28
Cl ⁻	379 \pm 765	179	43	656 \pm 1225	302	106	311 \pm 282	232	27
SO ₄ ²⁻	28 ^c \pm 61	19 ^c	38	75 \pm 152	45	84	33 \pm 30	23 ^c	28
Br ⁻	2.0 \pm 1.0	1.9	42	1.5 ^c \pm 3.0	0.7 ^c	98	0.5 ^c \pm 0.6	0.5 ^c	23
$DF_{SO_4^{2-}}$	0.29 \pm 0.57	0.48	38	0.07 \pm 0.94	0.29	74	0.12 \pm 0.60	0.21	27
DF_{Na^+}	-0.08 \pm 0.29	-0.03	43	-0.46 \pm 2.29	-0.04	97	-0.02 \pm 0.19	-0.01	27
DF_{Br^-}	-1.66 \pm 1.86	-1.86	36	0.04 \pm 0.96	0.37	89	0.05 \pm 1.26	0.49	23

^asample size ^bsea salt concentration is derived by multiplying the Na⁺ concentration by 3.262 based on the Na⁺ mass fraction in reference seawater after Millero et al., 2008 ^cbelow the estimated LOD (Table 3)

Table 2: manuscript Table 5 - Descriptive statistics of the volume-integrated snow chemistry during ANT-XXIX/6 on first-year sea ice (FYI) at ice stations S1-S6, on multi-year sea ice (MYI) at ice stations S7-9, and for snow layers within 10 cm of the snow surface (TOP10). Ion and sea salt concentrations are in units of $\mu\text{g g}^{-1}$. See section 2.4 for definition of depletion factors DF .

Parameter	FYI			MYI			TOP10		
	mean $\pm\sigma$	median	N^a	mean $\pm\sigma$	median	N^a	mean $\pm\sigma$	median	N^a
snow depth (cm)	20.9 \pm 8.3	19.0	17	50.0 \pm 32.2	33.0	7	-	-	-
S_p (psu)	1.40 \pm 3.99	0.11	110	0.82 \pm 4.31	0.02	104	0.31 \pm 0.90	0.06	96
sea salt ^b	1176 \pm 3518	83	86	590 \pm 3157	22	95	249 \pm 729	58	80
Na^+	361 \pm 1079	26	86	181 \pm 968	7	95	76 \pm 223	18	80
Cl^-	680 \pm 2035	48	87	305 \pm 1842	13	98	141 \pm 415	34	81
SO_4^{2-}	61 \pm 182	6	87	30 \pm 166	1	98	17 \pm 62	3	81
Br^-	4.28 \pm 12.23	0.18	85	1.76 \pm 10.92	0.07	90	1.01 \pm 3.72	0.12	78
$DF_{\text{SO}_4^{2-}}$	0.19 \pm 0.41	0.24	86	0.33 \pm 0.44	0.35	94	0.27 \pm 0.39	0.27	80
DF_{Na^+}	0.01 \pm 0.38	0.06	86	-1.09 \pm 8.88	0.07	94	-0.11 \pm 0.99	0.06	80
DF_{Br^-}	-0.25 \pm 0.98	0.05	83	-0.28 \pm 1.16	-0.01	86	-0.21 \pm 0.99	0.04	76

^asample size ^bsea salt concentration is derived by multiplying the Na^+ concentration by 3.262 based on the Na^+ mass fraction in reference seawater after Millero et al., 2008

Table 3: manuscript Table S1 - Descriptive statistics of airborne snow particles observed for 8 June to 12 August 2013 (ANT6) and for 14 August to 16 October 2013 (ANT7): total number densities N_{46-478} and particle diameter d_p . Statistics refer to periods when airborne snow particles were present, i.e. times with no snow particles observed were removed prior to averaging.

Parameter	ANT6		ANT7	
	at 0.2 m	at 29 m ^a	at 29 m	at 29 m
N_{46-478} (m^{-3})				
mean	2.6×10^5	4.0×10^3	8.7×10^3	7.2×10^3
σ	7.4×10^3	9.5×10^3	2.7×10^4	2.2×10^4
median	4.7×10^3	7.7×10^2	9.9×10^2	1.3×10^3
d_p (μm)				
mean	138	132	133	143
σ	59	59	53	53
median	132	117	124	136
N^b	8608	11766	42959	37123
sampling time (days) ^c	6	8	30	26

^a for direct comparison of vertical differences statistics of the 29 m measurements only for times when sea ice observations at 0.2 m were available ^bsample size ^ctotal aggregated time during which airborne snow particles were detected

Table 4: manuscript Table S2 - Descriptive statistics of aerosol observed during 8 June - 26 July 2013 (ANT6): total number densities $N_{0.4-12}$ and particle diameter d_p .

Parameter	at 0.2 m	at 2 m	at 29 m ^a	at 29 m
$N_{0.4-12}$ (m ⁻³)				
mean	1.7×10^6	1.4×10^6	1.4×10^6	2.1×10^6
σ	2.5×10^6	1.9×10^6	1.6×10^6	6.4×10^6
median	8.6×10^5	5.6×10^5	8.0×10^5	1.1×10^6
d_p (μm)				
mean	0.67	0.60	0.67	0.69
σ	0.11	0.06	0.11	0.14
median	0.66	0.60	0.65	0.66
N^b	13077	14907	9963	48892
sampling time (days) ^c	9	10	7	34

^a for direct comparison of vertical differences statistics of the 29 m measurements only for times when sea ice observations at 2 m were available ^bsample size ^ctotal aggregated sampling time