



1 **Spatial and temporal variability of sea-salts in ice cores**
2 **and snow pits from Fimbul Ice Shelf, Antarctica**

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21 **Abstract**

22 Major ions were analysed in three firn cores from different ice rises located at Fimbul
23 Ice Shelf (FIS): Kupol Ciolkovskogo (KC), Kupol Moskovskij (KM), and Blåskimen
24 Island (BI), a 100 m long core drilled near the FIS edge (S100), and five snow pits
25 (M1, M2, G3, G4, and G5) sampled on the ice shelf. These sites are distributed over
26 the entire FIS area so that they provide a variety of elevation and distance to the
27 sea. Sea-salt species (mainly Na⁺ and Cl⁻) generally dominate the precipitation
28 chemistry in the study region. Concentrations of these ions were found to decrease
29 with latitude and distance from the sea. We associate a significant six-fold increase



1 in sea-salts observed in the S100 core after the 1950s with a change in deposition
2 regime. This increase in sea-salt concentrations is synchronous with a shift in non-
3 sea-salt sulfate (nssSO_4^{2-}) toward negative values, suggesting a possible
4 contribution of fractionated aerosol to the sea-salt load in the S100 core most likely
5 by dry deposition. In contrast, wet deposition of atmospheric sea-salts is dominant
6 in the three ice rises cores, and no evidence of a significant contribution of
7 fractionated sea-salt to these sites was found. In summary, these results suggest
8 that the S100 core contains a more local sea-salt signal, dominated by processes
9 during sea-ice formation in the neighbouring waters. In contrast, the ice rises cores
10 register the larger-scale signal of atmospheric flow conditions and transport of sea-
11 salt aerosols produced over open water rather than local changes in sea-ice, wind-
12 blown snow accumulated over sea-ice, and frost flower formation. These findings
13 are a contribution to the understanding of the mechanisms behind sea-salt aerosol
14 production, transport and deposition at coastal Antarctic sites, and for the
15 improvement of the current Antarctic sea-ice reconstructions based on sea-salt
16 chemical proxies obtained from ice cores.

17 **1 Introduction**

18 Antarctic ice and firn cores contain valuable information about the climate and
19 atmospheric chemical composition of the past and provide evidence for the
20 important role of Antarctica in the global climate system. Numerous ice and firn
21 cores have been drilled in Antarctica during the past decades (Stenni et al., 2017).
22 However, relatively few cores were drilled in coastal regions, which are more
23 sensitive to changes in climate than the dry and cold interior of Antarctica. In fact,
24 two recent review papers point out the lack of ice core data from low elevation
25 coastal areas when discussing Antarctic climate variability (Stenni et al., 2017;
26 Thomas et al., 2017). In an effort to understand the role of ice shelves in stabilizing
27 the Antarctic ice sheet, particular focus has been laid on the investigation of ice rises
28 and ice rumples as buttressing elements within the ice sheet - ice shelf complex
29 (Paterson, 1994; Matsuoka et al., 2015). Furthermore, due to their radial ice flow
30 regime, generally low ice velocities, and relatively high surface mass balance
31 (SMB), ice rises are potentially useful sites for ice core retrieval (Philippe et al., 2016;



1 Vega et al., 2016). Firn and ice cores drilled at ice rises allow obtaining high-
2 resolution climate records to investigate subannual and long-term temporal changes
3 in the loads of different chemical compounds found in the snow, providing
4 information about their sources and transport, particularly of sea-salt ions, such as
5 sodium (Na^+) and chloride (Cl^-), which are strongly modulated by sea-ice extent and
6 meteorological conditions. Recent modelling efforts to study the use of sea-salts as
7 proxies for past sea-ice extent have shown that, under present climate conditions
8 and on interannual timescales, meteorological conditions rather than sea-ice extent
9 are the dominant factor modulating atmospheric sea-salt concentrations that are
10 deposited at the interior and coastal sites in Antarctica (Levine et al., 2014).
11 However, sea-salts have the potential as proxy for sea-ice extent at glacial-
12 interglacial scales when large changes in sea-ice extent took place (Levine et al.,
13 2014).

14 At most Antarctic sites, atmospheric sea-salt concentrations present maxima during
15 austral winter (Wagenbach et al., 1998; Weller and Wagenbach, 2007; Jourdain et
16 al., 2008; Udisti et al., 2012), with the exception of Dumont D'Urville where maxima
17 occur during summer (Wagenbach et al., 1998). Similarly, sea-salt fluxes obtained
18 from Antarctic ice cores also show winter maxima (Abram et al. 2013 and references
19 therein). However, in some recent core records from coastal sites no clear
20 seasonality is observed, e.g. at Mill Island during the period 1934–2000 (Inoue et
21 al., 2017). Abram et al. (2013) conclude that despite the seasonal signal registered
22 in different Antarctic ice cores, sea-salt fluxes do not show a consistent relationship
23 with sea-ice extent on interannual timescales, and on the contrary, are highly
24 dependent on atmospheric transport, and/or the presence of polynyas.

25 Hitherto, two main sources of increased winter sea-salt aerosols have been
26 proposed: (i) increased storminess leading to an enhancement of sea-salt aerosols
27 above the open ocean with possibly faster meridional transport (Petit et al., 1999;
28 Fischer et al. 2007), and (ii) a direct input of sea-salts associated to increases in
29 sea-ice, overcoming source (i), e.g. due to frost flowers (Rankin and Wolff, 2002,
30 2004; Roscoe et al., 2011), brine (Rankin et al., 2000), and the contribution of snow
31 transported over sea-ice by wind (Yang et al., 2008, 2010; Huang and Jaeglé, 2017).



1 In the review by Abram et al. (2013), the authors suggest that the brine-frost flower
2 system is a plausible source of sea-salt aerosols to coastal Antarctic sites. This
3 evidence consists of negative non sea-salt sulfate (nssSO_4^{2-}) values both in winter
4 aerosol and fresh snow sampled at coastal sites (Hall and Wolff, 1998; Wagenbach
5 et al., 1998; Curran et al., 1998; Rankin and Wolff, 2002; and Rankin and Wolff,
6 2003), and also in ice cores from both inland (Wagenbach et al., 1994, Kreutz et al.,
7 1998) and coastal sites (Inoue et al., 2017). These negative values suggest that the
8 sea-salt source was highly fractionated compared to seawater.

9 During the process of sea-ice formation, ions present in the water are not
10 incorporated in the ice crystal matrix, but remain as highly concentrated brine in
11 brine pockets or channels. The brine can be transported by capillary effects through
12 brine channels to the newly formed ice surface, resulting in a thin layer of highly
13 saline surface brine. This fractionated brine is unlikely to be a direct source of sea-
14 salts because it usually quickly gets covered by snow or frost flowers, and no clear
15 mechanism has been found to explain how this brine could become airborne (Abram
16 et al., 2013). With further cooling of the ice, the volume of brine decreases and
17 consequently, its salinity increases, leading to the precipitation of different saline
18 compounds. This depends on the temperature, e.g. sodium sulfate or mirabilite
19 ($\text{Na}_2\text{SO}_4 \cdot 10 \text{H}_2\text{O}$) starts to precipitate at temperatures below $-8 \text{ }^\circ\text{C}$, and sodium
20 chloride (NaCl) at temperatures below $-26 \text{ }^\circ\text{C}$. Consequently, the remaining brine is
21 depleted in sodium and sulfate ions via precipitation of mirabilite at relatively mild
22 polar temperatures. Frost flowers can form from this brine when meteorological
23 conditions are adequate, i.e. at low intensity winds, which allows these delicate
24 structures to grow without breaking apart, and on very thin ice where a strong
25 temperature gradient is present between the ice surface and the overlying air
26 (Rankin et al., 2000; Rankin and Wolff, 2002 and references therein). Thus, frost
27 flowers formed at temperatures below $-8 \text{ }^\circ\text{C}$ will be depleted in sodium and sulfate
28 relative to other ions present in seawater (Rankin et al., 2000; Rankin and Wolff,
29 2002), evidenced by negative nssSO_4^{2-} values measured in aerosols and snow (see
30 section 2.3 for more details on the calculation of the nss-fractions).



1 For most of the last decade, frost flower formation, transport and deposition, has
2 been considered the most plausible mechanism behind the fractionated aerosol
3 detected at coastal areas. However, Yang et al. (2008 and 2010), and Huang and
4 Jaeglé (2017) proposed an alternative mechanism: the origin of sea-salt aerosol
5 could be due to the sublimation of blowing salty snow. This salty snow could be a
6 result of frost flower formation, upward migration of brine within the snow (Massom
7 et al., 2001), or by the input of sea-spray from the open ocean or nearby leads or
8 polynyas (Dominé et al., 2004). Flooding of sea-ice under the weight of accumulated
9 snow can also induce increased salinity of snow (Massom et al., 2001). As the snow
10 can be contaminated or wetted with fractionated brine or frost flowers, it could be
11 expected that this salty snow also shows such fractionation. As pointed by Yang et
12 al. (2008 and 2010), this salty snow can be transported by wind and if the air is not
13 saturated, the snow particles may lose water by sublimation and become sea-salt
14 aerosols. These aerosols could then be transported and deposited either by dry or
15 wet deposition, depending on local meteorology.

16 According to Abram et al. (2013), the idea proposed by Yang et al. (2008) is
17 plausible for coastal sites, along with the frost flower mechanism. Consequently,
18 snow present on new sea-ice and frost flowers are important features that,
19 combined with wind transport, need to be taken into account when interpreting the
20 sea-salt record of coastal ice and firn cores.

21 This study discusses sub-annual and long-term temporal changes in sea-salt and
22 major ion concentration measured in three recently drilled firn cores from different
23 ice rises located at Fimbul Ice Shelf (FIS): Kupol Ciolkovskogo, Kupol Moskovskij,
24 and Blåskimen Island, a 100 m long core drilled near the FIS edge (S100), and five
25 snow pits sampled along the ice shelf (Figure 1). The main goals of the present
26 study are to investigate possible mechanisms behind deposition, subannual, and
27 spatial variability of sea-salts in this coastal region. The results presented here
28 contribute to bridging the data gap existent at coastal Antarctic sites, and to the
29 improvement of current Antarctic sea-ice reconstructions based on sea-salt
30 chemical proxies.



1 **2 Methods**

2 **2.1 Study area**

3 With an extent of approximately 36 500 km², FIS is the largest ice shelf in the
4 Haakon VII Sea (Figure 1). Fed by Jutulstraumen, the largest outlet glacier in DML,
5 FIS is divided into a fast moving ice tongue, Trolltunga, directly feeding the central
6 part of the ice stream, and slower surrounding parts. Several ice rises are found at
7 FIS, varying in size from 15 to 1200 km², and located approximately 200 km apart.
8 Early investigations in this area began during the International Geophysical Year
9 (IGY) 1956/57 (Swithinbank, 1957; Lunde, 1961; Neethling, 1970) and continued
10 during the last decades with focus on surface mass balance (SMB) variability in
11 space and time (Melvold et al., 1998; Melvold, 1999; Rolstad et al., 2000; Isaksson
12 and Melvold, 2002; Kaczmarska et al., 2004; Kaczmarska et al., 2006; Divine et al.,
13 2009; Sinisalo et al., 2013; Schlosser et al., 2012, 2014; Langley et al., 2014; Vega
14 et al., 2016). However, studies on spatial and temporal variability of chemical
15 composition of snow and ice from this area are limited to water stable isotopes
16 interpretations (Kaczmarska et al., 2004; Schlosser et al., 2012, 2014; Vega et al.,
17 2016).

18 SMB obtained from the S100 core (Figure 1) retrieved at FIS shows a mean long-
19 term accumulation rate of 0.3 m water equivalent per year (m w.e. yr⁻¹) for the period
20 1737–2000, with a significant negative trend in SMB for the period 1920–2000
21 (Kaczmarska et al., 2004). This negative trend in SMB has been reported in several
22 shorter firn cores from the region (Isaksson and Melvold, 2002; Divine et al., 2009;
23 Schlosser et al, 2014), including one record from the Kupol Ciolkovskogo ice rise
24 (Vega et al., 2016).

25 More detailed information on previous campaigns, glaciological and meteorological
26 conditions at FIS and the core sites at the ice rises, can be found in Vega et al.
27 (2016) and Goel et al., (2017), and references therein, whereas an overview on
28 Antarctic ice rises is given in Matsuoka et al. (2015).

29 **2.2 Sampling**

30 Three shallow firn cores were retrieved at different ice rises (Kupol Ciolkovskogo
31 (KC), Kupol Moskovskij (KM), and Blåskimen Island (BI), Figure 1, Table 1), located



1 at FIS between January 2012 and January 2014 during field expeditions organized
2 by the Norwegian Polar Institute (NPI). Location, elevation, and length of the
3 different ice rises cores are presented in Table 1. Each core was drilled from the
4 bottom of a 2 m snow pit (not sampled for major ions). The firn density was
5 determined as bulk density of each sub-core piece (average length of 45 cm), and
6 of each snow pit interval (20 cm). The samples were collected following clean
7 protocols (Twickler and Whitlow, 1997), shipped frozen to NPI, and later to the Paul
8 Scherrer Institute (PSI), Switzerland, for cutting and chemical analysis. Sample
9 length varied between 4 and 8 cm depending on sample depth and density.
10 Thickness of ice lenses, water stable isotope ratios and SMB for the three ice rises
11 are reported in Vega et al. (2016). Additionally, unpublished major ion
12 concentrations measured in the 100 m deep S100 core drilled in austral summer
13 2000/2001 (Kaczmarek et al., 2004) were included in this study (Figure 1, Table 1).
14 In addition to the firn and ice cores, five snow pits (M1, M2, G3, G4, and G5, with
15 additional re-sampling of G4 and G5 as G4a and G5a) were sampled at different
16 sites at FIS during Nov.–Dec. 2009 by a field team from NPI (Figure 1, Table 1). The
17 snow pits were sampled at 20 cm (M1, M2, and G3, G4, and G5) and 4 cm (G4a
18 and G5a) intervals; samples were transported and kept frozen until analysis. The
19 previous summer surface at the snow pit sites was estimated to be located at a
20 depth between 120 and 160 cm (Sinisalo et al., 2013).

21 **2.3 Chemical analyses**

22 Major ions (methanesulfonic acid (MSA), Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, K⁺, Mg²⁺ and Ca²⁺)
23 present in the three firn cores from the ice rises were analysed at PSI using a
24 Metrohm ProfIC 850 ion chromatograph combined with an 872 Extension Module
25 and auto-sampler. The precision of the method was within 5 % and detection limits
26 (D.L.) were below 0.02 μmol L⁻¹ for each ion (Wendl et al., 2014). Ion concentrations
27 (MSA, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, K⁺, Mg²⁺ and Ca²⁺) in the S100 core were measured at
28 the British Antarctic Survey (BAS) using fast ion chromatography (Littot et al., 2002).
29 The reproducibility of the measurements was 4–10 %.



1 In addition, major ions (Cl^- , Br^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} and Ca^{2+}) were measured
2 in the FIS snow pits. Triplicate snow samples were taken at each depth interval.
3 Hence, average concentrations of each ion at each depth interval are reported here.
4 Additionally, pits G4 and G5 were sampled at 2 cm intervals, with one sample per
5 depth interval (pits G4a and G5a). For these snow pits sampled at high resolution
6 only Cl^- , NO_3^- , and SO_4^{2-} are available. All snow pit samples were transported frozen
7 from the sampling site to NPI and then to the Department of Earth Sciences,
8 Uppsala University, where major ions were quantified using a Metrohm ProfIC 850
9 ion chromatograph. The reproducibility of the measurements was within 5 % and
10 detection limits (DL) were below $0.3 \mu\text{mol L}^{-1}$ for each ion.

11 Hereafter, brackets are used to denote ionic concentrations expressed in $\mu\text{mol L}^{-1}$.
12 Non sea-salt fractions were calculated from the mean seawater composition using
13 Na^+ as standard ion, using:

$$14 \quad [\text{nssX}] = [\text{X}]_{\text{total}} - k \times [\text{Na}^+]_{\text{total}},$$

15 where

$$16 \quad k = \frac{[\text{X}]_{\text{seawater}}}{[\text{Na}^+]_{\text{seawater}}},$$

17 using the standard mean chemical composition of seawater with ion concentration
18 expressed in $\mu\text{mol L}^{-1}$.

19 In addition, water stable isotopes analyses of the KC, KM and BI cores are described
20 in Vega et al. (2016); while analysis of the S100 core is described in (Kaczmarek
21 et al., 2004). Water stable isotope analyses were not performed in the FIS snow
22 pits.

23 **2.4 Firn and ice core timescales**

24 The timescales of the KM and BI cores were obtained based on annual layer
25 counting of water stable isotope ratios ($\delta^{18}\text{O}$), and found to cover the periods
26 between austral winter-1995(96) and summer-2014, respectively. The error in the
27 dating was estimated as ± 1 year for both of these cores (Vega et al., 2016). Both
28 KC and the S100 cores were dated using a combination of annual layer counting of
29 $\delta^{18}\text{O}$ and identification of volcanic horizons (i.e. by using the SO_4^{2-} , dielectric profiling
30 (DEP), and electrical conductivity measurements (ECM)), with timescales covering



1 the time period 1958–2012 (± 3 years) at KC (Vega et al., 2016), and 1737–2000
2 (± 3 years) at S100 (Kaczmarek et al., 2004).

3 **2.5 Snow pits timescales**

4 Snow pits were dated by visually identifying the summer layer previous to the
5 digging, since no water stable isotope data were available. Consequently, a highly
6 precise dating (e.g. with monthly resolution) of the snow layers was not possible.
7 The previous summer layer in pits M1, M2 and G3–G5 was identified at 120–160 cm
8 depth according to Sinisalo et al. (2013); however, chemical sampling was done just
9 down to a depth of 60–90 cm in each snow pit reported in this study. Consequently,
10 the snow depth at the different snow pit sites in which ions were analysed represents
11 less than a year of accumulation (Table 1). The snow accumulation between top
12 and bottom in each snow pit agree with previously documented values estimated
13 for the most recent years at the different pit sites (Schlosser et al., 2014), being
14 slightly below the average annual accumulation, with the exception of pit G3, which
15 showed a higher accumulation between 0–90 cm depth than the average
16 accumulation rate at the site for the period 1993–2009 (Schlosser et al., 2014).

17 **3 Results**

18 **3.1 Ion concentrations and sources**

19 Median concentration values for all ions measured in the cores and snow pits are
20 shown in Table 2. In general, concentrations in the KM core are higher than in the
21 other ice rises cores, and snow pits, e.g. about eight-fold higher concentrations of
22 Na^+ , K^+ , Mg^{2+} and Cl^- in KM than in the KC core are found for the period 1995–2012.
23 The relatively high Na^+ and Cl^- concentrations observed in the KM core are also
24 detected in the upper meters of the S100 core (in the periods 1995–2000, and 1950–
25 2000, respectively, Table 2). Similarly high values have been reported in several
26 snow and firn samples from other western DML coastal sites (Kärkäs et al., 2005),
27 and in Mill Island, Wilkes Land (Inoue et al., 2017).

28 Ion concentrations in the snow pits are in reasonable agreement with firn and ice
29 core values reported in Table 2 and with ion concentration ranges for snow pits



1 previously sampled at FIS (Mulvaney et al., 1993). Temporal and spatial variability
2 of ion concentrations are explored in more detail in the following sections.
3 In order to assess the most important sources explaining the total variance in the
4 glacio-chemical records from FIS, a principal component analysis (PCA) was
5 applied to the different ion series measured at the KC, KM, BI, and S100 cores.
6 Years, in which no sub-annual concentrations were available in the S100 (1793,
7 1841, 1866, 1918, and 1944) due to low resolution, were filled in by linearly
8 interpolating between the annual means of the previous and following year. For the
9 PCA analysis, ion concentrations were logarithmized (at sub-annual (using the raw
10 values as input) and annual resolutions) and standardized by subtracting the mean
11 of the data series from each data point and then dividing the result by the standard
12 deviation of the data series. Due to the sampling resolution, only the KM and BI
13 cores were comparable at a sub-annual level. PCA analyses were performed for
14 three different periods of the S100 core: for the entire time interval spanning 1737–
15 2000, for the subsection between 1737–1949, and between 1950–2000.
16 The sum of the variances of the first three principal components (PC1, PC2 and
17 PC3) was $\geq 80\%$ of the total variance of the original sub-annual and annual data in
18 all cores. Since the results of the sub-annual and annual PCA analysis are similar
19 only the annual results are considered. The loadings of the first three (KC) and two
20 (KM, BI, and S100) principal components are shown in Table 3. PCA results are
21 consistent between the different cores. Consequently, the ions can be separated in
22 two main groups: sea-salts species (Na^+ , Cl^- , K^+ , Mg^{2+} , and Ca^{2+}) and marine-
23 biogenic/mixed (MSA, SO_4^{2-} , including NO_3^-) (Table 3).
24 Generally, our results indicate that the major sources of the ions at the different sites
25 are the same, independent of the core site and mean concentrations of ions in the
26 cores. Only at the KC site the PCA results imply an additional input of Ca^{2+} from
27 other sources than sea-salt, as for instance mineral dust. The high loadings of NO_3^-
28 and MSA in PC2, and thus, coherence between both species, was already observed
29 in an ice core from Lomonosovfonna, Svalbard (Wendl et al., 2015), and a fertilizing
30 effect was proposed as explanation. Enhanced atmospheric NO_3^- concentrations
31 and the corresponding nitrogen input to the ocean can trigger the growth of dimethyl-



1 sulfide-(DMS)-producing phytoplankton. However, there is a variety of possible NO_3^-
2 sources and the relative importance of these sources at certain locations and time
3 is still in discussion (Mulvaney and Wolff, 1993; Savarino et al., 2007; Wolff et al.
4 2008; Weller et al. 2011; Pasteris et al., 2014; Sofen et al. 2014).

5 **3.2 Long-term variability of ion concentrations**

6 We use the two longest available records for FIS (KC and S100) to explore the long-
7 term temporal variability of major ions, with special focus on sea-salts, represented
8 by Cl^- and Na^+ (Figure 2). In the S100 core, Na^+ , Cl^- , K^+ , SO_4^{2-} , Mg^{2+} , and Ca^{2+}
9 concentrations show a marked six-fold increase after the 1950s. However, there is
10 no significant increase of the concentration of these species in the KC core. Due to
11 its limited time coverage it cannot be determined if there was a substantial relative
12 increase in concentrations at this site after the 1950s. MSA and NO_3^- concentrations
13 do not show such marked increase in the S100 core and values agree between both
14 cores after the 1950s (Figure S1 in the supplementary material). Consequently,
15 three periods can be distinguished in the S100 record: (i) the period between 1995–
16 2000, comparable to the time covered by the KM and BI cores; (ii) the period
17 between 1737–1949, where ion concentrations remain low; and, (iii) the period
18 between 1950–2000, where sea-salt concentrations increased (Table 2).

19 With the exception of MSA, all ions show a positive trend (significant at the 95 %
20 confidence level) during the 1950–2000, although the slope for NO_3^- is three orders
21 of magnitude smaller than for the other ions (slope and error of the linear regression
22 are shown in Table S1 in the supplementary material). Such a linear trend was not
23 observed in the ion records from the KC core over the same period.

24 Ions, with the exception of MSA, also show a positive and significant trend between
25 1737–1949, (Table S1), however, the increase is less marked than during 1950–
26 2000 period.

27 **3.3 Sub-annual variability of ion concentrations**

28 To investigate the seasonality of the different ion groups in the KM, BI, and S100
29 cores, a composite year of data was produced for each ion species, i.e. for each



1 year, a linear interpolation was done in the concentration domain of each ion at a
2 resolution of 0.01-years (3.65 days). The seasonal concentration averages were
3 then calculated for the months MAM (autumn), JJA (winter), SON (spring) and DJF
4 (summer). All ion concentrations were expressed in $\mu\text{mol L}^{-1}$ and were
5 logarithmized. Ion concentrations were not available at the top 2 m (snow pit data),
6 therefore, the composite year consisted of 16 (1996–2011) and 15 (1997–2011)
7 complete years for the KM and BI cores, respectively. In the S100 core,
8 seasonalities were investigated only during the period 1995–2000, where the
9 concentrations have sufficient temporal resolution for subannual analyses. The
10 resulting seasonal average concentrations in the cores are presented in Figure 3.
11 Sea-salt species (Na^+ and Cl^- , Figure 3a) show minimum concentrations during
12 spring in the KM core, whereas in the S100 core minimum values are reached during
13 summer. Maximum concentrations are reached during summer (KM) and
14 spring/autumn (S100). In the BI core, sea-salt seasonalities are less pronounced.
15 Both Mg^{2+} and Ca^{2+} (Figure 3b) show high values in summer in the KM core, and in
16 spring in the S100 core, whereas, again, a less marked seasonality is observed in
17 the BI core. MSA concentrations (Figure 3c) show consistent and marked
18 spring/summer maxima in all three cores, and a marked winter minima in the BI
19 core, which is in agreement with the main source of MSA (marine-biogenic) most
20 active during the warmer months. The MSA winter minimum is not as pronounced
21 in the KM core as in the BI core, while the MSA minimum is reached in autumn in
22 the S100 core. NO_3^- and SO_4^{2-} concentrations (Figure 3d) show a distinct increase
23 toward the spring/summer in the BI core, which is also observed in the KM core,
24 although less defined. In the S100 core, higher NO_3^- and SO_4^{2-} concentrations are
25 found during the spring.

26 3.4 Ions spatial variability

27 In order to investigate ion spatial variability at FIS, we used median ion
28 concentrations in the different snow pits (M1, M2, G3, G4, and G5), and ice rises
29 cores (KM and BI) for the year 2009, and compared them with latitude, longitude,
30 and distance from the sea (Table 4). Since it has been found that the chemical



1 records in the KM and BI cores appear to be affected by surface topography and
2 local meteorological conditions (Vega et al., 2016), we also calculated the
3 correlation coefficients (R values) between concentration and latitude, longitude,
4 and distance from the sea, only using the snow pit data.
5 Concentrations of Br⁻, Na⁺, K⁺, Mg²⁺ and Ca²⁺ show a significant decrease at the
6 95 % confidence level with increased latitude, i.e. with increasing distance from the
7 coast. Correlation coefficients are high for Cl⁻ and SO₄²⁻, however, not significant.
8 No significant relationship is found between the median ion concentrations and
9 longitude for any of the species. When comparing concentrations with the distance
10 from the sea (obtained from the GIS package Quantarctica, www.quantarctica.org),
11 it was found that Br⁻, SO₄²⁻, Na⁺, and Ca²⁺ show a significant decrease with
12 increasing distance from the sea when only the snow pits are considered in the
13 analysis; i.e. significance is lost when the KM and BI 2009 median values are
14 included. These findings agree with previous studies from western Dronning Maud
15 Land (WDML) (Stenberg et al., 1998), confirming that sea-salt concentrations are
16 strongly dependent on the distance from the sea in this area. This relationship loses
17 significance when the records from the ice rises KM and BI are included, reflecting
18 the influence of local effects on annual SMB at these sites (Vega et al., 2016).
19 Since the elevation differences between the snow pits are only in the order of tens
20 of meters, we did not investigate the elevation dependence of mean ion
21 concentrations.

22 3.5 Deposition regimes

23 In order to explain the marked increase in ion concentrations after the 1950s, and
24 the negative trend observed in SMB for the S100 and KC cores for the second half
25 of the 20th century (Figure 2e and f) (Vega et al., 2016), we investigated the
26 deposition regimes at FIS. We employed correlation analysis to assess the
27 relationship between the annual average concentrations of all ions and annual SMB
28 at the different core sites for the respective period covered by each core (as noted
29 in Table 2). Correlation coefficients (R_{conc} , Table 5) were < 0.57 and non-significant
30 at the 95 % confidence interval (with the exception of NO₃⁻ and SO₄²⁻ in the S100



1 core), which indicates that accumulation rates, and thus, wet deposition is not
2 controlling the ion concentration at the different core sites at FIS. We also
3 investigated the relationship between chemical flux, expressed as $\mu\text{mol cm}^{-2} \text{yr}^{-1}$
4 and annual snow accumulation at the different core sites (R_{flux} , Table 5). A
5 significant positive correlation between fluxes and accumulation exists for almost all
6 ion species analysed in the different cores with the exception of sea-salts in the
7 S100 core, with significant R_{flux} values larger than 0.5 for most ions (with the
8 exception of MSA in the KC and S100 cores, and SO_4^{2-} , Mg^{2+} and Ca^{2+} in the S100
9 core). This indicates that most of the variance in the ion flux can be explained by
10 changing snow accumulation rate at each site for most of ions, with the exception
11 of sea-salts in the S100 core. Therefore, dry deposition of sea-salt aerosols would
12 be the controlling factor in sea-salt fluxes at the S100 site. To confirm that the
13 increase in sea-salt fluxes observed after the 1950s is due to a change in sea-salt
14 deposition mechanism, we calculated the R_{conc} and R_{flux} values for the periods
15 1737–1949 and 1950–2000 (Table 5). While R_{conc} values for both periods were not
16 significant and low, during 1737–1949, all R_{flux} values were significant ($R > 0.41$). This
17 indicates that during the period 1737–1949, most of the variance in all ion fluxes can
18 be related to SMB at the S100 site. Sea-salt R_{flux} values decreased (with the
19 exception of R_{flux} of NO_3^-) and became non-significant in the case of Cl^- , SO_4^{2-} , and
20 Ca^{2+} during the 1950–2000 period, which points to a change in the deposition
21 regime after the 1950s.

22 3.6 Evidence for increased fractionated nss- SO_4^{2-} after 1950s

23 In order to evaluate the possible effect of fractionated aerosols as a source of sea-
24 salts to the snow on FIS, we calculated the nss SO_4^{2-} fraction as described in section
25 2.3, using k values of 0.06 (see supplementary material in Vega et al., 2015). The
26 percentage of nss SO_4^{2-} relative to total SO_4^{2-} is two- to three-times higher in the KC
27 core than in the other ice rises cores, KM and BI. Negative median nss SO_4^{2-} values
28 were obtained in the S100 core and snow pits M1, M2, G3, and G4. These negative
29 values, i.e. nss SO_4^{2-} is strongly depleted in SO_4^{2-} relative to Na^+ (Rankin and Wolff,
30 2002), suggest a possible role of frost flowers and wind-blown salty snow as source



1 of sea-salts to the S100 core (Figure 4a y b, black line). We therefore followed the
2 approach by Inoue et al. (2017), to obtain a new value of k , k' . This k' was calculated
3 by subtracting the slope of the linear regression (slope= 0.02, in Figure 5) for the
4 nssSO_4^{2-} concentrations vs. Na^+ concentrations (for the whole period, i.e. 1737–
5 2000) from the constant k (calculated as described in section 2.3), i.e. $k'= 0.04$ (in
6 $\mu\text{mol L}^{-1}$). Similarly, we calculated k'' for the period 1737–1949 ($k''=0.02$), and k'''
7 for the period 1950–2000 ($k'''= k'=0.04$). The nssSO_4^{2-} fraction without the effect of
8 sulfate fractionation was recalculated using the values of k' , and k'' (Table 6, and
9 Figure 4a y b, red and blue lines). nssSO_4^{2-} for the KC, KM, and BI cores is shown
10 in Figure 4c (calculated using $k=0.06$). The k' value recalculated for the S100 core
11 is similar to the k' values described by Palmer et al. (2002), and Plummer et al.
12 (2012) at Law Dome, while k'' is similar to the value obtained by Inoue et al. (2017)
13 for a Mill Island coastal core. Negative nssSO_4^{2-} values are more pronounced after
14 the 1950s, which will be discussed in the following sections.

15 **4 Discussion**

16 From the spatial and temporal variability of sea-salt concentrations in the different
17 FIS cores and snow pits discussed here, it seems that more than one mechanism
18 is contributing to the load of sea-salts at FIS, in agreement with the findings by
19 Abram et al. (2013). The ice core data from S100 also suggest that there was a
20 change in sea-salt deposition regime after the 1950s evidenced by an increase, up
21 to six-fold, of median sea-salt concentrations after the 1950s in comparison with the
22 previous 200 years. This increase in concentration is accompanied by a clear shift
23 in nssSO_4^{2-} toward negative values, indicative of SO_4^{2-} fractionation in sea-salts.
24 The negative nssSO_4^{2-} values found in the FIS records could be explained by an
25 enhanced input of sea-salt from (i) windblown frost flowers and/or (ii) aerosol formed
26 after fractionated salty-snow sublimation, with both (i) and (ii) being formed in the
27 neighbouring waters at the eastern flank of FIS. Yang et al. (2008) have reported
28 that aerosol production via (ii) can be more than one-fold larger per unit area than
29 sea-salt production from the open ocean. There is no or very limited amount of multi-
30 annual sea-ice near FIS, and young sea-ice formed during winter in the vicinity of



1 the S100 site is quickly covered by snow due to cyclonic activity. Trajectory studies
2 of air with high sea-salts concentrations and low sulfate/sodium ratios arriving at
3 Halley station, showed that these air masses mainly originate at regions where
4 young sea-ice and frost flowers are formed (Hall and Wolff, 1998; Rankin and Wolff,
5 2002). However, conditions at Halley are not comparable to FIS, since the main
6 easterly or north-northeasterly wind direction prevailing at Halley means an off-land
7 air flow, thus creation of polynyas with open water and consecutive new ice
8 formation, whereas at FIS, and most of the Dronning Maud Land coast, the wind is
9 mainly parallel to the coast or even slightly towards the coast. In particular, a
10 quantification of the areas covered by frost flowers is still missing. It is possible that
11 those areas are comparatively small due to the generally high wind speeds
12 prevailing above the Southern Ocean, resulting in a high percentage of frazil ice,
13 and synoptic conditions lead to the quick development of a snow cover on the young
14 sea-ice. Although it is not possible to apportion the contribution of fractionated sea-
15 salts via (i) or (ii) with the current data, it is plausible that a larger contribution of
16 fractionated aerosol formed from salty-snow than by frost flowers, in agreement with
17 dry deposition of sea-salts as pointed in section 3.5. In addition, frequent stormy
18 conditions in the area are detrimental for the formation of frost flowers, which form
19 under quiet, undisturbed conditions, usually only in leads or small polynyas under
20 the influence of anticyclonic weather. This also means low wind speeds and thus
21 not much transport of frost flowers to the sampling sites at FIS. Thus, mechanism
22 (ii), blowing salty snow formed on thin sea-ice that sublimates during transport to
23 form sea-salt aerosols, appears as a much more probable explanation considering
24 the local meteorological conditions in the study area.

25 Considering the dominant wind direction at FIS (ENE), and the size of frost flowers
26 (10–20 mm) and aerosol formed via (ii) (size >0.95 μm) (Seguin et al., 2014), wind
27 transport would be more efficient from the neighbouring waters to the low elevation
28 sites (e.g. S100 and the snow pit sites) than to the more elevated ice rises sites.
29 This hypothesis is reinforced by the fact that negative nssSO_4^{2-} values are mostly
30 registered in the low elevation sites S100, M1, M2, G3, and G4, but not in the more
31 elevated ice rises sites KC, KM, and BI, where the median nssSO_4^{2-} values for the



1 cores are all positive, and sea-salt concentrations considerably lower than at S100
2 (with the exception of KM, which will be discussed below).
3 The dramatic increase in fractionated sea-salt in the S100 core after 1950s could
4 be associated with an enhanced production of fractionated aerosol and frost flowers
5 in the waters at the eastern flank of FIS as a response to the calving of ice at the tip
6 of FIS (Figure 1), which occurred in 1967 (Vinje, 1975). The longer Trolltunga
7 present before the calving event could have formed a larger bay to the east of it,
8 where compaction of the sea ice occurred due to prevailing easterly winds, resulting
9 in thicker, longer-lasting sea-ice, which limited the sea-spray formation. Such thick
10 sea-ice does not form under post-calving event conditions, e.g. with a shorter
11 tongue. Post-calving event conditions would mean that more sea-spray could be
12 formed and deposited at the FIS sites compared with pre-calving periods. However,
13 sea-spray enhancing alone cannot account for either the increase of sea-salt
14 concentrations or the negative nssSO_4^{2-} found in snow and ice samples. In order to
15 explain the fractionated sea-salt values detected in the S100 cores, there must be
16 an enhanced source of fractionated sea-salts after the calving event. This would be
17 the case if young sea-ice (where fractionation of sea-salts can take place) formed
18 nearby the S100 site as a result of the greater area of open sea available after the
19 calving event. This hypothesis could be tested by closer analysis of satellite and
20 historical sea-ice data, which, however, is beyond the scope of the present study.
21 Other possible mechanisms, such as deposition of sea salts with rime or windblown
22 snow present over multi-annual sea ice, can explain neither the increase in sea salt
23 concentration nor the fractionation observed in S100 after the 1950s. Additionally,
24 annual averages of monthly zonal and meridional wind speeds (ERA40, Uppala et
25 al., 2005) for the area (69°S–71°S, 3.5°W–5°E) between 1955–2001 (Figure 6)
26 show no significant positive trends, thus evidencing that the S100 sea-salt increase
27 after 1950s cannot be related to enhanced transport by wind.
28 Due to the limited time coverage of the KC, KM, and BI cores, we do not know
29 whether there was a relative increase in sea-salt concentrations in the ice rises
30 cores after the 1950s. There is no signal of fractionated sea-salts in any of the ice
31 rises cores (e.g. no significant negative nssSO_4^{2-} values), which suggests that the



1 main source of sea-salts at these sites is wet deposition of sea-spray. Relatively
2 higher sea-salt concentrations measured in the KM core in comparison to the other
3 ice rises cores could be explained by a combination of distance to the sea and the
4 prevailing precipitation and wind conditions in the area: precipitation on FIS is mainly
5 caused by frontal systems of cyclones in the circumpolar trough that move
6 eastwards north of the coast, thus leading to easterly or east-north-easterly surface
7 winds on FIS (Schlosser et al., 2008). This means that even though BI is equally
8 close to the sea as KM (Figure 1), KM has by far the shortest distance to the source
9 of marine aerosols of all three cores, which could explain the comparatively high
10 sea-salt concentrations (Table 1).

11 **5 Conclusions**

12 This study reports subannual and long-term temporal sea-salt and major ion
13 concentration changes measured in three recently drilled firn cores from different
14 ice rises located at Fimbul Ice Shelf (FIS): Kupol Ciolkovskogo, Kupol Moskovskij,
15 and Blåskimen Island, a 100 m long core drilled near the FIS edge (S100), and five
16 snow pits sampled along the ice shelf. We found that Na^+ concentrations decrease
17 with latitude and distance from the sea when low elevation sites are considered
18 (S100 and the FIS snow pits). A significant increase in sea-salts is observed in the
19 S100 core after the 1950s, which is associated with a change in sea-salt sources.
20 This increase in sea-salt concentrations was accompanied by a shift in nssSO_4^{2-}
21 toward negative values, suggesting the input of fractionated sea-salts to the ion load
22 in the S100 core most likely by dry deposition. Such dry deposition regime is not
23 observed in any of the ice rises cores, and evidence of a significant contribution of
24 fractionated sea-salt aerosols to these sites was not found. Consequently, the
25 results of this study suggest that the S100 record contains a more local sea-salt
26 signal, dominated by processes during sea-ice formation in the neighbouring waters,
27 whereas the ice rises cores record the signal of larger-scale conditions of
28 atmospheric flow and transport of sea-salt aerosols produced over open water.
29 These findings are of vital importance for the understanding of the mechanisms of
30 sea-salt aerosol production, transport and deposition at coastal Antarctic sites, and



1 for the improvement of the current Antarctic sea-ice reconstructions based on sea-
2 salt chemical proxies.

3 **6 Data availability**

4 For the chemistry profiles of the KC, KM, BI, and S100 cores, and FIS snow pits,
5 please contact E. Isaksson (elisabeth.isaksson@npolar.no).

6 ERA40 reanalysis data is available at [https://climatedataguide.ucar.edu/climate-](https://climatedataguide.ucar.edu/climate-data/era40)
7 [data/era40](https://climatedataguide.ucar.edu/climate-data/era40) (Uppala, et al., 2005).

8

9 **Acknowledgements**

10 We are grateful to those who helped to collect, transport, sample and analyse the
11 firn cores and snow pits at FIS. We would like to thank V. Goel and J. van Oostveen
12 for providing the 50-m contours and the pre-calving extent of Trolltunga,
13 respectively, used in Figure 1, and T. Maldonado for processing the data for
14 Figure 6. Financial support came from Norwegian Research Council through NARE
15 and the Centre for Ice, Climate and Ecosystems (ICE) at the Norwegian Polar
16 Institute in Tromsø. Additional support was received from University of Costa Rica,
17 network ISONet (project B6-774).



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1 Tables

2 Table 1. Cores (KC, KM, BI, S100), and snow pits (M1, M2, G3–G5) locations and
 3 sampling details. Distances of the core and pit locations to the ice shelf side were
 4 obtained using the GIS package Quantarctica (www.quantarctica.org). (*) refers to
 5 Kaczmarek et al. (2004), and (§) to Vega et al. (2016).

Site	Location	Elevation (m a.s.l.)	Core length Ice depth Ice temp. at 10 m (m)	Distance from the coast (km)	Time coverage (years)	Average SMB (m w.e. y ⁻¹)
KC [§]	70°31'S, 2°57'E	264	20.0 460 -17.5	42	(1958–2012) ± 3	0.24
KM [§]	70°8'S, 1°12'E	268	19.6 410 -15.9	12	(1995–2014) ± 1	0.68
BI [§]	70°24'S, 3°2'W	394	19.5 460 -16.1	10	(1996–2014) ± 1	0.70
S100*	70°14'S, 4°48'E	48	100 - -17.5	3	(1737–2000) ± 3	0.30
Snow pit sampling depth (m)						
M1	70°0'S, 1°2'W	55	0.6	21	Aut.2009– 2009/11/28	0.38 (for the period
M2	70°19'S, 0°7'W	73	0.7	64	Aut.2009– 2009/12/17	1983–2009
G3	69°49'S, 0°37'W	57	0.9	27	Aut.2009– 2009/12/21	inferred from a
G4(G4a)	70°54'S, 0°24'W	60	0.7	117	Aut.2009– 2009/12/19	composite core, see
G5(G5a)	70°33'S, 0°2'W	82	0.6	83	Aut.2009– 2009/12/03	Vega et al. 2016)

6



1 Table 2. Median ion concentrations (in $\mu\text{mol L}^{-1}$) in the KC, KM, BI, and S100 firn/ice
 2 cores and in snow pits M1, M2, G3–G5. Ion concentrations at the top 2 m of the KC,
 3 KM, and BI cores were not measured. Non-detected concentrations were set as half
 4 the detection limit of each ion. Note: (*) the period is 1958.5–2012, (-) not measured.
 5 Median values of water stable isotopes and deuterium excess for the KC, KM, and BI
 6 are reported by Vega et al. (2016).

Site	Period (years)	Median ($\mu\text{mol L}^{-1}$)								
		MSA	Cl ⁻	Br ⁻ ($\times 10^3$)	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
KC	1958–2007	0.2	10.0	-	0.6	1.8	9.4	0.2	0.9	0.5
KM	1995–2012	0.3	71.3	-	0.4	4.5	57.7	1.5	6.3	1.6
BI	1996–2012	0.4	23.1	-	0.4	1.9	19.0	0.5	2.0	0.6
S100	1737–2000	0.1	20.9	-	0.5	1.2	20.7	0.4	2.0	0.7
S100	1995–2000	0.1	132.4	-	0.6	3.2	144.0	3.3	10.7	3.0
S100	1737–1949	0.1	16.0	-	0.6	1.0	15.1	0.3	1.4	0.5
S100	1950–2000	0.1	88.5	-	0.5	2.8	98.2	2.0	7.9	1.9
M1		-	55.9	7.5	0.5	2.1	46.4	0.8	5.6	1.2
M2		-	26.2	5.7	0.9	1.2	18.3	0.3	2.7	0.8
G3	aut.2009–	-	27.1	7.1	0.5	1.4	61.9	1.5	6.9	2.0
G4	sum.2010	-	7.4	2.9	0.4	0.4	5.3	0.1	0.7	0.5
G5	for all snow	-	7.6	3.9	0.4	0.5	5.4	0.2	0.9	0.5
G4a	pits	-	5.6	6.1	0.5	0.3	-	-	-	-
G5a		-	8.1	8.1	0.3	0.4	-	-	-	-



1

Table 3. PCA loadings of the first three (KC) and two (KM, BI, and S100) principal components calculated at an annual resolution in a set of 8 different ions measured in the ice rises and S100 cores. PCA loadings were obtained at three different time intervals in the S100 core: 1737–2000, 1737–1949, and 1950–2000. Sources related to the different components are displayed in the bottom row.

Core	KC			KM		BI		S100					
	Annual			Annual		annual		annual (1737–2000)		annual (1737–1949)		annual (1950–2000)	
Resolution	PC1	PC2	PC3	PC1	PC2								
Loadings	0.17	0.52	-0.19	-0.20	0.64	0.03	0.65	0.16	0.54	0.23	0.44	0.03	0.73
MSA	0.46	-0.17	-0.19	0.40	0.03	0.43	-0.07	0.42	-0.07	0.43	-0.11	0.42	-0.08
Cl	0.13	0.59	0.35	-0.26	0.56	-0.03	0.56	-0.06	0.79	-0.08	0.74	0.14	0.60
NO ₃ ⁻	0.33	0.47	0.08	0.30	0.50	0.30	0.48	0.37	0.23	0.30	0.45	0.38	0.23
SO ₄ ²⁻	0.44	-0.11	-0.22	0.40	0.07	0.43	-0.06	0.42	-0.09	0.43	-0.13	0.42	-0.10
Na ⁺	0.46	-0.19	-0.11	0.40	0.03	0.40	-0.10	0.41	-0.06	0.41	-0.10	0.42	-0.05
K ⁺	0.45	-0.15	0.11	0.39	0.08	0.43	-0.08	0.41	-0.10	0.41	-0.11	0.40	-0.17
Mg ²⁺	0.17	-0.24	0.85	0.40	0.10	0.43	-0.03	0.39	0.02	0.36	0.05	0.39	-0.07
Ca ²⁺	51	22	12	76	18	65	24	70	15	60	17	69	16
Explained Variance (%)	sea-salts	biogenic mixed	terrestrial	sea-salts terrestrial	biogenic mixed								
Source	sea-salts	biogenic mixed	terrestrial	sea-salts terrestrial	biogenic mixed								



1 Table 4. Correlation coefficients (R) for the median ion concentrations at the different
 2 FIS snow pits vs. latitude, longitude, and distance from the ice shelf edge. Numbers in
 3 italics show the R values including the ion median concentration values in the KM and
 4 BI cores for the year 2009. Significant values at the 95 % confidence interval are shown
 5 in bold.

R	Cl ⁻	Br ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
Latitude (°S)	-0.74	-0.96	-0.10	-0.84	-0.93	-0.90	-0.90	-0.95
	<i>-0.62</i>	-	<i>-0.07</i>	<i>-0.45</i>	-0.87	-0.83	-0.88	-0.91
Longitude (°W)	0.81	0.67	-0.40	0.77	0.72	0.59	0.57	0.72
	<i>0.16</i>	-	<i>-0.22</i>	<i>0.30</i>	<i>0.08</i>	<i>0.13</i>	<i>-0.04</i>	<i>0.02</i>
Distance from the sea (km)	-0.84	-0.99	-0.07	-0.91	-0.90	-0.83	-0.83	-0.93
	<i>-0.67</i>	-	<i>0.05</i>	<i>-0.64</i>	<i>-0.70</i>	<i>-0.70</i>	<i>-0.61</i>	<i>-0.70</i>

6



- 1 Table 5. Correlation coefficients (R) for the annual mean chemical concentrations
- 2 (R_{conc}) and chemical fluxes (R_{flux}) vs. annual snow accumulation in the KC and S100
- 3 cores. Significant values at the 95 % confidence interval are shown in bold.

R_{conc}	MSA	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
KC	-0.18	-0.17	-0.24	-0.16	0.05	-0.08	-0.21	-0.12
KM	0.14	-0.06	-0.02	0.01	-0.07	-0.04	0.08	-0.04
BI	-0.31	-0.11	-0.57	-0.52	-0.14	-0.19	-0.13	-0.11
S100	0.07	-0.12	-0.04	-0.10	-0.12	-0.10	-0.13	-0.09
S100 (1737–1949)	0.11	-0.11	-0.05	-0.05	-0.11	-0.12	-0.05	-0.05
S100 (1950–2000)	0.16	-0.09	-0.06	-0.10	-0.08	-0.03	-0.15	-0.03
R_{flux}	MSA	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺
KC	0.45	0.76	0.72	0.63	0.59	0.75	0.74	0.01
KM	0.59	0.65	0.57	0.71	0.64	0.68	0.74	0.73
BI	0.43	0.55	0.42	0.55	0.53	0.49	0.56	0.73
S100	0.18	0.07	0.63	0.18	0.08	0.10	0.16	0.13
S100 (1737–1949)	0.52	0.45	0.61	0.56	0.41	0.44	0.48	0.43
S100 (1950–2000)	0.22	0.25	0.73	0.24	0.30	0.36	0.44	0.27



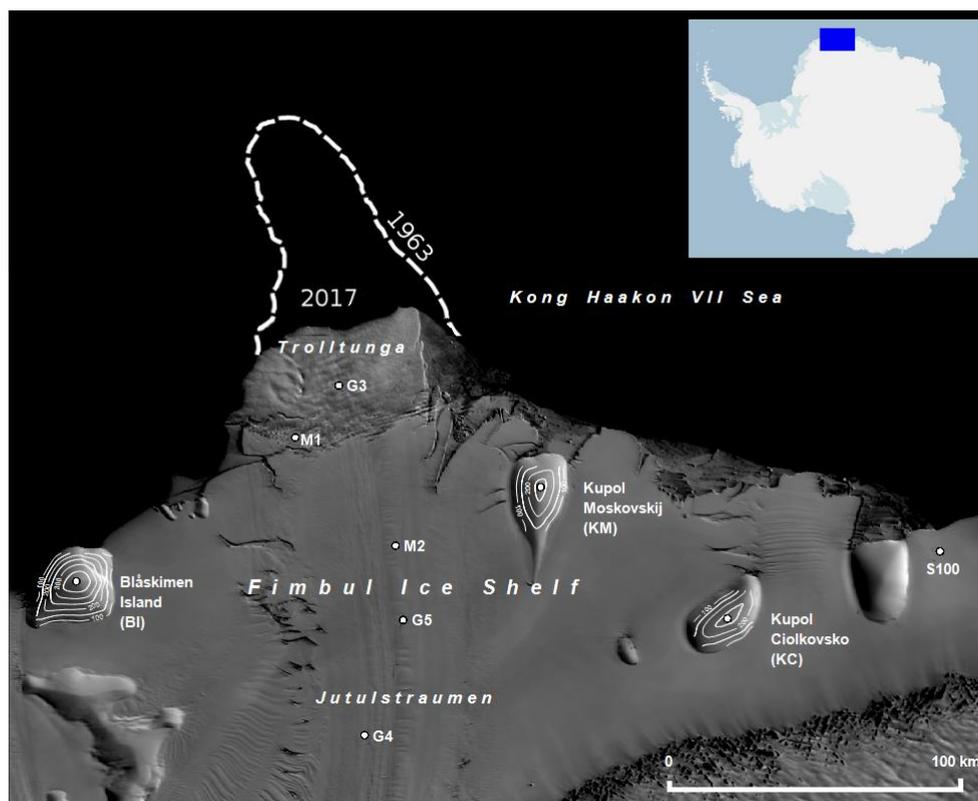
- 1 Table 6. Median nssSO_4^{2-} concentrations (in $\mu\text{mol L}^{-1}$) in the KC, KM, BI, and S100
- 2 firn/ice cores and in snow pits M1, M2, G3–G5. (-) Not re-calculated.

Site	Period (years)	Median ($\mu\text{mol L}^{-1}$) nssSO_4^{2-} $k=0.06$	Median ($\mu\text{mol L}^{-1}$) nssSO_4^{2-} $k=0.04$	Median ($\mu\text{mol L}^{-1}$) nssSO_4^{2-} $k'=0.02$
KC	1958–2007	1.1	-	-
KM	1995–2012	0.7	-	-
BI	1996–2012	0.7	-	-
S100	1737–2000	-7×10^{-2}	0.3	0.7
S100	1995–2000	-4.8	-0.9	0.7
S100	1737–1949	0.1	0.4	0.7
S100	1950–2000	-3.1	-0.7	0.7
M1	Aut.2009–	-0.7	-	-
M2	Sum.2010	-0.3	-	-
G3	for all snow pits	-3.1	-	-
G4		-2×10^{-2}	-	-
G5		0.2	-	-

3

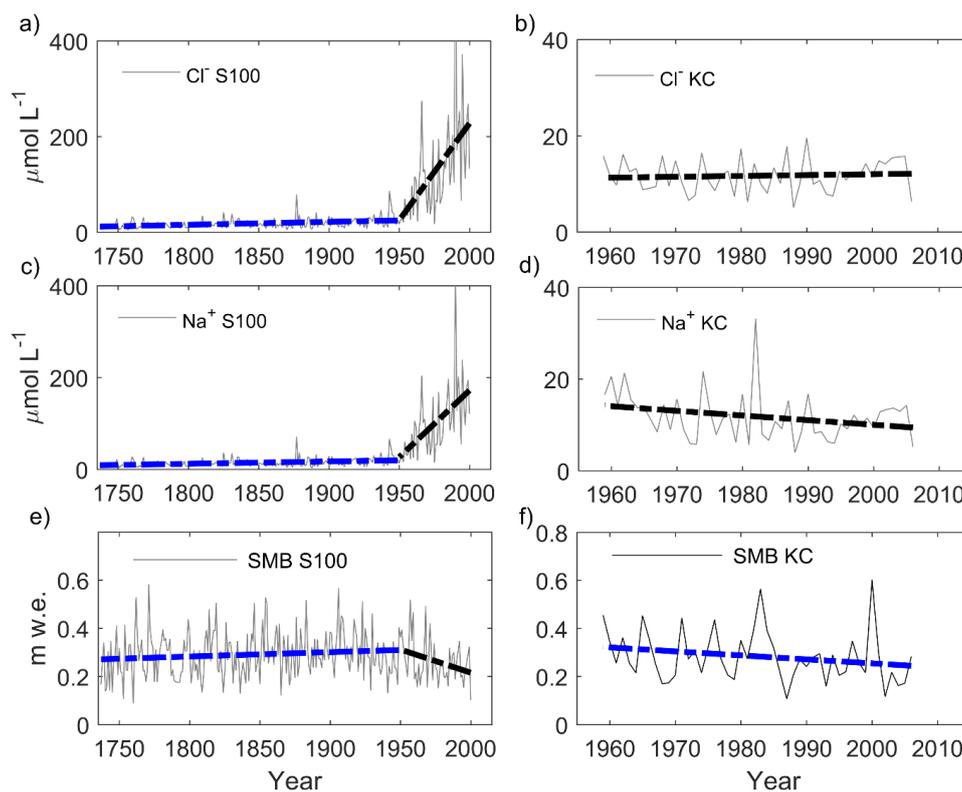


1 Figures



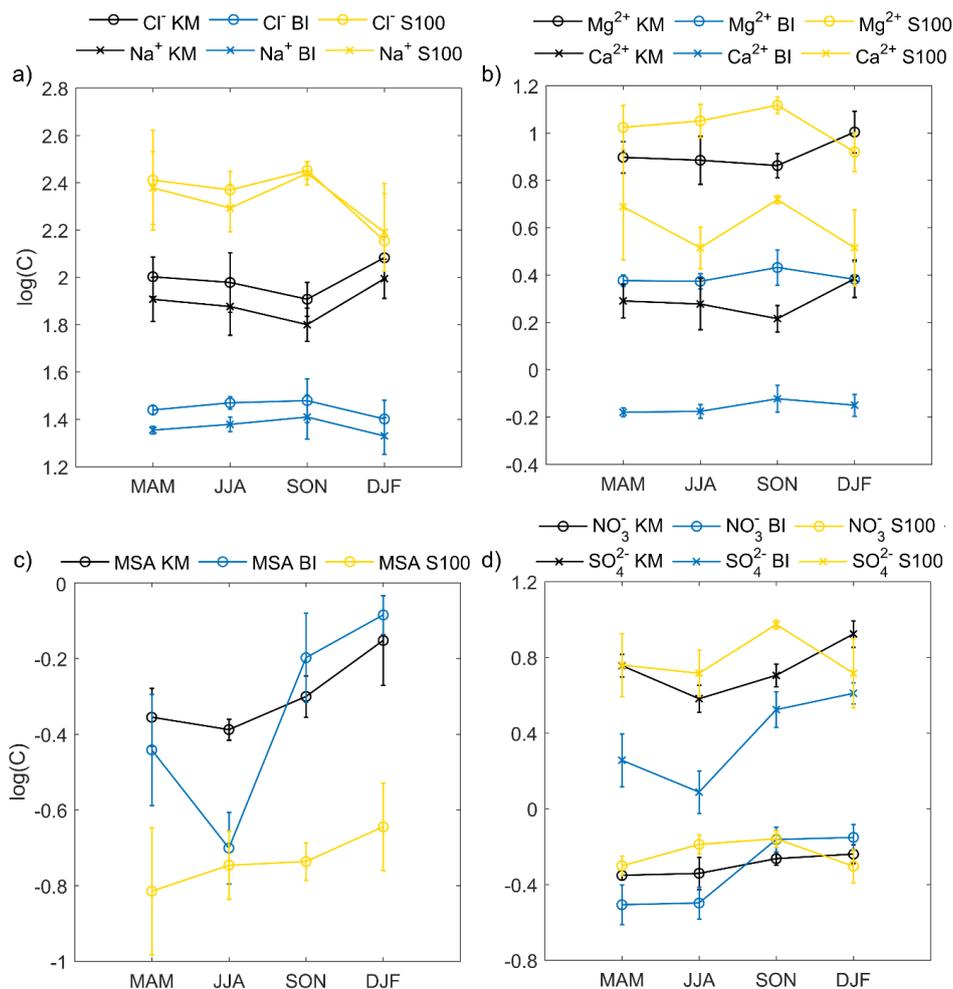
2

3 Figure 1. Satellite image of Fimbul Ice Shelf (FIS) showing the KC, KM, BI, and S100
4 core sites, the M1, M2, G3, G4, and G5 snow pit sites, Jutulstraumen, and Trolltunga.
5 In addition, 50-m contours are shown at each ice rise, as derived from GPS profiles
6 (V. Goel, personal communication, 2016). In addition, the dashed line shows the extent
7 of Trolltunga according to Corona Satellite data from 1963 (J. van Oostveen, personal
8 communication, 2017). Map image is from the MODIS Mosaic of Antarctica
9 (MOA). Map image is from the MODIS Mosaic of Antarctica (MOA). Additional
10 information regarding the sampling sites and traverses in FIS can be found in
11 Schlosser et al. (2014) and Vega et al. (2016).

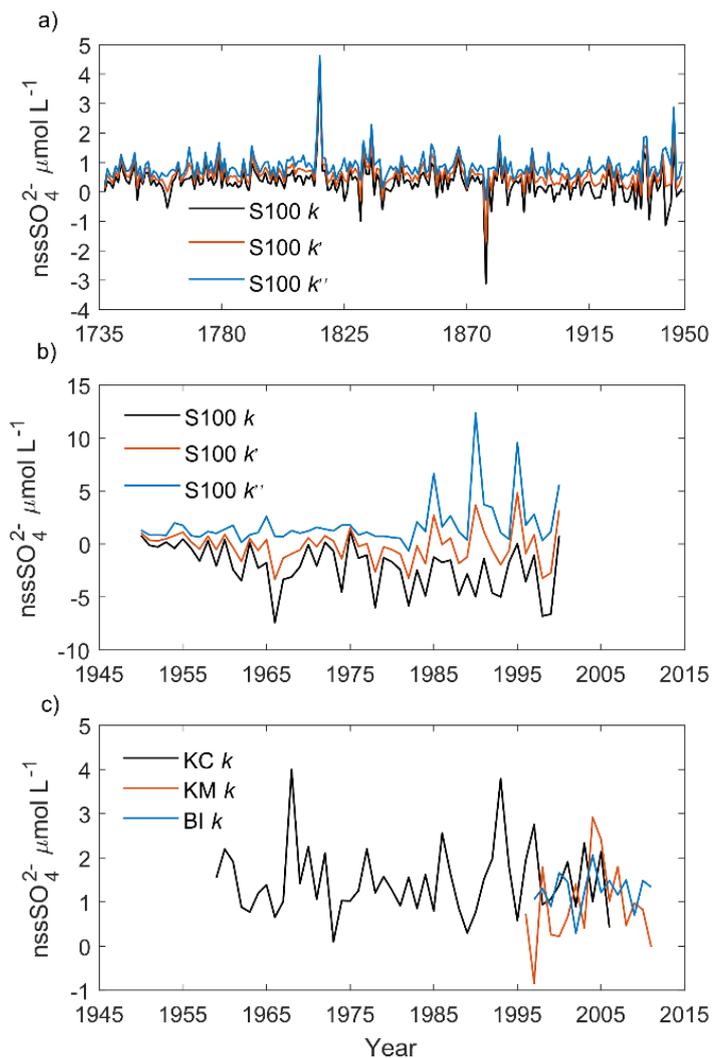


1

2 Figure 2. Annual sea-salt (Cl^- and Na^+) concentrations and surface mass balance
3 (SMB) in the two longest records retrieved at Fimbul Ice shelf, S100, (a), (c), and (e),
4 and KC, (b), (d) and (f). Linear trends in Cl^- and Na^+ concentration, and SMB measured
5 in the S100 core are shown for two different periods: 1737–1949 (blue dashed line)
6 and 1950–2000 (black dashed line) in (a), (c), and (e), respectively. Linear trends in
7 Cl^- and Na^+ concentrations, and SMB measured in the KC core are shown for the
8 period 1958–2007 (black dashed line) in (b), (d) and (f), respectively.

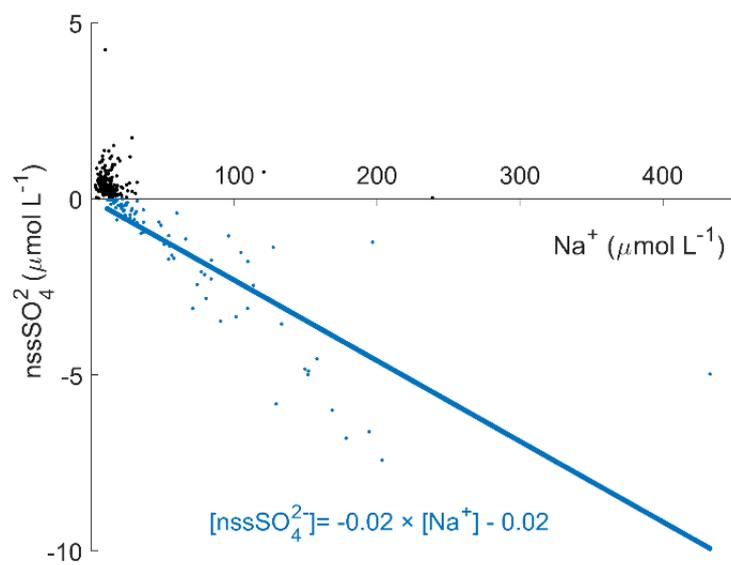


1
 2 Figure 3. Seasonal variability of selected ions, Cl⁻ and Na⁺ (a), Mg²⁺ and Ca²⁺ (b), MSA
 3 (c) and NO₃⁻ and SO₄²⁻ (d) in cores KM, BI, and S100. Average seasonal values were
 4 calculated for the trimesters MAM (autumn), JJA (winter), SON (spring), and DJF
 5 (summer) from the interpolated composite record (0.01-year resolution) of 16, 15, and
 6 5 years in the KM, BI, and S100 cores, respectively.



1

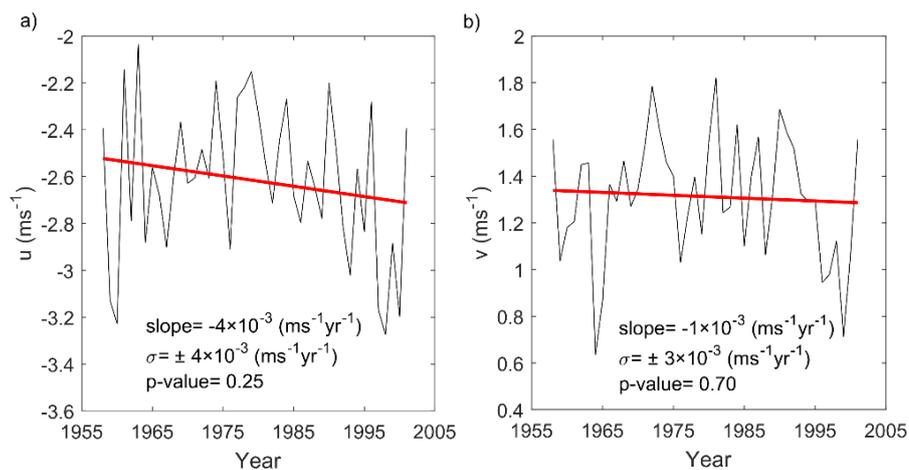
2 Figure 4. Annual nssSO_4^{2-} concentrations in the S100 core between a) 1737–1949, b)
3 1950–2000, and c) in the KC, KM, and BI cores. nssSO_4^{2-} recalculated using $k=0.06$,
4 $k'=0.04$ and $k''=0.02$ are shown in panels a) and b) with black, red and blue lines,
5 respectively. nssSO_4^{2-} in the KC, KM, and BI cores was calculated using $k=0.06$.



2 Figure 5. Scatter plot of annual non-sea-salt SO_4^{2-} vs. Na^+ concentrations in the S100
3 core. nssSO_4^{2-} was calculated using the seawater ratio as described in section 2.3 and
4 using a $k=0.06$ (in $\mu\text{mol L}^{-1}$). Positive nssSO_4^{2-} values are denoted with black dots,
5 while negative values are denoted with blue dots. A linear regression was calculated
6 using the negative nssSO_4^{2-} values to infer a new value of k , k' , following the approach
7 by Inoue et al. (2017).



1



2

3 Figure 6. Annual averages of monthly a) zonal, and b) meridional wind speeds
4 (ERA40) for the area (69°S–71°S, 3.5°W–5°E) between 1958–2001. Slope, standard
5 deviation, and p -value of the linear regression are shown in the figure.

6