*Letter to the Editor regarding the referees' comments on "*Spatial and temporal variability of sea-salts in ice cores and snow pits from Fimbul Ice Shelf, Antarctica*" by C. P. Vega et al.*

Dear Editor:

The authors would like to thank again the referees and the editor for the time taken to revise this manuscript. We agree with the referees comments and suggestions, and consequently, we have done the correspondent modifications in the manuscript text to include their valuable suggestions. We have now provided evidence that the ice shelf in front S100 has moved during the 1950–2000 period bringing the S100 site closer to the ice shelf edge. This was a major concern of referee #1 that we did not address in a previous revision of the manuscript because we did not clearly understand what exactly the petition of the referee was on this point. We have now considered this change in the S100 distance to the FIS edge in the paper discussion and conclusion (sections 4 and 5), and changed the abstract accordingly. We have also considered the correction of the ss- and nss-fractions as suggested by referee #2. In fact, using the equations system to obtain $ssNa^+$, $nssNa^+$, $ssCa^{2+}$, $nssCa^{2+}$ was simple to implement and all ss- and nss-fraction for major ions were re-calculated and corrected in the manuscript Tables, Figures and text. We appreciate the referee's suggestion regarding the calculation and we think that it has considerably improved the method section of the manuscript. In effect, as referee #2 mentioned, our previous assumption of Ca^{2+} having only a crustal origin led us to overestimate $nssNa^+$, however, not in a significant way, and all recalculated median, mean, maxima, and minima ss- and nss- concentrations remained similar. Therefore, the conclusions of the manuscript remain the same after the recalculation of the fractions.

We have included in this letter, a detailed response to the referees' comments done in January 2018. Authors' responses are noted as **CV**. In addition, we provide a marked up version of the manuscript back-tracking the changes made, and a final version of the manuscript without tracked changes.

We consider that all the points made by referees have been covered in this revision, and we hope that you will consider this manuscript now as ready to be published in The Cryosphere.

Sincerely,

C. P. Vega, on behalf of all co-authors

Response to Anonymous Referee #1

Ref. #1. The authors have taken on board most of the comments I made on the earlier version, and although I don't always agree with the conclusions they reach (for example about nitrate as a fertiliser), most of them are now reasonable.

CV. We would like to point out that we have never mentioned nitrate as a fertilizer as a conclusion of the work presented in this manuscript (in any of its versions). We only have referred to previous work done in the Arctic by Wendl et al. (2015) in which they suggest a fertilizing effect of nitrate; those are not conclusions of this manuscript (Vega et al., in revision). We decided to mention Wendl et al. (2015) findings together with findings of other authors (i.e. Mulvaney and Wolff, 1993; Savarino et al., 2007; Wolff et al. 2008; Weller et al. 2011; Pasteris et al., 2014; Sofen et al. 2014) to stress the fact that the interpretation of nitrate concentrations in ice cores is a topic which is still in discussion. We have now re-written the paragraph in order to clearly state what it is explained above. The paragraph in section 3.1 now reads: "Table 3 shows high loadings of NO_3^- and MSA in PC2, and thus, coherence between both species. This correspondence has been previously observed in an ice core from Lomonosovfonna, Svalbard, and a fertilizing effect was proposed as explanation for those findings (Wendl et al., 2015). Wendl et al. (2015) suggest that enhanced atmospheric NO_3^- concentrations

and the corresponding nitrogen input to the ocean can trigger the growth of dimethyl-sulfide-(DMS)-producing phytoplankton. However, there is a variety of possible NO_3^- sources to polar sites, and the relative importance of these sources at certain locations and time is still in discussion (Mulvaney and Wolff, 1993; Savarino et al., 2007; Wolff et al. 2008; Weller et al. 2011; Pasteris et al., 2014; Sofen et al. 2014)."
We hope that it is understood that we are not saying that nitrate has a fertilizing effect in the context of our results, but only refer to Wendl et al. (2015) in the context of nitrate source identification.
We therefore think that we are not reaching any conclusions that referee #1 would not agree with.

Ref. #1. The conclusion about the cause of the increasing concentration at S100 needs further work. Apart from that, the remaining issues with the paper are mainly to do with readability as the changes made are quite rambling and confusing and I suspect many readers will not be able to follow the arguments. In particular:
Section 3.4 and Table 4 should be deleted. The authors have agreed that the pit data cannot be used because they do not represent even a full year. This means that now they are trying to regress 4 data points against 4 variables which obviously cannot be done in any meaningful way. Even where you find significance it looks spurious: for example sulfate is significantly related to latitude because S100 and KM are higher values than the other two sites, but the latitude difference between them is a matter of minutes and it is not credible that this is actually what is causing the difference in sulfate. Simply state that you cannot see any significant relationship between chemistry and the variables assessed, although sea salt is clearly lowest at KC, which is further inland than the other sites.

CV. We agree with the points made by referee #1. Consequently, we have removed section 3.4 and Table 4 (appearing in the previous version of the manuscript). Since the section on ions spatial variability has now been removed, we consider that the title of the paper should be modified. We suggest the title *Variability of sea-salts in ice/firn cores from Fimbul Ice Shelf, Dronning Maud Land – DML, Antarctica*, and we are open to any suggestions the editor may have about it. Regarding ion spatial variability, we have kept the following lines as suggested by referee #1, now in section 3.1: "We found no significant relationship between the median annual ion concentration and latitude, site elevation, and distance from the sea for most of the species, with the exception of annual SO_4^{2-} and annual MSA concentrations which show a significant decrease (at the 95 % confidence level) with latitude, and east longitude, respectively. However, additional ice cores from Fimbul are needed to obtain a robust conclusion on spatial distribution of major ions." We have also added the following sentence in parenthesis in section 3.1, as suggested by the referee: "In general, median concentrations in the KM core are higher than in the other ice-rises cores, e.g. six to eight-fold higher concentrations of Na^+, K^+, Mg^{2+} and Cl^- in the KM core than in the KC core (which is further inland than the other sites) are found for the period 1995–2012.".

Ref. #1. You have now added a lot of SI about the snowpits, which are not used in the text. I appreciate you probably put work into them and are attached to them, but they add nothing to the paper and complicate it for the reader. I propose removing Table S1, S3, and Figs S2 and S3, plus any mention of them in the text.

CV. We thought it would be adequate to keep the snow pit data in the supplementary material, but we agree with the referee that including the data might be confusing for the reader. Consequently, we have now removed the snow pit data from the supplementary material, and do not mention them in the main text anymore.

Ref. #1. The new tables 2, 5, 6 and 7 are unreadably complicated because of the way that different statistics are piled vertically on top of each other. I propose that the authors present these tables with only the mean values (or the medians if they prefer) in the main text. If they really want to show off the other statistics (which are really thesis material and not needed in the paper), then place them in the supplement.

CV. We have followed the referee's suggestion and we placed mean, maximum, minimum, and standard deviations found in tables 2, 5, 6, and 7, in the supplementary material. We only keep median values in Table 2, 4, 5, and 6, as the referee suggested.

Ref. #1. Sections 3.5 and 3.6 are very long and confusing. They need carefully evaluating by all authors who should then try and express the concepts more concisely, pointing carefully to the reduced Tables 5 and 7.

CV. Sections 3.5 and 3.6 are now sections 3.4 and 3.5, respectively. We have edited these sections as much as possible following the referees suggestions.

Ref. #1. The conclusion about the increase at S100 after 1950 is now more credible, but still ignores what seems the obvious answer, and the authors still did not address what I asked about. While it could be that the loss of the ice tongue somehow influenced sea ice formation and caused the change it is not obvious to me why this would affect S100 so strongly but not KC or KM. What I asked is whether the authors are sure that the ice shelf immediately in front of S100 has not retreated over the last 50 years, putting S100 much closer to the sea (something that cannot affect the fixed points at KM and KC). Although the authors point me to glaciological data there is nothing I can see that answers this question, because there is no information about the ice front position, but I can make a guess. S100 is apparently 3 km from the sea today. I would guess the ice front position is almost fixed relative to the ice rise adjacent to it, with regular calving events keeping up with the ice velocity behind it (high resolution images seem to show crevassing near the ice front so this seems likely). The authors say that the ice velocity is "10s-100s of m/a", but if one blows up the Rignot map the velocity in the free-running parts of Fimbul all appear to be in blue shades, ie at least 100 m/a. Then over 50 years the site S100 has moved at least 5km, and is now that much closer to the ice front, and out of any "shadow" from the unnamed ice rise next to it. A move from eg 8 km to 3 km from the sea might well be enough to explain the data. (As an example a paper by Gorlach et al (FRISP report 2, page 48) shows sea salt concentration increasing by a factor 4 over about 10 km near Neumayer, moving from what I estimate to be about 13 to 3 km from the ice edge).

CV. The referee points to a very important issue, and we have now included a contour line in Figure 1 that shows the extent of FIS in 1963, not only around Trolltunga (as in the previous version of the manuscript), but also down to the S100 site. As it can be seen in the FIS edge contours shown in Figure 1, by 1963 the FIS ice front in the vicinity of the S100 site was located about 17 km further north than at present. As the referee pointed out, S100 would also have got closer to the ice edge, moving at least 5 km closer to the sea over the 1950–2000 period. We argue that the calving of Trolltunga in 1967, the change in the ice front position and the ice advance in the vicinity of the S100 site led to the increase in sea-salts observed in the S100 core after 1950, but did not significantly change sea-salt concentrations in the ice-rises cores, which are located at fixed points on FIS. We have now included this hypothesis in the discussion and conclusions, and changed the abstract accordingly, and we think that the point made by the referee has now been addressed properly. We also would like to note that we have no access to the reference suggested by the referee, i.e. Gorlach et al (FRISP report 2, page 48) and therefore, we could not include it.

Ref. #1. This suggests that this section of the paper needs a further iteration as this seems a much simpler explanation than yours. I know that you are trying to explain a change in the nss-sulfate contribution as well, but this is actually poorly constrained. The nss sulfate is very obviously negative in the last 50 years because the sea salt is so high in concentration and dominates. In the pre-1950 period, the partitioning of sulfate between ss and nss is much less safe, and it remains plausible that the sea salt part may still be depleted. In any case whatever you choose to say, you cannot just ignore the likelihood that S100 is just a much more coastal site as time progresses.

CV. As we mentioned above, we think that we have now addressed the point made by the referee regarding the change in distance between the S100 site and the ice edge. Changes have been done accordingly in section 4, 5 and in the abstract.

Response to Anonymous Referee # 2

Ref. #2. I'm happy to see that Authors made almost all the changes I suggested and the text was improved accordingly. For this reason, I think that the manuscript is now ready to be published on The Cryosphere journal. However, I have to note a potentially relevant error in calculating the ss-and nss-fractions of Na and Ca. If I have well understood, Authors used totCa as a marker of the crustal source and calculated nss-Na by the Ca/Na ratio in the Earth crust. This is not correct because also Ca can have two main sources: mineral dust and sea spray. This is particularly true for coastal sites, where the ssCa contribution could be relevant or even dominant. In order to calculate more reliable ss- and nss-fractions of Na and Ca, Authors should use the following 4-equation system:
totNa = ssNa + nssNa
totCa = ssCa + nssCa
ssNa = totNa - 0.562 nssCa
nssCa = totCa - 0.038 ssNa
where 0.562 = Na+/Ca2+ (w/w) in the crust (Bowen, 1979), and 0.038 = Ca2+/Na+ (w/w) in seawater (Nozaki, 1997).
Bowen, H.J.M., 1979. Environmental Chemistry of the Elements. Academic Press, London.
Nozaki, Y., 1997. A fresh look at element distribution in the North Pacific. http:/www.agu.org/eos_elec/97025e.html.
By solving the equation system (4 equations, 4 unknown variables), a more correct evaluation of ss-Na can be obtained.
If Authors consider that all the Ca comes from the crustal sources, nssNa fraction can be over-estimated. As a consequence, the critical parameter ssNa can be significantly under-estimated and the error can propagate to the nss-SO4 calculation.
I'm aware that using the correct procedure to calculate a reliable ss-Na fraction could imply many changes in the manuscript figures and text, if ss-Na values obtained with the equation system are significantly different from those obtained by using totCa as crustal marker. Therefore, I'd suggest that the Authors test the differences in ss-Na calculation with the two different procedures, at least for a sub-dataset of samples in which the contribution of ss-Ca appears to be relevant (i.e., for samples enriched in sea salt, for which totCa cannot be used a-priori as crustal marker). If this test is positive (i.e., if the ss-Na values calculated with the two methods are quite close each other), Authors could leave unchanged figures and text. On the contrary, ss- and nss-fraction of all the components (Na, Ca, sulfate etc.) should be revised.

CV. We appreciate the correction made by the referee regarding the calculation of $ssNa^+$ and $nssCa^{2+}$. We have now implemented the equations system and therefore we recalculated all ss- and nss-fractions using this approach. We explain the methodology in section 2.3. We used seawater and crustal composition given by Summerhayes and Thorpe (1996), and Lutgens and Tarbuck (2012). As mentioned before in this letter, the fractions did not significantly change, and therefore the main results of the manuscript remained the same. We appreciate the referee's suggestion regarding the ss- and nss-fraction calculations, and we think that the methods section of the manuscript has been improved accordingly.

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~~Spatial and temporal v~~Variability of sea-salts in ice/firn cores from Fimbul Ice Shelf, Dronning Maud Land – DML, Antarctica

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Abstract.

Major ions were analysed in firn/ice cores located at Fimbul Ice Shelf (FIS), Dronning Maud Land – DML, Antarctica. FIS is the largest ice shelf in the Haakon VII Sea, with an extent of approximately 36 500 km^2. Three shallow firn cores (about 20 m deep) were retrieved in different ice-rises, Kupol Ciolkovskogo (KC), Kupol Moskovskij (KM), and Blåskimen Island (BI), while a 100 m long core (S100) was drilled near the FIS edge. These sites are distributed over the entire FIS area so that they provide a variety of elevation (50–400 m a.s.l.) and distance (3–42 km) to the sea. Sea-salt species (mainly Na^+ and Cl^-) generally dominate the precipitation chemistry in the study region. We associate a significant six-fold increase in median sea-salt concentrations~~s~~, observed in the S100 core after the 1950s,~~-~~ to an enhanced exposure of the S100 site to primary sea-salt aerosol due to a shorter distance from the S100 site to the ice front, and ~~with~~to enhanced sea-salt aerosol production from blowing salty snow over sea ~~-~~ice, most likely related to the calving of Trolltunga occurred during the 1960s. This increase in sea-salt concentrations is synchronous with a shift in non-sea-salt sulfate ($nssSO_4^{2-}$) toward negative values, suggesting a possible contribution of fractionated aerosol to the sea-salt load in the S100 core most likely originating from salty snow found on sea~~-~~ ice. In contrast, there is no evidence of a significant contribution of fractionated sea-salt to the ice-rises sites, where

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the signal would be most likely masked by the large inputs of biogenic sulfate estimated for these sites. In summary, these results suggest that the S100 core contains a sea-salt record dominated by the proximity of the site to the ocean, and processes of sea~~-~~ ice formation in the neighbouring waters. In contrast, the ice-rises firn cores register a~~the~~ larger-scale signal of atmospheric flow conditions and a less efficient transport of sea-salt aerosols to these sites. These findings are a contribution to the understanding of the mechanisms behind sea-salt aerosol production, transport and deposition at coastal Antarctic sites, and ~~for~~ the improvement of the current Antarctic sea~~-~~ ice reconstructions based on sea-salt chemical proxies obtained from ice cores.

1 Introduction

Antarctic ice and firn cores contain valuable information about the climate and atmospheric chemical composition of the past and provide evidence for the important role of Antarctica in the global climate system. Numerous ice and firn cores have been drilled in Antarctica during the past decades (Stenni et al., 2017). However, relatively few cores were drilled in coastal regions, which are more sensitive to changes in climate than the dry and cold interior of Antarctica. In fact, two recent review papers point out the lack of ice core data from low elevation coastal areas when discussing Antarctic climate variability (Stenni et al., 2017; Thomas et al., 2017). In an effort to understand the role of ice shelves in stabilizing the Antarctic ice sheet, particular focus has been laid on the investigation of ice-rises and ice~~-~~-rumples as buttressing elements within the ice sheet - ice shelf complex (Paterson, 1994; Matsuoka et al., 2015). Furthermore, due to their radial ice flow regime, generally low ice velocities, and relatively high surface mass balance (SMB), ice-rises are potentially useful sites for ice core retrieval (Philippe et al., 2016; Vega et al., 2016). Firn and ice cores drilled at ice-rises allow obtaining high-resolution climate records to investigate sub-annual and long-term temporal changes in the loads of different chemical compounds found in the snow, providing information about their sources and transport, particularly of sea-salt ions, such as sodium (Na^+) and chloride (Cl^-), which are strongly modulated by sea~~-~~ ice extent and meteorological conditions. Recent modelling efforts to study the use of sea-salts as proxies for past se~~a-~~a ice extent have shown that, under present climate conditions and on interannual timescales, meteorological conditions rather than sea~~-~~ ice extent are the dominant factor modulating atmospheric sea-salt concentrations that are deposited at the interior and coastal sites in Antarctica (Levine et al., 2014). However, sea-salts have the potential as proxy for sea ~~-~~ice extent at glacial-interglacial scales when large changes in sea~~-~~ ice extent took place (Levine et al., 2014).

At most Antarctic sites, atmospheric sea-salt concentrations present maxima during austral winter (Wagenbach et al., 1998; Weller and Wagenbach, 2007; Jourdain et al., 2008; Udisti et al., 2012), with the exception of Dumont D'Urville where maxima occur during summer (Wagenbach et al., 1998). Similarly, sea-salt fluxes obtained from Antarctic ice cores also show winter maxima (Abram et al. 2013 and references therein). However, in some recent core records from coastal sites, no clear seasonality is observed, e.g. at Mill Island during the period 1934–2000 (Inoue et al., 2017). Abram et al. (2013) conclude that despite the seasonal signal registered in different Antarctic ice cores, sea-salt fluxes do not show a consistent relationship with

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sea ~~-~~ice extent on inter-annual timescales, and on the contrary, are highly dependent on atmospheric transport, and/or the presence of polynyas.

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

In the review by Abram et al. (2013), the authors suggest that the brine-frost flower system is a plausible source of sea-salt aerosols to coastal Antarctic sites. This hypothesis is supported by the experimental evidence that the original seawater SO_4^{2-}/Na^+ ratio cannot be used in the non~~-~~-sea-salt sulfate ($nssSO_4^{2-}$) calculations, leading to negative $nssSO_4^{2-}$ values both in winter aerosol and fresh snow sampled at coastal sites (Hall and Wolff, 1998; Wagenbach et al., 1998; Curran et al., 1998; Rankin and Wolff, 2002 and 2003), and also in ice cores from both inland (Wagenbach et al., 1994, Kreutz et al., 1998) and coastal sites (Inoue et al., 2017). These negative values indicate that a lower SO_4^{2-}/Na^+ ratio has to be used in $nssSO_4^{2-}$ calculations, i.e., a depletion of SO_4^{2-} with respect to seawater composition~~,~~ occurred in wet and dry deposition.

During the process of sea ~~-~~ice formation, ions present in the water are not incorporated in the ice crystal matrix, but remain as highly concentrated brine in brine pockets or channels. The brine can be transported by capillary effects through brine channels to the newly formed ice surface, resulting in a thin layer of highly saline surface brine. This fractionated brine is unlikely to be a direct source of sea-salts because it usually quickly gets covered by snow, and no clear mechanism has been found to explain how this brine could become airborne (Abram et al., 2013). With further cooling of the ice, the volume of brine decreases and consequently, its salinity increases, leading to the precipitation of different saline compounds. This depends on temperature, e.g. sodium sulfate or mirabilite ($Na_2SO_4 \cdot 10\ H_2O$) starts to precipitate at temperatures below −8 °C, ~~and~~ while sodium chloride (NaCl) at temperatures below −26 °C. Consequently, the remaining brine is depleted in sodium and sulfate ions via precipitation of mirabilite at relatively mild polar temperatures. Frost flowers can form from this brine when meteorological conditions are adequate, i.e. at low intensity winds, which allows these delicate structures to grow without breaking apart, and on very thin ice where a strong temperature gradient is present between the ice surface and the overlying air (Rankin et al., 2000; Rankin and Wolff, 2002, and references therein). Thus, frost flowers formed at temperatures below −8 °C will be depleted in sodium and sulfate relative to other ions present in seawater (Rankin et al., 2000; Rankin and Wolff, 2002), evidenced by negative $nssSO_4^{2-}$ values measured in aerosols and snow (see section 2.3 for more details on the calculation of the nss-fractions).

For most of the last decade, frost flower formation, transport and deposition, has been considered the most plausible mechanism behind the fractionated aerosol detected at coastal areas. However, Yang et al. (2008 and 2010), and Huang and Jaeglé (2017) proposed an alternative mechanism: the origin of sea-salt aerosol could be due to the sublimation of blowing salty snow. This salty snow could be a result of frost flower formation, upward migration of brine within the snow (Massom et al., 2001), or by

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the input of sea-spray from the open ocean or nearby leads or polynyas (Dominé et al., 2004). Flooding of sea~~-~~ ice under the weight of accumulated snow can also induce increased salinity of snow (Massom et al., 2001). As the snow can be contaminated or wetted with fractionated brine or frost flowers, it could be expected that this salty snow also shows such fractionation. As pointed by Yang et al. (2008 and 2010), this salty snow can be transported by wind and if the air is not saturated, the snow particles may lose water by sublimation and become sea-salt aerosols. These aerosols could then be transported and deposited either by dry or wet deposition, depending on local meteorology.

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

This study discusses sub-annual and long-term temporal changes in sea-salt and major ion concentration measured in three recently drilled firn cores from different ice-rises located at Fimbul Ice Shelf (FIS): Kupol Ciolkovskogo, Kupol Moskovskij, and Blåskimen Island, a 100 m long core drilled near the FIS edge (S100)~~, and five snow pits (Table S1, Supplementary material) sampled along the ice shelf~~ (Figure 1). The main goals of the present study are to investigate possible mechanisms behind deposition, sub-annual, and spatial variability of sea-salts in this coastal region. The results presented here contribute to bridging the data gap existent at coastal Antarctic sites, and to the improvement of current Antarctic sea~~-~~ ice reconstructions based on sea-salt chemical proxies.

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2 Methods

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2.1 Study area

With an extent of approximately 36 500 km^2, FIS is the largest ice shelf in the Haakon VII Sea (Figure 1). Fed by Jutulstraumen, the largest outlet glacier in DML, FIS is divided into a fast moving ice tongue, Trolltunga, directly feeding the central part of the ice stream, and slower surrounding parts. Several ice-rises (250–400 m a.s.l.; 10–42 km from the coast) are found at FIS, varying in size from 15 to 1200 km^2, and located approximately 200 km apart.

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Early investigations in this area began during the International Geophysical Year (IGY) 1956/57 (Swithinbank, 1957; Lunde, 1961; Neethling, 1970) and continued during the last decades with focus on surface mass balance (SMB) variability in space and time (Melvold et al., 1998; Melvold, 1999; Rolstad et al., 2000; Isaksson and Melvold, 2002; Kaczmarska et al., 2004; Kaczmarska et al., 2006; Divine et al., 2009; Sinisalo et al., 2013; Schlosser et al., 2012, 2014; Langley et al., 2014; Vega et al., 2016). However, studies on spatial and temporal variability of chemical composition of snow and ice from this area are limited to water stable isotopes interpretations (Kaczmarska et al., 2004; Schlosser et al., 2012, 2014; Vega et al., 2016).

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

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

2.2 Sampling

Three shallow firn cores (about 20 m deep) were retrieved at different ice-rises (Kupol Ciolkovskogo (KC), Kupol Moskovskij (KM), and Blåskimen Island (BI), Figure 1, Table 1), located at FIS between January 2012 and January 2014 during field expeditions organized by the Norwegian Polar Institute (NPI). Location, elevation, and length of the different ice-rises cores are presented in Table 1. Each core was drilled from the bottom of a 2 m snow pit (not sampled for major ions). The firn density was determined as bulk density of each sub-core piece (average length of 45 cm)~~, and of each snow pit interval (20 cm)~~. The samples were collected following clean protocols (Twickler and Whitlow, 1997), shipped frozen to NPI, and later to the Paul Scherrer Institute (PSI), Switzerland, for cutting and chemical analysis. Sample resolution varied between 4 and 8 cm depending on sample depth and density. Thickness of ice lenses, water stable isotope ratios and SMB for the three ice-rises are reported in Vega et al. (2016). Additionally, unpublished major ion concentrations measured in the 100 m deep S100 core drilled in austral summer 2000/2001 (Kaczmarska et al., 2004) were included in this study (Figure 1, Table 1). The S100 core was sampled at 5 cm resolution between top and 6 m deep, and then at 25 cm resolution between 6 m to 100 m deep.

2.3 Chemical analyses

Major ions (methanesulfonic acid (MSA), Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+} and Ca^{2+}) present in the three firn cores from the ice-rises were analysed at PSI using a Metrohm ProfIC 850 ion chromatograph combined with an 872 Extension Module and auto-sampler. The precision of the method was within 5 % and detection limits (D.L.) were below 0.02 μmol L^{-1} for each ion (Wendl et al., 2014). Ion concentrations (MSA, Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+} and Ca^{2+}) in the S100 core were measured at the British Antarctic Survey (BAS) using fast ion chromatography (Littot et al., 2002). The reproducibility of the measurements was 4–10 %.

Major ion concentrations were separated into sea-salt (ss) and non-sea-salt (nss) fractions calculated from the mean seawater composition using $ssNa^+$ as standard ion, as:

$[nssX]=[X]_{total} - k_{seawater} \times [ssNa^+]$, and

$[ssX] = k_{seawater} \times [ssNa^+]$

where

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$$k_{\mathrm{seawater}} = \frac{[\mathrm{X}]_{\mathrm{seawater}}}{[\mathrm{Na}^{+}]_{\mathrm{seawater}}}$$

with k_{seawater} values listed in Table S1 in the Supplementary material.

To calculate the $ssNa^{+}$ fraction, we used the following equations system, in which $nssCa^{2+}$ was employed as reference ion to obtain the $nssNa^{+}$ associated to Earth's crustal material:

$$[\mathrm{Na}^{+}]_{\mathrm{total}} = [\mathrm{ssNa}^{+}] + [\mathrm{nssNa}^{+}]$$

$$[\mathrm{Ca}^{2+}]_{\mathrm{total}} = [\mathrm{ssCa}^{2+}] + [\mathrm{nssCa}^{2+}]$$

$$[\mathrm{ssNa}^{+}]_{\mathrm{total}} = [\mathrm{Na}^{+}]_{\mathrm{total}} - k_{\mathrm{crustal}}[\mathrm{nssCa}^{2+}]$$

$$[\mathrm{nssCa}^{2+}]_{\mathrm{total}} = [\mathrm{Ca}^{2+}]_{\mathrm{total}} - k_{\mathrm{seawater}}[\mathrm{ssNa}^{+}]$$

where $k_{\mathrm{crustal}} = \frac{[\mathrm{Na}^{+}]_{\mathrm{crust}}}{[\mathrm{Ca}^{2+}]_{\mathrm{crust}}} = 1.40$ (using molar concentrations) (Lutgens and Tarbuck, 2012), and $k_{\mathrm{seawater}} = \frac{[\mathrm{Ca}^{2+}]_{\mathrm{seawater}}}{[\mathrm{Na}^{+}]_{\mathrm{seawater}}} = 0.02$, (using molar concentrations, Table S1) (Summerhayes and Thorpe, 1996).

~~Cl^{-}, SO_4^{2-}, K^{+}, and Mg^{2+} non sea-salt fractions (nss) were calculated from the mean seawater composition using the sea-salt Na^{+} fraction ($ssNa^{+}$) as standard ion (section 3.5), using:~~

~~$[\mathrm{nssX}] = [\mathrm{X}]_{\mathrm{total}} - k \times [\mathrm{ssNa}^{+}]$,~~

~~where~~

~~$k = \frac{[\mathrm{X}]_{\mathrm{seawater}}}{[\mathrm{Na}^{+}]_{\mathrm{seawater}}}$,~~

~~using the standard mean chemical composition of seawater with ion concentration expressed in □mol L^{-1} (k values are listed in Table S2), and where~~

~~$[\mathrm{ssX}] = k \times [\mathrm{ssNa}^{+}]$~~

Due to the low concentrations of NO_3^{-} in standard seawater (Summerhayes and Thorpe, 1996), NO_3^{-} was not separated into nss- and ss-fractions (i.e., NO_3^{-} was assumed to have a nss-origin only, as well as MSA). ~~The $nssNa^{+}$ and $ssNa^{+}$ fractions were calculated using Ca^{2+} as reference ion and k= 1.40 for Earth's crust composition (Lutgens and Tarbuck, 201200) (section 3.5).~~ In addition, water stable isotopes analyses of the KC, KM and BI cores are described in Vega et al. (2016); while analysis of the S100 core is described in (Kaczmarska et al., 2004).

2.4 Firn and ice core timescales

The timescales of the KM and BI cores were obtained based on annual layer counting of water stable isotope ratios (δ~~□~~^{18}O), and found to cover the periods between austral winter-1995(96) and summer-2014, respectively. The error in the dating was estimated as ± 1 year for both of these cores (Vega et al., 2016). Both KC and the S100 cores were dated using a combination of annual layer counting of δ~~□~~^{18}O and identification of volcanic horizons, ~~(~~i.e. by using the SO_4^{2-}, dielectric profiling (DEP),

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and electrical conductivity measurements (ECM)~~)~~, with timescales covering the time period 1958–2012 (± 3 years) at KC (Vega et al., 2016), and 1737–2000 (± 3 years) at S100 (Kaczmarska et al., 2004).

3 Results

3.1 Ion concentrations and sources

Table 2 shows ~~M~~median~~,~~ ~~mean, maximum, minimum, and standard deviation (□) of~~ concentrations for all ions measured in the cores~~.~~. In addition, mean~~are shown in Table 2~~, maximum, minimum, and standard deviation (σ) are shown in Table S2, and~~.~~ ~~B~~box-plots of raw ion concentrations in the different cores are shown in Figure S1 in the Supplementary material. ~~In addition, median, mean, maximum, minimum, and □□ of concentrations for all ions measured in the FIS snow pits are shown in Table S3, while boxplots are shown in Figures S2 and S3 in the Supplementary material.~~

In general, median concentrations in the KM core are higher than in the other ice-rises cores, ~~and snow pits,~~ e.g. ~~about~~ six to eight-fold higher concentrations of Na^+, K^+, Mg^{2+} and Cl^- in the KM core than in the KC core (which is further inland than the other sites) are found for the period 1995–2012. The relatively high Na^+ and Cl^- concentrations observed in the KM core are also detected in the upper meters of the S100 core (in the periods 1995–2000, and 1950–2000, respectively, Table 2). Similarly high values have been reported in several snow and firn samples from other western DML coastal sites (Kärkäs et al., 2005), and in Mill Island, Wilkes Land (Inoue et al., 2017). We found ~~Ion concentrations in the snow pits (Table S3) are in reasonable agreement with firn and ice core values and with ion concentration ranges for snow pits previously sampled at FIS (Mulvaney et al., 1993).~~ no significant relationship between median annual ion concentration and latitude, site elevation, and distance from the sea for most of the species, with the exception of annual SO_4^{2-} and annual MSA concentrations which show a significant decrease (at the 95 % confidence level) with latitude, and east longitude, respectively. However, additional ice cores from Fimbul are needed to obtain a robust conclusion on spatial distribution of major ions.

~~Temporal and spatial variability of ion concentrations are explored in more detail in the following sections.~~

In order to assess the most important sources explaining the total variance in the glacio-chemical records from FIS, a principal component analysis (PCA) was applied to the different ion series measured ~~at~~ in the KC, KM, BI, and S100 cores. Years~~,~~ in which no sub-annual concentrations were available in the S100 (1793, 1841, 1866, 1918, and 1944) due to low resolution, were filled in by linearly interpolating between the annual means of the previous and following year. For the PCA analysis, the logarithms of the raw concentrations were used, ~~(~~at sub-annual (using the raw values as input) and annual resolutions,~~)~~ and standardized by subtracting the mean of the data series from each data point and then dividing the result by the standard deviation of the data series. Due to the sampling resolution, only the KM and BI cores were comparable at a sub-annual level. PCA analyses were performed for three different periods of the S100 core: for the entire time interval spanning 1737–2000, for the subsection between 1737–1949, and between 1950–2000.

The sum of the variances of the first three principal components (PC1, PC2 and PC3) was ≥ 80 % of the total variance of the original sub-annual and annual data in all cores. Since the results of the sub-annual and annual PCA analysis are similar only

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the annual results are considered. The loadings of the first three (KC) and two (KM, BI, and S100) principal components are shown in Table 3. PCA results are consistent between the different cores. Consequently, the ions can be separated in two main groups: sea-salts species (Na^+, Cl^-, K^+, Mg^{2+}, and Ca^{2+}) and marine-biogenic/mixed (MSA, SO_4^{2-}, including NO_3^-) (Table 3). Generally, our results indicate that the major sources of ~~the~~ ions at the different sites are the same, independent of the core site and mean concentrations of ions in the cores. Only at the KC site the PCA results imply an additional input of Ca^{2+} from other sources than sea-salt, as for instance mineral dust. Table 3 shows ~~H~~high loadings of NO_3^- and MSA in PC2, and thus, coherence between both species~~,~~. This correspondence ~~have~~ has been previously observed in an ice core from Lomonosovfonna, Svalbard ~~(Wendl et al., 2015)~~, and a fertilizing effect was proposed as explanation for those findings (Wendl et al., 2015). Wendl et al. (2015) suggest that enhanced atmospheric NO_3^- concentrations and the corresponding nitrogen input to the ocean can trigger the growth of dimethyl-sulfide-(DMS)-producing phytoplankton. However, there is a variety of possible NO_3^- sources to polar sites, and the relative importance of these sources at certain locations and time is still in discussion (Mulvaney and Wolff, 1993; Savarino et al., 2007; Wolff et al. 2008; Weller et al. 2011; Pasteris et al., 2014; Sofen et al. 2014).

~~In order to investigate ion spatial variability at FIS, we used median annual ion concentrations in the different ice-rises cores (KC, KM and BI), and S100 for the overlapping period between 1997 and 2000, and compared them with latitude, longitude, site elevation, and distance from the sea (obtained from the GIS package Quantarctica, www.quantarctica.org) (Table 4). Only annual SO_4^{2-} and annual MSA concentrations show a significant decrease (at the 95 % confidence level) with latitude, and east longitude, respectively. No significant relationship is found between the median annual ion concentrations and latitude, site elevation, and distance from the sea for any of the species. These findings contrast with previous studies from western Dronning Maud Land (WDML) (Stenberg et al., 1998), where a strong correlation between sea-salt concentrations and distance from the sea was found in this area. We attribute the lack of significance for the correlations presented in Table 4 to the local effects on annual SMB due to topography and local meteorology at the KM and BI sites, reported by Vega et al. (2016).~~

~~3.3~~3.2 Long-term variability of ion concentrations

We use the two longest available records for FIS (KC and S100) to explore the long-term temporal variability of major ions, with special focus on sea-salts, represented by Cl^- and Na^+ (Figure 2). In the S100 core, Na^+, Cl^-, K^+, and Mg^{2+} median concentrations show a marked six-fold increase after the 1950s. However, there is no significant increase of the concentration of these species in the KC core. Due to its limited time coverage it cannot be determined if there was a substantial relative increase in concentrations at this site after the 1950s. MSA and NO_3^- concentrations do not show such marked increase in the S100 core and values agree between both cores after the 1950s (Figure S2~~4~~ in the Supplementary material). Consequently,

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three periods can be distinguished in the S100 record: (i) the period between 1995–2000, comparable to the time covered by the KM and BI cores; (ii) the period between 1737–1949, where ion concentrations remain low; and, (iii) the period between 1950–2000, where sea-salt concentrations increased (Table 2 and Table S2).

With the exception of MSA, all ions show a positive trend (significant at the 95 % confidence level) during the period 1950–2000, although the slope for NO_3^- is three orders of magnitude smaller than for the other ions~~ (slope and error of the linear regression are shown in Table S4 in the Supplementary material)~~. Such significant linear trend was not observed in the KC ion record over the same period (slope and error of the linear regression are shown in Table S3).~~.~~

Ions, with the exception of MSA, also show a positive and significant trend between 1737–1949~~,~~ (Table S~~4~~3), however, the increase is less marked than during the 1950–2000 period.

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~~3.4~~3.3 Sub-annual variability of ion concentrations

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The lack of extensive precipitation measurements at sub-annual resolution near the sampling sites at FIS, makes a precise reconstruction of the precipitation regime at the area difficult. To obtain a time scale for the KC, KM, and BI ice-rises cores, Vega et al. (2016) employed $\delta^{18}O$ winter minima and summer maxima, and assumed uniform precipitation throughout the year at the core sites. The assumption was made on the basis of precipitation data for DML reported by Schlosser et al. (2008), which showed high temporal variability in the monthly sums due to the influence of cyclone activity affecting both, coastal and inland regions. In addition, at Neumayer station (70º 39' S, 8º 15' W), the closest to the ice-rises core sites, two precipitation maxima (April and October) are identifiable for the period 2001–2006, ~~possibly~~ a manifestation of the semi-annual oscillation of the circumpolar trough (Schlosser el al., 2008). Considering the above, to investigate the sub-annual variability of the different ion groups in the KM, BI, and S100 cores, we associated the winter minima and summer maxima in $\delta^{18}O$ determined in the KC, KM, and BI cores (Vega et al., 2016), and in the S100 core (Kaczmarska et al., 2004), with the months of July and January, respectively. The values for April and October were derived by interpolation between January–July, and July–January, respectively, in each core time scale. We defined *summer samples*, as samples within November and April (NDJFMA), and *winter samples*, as samples within May and October (MJJASO). Summer and winter mean concentrations were then calculated based on logarithms of raw ion concentrations expressed in µmol L^{-1}. Ion concentrations were not available at the top 2 m (removed before drilling), therefore, the composite year consisted of 16 (1996–2011) and 15 (1997–2011) complete years for the KM and BI cores, respectively. In the S100 core, sub-annual variability were investigated only during the period 1995–2000, where the concentrations have sufficient temporal resolution. The resulting summer and winter mean concentrations in the cores are presented in Figure 3.

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Sea-salt species (Na^+ and Cl^-, Figure 3a) show lower concentrations during summer in the BI, and S100 core, whereas in the KM core summer and winter show similar means. Both Mg^{2+} and Ca^{2+} (Figure 3b) show similar means in both summer and winter. MSA concentrations (Figure 3c) show summer maxima in all three cores, with a higher summer to winter difference

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in the BI core, compared with the KM, and S100 cores. These summer maxima are in agreement with the main source of MSA (marine-biogenic), most active during the warmer months. The MSA winter minimum is not as pronounced in the KM core as in the BI core, while the lowest MSA minimum is reached in the S100 core. NO_3^- and SO_4^{2-} concentrations (Figure 3d) show a distinct increase toward the summer in the BI core, which is also observed in the KM core, although less mark~~defin~~ed. KM, and BI SO_4^{2-} concentrations are higher in the summer, while both NO_3^- and SO_4^{2-} summer and winter means are similar in the S100 core.

~~3.5 Ions spatial variability~~

~~In order to investigate ion spatial variability at FIS, we used median annual ion concentrations in the different ice-rises cores (KC, KM and BI), and S100 for the overlapping period between 1997 and 2000, and compared them with latitude, longitude, site elevation, and distance from the sea (obtained from the GIS package Quantarctica, www.quantarctica.org) (Table 4). Only annual SO$_4^{2-}$ and annual MSA concentrations show a significant decrease (at the 95 % confidence level) with latitude, and east longitude, respectively. No significant relationship is found between the median annual ion concentrations and latitude, site elevation, and distance from the sea for any of the species. These findings contrast with previous studies from western Dronning Maud Land (WDML) (Stenberg et al., 1998), where a strong correlation between sea-salt concentrations and distance from the sea was found in this area. We attribute the lack of significance for the correlations presented in Table 4 to the local effects on annual SMB due to topography and local meteorology at the KM and BI sites, reported by Vega et al. (2016).~~

~~3.8~~3.4 Sea-salt and ~~non~~non-~~-~~sea-salt fractions

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PCA results presented in section 3.1 show two main groups in which ions can be separated: sea-salts (ss-fraction), and marine biogenic/mixed (nss-fraction). In order to confirm the common sea-salt source for Na^+ and Cl^-, we calculated the Cl^-/Na^+ ratio, and ion sea-salt and ~~non-sea~~non-sea-salt fractions. Table 4~~5~~ shows median~~the~~ Cl^-/Na^+ ratios (~~expressed in~~using concentrations in µ~~□~~mol L^{-1}) in the KC, KM, BI, and S100 cores. Medians of the Cl^-/Na^+ ratio in the ice-rises cores are equal (KC, and BI) or slightly higher (KM) than the expected ratio in sea-water (i.e., $Cl^-/Na^+= 1.2$), while Cl^-/Na^+ medians in the S100 core are lower than the expected ratio in sea-water, both before and after 1950. Maxima in the Cl^-/Na^+ ratio vary between 1.5–3.8, and minima between 0.1–0.9 (Table S4). These results show a clear difference in the Cl^-/Na^+ ratio~~s~~ between the ice-rises cores and the S100 core, i.e. a Cl^- to Na^+ ~~unbalance~~ imbalance in the S100 core associated to an excess of Na^+. This excess of Na^+ can be due to the recombination of biogenic SO_4^{2-} with ssNa^+, and/or to additional nssNa^+ sources (Legrand and Delmas, 1988). This ~~unbalance~~ imbalance can further be enhanced by a depletion of Cl^- due to shorter sea-salt atmospheric residence time~~s~~, and HCl loss from snow (Legrand and Delmas, 1988; Wagnon et al., 1999). HCl loss becomes significant at relatively low snow accumulation rates (Röthlisberger et al., 2003; Benassai et al., 2005), below the accumulation rate reported for the S100 site, therefore, it is unlikely that HCl loss is a dominant factor that could account for the low Cl^-/Na^+ ratios at this site. Cl^-

depletion by recombination of ssCl^- with atmospheric acids is dependent on the acidic condition of the atmosphere, especially sulfuric acid (H_2SO_4), linked to marine biogenic emissions. Due to the seasonality of~~ ~~ biogenic sulphur~~ biogenic emissions in polar regions~~, ~~it is expected that~~ the Cl^-/Na^+ ratio would present lower values ~~predominantly~~ during the summer months compared to the winter season (Jourdain and Legrand, 2002). Sub-annual Cl^-/Na^+ ratios (estimated as explained in section 3.3) in the S100 core show values of 1.4 ± 0.5 for the winter period, and 1.2 ± 0.1 for the summer period. Since the temporal resolution of the S100 core only allows sub-annual values for the period 1995–2000, is not possible to assess a sub-annual pattern on the Cl^-/Na^+ ratio, and Cl^- depletion by acidification cannot be ruled out as mechanism to explain the low ratios registered in the S100 core during the last centuries. In addition to Cl^- loss, low Cl^-/Na^+ ratios can also be a product of excess Na^+ from ~~non sea~~non-sea-salt sources (nssNa^+), as for example crustal material from snow-free coastal areas, nunataks, or dust transported from other continents. ~~In order to estimate nssNa^+, ssNa^+, and the percentage of crustal nssNa^+ to total Na^+, we used Ca^{2+} as reference ion, therefore, assuming Ca^{2+} only has a crustal origin (Mahalinganathan et al., 2012) and using a Na^+/Ca^{2+} ratio of 1.40 (with concentrations expressed in □mol L^{-1}) for Earth's crust (Lutgens and Tarbuck, 201200). This assumption will introduce an overestimation of the nssNa^+ fraction proportional to the ratio Ca^{2+}/Na^+= 0.02 (with concentrations expressed in □mol L^{-1}) in standard seawater, that is not considered when Ca^{2+} is assumed to only have crustal origin. This procedure offers an alternative to obtain nssNa^+, without using Cl^- as reference ion, and the ratio Na^+/Cl^- in bulk seawater.~~ Table ~~5~~4 shows the~~ nssNa^+,~~ ssNa^+, nssNa^+, (calculated as explained in section 2.3) and percentage of mean nssNa^+ to mean total Na^+ in the different cores. Since some of the calculated ssNa^+ values in the KC core were negative (5 % of the values), ssNa^+ statistics in Table ~~5~~4 and Table S4 are shown considering all data points, and only positive ssNa^+ values. The KC core presents the largest contribution of nssNa^+ to total Na^+ with a 19~~21~~ % in comparison to the KM, BI, and S100 cores (0.6~~3~~ %, 1.4 %, and ~~5~~0.5 %, respectively), which is in agreement with PC3 in Table 3 pointing to a strong source of Ca^{2+} to the KC site.

As mentioned in section 2.3, we used the ssNa^+ fraction ~~obtained above~~ to calculate nss- and ss-fractions for Cl^-, SO_4^{2-}, K^+, and Mg^{2+}, while ssCa^{2+} and nssCa^{2+} were obtained using the equations system described in section 2.3. ~~(~~Table ~~6~~5~~)~~ shows median concentrations of ss- and nss-fractions for Cl^-, SO_4^{2-}, K^+, and Mg^{2+}, and Ca^{2+}, while ssNa^+ and nssNa^+ median concentrations are shown in Table 4. Complementary statistics are shown in Table S4 and Table S5. The sea-salt fraction clearly dominates in all ions, with the exception of SO_4^{2-} in the KC~~, and BI~~ core,~~s,~~ which shows almost three times more nssSO_4^{2-} than ssSO_4^{2-}. Nss-fractions often have negative values which can be associated to an ssNa^+ enrichment or to a depletion of major ions found in snow in comparison to bulk seawater, i.e. ion fractionation. Negative nss-fractions represent a higher percentage of total values at the S100 core compared to the ice-rises cores, with values up to ~~90~~ 93 % for the S100 (1950–2000) (for nssCl^-), and up to 51~~43~~ % for the ~~KM~~ BI core (for nssMg^{2+}).

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~~3.9~~3.5 Evidence for increased fractionated nss-SO$_4^{2-}$ after 1950s

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The nssSO$_4^{2-}$ fraction contains all SO$_4^{2-}$ sources besides sea-salts, e.g. marine biogenic emissions, and volcanic emissions. In coastal regions, most of the nssSO$_4^{2-}$ can be attributed to marine biogenic activity via DMS oxidation (Legrand et al., 1992) with maxima in concentrations during the summer (Minikin et al., 1998). To evaluate if ion fractionation is evidenced in the core records, i.e. ~~nssSO$_4^{2-}$~~ the snow is strongly depleted in ssSO$_4^{2-}$ relative to ssNa$^+$ (Rankin and Wolff, 2002), leading to an underestimation of nssSO$_4$~~$^{2-}$~~, it is necessary to account for the biogenic contribution to total nssSO$_4^{2-}$ at each core. In sites where biogenic SO$_4^{2-}$ production is high, this fraction could mask a ssSO$_4^{2-}$ depletion. Legrand and Pasteur (1998) have estimated MSA/nssSO$_4^{2-}$ ratios of 0.18 (annual), 0.29 (summer), and 0.86 (winter) (with concentration in µ~~□~~mol L^{-1}) in aerosol collected at Neumayer station, Antarctica. Median MSA/nssSO$_4^{2-}$ ratios calculated in the KC, KM, BI, and S100 cores (Table 6; complementary statistics are shown in Table S6~~7~~) span a range between 0.~~1~~ and 0.3, therefore, closer to the annual and summer values reported by Legrand and Pasteur (1998). Using an annual MSA/nssSO$_4^{2-}$ ratio of 0.18 (Legrand and Pasteur, 1998) and the MSA concentrations measured in the KC, KM, BI, and S100 cores, we estimated the biogenic portion of nssSO$_4^{2-}$ (bio-nssSO$_4^{2-}$) in all the cores to assess the percentage of mean bio-nssSO$_4^{2-}$ to mean total SO$_4^{2-}$ (Table ~~7~~6). In the ~~KM and~~ BI core~~s~~, the estimation of bio-nssSO$_4^{2-}$ surpasses the total SO$_4^{2-}$ observed in the~~se~~ core~~s~~, while in the KC, and KM cores the bio-nssSO$_4^{2-}$ would represent about 5~~0~~8 % and 46 % of total SO$_4^{2-}$, respectively. These high percentages were expected especially in the KC core ~~, and BI cores,~~ in which the nssSO$_4^{2-}$ fraction dominates over ssSO$_4^{2-}$ (section 3.~~5~~4). In the S100 core, bio-nssSO$_4^{2-}$ varies according to the time period considered with percentages three times higher during the period 1737–1749 (72 %), than the period 1950–2000 (24 %). It is important to bear in mind the estimation of bio-nssSO$_4^{2-}$ when assessing the possible effect of fractionated aerosols as a source of sea-salts to the snow. In the ice-rises cores, the high estimated bio-nssSO$_4^{2-}$ percentages would most likely mask any ssSO$_4^{2-}$ depletion in sea-salt aerosols, making fractionation hard to evidence; consequently, fewer negative nssSO$_4^{2-}$ values or the absence of them in the ice-rises cores would not directly indicate that there is no input of ~~SO$_4^{2-}$~~ fractionat~~ion~~ed ~~in~~ sea-salts ~~found in snow~~to the sites but rather reflect the dominance of the bio-nssSO$_4^{2-}$ fraction in these sites. In the S100 core, this could be relevant for the pre-1950 period in which estimated bio-nssSO$_4^{2-}$ accounts for 72 % of total SO$_4^{2-}$.

In order to evaluate the possible effect of fractionated aerosols as a source of sea-salts to the snow on FIS, we use the ~~used~~ calculated ~~the~~ nssSO$_4^{2-}$ fraction (section 2.3) ~~calculated as described in section 2.3, using *k* values of 0.06 (Table S2, Supplementary material)~~. The percentage of mean nssSO$_4^{2-}$ relative to mean total SO$_4^{2-}$ is one and a half- to three-times higher in the KC core than in the other ice-rises cores, KM and BI. Negative median nssSO$_4^{2-}$ values were obtained in the S100 core, ~~and snow pits M1, M2, and G3 (not shown),~~ with negative nssSO$_4^{2-}$ values being more pronounced after the 1950s (Table 5). These negative values found in the snow, i.e. the sea-salt content in snow is strongly depleted in ssSO$_4^{2-}$ relative to seawater composition, suggest a possible role of frost flowers and wind-blown salty snow as source of sea-salts (Rankin and Wolff, 2002) to the S100 core (Figure 4a y b, black line). To assess the degree of fractionation of ssSO$_4^{2-}$ in the cores in respect to seawater, we obtained the linear regression between annual nssSO_4^{2-} (both positive and negative nssSO_4^{2-} data points) and annual ssNa^+ for the periods 1737–2000 ~~(Figure 5)~~, 1737–1949, and 1950–2000, using a robust regression method that is known to be less sensitive to a possible heteroscedasticity and non-Gaussianity of the model residuals (which is a common problem for ion concentration data) than the usual least squares method. We obtained negative slope values of 0.04, 0.03, and 0.04 for the 1737–2000, 1737–1949, and 1950–2000 periods, respectively. Figure 5 shows a scatter plot of annual nssSO_4^{2-} vs. ssNa^+ for the 1737–2000 period. Following the approach by Wagenbach et al. (1998), we calculated corrected ~~k~~ $k_{seawater}$ values (k') by subtracting the absolute value of the linear regression slope from the constant $k_{seawater}$ ~~k~~ $= \frac{[SO_4^{2-}]}{Na^+}$ in seawater (Table S~~2~~1), i.e. $k'_{1737–2000}$ = 0.02, $k'_{1737–1949}$ = 0.03, and $k'_{1950–2000}$ = 0.02. The k' values recalculated for the S100 core are lower than k' values described by Palmer et al. (2002), and Plummer et al. (2012) at Law Dome ($k'_{Law\ Dome}$ = 0.04, with concentrations expressed in µmol L^{-1}), and similar to the k' value obtained by Inoue et al. (2017) for a Mill Island coastal core ($k'_{Mill\ Island}$ = 0.03, with concentrations expressed in µmol L^{-1}). Wagebach et al. (1998) reported winter k' of 0.02 (with concentrations expressed in µmol L^{-1}) associated to airborne sea-salt particles experiencing ssSO_4^{2-} depletion in respect to seawater, with a depletion factor ($k = k/k'$) of 5.5 for a firn core drilled at eastern Ronne Ice Shelf. The S100 core presents depletion factors of two for the period 1737–1949, and three for the period 1950–2000.

The annual nssSO_4^{2-} fraction, without the effect of sulfate fractionation, was then recalculated in the S100 core using the values for k' of 0.02 and 0.03 (Table ~~8~~7, and Figure 4a y b, red and blue lines, respectively).

4 Discussion

From the spatial and temporal variability of sea-salt concentrations in the different FIS cores discussed here, it ~~seems~~ was found that ~~more than one~~several mechanisms ~~is~~are contributing to the load of sea-salts at FIS, in agreement with the findings by Abram et al. (2013). The ice core data from S100 also suggest that there was a change in sea-salt deposition regime after the 1950s evidenced by an increase, up to six-fold, of median sea-salt concentrations after the 1950s in comparison with the previous 200 years. Although a negative trend in SMB has been observed in the S100 and KC cores for the second half of the 20th century (Figure 2e and f) (Vega et al., 2016), the 0.2 % m w.e. y^{-1} decrease in accumulation registered in the S100 core after 1950 (Table S~~43~~2) cannot account for the increase observed in sea-salt concentrations after 1950s. This increase in concentration is accompanied by a clear shift in nssSO_4^{2-} toward negative values, indicative of ssSO_4^{2-} depletion in sea-salts measured in the core in comparison to bulk seawater, with ssSO_4^{2-} depletion factors of two for the period 1737–1949, and three for the period 1950–2000.

The negative nssSO_4^{2-} values found in the FIS records could be explained by an enhanced input of sea-salts from (i) windblown frost flowers and/or (ii) aerosol formed after fractionated salty-snow sublimation, with both (i) and (ii) being formed in the neighbouring waters at the eastern flank of FIS. Yang et al. (2008) have reported that aerosol production via (ii) can be more than one-fold larger per unit area than sea-salt production from the open ocean. There is no or very limited amount of multi-annual sea~~-~~ ice near FIS, and young sea~~-~~ ice formed during winter in the vicinity of the S100 site is quickly covered by snow

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due to cyclonic activity. Trajectory studies of air with high sea-salt~~s~~ concentrations and low SO_4^{2-}/Na^+ ratios arriving at Halley station, showed that these air masses mainly originate at regions where young sea~~-~~ ice and frost flowers are formed (Hall and Wolff, 1998; Rankin and Wolff, 2002). However, conditions at Halley are not comparable to FIS, since the main easterly or north-northeasterly wind direction prevailing at Halley means an off-land air flow, thus creation of polynyas with open water and consecutive new ice formation, whereas at FIS, and most of the Dronning Maud Land coast, the wind is mainly parallel to the coast or even slightly towards the coast. In particular, a quantification of the areas covered by frost flowers is still missing. It is possible that those areas are comparatively small due to the generally high wind speeds prevailing above the Southern Ocean, resulting in a high percentage of frazil ice, and synoptic conditions lead to the quick development of a snow cover on the young sea~~-~~ ice. Although it is not possible to apportion the contribution of fractionated sea-salts via (i) or (ii) with the current data, it is plausible that a larger contribution of fractionated aerosol formed from salty-snow than by frost flowers, based on recent experimental evidence that frost flowers would not be a direct source of sea-salt aerosols (Yang et al., 2017). In addition, frequent stormy conditions in the area are detrimental for the formation of frost flowers, which form under quiet, undisturbed conditions, usually only in leads or small polynyas under the influence of anticyclonic weather. This also means low wind speeds and thus not much transport of frost flowers to the sampling sites at FIS. Thus, mechanism (ii), blowing salty snow formed on thin sea~~-~~ ice that sublimates during transport to form sea-salt aerosols, appears ~~as~~ to be a much more probable explanation considering the local meteorological conditions in the study area.

Considering that we found no correlation between ion concentrations~~s~~ and site elevation~~ (section 3.4)~~, a decrease in wind transport efficiency of frost flowers (size of 10–20 mm) and aerosol formed via (ii) (size >0.95 μ~~□~~m) (Seguin et al., 2014) due to increased elevation cannot be addressed to explain the lower sea-salt values observed at the ice-rises compared to the S100 site. ~~As mentioned in section 3.4, l~~Local effects on annual SMB due to topography and meteorology at the KM and BI sites~~,~~ reported by Vega et al. (2016)~~,~~ are most likely involved in the different load of sea-salt to these sites.

The dramatic increase in fractionated sea-salt in the S100 core after the 1950s could be associated with a~~n~~ ~~enhanced~~ greater exposure of the S100 site to primary aerosol, in addition to an~~d~~ enhanced production of fractionated aerosol, evidenced by a dominance of negative $nssSO_4^{2-}$ values after 1950. Figures 2~~-~~a and c show that sea-salts started to increase after 1950 with a marked peak corresponding to the year 1966 (± 3 years). According to Rignot et al. (2011), ice velocities near S100 were in the order of ~~10s~~ 100s m y^{-1} for the period 2007–2009, therefore, the S100 site has moved at least 5 km closer to the ice front between 1950 and 2000. ~~, We hypothesize that the increase observed in sea-salts from 1950 could be linked to an increase in ice velocities in comparison to the 1737–1949 period~~In addition~~; and that~~, as ~~it~~ can be ~~observed~~seen in the FIS contours shown in Figure 1, by 1963 the ice front ~~at~~in the vicinity of the S100 site was located about 17 km further north than its present position~~north than the present location of the ice front~~. As a consequence, the distance from the S100 site to open water would have changed through time and~~This change in ice front position, and the ice advance in the vicinity of the S100 site,~~ this would likely caused ~~led to~~ a~~n~~ ~~enhanced~~greater input of sea-spray to the S100 ~~core~~site by shortening the distance to the open sea. However, sea-spray enhancing alone cannot account for the increase of sea-salt concentrations and the negative $nssSO_4^{2-}$ found

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in the S100 samples. We hypothesize that the calving of Trolltunga, which occurred in 1967 (Vinje, 1975) (Figure 1), ~~the calving event occurred at Trolltunga in 1967 (Vinje, 1975) (Figure 1), enhanced the input of fractionated sea-salts to the S100 core by~~ modified~~ying~~ the sea~~-~~ ice conditions north of the ~~around~~ S100 coring site, enhancing the input of fractionated sea-salts which then ~~, leading~~caused ~~to~~ the marked ~~peak found in~~ sea-salt peak appearing ~~s~~ in 1966 (± 3 years). Furthermore, additional ~~This could be~~ support for an effect of the Trolltunga calving event is~~ted by~~ the fact that negative $nssSO_4^{2-}$ ~~values~~ slowly decreased between 1950–1966, showing a marked minimum around 1966 (± 3 years) (Figure 4~~-~~b)~~, which could have been caused by the Trolltunga calving event~~. The ~~longer~~ much extended Trolltunga which was present before the calving event would have ~~could have~~ formed a larger bay to the east~~ of it~~, favourable for a ~~where compaction of the~~higher concentration of sea ice ~~occurred due~~caused by ~~to~~ the prevailing easterly winds, resulting in both thicker and~~,~~ longer-lasting sea~~-~~ ice~~,~~ which would ~~which limited~~limit~~ing~~ ~~the~~ sea-spray formation. Such thick sea ~~-~~ice does not seem to form under post-calving event conditions, e.g. with a shorter tongue. ~~Post-calving event conditions would mean that more sea-spray could be formed and deposited at the FIS sites compared with pre-calving periods.~~ ~~However, sea-spray enhancing alone cannot account for either the increase of sea-salt concentrations or the negative $nssSO_4^{2-}$ found in snow and ice samples.~~ In order to explain the fractionated sea-salt values detected in the S100 cores, there must be an enhanced source of fractionated sea-salts after the calving event. This would be the case if young sea~~-~~ ice (where fractionation of sea-salts can take place) formed near~~by~~ the S100 site as a result of the greater area of open sea available after the calving event. Thicker, long-lasting sea~~-~~ ice present before the calving event would have been a more stable substrate, prone to less flooding through cracks and leads, and ~~most likely~~ w~~ill~~ould also likely ~~present~~have red~~a red~~uced ~~snow~~ salinity content in the snow ~~in comparison~~compared to young sea-ice (Massom et al., 2001). ~~Following the same supposition argument as~~Considering the conjecture by Rhodes et al. (2017), i.e. that young sea~~-~~ ice ~~would be~~is more saline than multi-year ice, it can be expected that sea-salt aerosols produced by blowing snow over sea~~-~~ ice would have higher sea-salt concentrations when young-ice is formed than when multi-year sea~~-~~ ice is formed, in coherence with the proposed hypothesis. The higher sea-salt concentrations ~~-~~and the negative $nssSO_4^{2-}$ values ~~registered~~ found in S100 after the Trolltunga calving~~detachment~~, could thus be explained by a combination ~~an enhanced contribution~~ of shortening the distance from~~of~~ the S100 site to the ice shelf front, and an enhanced contribution of sea-salt aerosols entrained by blowing salty snow found over young sea~~-~~ ice formed near the S100 site. If the air is unsaturated, water in these snow particles will sublimate producing fractionated sea-salt aerosol. As schematized in Figure 2 in Rhodes et al. (2017), the sea-salt aerosol can be transported inland and be deposited either by dry or wet deposition. Since sea-salt concentrations are much higher at the S100 core than in the ice-rises cores, it is plausible that most of the flux of sea-salts at the S100 site is due to dry deposition, due to the short distance from the coast and low elevation, while deposition at the ice-rises would be balanced between the wet and dry regimes. Rhodes et al. (2017) found a marked gradient in the ratio between sea~~-~~ ice sea-salts and~~to~~ oceanic sea-salts (produced by bubble bursting) with distance to the source, ~~ratio~~ in Arctic sites, with~~hit~~ higher ratios closer to the sea ~~-~~ice source and when the sample location is in the path between sea~~-~~ ice and prevailing winds. To test the hypothesis presented here, a closer analysis of satellite and historical sea~~-~~ ice data and a model-based study to

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estimate the spatial and elevation gradient of sea~~-~~ ice sea-salts to FIS can be done, which, however, is beyond the scope of the present study.

Other possible mechanisms, such as deposition of sea-salts with rime or windblown snow present over multi-annual sea~~-~~ ice, can explain neither the increase in sea salt concentration nor the fractionation observed in S100 after the 1950s. Additionally, annual averages of monthly zonal and meridional wind speeds (ERA40, Uppala et al., 2005) for the area (69°S–71°S, 3.5°W–5°E) between 1955–2001 (Figure 6) show no significant positive trends, thus evidencing that the S100 sea-salt increase after 1950s cannot be related to enhanced transport by wind.

Due to the limited time coverage of the KC, KM, and BI cores, we do not know whether there was a relative increase in sea-salt concentrations in the ice-rises cores after the 1950s influenced by the Trolltunga calving. Due to the large input of bio-nssSO_4^{2-} to the ice-rises sites, any possible signal of fractionated sea-salts in any of the ice-rises cores could be easily masked by the biogenic fraction (e.g. no significant negative nssSO_4^{2-} values would be observed). Relatively higher sea-salt concentrations measured in the KM core in comparison to the other ice-rises cores could be explained by a combination of distance to the sea and the prevailing precipitation and wind conditions in the area: precipitation on FIS is mainly caused by frontal systems of cyclones in the circumpolar trough that move eastwards north of the coast, thus leading to easterly or east-north-easterly surface winds on FIS (Schlosser et al., 2008). This means that even though BI is equally close to the sea as KM (Figure 1), KM has by far the shortest distance to the source of marine aerosols of all three ice~~-~~-rises cores, which could explain the comparatively high sea-salt concentrations (Table 2~~1~~).

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5 Conclusions

This study reports sub-annual and long-term temporal sea-salt and major ion concentration changes measured in three recently drilled firn cores from different ice-rises located at Fimbul Ice Shelf (FIS): Kupol Ciolkovskogo, Kupol Moskovskij, and Blåskimen Island, and a 100 m long core drilled near the FIS edge (S100). ~~No significant relationship is found between the median annual ion concentrations and latitude, site elevation, and distance from the sea for any of the species, and only annual SO_4^{2-} and MSA concentrations show a significant decrease (at the 95 % confidence level) with latitude and east longitude, respectively.~~ A significant increase in sea-salts is observed in the S100 core after the 1950s, which is associated with an enhanced exposure of the S100 site to primary sea-salt aerosol associated with a shorter distance from the S100 site to the ice front during the last decades, and an enhanced input of fractionated sea-salts. This increase in sea-salt concentrations was accompanied by a shift in nssSO_4^{2-} toward negative values, which suggest~~ings the~~ input of fractionated sea-salts to the ion load in the S100 core most likely by enhanced ~~ing~~ sea-salts production by blowing salty snow over sea~~-~~ ice. Due to the large input of bio-nssSO_4^{2-} to the ice-rises cores, it is hard to assess the degree of ssSO_4^{2-} depletion in snow in comparison to bulk seawater at these sites. Consequently, the results of this study suggest that the S100 record contains a sea-salt record dominated by processes of sea ~~-~~ice formation in the neighbouring waters, and a strong component associated to the concentration gradient between the S100 site and the FIS edge. ~~whereas~~~~Whereas~~In contrast, the ice-rises cores record the signal of larger-scale

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conditions of atmospheric flow, large inputs of bio-nssSO_4^{2-}, and less efficient transport of sea-salts evidenced by lower median~~mean~~ concentrations ~~in comparison~~than at ~~to~~ the S100 site. These findings are of vital importance for the understanding of the mechanisms of sea-salt aerosol production, transport and deposition at coastal Antarctic sites, and for the improvement of the current Antarctic sea~~-~~ ice reconstructions based on sea-salt chemical proxies.

6 Data availability

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

MODIS Mosaic of Antarctica (MOA) image is available through the GIS package Quantarctica, version 2.0 at http://quantarctica.npolar.no/.

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

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Tables

Table 1. Cores (KC, KM, BI, S100) locations and sampling details. Distances of the core locations to the ice shelf side were obtained using the GIS package Quantarctica (www.quantarctica.org). (*) refers to Kaczmarska et al. (2004), and (§) to Vega et al. (2016). SMB is Surface Mass Balance.

Site	Location	Elevation	Core length *Ice depth* Ice temp. at 10 m	Distance from the coast	Time coverage	Average SMB
		(m a.s.l.)	(m)	(km)	(years)	(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

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Table 2. Median~~, mean, maximum, minimum, and standard deviation (□) of~~ ion concentrations (in μ~~□~~mol L^{-1}) in the KC, KM, BI, and S100 firn/ice cores. Ion concentrations at the top 2 m of the KC, KM, and BI cores were not measured. Non-detected concentrations were set as half the detection limit of each ion.~~ Note: (*) the period is 1958.5–2012, (-) not measured.~~ Values of water stable isotopes and deuterium excess for the KC, KM, and BI are reported by Vega et al. (2016).

					~~Median~~ ~~Mean~~ ~~Maximum~~ ~~Minimum~~ ~~□i(□mol L^{-1})~~				
Site	Period ~~(years)~~ (years)	MSA	Cl$^-$	NO$_3^-$	SO$_4^{2-}$	Na$^+$	K$^+$	Mg^{2+}	Ca^{2+}
					(μmol L^{-1})				
KC	1958–2007	0.2	10.0	0.6	1.8	9.4	0.2	0.9	0.5
		~~0.2~~	~~11.3~~	~~0.7~~	~~2.1~~	~~12.1~~	~~0.2~~	~~1.0~~	~~1.8~~
		~~0.9~~	~~59.3~~	~~1.8~~	~~10.3~~	~~162.6~~	~~1.5~~	~~4.2~~	~~62.7~~
		~~0.0~~	~~1.7~~	~~0.2~~	~~0.1~~	~~1.1~~	~~0.0~~	~~0.2~~	~~0.2~~
		~~0.2~~	~~7.0~~	~~0.4~~	~~1.5~~	~~12.2~~	~~0.2~~	~~0.6~~	~~6.5~~
KM	1995–2012	0.3	71.3	0.4	4.5	57.7	1.5	6.3	1.6
		~~0.5~~	~~119.7~~	~~0.5~~	~~6.2~~	~~88.6~~	~~2.0~~	~~8.8~~	~~2.2~~
		~~9.4~~	~~571.6~~	~~5.4~~	~~84.5~~	~~654.8~~	~~16.1~~	~~45.5~~	~~10.6~~
		~~0.0~~	~~6.9~~	~~0.1~~	~~0.5~~	~~2.9~~	~~0.1~~	~~1.0~~	~~0.4~~
		~~0.6~~	~~104.4~~	~~0.4~~	~~7.5~~	~~92.6~~	~~1.8~~	~~7.4~~	~~1.8~~
BI	1996–2012	0.4	23.1	0.4	1.9	19.0	0.5	2.0	0.6
		~~0.5~~	~~27.0~~	~~0.5~~	~~2.5~~	~~22.5~~	~~0.6~~	~~2.4~~	~~0.7~~
		~~2.0~~	~~185.8~~	~~2.3~~	~~11.2~~	~~161.8~~	~~5.0~~	~~15.9~~	~~3.5~~
		~~0.0~~	~~1.8~~	~~0.1~~	~~0.3~~	~~1.7~~	~~0.0~~	~~0.3~~	~~0.3~~
		~~0.4~~	~~20.3~~	~~0.4~~	~~1.9~~	~~17.3~~	~~0.5~~	~~1.7~~	~~0.4~~
S100	1737–2000	0.1	20.9	0.5	1.2	20.7	0.4	2.0	0.7
		~~0.2~~	~~78.2~~	~~0.6~~	~~2.6~~	~~75.5~~	~~1.6~~	~~4.4~~	~~1.8~~
		~~5.6~~	~~2174.1~~	~~1.8~~	~~56.0~~	~~1315.5~~	~~39.6~~	~~35.9~~	~~40.0~~
		~~0.0~~	~~3.7~~	~~0.1~~	~~0.2~~	~~3.7~~	~~0.1~~	~~0.0~~	~~0.1~~
		~~0.3~~	~~187.7~~	~~0.3~~	~~5.2~~	~~149.8~~	~~3.4~~	~~5.4~~	~~3.6~~
S100	1995–2000	0.1	132.4	0.6	3.2	144.0	3.3	10.7	3.0
		~~0.2~~	~~220.8~~	~~0.6~~	~~6.0~~	~~209.0~~	~~4.4~~	~~10.8~~	~~4.2~~
		~~1.0~~	~~2174.1~~	~~1.4~~	~~35.8~~	~~1315.5~~	~~39.6~~	~~35.9~~	~~40.0~~
		~~0.0~~	~~11.3~~	~~0.1~~	~~0.8~~	~~11.0~~	~~0.3~~	~~1.5~~	~~0.5~~
		~~0.2~~	~~332.1~~	~~0.3~~	~~7.4~~	~~232.0~~	~~5.9~~	~~6.2~~	~~6.0~~
S100	1737–1949	0.1	16.0	0.6	1.0	15.1	0.3	1.4	0.5
		~~0.1~~	~~18.8~~	~~0.6~~	~~1.1~~	~~18.4~~	~~0.4~~	~~1.6~~	~~0.6~~
		~~0.8~~	~~120.9~~	~~1.8~~	~~4.8~~	~~138.8~~	~~2.1~~	~~6.3~~	~~5.7~~
		~~0.0~~	~~3.7~~	~~0.1~~	~~0.2~~	~~3.7~~	~~0.1~~	~~0.3~~	~~0.1~~
		~~0.1~~	~~12.9~~	~~0.3~~	~~0.5~~	~~14.2~~	~~0.2~~	~~0.9~~	~~0.5~~
S100	1950–2000	0.1	88.5	0.5	2.8	98.2	2.0	7.9	1.9

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~~0.2~~	~~179.2~~	~~0.6~~	~~5.2~~	~~172.6~~	~~3.6~~	~~9.1~~	~~3.7~~
~~5.6~~	~~2174.1~~	~~1.5~~	~~56.0~~	~~1315.5~~	~~39.6~~	~~35.9~~	~~40.0~~
~~0.0~~	~~9.1~~	~~0.1~~	~~0.4~~	~~8.6~~	~~0.2~~	~~0.0~~	~~0.3~~
~~0.5~~	~~280.9~~	~~0.3~~	~~7.9~~	~~213.2~~	~~5.0~~	~~6.4~~	~~5.4~~

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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					
Resolution	Annual			Annual		Annual		annual (1737–2000)		annual (1737–1949)		annual (1950–2000)	
Loadings	PC1	PC2	PC3	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
MSA	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
Cl^-	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
NO_3^-	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
SO_4^{2-}	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
Na^+	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
K^+	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
Mg^{2+}	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
Ca^{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
Explained Variance (%)	51	22	12	76	18	65	24	70	15	60	17	69	16
Source	sea-salts	biogenic mixed	terrestrial	sea-salts terrestrial	biogenic mixed	sea-salts terrestrial	biogenic mixed	sea-salts terrestrial	biogenic mixed	sea-salts terrestrial	biogenic mixed	sea-salts terrestrial	biogenic mixed

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~~**R**~~	~~**MSA**~~	~~**Cl^-**~~	~~**NO_3^-**~~	~~**SO_4^{2-}**~~	~~**Na^+**~~	~~**K^+**~~	~~**Mg^{2+}**~~	~~**Ca^{2+}**~~
~~Latitude (˚S)~~	~~0.20~~	~~−0.84~~	~~0.89~~	~~**−0.98**~~	~~−0.84~~	~~−0.78~~	~~−0.91~~	~~−0.81~~
~~Longitude (˚W)~~	~~**−0.99**~~	~~0.6~~	~~0.15~~	~~0.28~~	~~0.58~~	~~0.60~~	~~0.51~~	~~0.60~~
~~Elevation (m a.s.l.)~~	~~0.94~~	~~−0.81~~	~~−0.04~~	~~−0.41~~	~~−0.81~~	~~−0.85~~	~~−0.73~~	~~−0.84~~
~~Distance from the sea (km)~~	~~−0.06~~	~~−0.70~~	~~0.54~~	~~−0.59~~	~~−0.70~~	~~−0.71~~	~~−0.72~~	~~−0.69~~

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Table 4. Median Cl$^-$/Na$^+$ ratio (expressed in µmol L^{-1}), ssNa$^+$, and nssNa$^+$, and percentage of mean nssNa$^+$ to mean total Na$^+$ in the KC, KM, BI, and S100 cores. Since some of the calculated ssNa$^+$ values in the KC core were negative, ssNa$^+$ statistics are shown considering all data points, and only positive ssNa$^+$ values (sample rejection percentage is shown in parenthesis).~~Table 5. Cl$^-$/Na$^+$ ratio (expressed in mol L^{-1}), nssNa$^+$, ssNa$^+$, and percentage of nssNa$^+$ to total Na$^+$ in the KC, KM, BI, and S100 cores. Since some of the calculated ssNa$^+$ values in the KC core were negative, ssNa$^+$ statistics are shown considering all data points, and only positive ssNa$^+$ values (sample rejection percentage is shown in parenthesis).~~

		~~Ratio~~				
Site	Period (years)	Cl$^-$/Na$^+$	ssNa$^+$ (µmol L^{-1})		nssNa$^+$ (crustal) (µmol L^{-1})	Mean nssNa$^+$ to mean total Na$^+$ (%)
			All values	Only positive values		~~(%)~~
KC	1958–2007	1.2 ~~1.1~~ ~~1.9~~ ~~0.1~~ ~~0.3~~	8.3	8.7 (5 %)	0.4~~7~~ ~~2.6~~ ~~87.7~~ ~~0.3~~ ~~9.1~~	19~~21~~
KM	1995–2012	1.3 ~~1.3~~ ~~3.8~~ ~~0.8~~ ~~0.2~~	56.6	56.6 (0%)	0.5~~2.2~~ ~~3.0~~ ~~14.8~~ ~~0.5~~ ~~2.5~~	0.6~~3~~
BI	1996–2012	1.2 ~~1.2~~ ~~1.5~~ ~~0.9~~ ~~0.1~~	18.6	18.6 (0 %)	0.3~~8~~ ~~1.0~~ ~~4.7~~ ~~0.4~~ ~~0.5~~	1.4~~4~~
S100	1737–2000	1.0 ~~1.0~~ ~~2.1~~ ~~0.1~~ ~~0.2~~	20.1	20.1 (0 %)	0.2~~1.0~~ ~~2.5~~ ~~56.0~~ ~~0.1~~ ~~5.1~~	0.5~~5~~
S100	1995–2000	1.0 ~~1.0~~ ~~2.1~~ ~~0.1~~ ~~0.2~~	143.7	143.7 (0 %)	-0.1~~9.1~~ ~~9.2~~ ~~30.5~~ ~~1.3~~ ~~5.2~~	0.0~~4~~
S100	1737–1949	1.0 ~~1.1~~	14.7	14.7 (0 %)	0.3~~1.2~~ ~~1.4~~	2.1~~8~~

		~~1.8~~			~~5.3~~	
		~~0.6~~			~~0.2~~	
		~~0.2~~			~~0.8~~	
		1.0			0.0~~6.7~~	
		~~1.0~~			~~7.7~~	
S100	1950–2000	~~2.1~~	96.3	96.3 (0 %)	~~30.5~~	0.2~~4~~
		~~0.1~~			~~0.0~~	
		~~0.2~~			~~5.6~~	

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Table ~~5~~6. Median~~, mean, maximum, minimum, and standard deviation~~ of ss- and nss-fractions in the KC, KM, BI, and S100 cores. Percentage of negative nss-values for each ion is shown in parenthesis. Negative ss-values in the KC core are due to the 5 % of ssNa$^+$ negative values obtained in section 3.~~5~~4 ~~(Table 5)~~.

							~~Median~~ ~~Mean~~ ~~Maximum~~ ~~Minimum~~ ~~□i□□mol L^{-1})~~				
Core	Period ~~(years)~~ (years)	Cl$^-$		SO_4^{2-}		K$^+$		Mg^{2+}		Ca^{2+}	
		ss	nss	ss	nss (μmol L^{-1})	ss	nss	ss	nss	ss	nss
KC	1958–2007	9.7~~4~~ ~~11.1~~ ~~184.6~~ ~~−78.6~~ ~~16.8~~	~~(28 %)~~ 0.6 (32 %)~~9~~ ~~0.2~~ ~~96.6~~ ~~−165.6~~ ~~15.4~~	0.5 ~~0.6~~ ~~9.6~~ ~~−4.0~~ ~~0.9~~	~~(3 %)~~ 1.2 (4 %) ~~1.6~~ ~~8.2~~ ~~−0.5~~ ~~1.4~~	0.2 ~~0.2~~ ~~3.2~~ ~~−1.4~~ ~~0.3~~	~~(27 %)~~ 0.0 (27 %) ~~0.1~~ ~~1.8~~ ~~−2.0~~ ~~0.2~~	0.9 ~~1.1~~ ~~17.5~~ ~~−7.4~~ ~~1.6~~	~~(41 %)~~ 0.0 (43 %)~~1~~ ~~0.0~~ ~~9.5~~ ~~−16.8~~ ~~1.6~~	0.2	0.3 (5%)
KM	1995–2012	65.6~~3.8~~ ~~99.3~~ ~~747.5~~ ~~2.8~~ ~~104.7~~	~~(5 %)~~ 4.7 (12 %)~~6.2~~ ~~10.4~~ ~~80.5~~ ~~−200.3~~ ~~19.4~~	3.4~~3~~ ~~5.1~~ ~~38.7~~ ~~0.1~~ ~~5.4~~	~~(38 %)~~ 0.7 (39 %)~~9~~ ~~1.1~~ ~~45.9~~ ~~−15.1~~ ~~4.5~~	1.1 ~~1.7~~ ~~12.9~~ ~~0.1~~ ~~1.8~~	~~(12 %)~~ 0.2 (13 %) ~~0.3~~ ~~13.8~~ ~~m8.4~~ ~~1.3~~	6.2~~1~~ ~~9.4~~ ~~70.9~~ ~~0.3~~ ~~9.9~~	~~(43 %)~~ 0.1 (48 %)~~2~~ ~~−0.7~~ ~~23.6~~ ~~−54.1~~ ~~6.7~~	1.1	0.4 (0 %)
BI	1996–2012	21.6~~0~~ ~~25.0~~ ~~181.9~~ ~~1.6~~ ~~19.5~~	~~(4 %)~~ 1.0 (11 %)~~7~~ ~~2.1~~ ~~15.3~~ ~~−1.7~~ ~~1.8~~	1.1 ~~1.3~~ ~~9.4~~ ~~0.1~~ ~~1.0~~	~~(26 %)~~ 0.8 (26 %) ~~1.3~~ ~~7.4~~ ~~−1.4~~ ~~1.7~~	0.4 ~~0.4~~ ~~3.1~~ ~~0.0~~ ~~0.3~~	~~(2 %)~~ 0.1 (4 %) ~~0.1~~ ~~4.5~~ ~~0.0~~ ~~0.3~~	2.0 ~~2.4~~ ~~17.3~~ ~~0.2~~ ~~1.9~~	~~(42 %)~~ 0.0 (51 %) ~~0.0~~ ~~2.0~~ ~~−3.0~~ ~~0.5~~	0.4	0.2 (0 %)
S100	1737–2000	23.3~~2.7~~ ~~84.7~~ ~~1460.9~~ ~~4.0~~ ~~168.4~~	~~(81 %)~~ −2.8~~1~~ (85 %) ~~−6.6~~ ~~713.2~~ ~~−583.3~~ ~~54.2~~	1.2 ~~4.4~~ ~~75.6~~ ~~0.2~~ ~~8.7~~	~~(51 %)~~ −0.1~~0.0~~ (53 %) ~~−1.7~~ ~~5.2~~ ~~−44.6~~ ~~5.0~~	0.4 ~~1.5~~ ~~25.2~~ ~~0.1~~ ~~2.9~~	~~(40 %)~~ 0.0 (45 %) ~~0.1~~ ~~14.4~~ ~~−2.7~~ ~~0.9~~	2.2 ~~8.0~~ ~~138.5~~ ~~0.4~~ ~~16.0~~	~~(74 %)~~ −0.3 (76 %) ~~−3.7~~ ~~3.7~~ ~~−102.7~~ ~~11.3~~	0.4	0.1 (0 %)
S100	1995–2000	166.7 ~~2.0~~ ~~235.6~~ ~~1460.9~~ ~~11.3~~ ~~259.9~~	~~(87 %)~~ −29.3~~3.7~~ (91 %) ~~−14.8~~ ~~713.2~~ ~~−583.3~~ ~~134.6~~	8.6~~4~~ ~~12.2~~ ~~75.6~~ ~~0.6~~ ~~13.4~~	~~(78 %)~~ −4.9~~5~~ (80 %) ~~−6.2~~ ~~3.2~~ ~~−39.8~~ ~~8.2~~	2.9~~8~~ ~~4.1~~ ~~25.2~~ ~~0.2~~ ~~4.5~~	~~(39 %)~~ 0.0~~1~~ (50 %) ~~0.3~~ ~~14.4~~ ~~−1.2~~ ~~2.1~~	15.8~~4~~ ~~22.3~~ ~~138.5~~ ~~1.1~~ ~~24.6~~	~~(74 %)~~ −5.5~~0~~ (74 %) ~~−11.5~~ ~~3.5~~ ~~−102.7~~ ~~18.9~~	2.9	−0.1 (56 %)
S100	1737–1949	17.0~~6.6~~	~~(75 %)~~ −1.5~~1~~ (80 %) ~~−1.6~~	0.9 ~~1.1~~	~~(33 %)~~ 0.1~~2~~ (36 %) ~~0.1~~	0.3 ~~0.4~~	~~(41 %)~~ 0.0 (45 %) ~~0.0~~	1.6 ~~1.9~~	~~(73 %)~~ −0.2 (76 %) ~~−0.3~~	0.3	0.2 (8 %)

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		~~20.4~~ ~~159.4~~ ~~4.0~~ ~~16.1~~	~~10.4~~ ~~−44.5~~ ~~4.5~~	~~8.3~~ ~~0.2~~ ~~0.8~~	~~4.1~~ ~~−7.2~~ ~~0.8~~	~~2.8~~ ~~0.1~~ ~~0.3~~	~~0.6~~ ~~−0.6~~ ~~0.1~~	~~15.1~~ ~~0.4~~ ~~1.5~~	~~3.7~~ ~~−14.1~~ ~~1.2~~		
S100	1950–2000	111.7~~08.6~~ ~~194.2~~ ~~1460.9~~ ~~9.1~~ ~~239.3~~	~~(90 %)~~ −18.6~~4.7~~ (93 %) ~~−15.0~~ ~~713.2~~ ~~−583.3~~ ~~88.5~~	5.8~~6~~ ~~10.1~~ ~~75.6~~ ~~0.5~~ ~~12.4~~	~~(81 %)~~ −2.8~~7~~ (83 %) ~~−4.8~~ ~~5.2~~ ~~−44.6~~ ~~7.1~~	1.9 ~~3.4~~ ~~25.2~~ ~~0.2~~ ~~4.1~~	~~(38 %)~~ 0.0~~1~~ (43 %) ~~0.2~~ ~~14.4~~ ~~−2.7~~ ~~1.4~~	10.6~~3~~ ~~18.4~~ ~~138.5~~ ~~0.9~~ ~~22.7~~	~~(74 %)~~ −2.6~~3~~ (76 %) ~~−9.3~~ ~~3.5~~ ~~−102.7~~ ~~17.0~~	1.9	0.0 (47 %)

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Table 6~~7~~. Median~~, mean, minimum, maximum, and standard deviation (□)~~ of MSA/nssSO_4^{2-} ratios, and bio-nssSO_4^{2-} in the KC, KM, BI, and S100 cores. Statistics for the MSA/nssSO_4^{2-} ratio are presented considering all values, and only positive values (sample rejection percentage is shown in parenthesis). In addition, the percentage of mean bio-nssSO_4^{2-} to mean total SO_4^{2-} is shown for all the cores.

Site	Period (years)	MSA/nssSO_4^{2-} All values	MSA/nssSO_4^{2-} Only positive values	bio-nssSO_4^{2-} (µmol L^{-1})	Mean bio-nssSO_4^{2-} to mean total SO_4^{2-} (%)
			~~Median~~ ~~Mean~~ ~~Maximum~~ ~~Minimum~~ ~~□~~		
KC			~~(3 %)~~		
	1958–2007	0.1 ~~0.3~~ ~~−17.2~~ ~~14.9~~ ~~1.8~~	0.1 (4 %) ~~0.4~~ ~~14.9~~ ~~0.0~~ ~~1.4~~	1.0 ~~1.2~~ ~~0.0~~ ~~5.2~~ ~~1.0~~	58
KM			~~(38 %)~~		
	1995–2012	0.1 ~~0.7~~ ~~−12.9~~ ~~138.8~~ ~~9.9~~	0.3 (39 %) ~~1.7~~ ~~138.8~~ ~~0.0~~ ~~12.4~~	1.9 ~~2.9~~ ~~0.1~~ ~~52.3~~ ~~3.5~~	~~136~~46
BI			~~(25 %)~~		
	1996–2012	0.3 ~~−0.4~~ ~~−245.1~~ ~~30.6~~ ~~13.1~~	0.3 (26 %) ~~0.7~~ ~~30.6~~ ~~0.0~~ ~~2.1~~	2.1 ~~2.7~~ ~~0.1~~ ~~11.3~~ ~~2.4~~	10~~8~~7
S100			~~(51 %)~~		
	1737–2000	0.0 ~~0.2~~ ~~−15.6~~ ~~11.3~~ ~~1.8~~	0.3~~3~~ (53 %) ~~1.0~~ ~~11.3~~ ~~0.0~~ ~~1.9~~	0.7 ~~1.0~~ ~~0.0~~ ~~31.4~~ ~~1.6~~	37
S100			~~(78 %)~~		
	1995–2000	0.0 ~~0.1~~ ~~−1.7~~ ~~9.5~~ ~~1.4~~	0.2 (80 %) ~~1.1~~ ~~9.5~~ ~~0.1~~ ~~2.7~~	0.5 ~~1.0~~ ~~0.1~~ ~~5.4~~ ~~1.2~~	17

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S100			~~(33 %)~~		
	1737–1949	0.2 ~~0.3~~ ~~−15.6~~ ~~10.3~~ ~~2.0~~	1.0 (36 %) ~~1.0~~ ~~10.3~~ ~~0.0~~ ~~1.7~~	0.7 ~~0.8~~ ~~0.0~~ ~~4.4~~ ~~0.6~~	72
S100			~~(81 %)~~		
	1950–2000	0.0 ~~0.1~~ ~~−2.6~~ ~~11.3~~ ~~1.4~~	0.2 (83 %) ~~1.2~~ ~~11.3~~ ~~0.0~~ ~~2.8~~	0.7 ~~1.2~~ ~~0.1~~ ~~31.4~~ ~~2.6~~	24

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Table 7~~8~~. Median annual nssSO$_4^{2-}$ concentrations (in µ~~□~~mol L^{-1}) in the KC, KM, BI, and S100 firn/ice cores. (-) ~~-~~Not re-calculated.

Site	Period (years)	nssSO$_4^{2-}$ ~~Median~~ ~~Median~~ ~~Median~~ (µ~~□~~mol L^{-1}) ~~(□mol L^{-1})~~ ~~(□mol L^{-1})~~ ~~nssSO$_4^{2-}$~~ ~~nssSO$_4^{2-}$~~ ~~nssSO$_4^{2-}$~~		
		k~~*K*~~$_{seawater}$=0.06	*k'*=0.02	*k'*=0.03
KC	1958–2007	1.4~~2~~	-	-
KM	1995–2012	0.8~~9~~	-	-
BI	1996–2012	1.3~~0.8~~	-	-
S100	1737–2000	0.3	0.9	0.7
S100	1995–2000	−2.3~~1~~	2.3~~4~~	1.2~~3~~
S100	1737–1949	0.3~~4~~	0.8	0.7
S100	1950–2000	−1.7~~5~~	1.2~~3~~	0.6~~7~~

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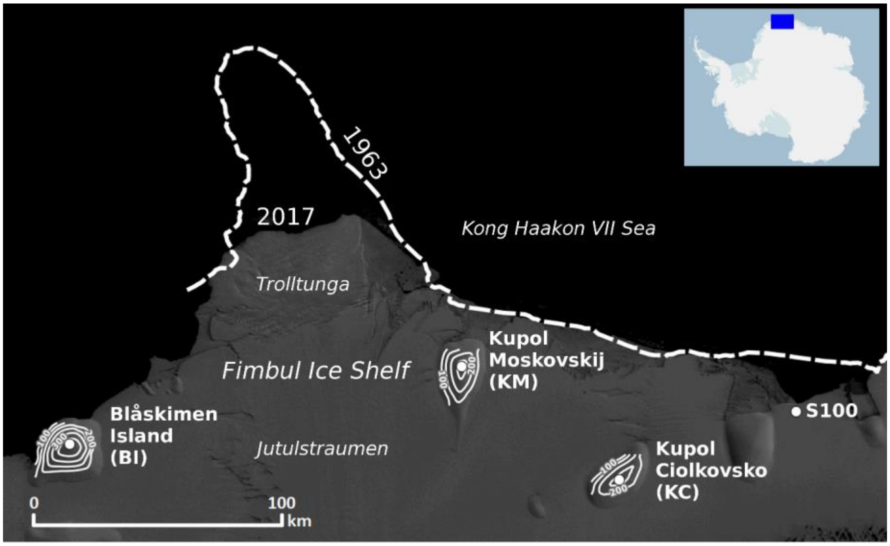

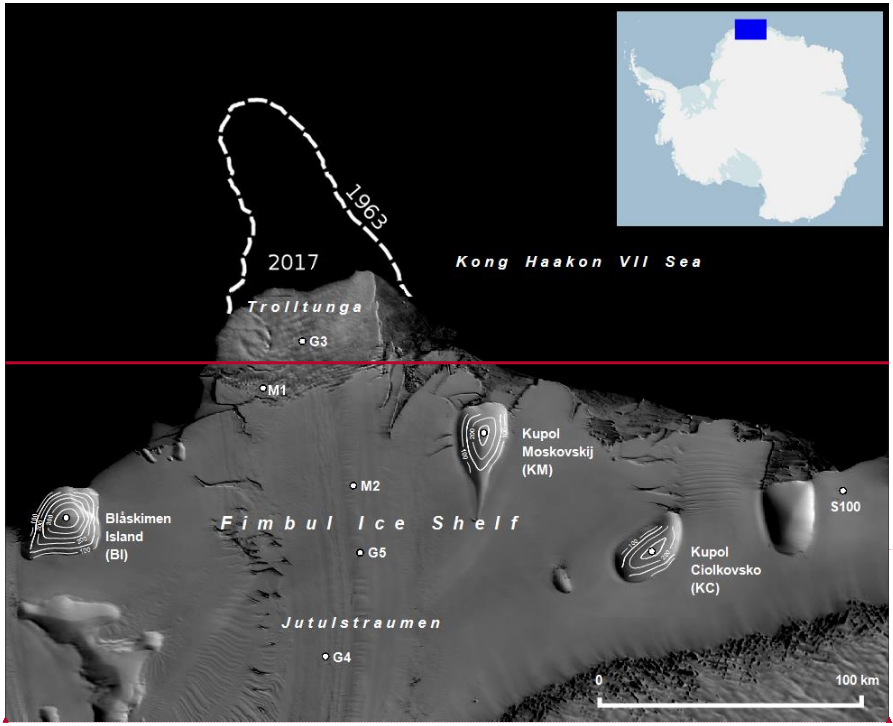

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Figure 1. Satellite image of Fimbul Ice Shelf (FIS) showing the KC, KM, BI, and S100 core sites, ~~the M1, M2, G3, G4, and G5 snow pit sites (Supplementary material),~~ Jutulstraumen, and Trolltunga. In addition, 50-m contours are shown at each ice-rise,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

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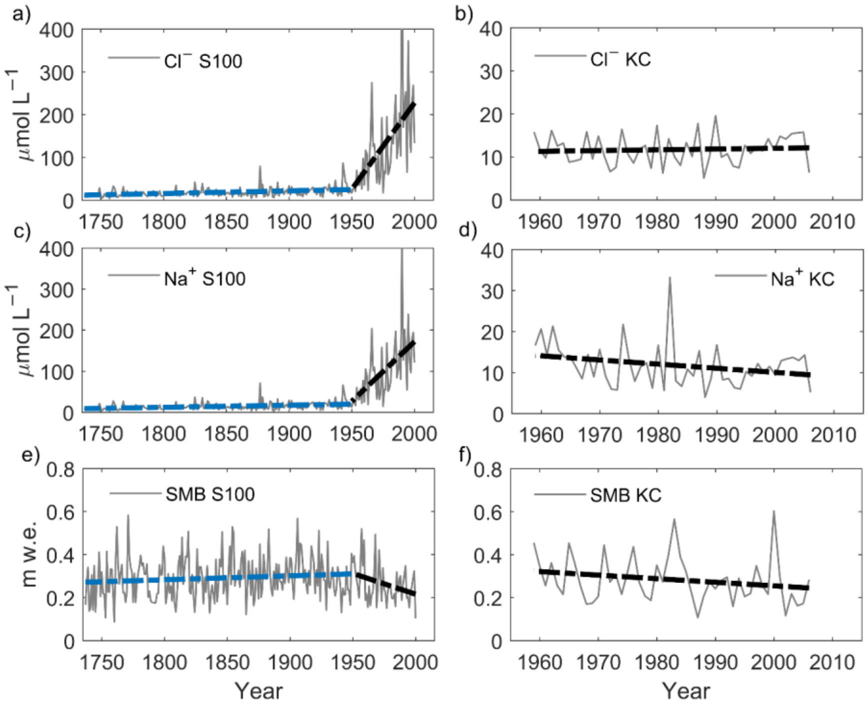

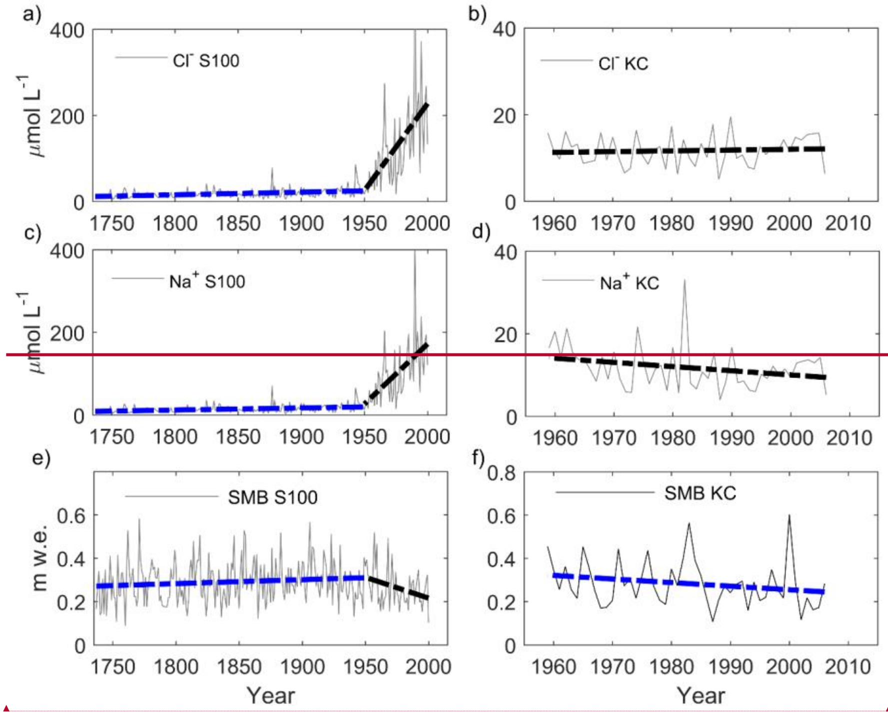

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Figure 2. Annual sea-salt (Cl^- and Na^+) concentrations and surface mass balance (SMB) in the two longest records retrieved at Fimbul Ice shelf, S100, (a), (c), and (e), and KC, (b), (d) and (f). Linear trends in Cl^- and Na^+ concentrations, and SMB measured in the S100 core are shown for two different periods: 1737–1949 (blue dashed line) and 1950–2000 (black dashed line) in (a), (c), and (e), respectively. Linear trends in Cl^- and Na^+ concentrations, and SMB measured in the KC core are shown for the period 1958–2007 (black dashed line) in (b), (d) and (f), respectively. Significance, slope, and standard error of the linear regressions are given in Table S~~4~~3.

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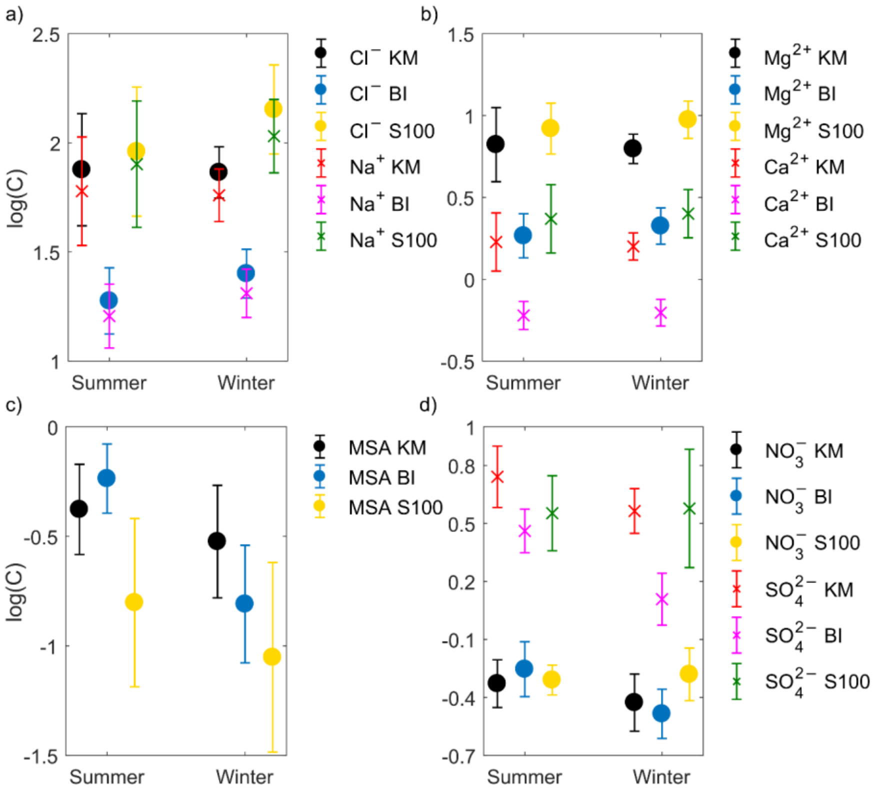

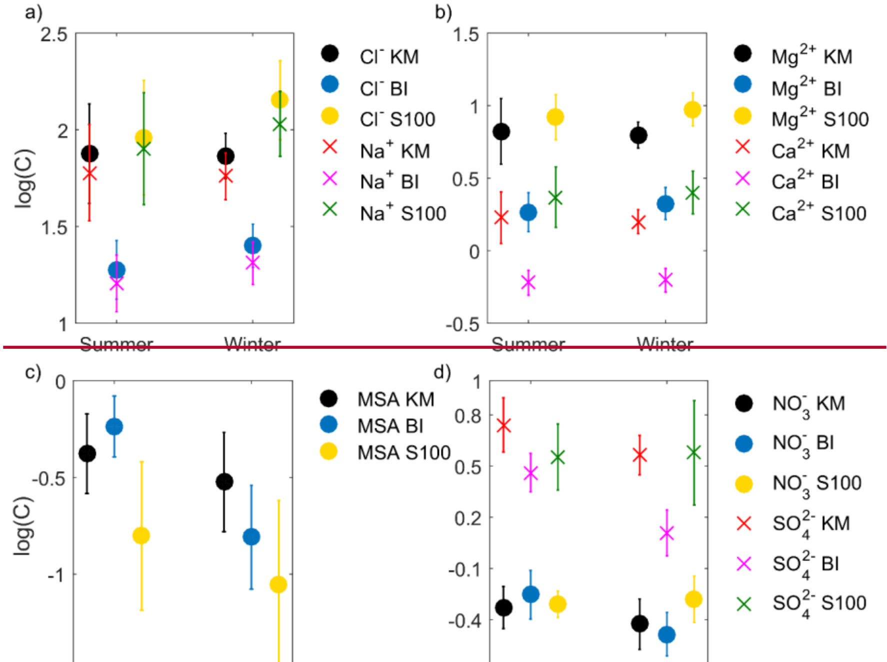

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Figure 3. Sub-annual variability of selected ions, Cl^{-} and Na^{+} (a), Mg^{2+} and Ca^{2+} (b), MSA (c) and NO_3^{-} and SO_4^{2-} (d) in cores KM, BI, and S100. Mean summer and winter concentrations were calculated for the months NDJFMA, and MJJASO, for a period of 16, 15, and 5 years in the KM, BI, and S100 cores, respectively.

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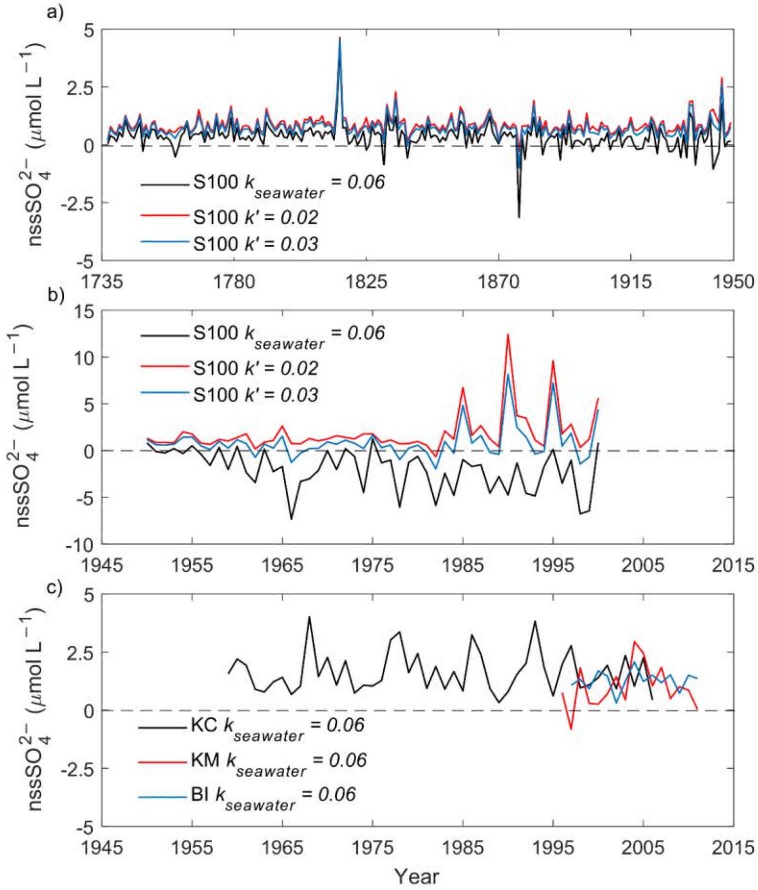

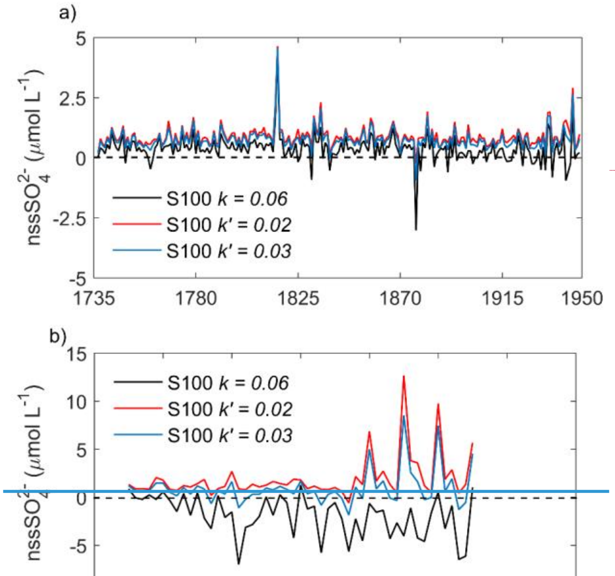

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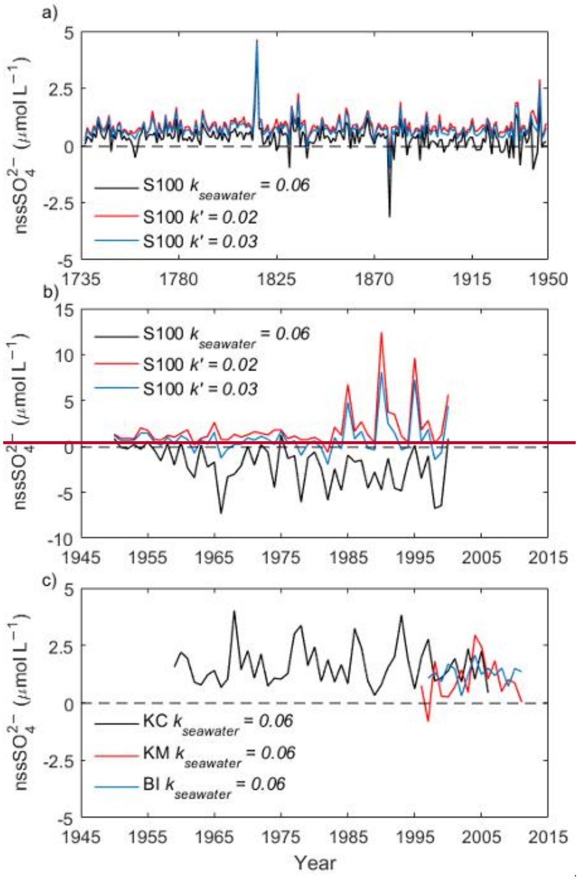

Figure 4. Annual nssSO$_4^{2-}$ concentrations in the S100 core between a) 1737–1949, b) 1950–2000, and c) in the KC, KM, and BI cores. nssSO$_4^{2-}$ recalculated using $k_{seawater}$= 0.06, k'=0.02 and k'=0.03 are shown in panels a) and b) with black, red and blue lines, respectively. nssSO$_4^{2-}$ in the KC, KM, and BI cores was calculated using k= 0.06.

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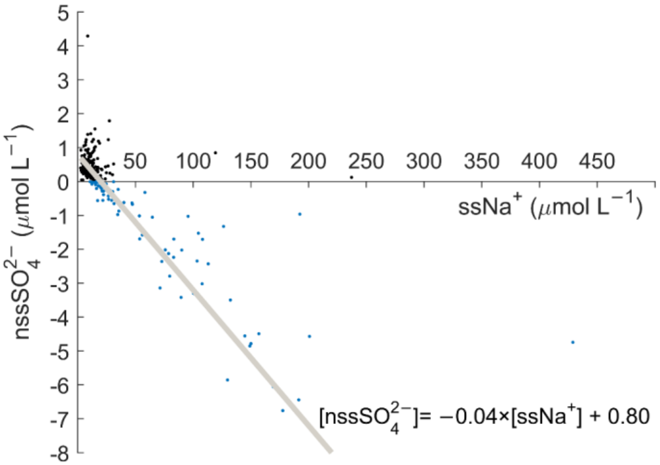

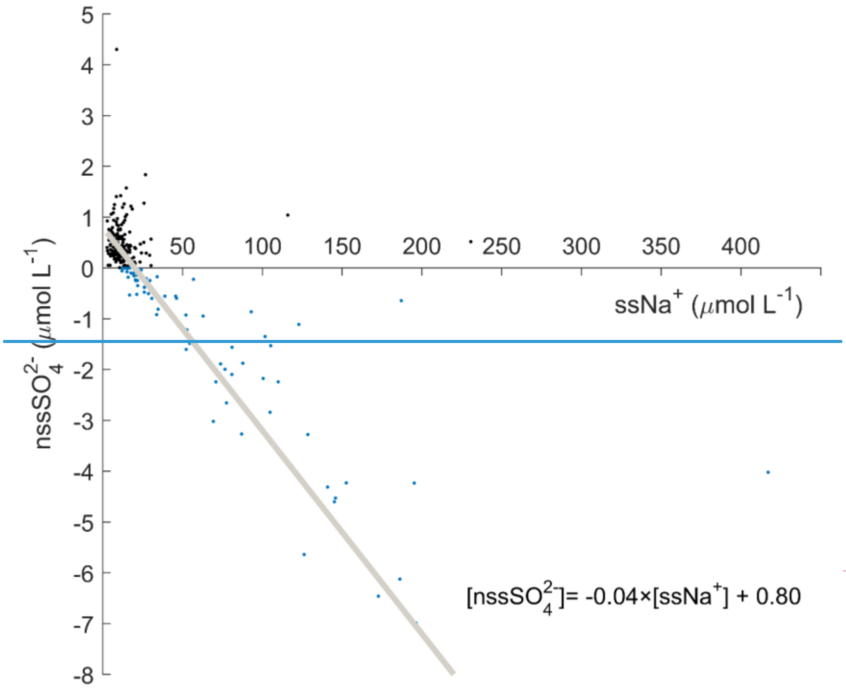

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Figure 5. Scatter plot of annual nssSO_4^{2-} vs. ssNa^+ concentrations in the S100 core. nssSO_4^{2-} was calculated using the seawater ratio as described in section 2.3 and using a $k_{seawater}$=0.06 (in µmol L^{-1}). Positive nssSO_4^{2-} values are denoted with black dots, while negative values are denoted with blue dots. A linear regression was calculated using all nssSO_4^{2-} data points to infer corrected *k* value (*k'*), following the approach by Wagenbach et al. (1998).

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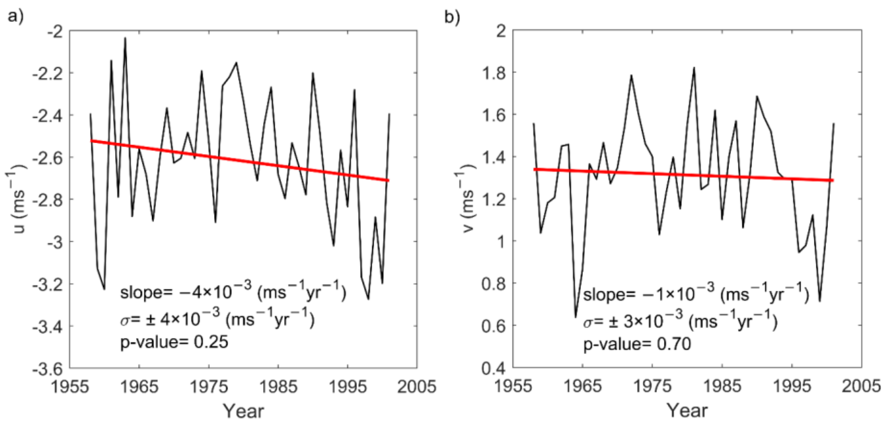

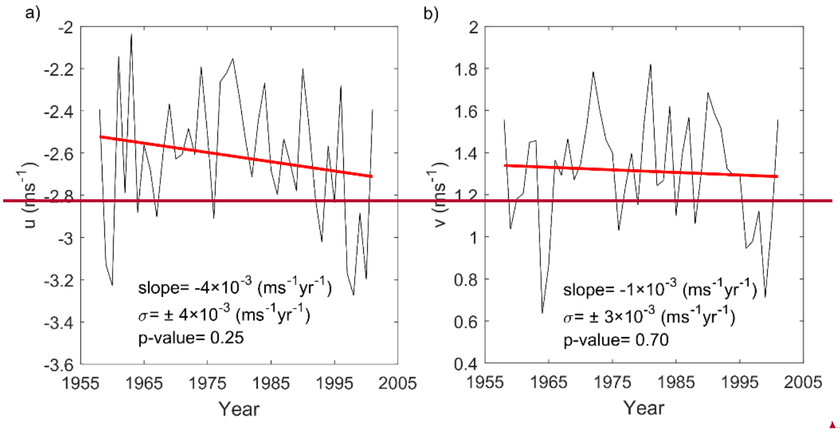

Figure 6. Annual averages of monthly a) zonal, and b) meridional wind speeds (ERA40) for the area (69°S–71°S, 3.5°W–5°E) between 1958–2001. Slope, standard deviation, and *p*-value of the linear regression are shown in the figure.

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