*Final author 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.*

To the referees:

The authors would like to thank the referees for the time they took to review this manuscript. The referees have provided valuable general and specific comments which have been very helpful to prepare a revised version of the manuscript. We agree in most of the referees comments, and consequently, we have done the correspondent modifications in the manuscript text to include their valuable suggestions, as long as it was possible. This letter contains a full response to the referees' comments, following a preliminary response uploaded on 2017-09-30 as part of the Discussion period.

We provide in this letter, a detailed point by point response to all referee comments. Referees comments are noted in italics, authors responses have been 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.

Response to Anonymous Referee #1

This paper presents new ice core chemical data for a coastal region of Antarctica. It interprets particularly the sea salt chemistry, and attempts to discuss the mechanisms behind sea salt production and deposition from the data. The data have some interest, particularly the unusual record from S100, and it may be possible to make a workable paper out of them. (The application of this paper is somewhat reduced because sites so close to the sea can be interesting but do not tell us too much about processes affecting inland sites.)

CV. As we have mentioned in the discussion period, the data presented in this manuscript contribute to fill in the existent gap in ice core records for low elevation coastal areas. The relevance of acquiring more data from the coastal areas has been stressed by recent research (Stenni et al., 2017, and Thomas et al., 2017), and it is also pointed out in the general comments by Referee #2. We agree with Referee #1 that those sites are interesting. However, information about inland sites cannot be expected from a study dealing with data from coastal sites only.

However at the moment the paper suffers from three very major flaws, and an omission that render most of the interpretations dubious:

CV. Points 1 to 3 have been addressed below. Referee #1 also mentions an 'omission' mostly associated to glaciological data between 1950s to 2000s. Relevant references for the glaciological settings of Fimbul Ice Shelf (FIS) have been included in the first version of the manuscript, however, we did not show the data in an explicit way when discussing the abrupt change in sea-salts after the 1950s. We have now included available glaciological data by Rignot et al. (2011) for FIS in the discussion part.

1. The snowpit data are all from samples covering less than a year of snowfall. This makes it impossible to use the average values generated quantitatively, both because they are not a real yearly value, and because interannual variability means that the average for one year should have a huge uncertainty on it. There was a time when we were so desperate for new data from unexplored parts of the continent that we would at least consider surface snow data from part-years but those days are over. Without the snowpit data, the discussion of spatial variability is impossible, so section 3.4, Table 4 and all discussion about spatial variability should be removed from the paper.

CV. The referee is correct about the snow pits use in the discussion in section 3.4; consequently, we have removed the snow pits from the manuscript main text and added them as Supplementary material. In order to keep the spatial variability discussion, we have now only used the core data (KC, KM, BI, and S100) for the years in which the cores overlap (1997–2000). We calculated R-values of the correlations between median annual concentrations for all ions versus latitude, longitude, site elevation, and distance from the sea. We found that only SO_4^{2-} and MSA showed a significant (at the 95 % confidence interval) correlation with latitude, and longitude, respectively, whereas all other correlations in Table 4 were not significant. We attributed these results to the local effects on annual SMB evidenced at the KM and BI sites reported by Vega et al. (2016) which would affect any spatial pattern, e.g., the dependency reported by Stenberg et al. (1998) between sea-salts and distance from the sea.

2. The authors seem to be under the impression that if they don't observe negative nss sulfate, then there is no fractionation and no sea ice source. Of course this is not correct: while sea ice fractionation removes sulfate and causes negative nss-sulfate values, biogenic sulfate gives positive nss-sulfate. Only if the former overwhelms the latter will net negative values be seen. At sites very near the coast where marine biogenic inputs are large, this makes diagnosing fractionation tricky. As a rough estimate, one can note that typical values of MSA/nss-sulfate in biogenic input are 20% (Legrand and Pasteur 1998). From that we can estimate for example that biogenic sulfate at BI could easily have contributed all the sulfate seen, so that fractionation must have occurred. Uncertainty on the MSA concentration and the ration MSA/nss-sulfate makes this calculation very uncertain, but just illustrates that any of these sites could be experiencing large proportions of fractionated aerosol. The nss-sulfate discussion is valuable but needs to be done in a much more sophisticated way.

CV. As we mentioned in the preliminary response to the referees, we do not think that no negative nss-sulfate values mean no fractionation. In order to clarify this point and present a more solid discussion on the topic, we have calculated the MSA/nssSO_4^{2-} ratio, and calculated the percentage of biogenic nssSO_4^{2-} (bio-nssSO_4^{2-}) to total SO_4^{2-} in all the cores (Table 7), and section 3.6 has been rewritten accordingly.

3. The authors use the correlations between concentration or flux and snow accumulation rate to try to diagnose the deposition mechanism. This could have some value if interpreted sensibly. However for S100 (1950-2000), it is obvious that the main feature is an immense rise in Na and Cl (factor 6) accompanied by a small drop (perhaps 20%) in accumulation rate. The relationship between these two trends will dominate any correlation but a 20% drop in snowfall cannot in itself cause more than a 20% increase in concentration even if dry deposition dominates completely. One simply cannot learn about dry and wet deposition for this site: something else is overwhelming the situation by causing a huge increase in sea salt to the site.

CV. The referee is correct. Consequently, we have removed the section and we focused more into explaining both the six-fold increase in sea-salts and the fractionated nssSO_4^{2-} in terms of an enhancing of sea-salts originated from the blowing of salty snow over sea ice. We have then introduced the above in sections 3.6 and 4.

4. The something else is causing huge sea salt concentration increases after 1950. It cannot be a change in the source to the ice shelf as a whole, since KC doesn't see it. I feel I am missing crucial information to allow me to interpret this. The obvious explanation would be that S100 has been getting closer to the ice shelf edge since 1950. But the paper gives no glaciological information that would allow us to interpret that. My assumption would be that the ice front at S100 occasionally calves icebergs, and that S100 is moving forwards at 10s to 100s of m/yr. The authors need to check and discuss what happened between 1950 and 2000. Did the S100 site simply get nearer the ice front?

CV. We have now included ice speed information (m y^{-1}) available for FIS. Ice velocities near S100 are in the order of 10s–100s m y^{-1} (Rignot et al., 2011) for the period 2007–2009. However, we cannot assume the same ice velocities for S100 for the pre-1950 and post-1950 periods. By using the S100 core sea-salt results we could 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; and that the calving event at Trolltunga in 1967 (Vinje, 1975), enhanced the input of fractionated sea-salts to the S100 core by modifying the sea-ice conditions around S100. This could be supported by the fact that negative $nssSO_4^{2-}$ values slowly decreased between 1950–1966, showing a marked minimum around 1966 (±3 years), which could have been induced by the calving event. We discuss this in section 4.

I'm afraid all these points call for a major rethink about the purpose and conclusions of the paper. I will discuss a few more detailed points, but clearly any revision will be close to a new paper (it's borderline between major revision and reject) and will need reviewing again.
Abstract, page 2, line 5. As discussed above, the authors cannot conclude about dry deposition from the method they used. The very high concentrations do suggest a very high atmospheric concentration above the site by the year 2000, which would likely be both wet and dry deposited (such local material would have large particle sizes so would deposit fast). But this cannot exactly be described as dry deposition in the conventional aerosol dry deposition sense.

CV. We appreciate the referee's suggestions. We consider that after the modifications done to the manuscript, this has now improved and the hypothesis of sea-ice sea-salts as source to the S100 is now described in clearer way.

Page 10. The MSA-nitrate connection is overdone here. They surely end up on the same PC mainly because they don't show the sea salt pattern. We are not shown data that would allow us to judge this. However, for sure trying to link nitrate to MSA as a fertiliser seems far-fetched for a number of reasons. The Southern Ocean is not generally considered to be nitrate-limited; it seems unlikely that nitrate in the ocean is dominated by local atmospheric deposition. If you want to make this point you need to show data that would make a convincing case that high nitrate really is associated with high MSA.

CV. We have stated in the paragraph that MSA and NO_3^- have also shown coherence in other cores (Wendl et al., 2015), and that a fertilizer effect was proposed to explain such coherence in those Svalbard cores. We then briefly explained Wendl et al. (2015) hypothesis for the MSA and NO_3^- coherence, and then we added "However, there is a variety of possible NO_3^- sources 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 did not attribute the coherence observed between MSA and NO_3^- in our cores (PC2) to the "fertilizing effect" proposed by Wendl et al. (2015). We have now corrected the paragraph so our point is clearly made. The sentence now reads: "High loadings of NO_3^- and MSA in PC2, and thus, coherence between both species, have been 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) 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)."

Page 11, line 8: they don't all show a 6 fold increase.

CV. The sentence has now been corrected and now reads: "In the S100 core, Na^+, Cl^-, K^+, and Mg^{2+} median concentrations show a marked six-fold increase after the 1950s."

Section 3.3. You are doing something very difficult here. Please start with a discussion about the caveats: that it is very difficult to reliably divide the annual layers into 4 sections of equal time so the uncertainty on this is very large.

CV. This is a good point, and had also been mentioned by referee #2 (Point 3). Consequently, we have now followed both referees' suggestions and section 3.3 has been modified accordingly (please refer to response to referee #2 for more details on this section).

Section 3.4 should be removed as discussed above. At the end of the section, you dismiss the importance of elevation, but this cannot be excluded as a factor for the 3 ice rises reaching 200 m.

CV. We have now modified this section, as mentioned in the response to Point 1 of referee #1.

Section 3.5 – see discussion above.

Section 3.6 is very confused. The best way to treat this is to use the slope of lines such as that in Fig 5 to estimate the degree of fractionation, rather than trying different ratios (0.06, 0.04, 0.02). However, the line in Fig 5 should be a best fit through all the data (not just the negative), and should not go through zero (because when there is no sea salt there is still nss-sulfate from biogenic sources). Treated this way, I guess the slope will be about -0.04, implying 66% fractionation for the whole dataset (not just the post 1950s unless you see a significantly different slope for the two time periods).

CV. We have now followed the referee's suggestions and we have modified section 3.6 and the discussion, accordingly.

Anonymous Referee #2

General Comments

The paper is concerning the study of sea-salt components in snow, firn and ice samples collected in a coastal area of East Antarctica. There is a lack of knowledge on environmental and climatic data from firn/ice core stratigraphies in Coastal Regions of Antarctica; therefore, the paper is interesting for the Antarctic Glaciology Community. The paper is well written and sufficiently concise (see Comments to address toward more synthetic sections). However, in my opinion, the manuscript contains some weak points that should be addressed before to be accepted for publication on The Cryosphere journal. These weak points are discussed in the Specific and Minor Comments section, but can be here listed:

1. Some experimental procedures should be clarified;

CV. We have addressed this general comment on the responses to the referee's minor comments.

2. Ss- and nss- fractions of most of the analyzed components (especially Na and Ca) should be calculated as more reliable markers of sea spray (ssNa) and crustal (nssCa) contributions;

CV. We have now calculated nssNa$^+$ using Ca^{2+} as reference, and assigning a crustal origin for the nssNa$^+$ in the cores. We then calculated the percentage of nssNa$^+$ to total Na$^+$, and estimated ssNa$^+$ removing the nss-fraction from the total Na$^+$. This is now presented in section 2.3 and discussed in section 3.5. The motivation to use Ca^{2+} as a reference ion to obtain nssNa$^+$ instead of using the Na$^+$/Cl$^-$ ratio in seawater, was on the basis of the lower Cl$^-$/Na$^+$ ratios found in the S100 core in comparison to standard seawater (presented in Table 5, and discussed in section 3.5).
We then calculated both ss-and nss-fractions for Cl$^-$, SO$_4^{2-}$, K$^+$, and, Mg^{2+} using ssNa$^+$ as reference ion and *k* values obtained from standard mean seawater composition (Table S2 in the Supplementary material), and the calculation has been explained in section 2.3. Due to the low concentrations of NO$_3^-$ in standard seawater (Summerhayes and Thorpe, 1996), we did not separate NO$_3^-$ into nss- and ss-fractions (i.e., NO$_3^-$ is assumed to only have a nss-origin, as well as MSA). Median, mean,

maximum, minimum, and standard deviation of nss- and ss-fractions are shown in Table 5 and Table 6.

3. Seasonal characterization of the sub-samples should be made taking into account the $\delta^{18}O$ profiles, instead of using an interpolation procedure;

CV. This point was also addressed by referee #1. We have now corrected this section following both referees' suggestions (please refer to the response to referee #2 in the section *Specific and Minor Comments* for more details on this procedure or directly to section 3.3 in the manuscript).

4. The evaluation of the spatial variability by snow pit data appears to be not significant, because of the short record (lower than 1-year deposition),

CV. Referee #1 also pinpointed this. In order to keep the spatial variability discussion, we have now only used the core data (KC, KM, BI, and S100) for the years in which the cores overlap (1997–2000) (please refer to response to point 1 of referee #1 for more details).

5. The evaluation of ss-sulfate depletion from negative nss-sulfate values has to be completely revised;

CV. This has been now done in section 3.6. We have also included a new section (3.5) in which we discuss the ss- and nss-fractions.

6. I'm not convinced about the explanation of abrupt changes in sea salt deposition since 1950 in the S100 ice core (see, Specific Comments);

CV. We have now improved our discussion around this point as suggested by both referees. For more details on this, please refer to our response to point 4 of referee #1, and to section 4 which has been rewritten accordingly.

For these reasons, in my opinion, the manuscript needs major revisions before to be published on the The Cryosphere journal.

CV. As we mentioned to referee #1, we consider that after the referees's comments and suggestions, the manuscript has now improved and the hypothesis of sea-ice sea-salts as source to the S100 is now described in clearer way.

Specific and Minor Comments

Title: I'd suggest adding the Antarctic Region where Fimbul Ice Shelf is located (I think DML – Dronning Maud Land). Besides, since three shallow firn cores were analyzed, I'd suggest changing "ice cores" in "ice/firn cores".

CV. The title has now been corrected to: "Spatial and temporal variability of sea-salts in ice/firn cores from Fimbul Ice Shelf, Dronning Maud Land – DML, Antarctica".

Abstract. Authors should add some basic information about the sampling area.

CV. We have now modified the first part of the abstract, and now it reads: "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."

Line 22, page 1. Please, change "three firn cores" in "three shallow firn cores (about 20 m deep)

CV. Since we modified the first part of the abstract, we have included this suggestion when we refer only to the ice-rises cores (please see response to the previous comment).

Lines 22-23, page 1. Please, add "(Dronning Maud Land – DML) to "Fimbul Ice Shelf (FIS)" location.

CV. This has been added.

Line 24, page 1. Please, change "five snow pits" in "five snow pits (60-90 cm deep)"

CV. Since we have removed the snow pit data from the main text (please refer to response to point 1 of referee #1 for more details), this has been modified and moved to the Supplementary material.

Line 27, page 1. Please, change "elevation and distance to the sea" in "elevation (50- 400 m a.s.l.) and distance (3-117 km) to the sea.

CV. Since we removed the snow pit data, the sentence now reads: "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."

Lines 28-29, page 1. As the same Authors say at Lines 6-7, page 13, latitude and distance from the sea are related one to the other. I'd suggest referring just to the distance from the sea, as the most significant parameter (other than altitude) for the site characterization.

CV. We have now removed the sentence "Concentrations of these ions were found to decrease with latitude and distance from the sea." found in the abstract in view of the changes to section 3.4.

Lines 7-9 page 2. See Specific Comments for the interpretation of the S100 changes in sea salt deposition.

CV. This has been addressed in that part of the response letter. Section 4 in the manuscript has been changed accordingly.

Line 9, page 2. Please change "ice rises cores" in "firn rises cores".

CV. Here we referred to the cores drilled at the ice rises, therefore "ice rises cores" should be interpreted as the *cores drilled at the ice rises*. In order clarify the text, we have now changed "ice rises" for "ice-rises", when referring to the topographical feature.

Introduction. This section seems to be too long and contains several information well known to the Glaciology Community. I'd suggest to summarize such information (especially those related to sea salt sources, specifically discussed in Section 4) focusing on the specific features of low altitude coastal sites, located in areas characterized by the presence of ice rises and ice rumples.

CV. We consider that it is important to keep the information that it has been included in the introduction; therefore, we would like to keep it as it is.

Lines 2-3, page 4. I'd suggest changing "This evidence : : :: : : values : : :.) in "This hypothesis is supported by the experimental evidence that the original seawater SO4/Na ratio cannot be used in nss-SO4 calculation, leading to negative values : : :.".

CV. The sentence now reads: "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…"

Lines 7-8, page 4. I'd suggest changing this sentence in: "These negative values indicate that a lower SO4/Na ratio has to be used in nss-SO4 calculation; i.e., a depletion of SO4, with respect to seawater composition, occurred in wet and dry depositions."

CV. The sentence has now modified, and it now reads: "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."

Lines 9, page 4 – Line 15, page 5. In my opinion, the discussion about the formation of brine, frost flower and other possible sources of fractionated sea-salt aerosol, although interesting, is too long in the introduction. This part should be summarized, eventually moving some key sentences in the Discussion Section.

CV. We consider that it is important to keep this information as it is in the introduction to facilitate the understanding of the manuscript; therefore, we would like to keep it as it is.

Line 14, page 4. I think that frost flower, due their fragile structure, cannot cover the fractionated brine, but constitute an alternative processes leading the sulfate fractionation (as successively well explained by the Authors). I would suggest deleting "frost flower" in this sentence.

CV. "frost flower" has now been deleted from the sentence.

Line 6, page 6. Authors should give some basic information on the ranges of altitude and distance from the sea of the FIS ice rises (even if specific data are reported in Table 1). For instance: "Several ice rises (250-400 m a.s.l.; 10-50 km from the sea) are found at FIS : : :..".

CV. The sentence now reads: "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."

Line 30, page 6. Please, add "about 20 m deep" after "Three shallow firn cores".

CV. This has been added.

Lines 8-9, page 7. I think 4-8 cm is the sample resolution for firn cores. Authors should clarify that.

CV. The referee is correct. We have changed "sample length" for "sample resolution".

Lines 11-13, page 7. Information on the sample resolution for the S100 ice core should be reported.

CV. We have now included the information as: "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."

Line 14, page 7. Please, change "five snow pits : : :" in "five snow pits (60-90 cm deep): : :".

CV. This has been now modified and moved to the Supplementary material.

Line 20, page 7. I'd suggest adding: ", therefore snow pits samples cover the last year deposition".

CV. This has now been added, and moved to the Supplementary material.

Line 22, page 7. There is a reason why ammonium was not determined together with the other cations?

CV. Ammonium was not measured in the S100 core due to analytical restrictions. Ammonium was measured in the KC, KM, and BI cores, and snow pits, but not reported because its concentrations were often close to the detection limit.

Line 4, page 8. Here, a resolution of 2 cm is indicated, while Lines 17-18 report a resolution of 4 cm. Authors are requested to clarify the resolution of G4a and G5a snow pits.

CV. The sample resolution of pits G4a y G5a was 4 cm. We have now corrected the sentence, and this part has been moved to the Supplementary material.

Lines 14-16, page 8. I'm aware that Na probably originates mainly from sea salt in this coastal area. However, Authors should use ss-Na as specific sea salt marker or justify the choice of using total Na by demonstrating that nss-Na is a negligible (for instance, lower than 5%) fraction of total Na. This could be especially relevant for KC site, where PCA shows a significant crustal contribution.

CV. The referee is correct. We have now included this in section 3.5 in which we discuss the Cl^-/Na^+ ratio, mechanisms of Cl^- and Na^+ enrichment/depletion, and calculated $nssNa^+$, $ssNa^+$, and percentage of $nssNa^+$ to total Na^+, as suggested by the referee. For more details on this, please refer to the response given to point 2 of referee #2.

Line 4 and Line 8, page 9. How the Authors identified the previous summer layers, if $\delta^{18}O$ measurements were not performed? By ice lens, different density, or other physical features? Authors should clarify their procedure, even if a reference is cited.

CV. The estimation of the summer layer in each pit was visually and stratifically done by Sinisalo et al. (2013). We have now clarified the paragraph (and as metioned before, moved the snow pit data to the Supplementary material), and it now reads: "The previous summer layer in pits M1, M2 and G3–G5 was visually and stratifically identified at 120–160 cm depth according to Sinisalo et al. (2013); however, chemical sampling was done just down to a depth of 60–90 cm in each snow pit reported in this study, and samples for water stable isotope were not collected. Consequently, the snow depth at the different snow pit sites in which ions were analysed represents less than a year of accumulation (Table S1), and a highly precise dating (e.g. with monthly resolution) of the snow layers was not possible."

Section 3.1. Firn cores and snow pits values are reported as median. This is correct but it was not possible to evaluate the data variability, in order to compare the different sea salt contributes in the different sampling site. If median is used, then 25th and 75th percentile have to be shown, at least. I'd suggest plotting box plots with median, percentiles and outlier for each data set and reporting mean, minimum, maximum, and standard deviation in Table 2. In this way, it will be easier to appreciate the significance of the inter-site comparison.

CV. We have now included mean, minimum, maximum, and standard deviation in Table 2 for all ions in the different cores (and snow pits in Table S3). Box-plots for the different ions in the cores and snow pits have now been included as Figure S1–S3 in the Supplementary material.

Line 4, page 10 and following. PCA analysis. Usually, PCA analysis is carried out on raw data. Authors should clarify why they used normalized values as input for PCA data matrices. Indeed, PCA analysis on raw data is able to give results independent site-by-site and comparable among them. I do not know if this can be a result of the normalization, but the factor loading in every PCx factor seems to be quite low (lower than 0.5 in the majority of the loadings). In every way, I agree with the PCA results: the factors are surely related to sea salt, biogenic emission (mixed to nitrate) and crustal (for the site farthest from the sea) sources.

CV. The PCA technique calculates a new projection of the input data set on the basis of the standard deviation of each variable. Consequently, variables must be normalized so standard deviations are weighted equally when using PCA analysis.

Lines 26-27, page 10. As a marker of the crustal source, the nss-Ca fraction has to be calculated at least for the KC site (but it could be useful to evaluate the ss- and nssfractions for all the components in every data record).

CV. ss-fractions have been now calculated for Cl^-, SO_4^{2-}, K^+, Mg^{2+}, and Na^+ in all the cores, and presented in Table 5 and Table 6. The ss-fraction results are discussed in section 3.5. Please refer to our response to point 2 of referee #2 for more details.

Line 29-30, page 10. A further explanation could be common transport processes or pathways from marine areas at lower latitude. I do not think that nitrate, as a major nutrient, is a limiting factor for phytoplanktonic bloom in the Antarctic marine regions.

CV. Referee #1 has also commented on this. The paragraph has been re-written in view of the referees' comments (please refer to the response to referee #1 for more details).

Line 8, page 11. The common sea-salt source between Cl and Na has to be confirmed (other than from PCA analysis) by calculating the Cl/ssNa ratios and comparing them with seawater composition. In this way, also a possible chloride depletion (for instance, by wet or dry deposition of aged sea spray aerosol) could be observed.

CV. We have now included a discussion on this in a new section (3.5). For more details, please refer to our response to point 2 of referee #2.

In particular, it has to be noted and discussed that Na and Cl have quite different temporal profiles in the KC firn core (constant or light increase for Cl; quite sharp decrease for Na).

CV. Figure 2 b and d show the Cl^- and Na^+ annual concentrations in the KC core. As noted in Table S4 the slopes of the linear regression for both ions are not significant at the 95 % confidence level, therefore we did not discuss the increase/decrease for Cl^- and Na^+ in the KC core.

Section 3.3. Seasonal pattern. I have some doubts about the linear interpolation procedure. Indeed, a simulated resolution of about 3 days has not a physical meaning, especially considering the variability of composition and temporal occurrence and frequency of snowfall events and dry deposition. Have the Authors information about the seasonal pattern of snowfalls in the studied area?

CV. As we noted in the response to the referees of the paper by Vega et al. (2016) (https://www.the-cryosphere-discuss.net/tc-2016-164/tc-2016-164-AC1.pdf), extensive precipitation records at the core sites at FIS are, to our knowledge, not existent. The KC, and KM time scales used in this manuscript were constructed by Vega et al. (2016) based on δ^{18}O winter minima and summer maxima, and assuming uniform precipitation throughout the year at the core sites. This assumption was made on the basis of the precipitation regime at DML reported by Schlosser et al. (2008) which showed high temporal variability of the precipitation monthly sums due to the influence of cyclone activity affecting both coastal and inland regions. At Neumayer, the closest station to the core sites, two precipitation maxima are identifiable for the period 2001–2006 (April and October) possibly due to the semi-annual oscillation of the circumpolar trough (Schlosser el al., 2008). We have now included this information in at the beginning of section 3.3.

I strongly suggest attributing the sample seasonality by using the raw data and the δ^{18}O profiles, identifying the four seasons by high, low or intermediate δ^{18}O values or, simply, classifying the samples as "summer" or "winter" samples, by assuming a threshold for summer/winter δ^{18}O values. If the results of this seasonal attribution are different from those reported in figure 3, all the section should be revised accordingly.

CV. Both referees have pointed to this, therefore we have re-done the analysis considering δ^{18}O winter minima and summer maxima reported by Vega et al. (2016) as tie points for the seasonal analysis. We have then separated the annual samples into "winter" and "summer" samples, and we have rewritten section 3.3 accordingly.

Section 3.4. Spatial variability. As the same Authors say (Line 6-7, page 13), differences in latitude reflect differences in distance from the coast. Besides, longitude variations are too little (at least for snow pits) to constitute a significant parameter for ion composition (as demonstrated by the not significant relationships, see Line 8-9, page 13). In the studied area, I think the only significant parameter is the distance from the sea and, possibly, the altitude (at least for the firn cores). Therefore, I suggest to discuss only the effect of the distance from the sea (and altitude and position with respect to the glacier tongue, if firn cores are included in the discussion) in this section.

CV. Referee #1 also pinpointed this. In order to keep the spatial variability discussion, we have now only used the core data (KC, KM, BI, and S100) for the years in which the cores overlap (1997–2000). The discussion has been changed accordingly, with no significant relationship between the median annual ion concentrations and latitude, site elevation, and distance from the sea for any of the species (except for SO_4^{2-} and MSA vs. latitude and longitude, respectively) (please refer to response to point 1 of referee #1 for more details).

Line 5, page 13. Probably, all these ions are mainly (or completely) coming from sea spray. It could be interesting to calculate their ss- and nss- fractions (at least for Na, Ca and Mg).

CV. ss-fractions have been now calculated for Cl^-, SO_4^{2-}, K^+, Mg^{2+}, and Na^+ in all the cores, and presented in Table 5 and Table 6. The ss-fraction results are discussed in section3.5.

Section 3.6. Authors have to be aware that the evaluation of sulfate depletion from the observation of negative values of nss-sulfate is a difficult and controversial task. In coastal areas, the contribution of nss-SO4 from phytoplanktonic emissions (marked by relatively high MSA concentrations) could be very relevant in spring/summer period. The nss-SO4 originated by the biogenic source can "cover" the ss-sulfate depletion by adding nss-SO4 to the sulfate budget. Therefore, a ss-sulfate depletion can occur even if no negative nss-SO4 values were found. The correction for biogenic nsssulfate can be made by considering aerosol size distribution, aerosol seasonality and the nssSO4/MSA ratio from DMS atmospheric oxidation. No discussion on this relevant topic is given in this manuscript.

CV. This was a common point between the referees. We followed the referees suggestions and in order to clarify this point and present a more solid discussion on the topic, we have calculated the

$MSA/nssSO_4^{2-}$ ratio, and calculated the percentage of biogenic $nssSO_4^{2-}$ (bio-$nssSO_4^{2-}$) to total SO_4^{2-} in all the cores (Table 7), and section 3.6 has been rewritten accordingly.

Line 23, page 13. Please, change: ": : : increase in ion concentrations after 1950s : : : " in ": : : increase in ion concentrations after 1950s in the S100 ice core: : :".

CV. We removed the sentence in the revised version of the manuscript.

Line 30, page 13. Nitrate and sulfate correlation coefficient can be statistically significant, probably thanks to the high number of samples, but their values are so low (-0.04 and -0.10) to exclude every real correlation.

CV. We have removed the "deposition regime" section in the revised version of the manuscript after adding section 3.5 on sea-salt and non sea-salt fractions, and modified sections 3.6 and 4.

Lines 11-12, page 14. A dominant role of dry deposition should be demonstrated also by a significant negative correlation between snow concentration and accumulation rate. By looking to Table 5, that does not occur for Na (Rconc = -0.12). I think that both wet and dry deposition are relevant for a site so near to the sea (3 km) and located at so low altitude (48 m a.s.l.) to be affected from snowfall deposition, as well as from direct sea-spray primary production (i.e., aerosol directly produced by wind action on open sea surface or on frost flowers/brine structures over the sea ice surface).

CV. As mentioned above, we removed the "deposition regime" section in the revised version of the manuscript.

Line 25, page 14. The value 0.06 is the well-known SO4/Na molar ratio in seawater (corresponding to the w/w ratio of 0.25) and the reference here cited is not pertinent.

CV. We have now referred to "Table S2, Supplementary material" for the current manuscript, and removed the Vega et al. (2015) reference.

Line 29, page 14. the negative values do not mean that "nssSO4 is strongly depleted in SO4 relative to Na", but that the sea-salt content in the snow is depleted in ss-sulfate with respect to seawater composition, so leading to an under-evaluation of nssSO4, if the 0.06 SO4/Na ratio is used. Indeed, if part of the original sns-sulfate is precipitated as mirabilite (in case of frost flower formation), the sea salt aerosol originated from the sea-ice surface is depleted in seawater sulfate.

CV. The referee is correct. We have now rewritten the sentence as: "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,…".

Line 1, page 15 and following. In order to better understand the meaning of the lower SO4/Na (k) values here calculated, the Authors are requested to compare these values (k', k", k"') with the SO4/Na ratio for the frost flowers (0.07 w/w, Wagenbach et al., 1998, corresponding to 0.017 mol/mol). In this way, Authors could evaluate the relative contributions of sea salt from seawater (k = 0 0.06) and from frost flowers (k = 0.017).

CV. We have now included this discussion in section 3.6.

Section 4. Discussion. The discussion is focused on the interpretation of the temporal trend of sea-salt components in the S100 ice core, showing a dramatic increase from 1950 to 2000. Indeed, this *increase is impressive and, in my knowledge, never observed (with this intensity) in coastal and inner area of Antarctica (with the exception, of course, of the interglacial/glacial changes). Authors report a 6-times concentration increase from 1750-1949 to 1950-2000 periods. This difference, however, is calculate on the long-period median values (also in this case, percentile values or mean values with standard deviations could help in evaluating the data variability).*

CV. We have now included mean, minimum, maximum, and standard deviation in Table 2 for all ions in the different cores (and snow pits in Table S3). Box-plots for the different ions in the cores and snow pits have now been included as Figure S1–S3 in the Supplementary material.

If we observe the increasing trend of Na in figure 2, we can note that the concentration increases follows a continuous and quite constant trend, up to values as high as 200 umol/L. In comparison with the quite constant concentrations measured along the 1750-1949 period (around 15 umol/L), the Na concentration increases of a factor higher than 13. Chloride follows a similar trend. These S100 sea-salt values are about 4 to 20 times higher than those measured in the three firn cores. Authors attribute this large variation to changes in extension and persistence of sea ice east of the glacier tongue, after the tongue breaking on 1967. The decrease of the glacial tongue could have been against the preservation of multi-annual sea ice, promoting annual fast sea ice, where the formation of fractionated sea-salt aerosol and of frost flowers is more efficient (Lines 3-6, page 17). This is possible (and probable), but some experimental evidences, in my opinion, make weak this hypothesis. 1. The increase is not related to abrupt changes starting on 1967, but Figure 2 shows a very gradual and progressive trend (at least seeing the "linear trends" plotted in figures 2a and 2c) since 1950, before the glacier tongue breaking.

CV. As suggested by the referees, we have now included some glaciological data that might help to support our hypothesis regarding the increase in sea-salt after 1950s, and the increase in the ssSO_4^{2-} depletion factor in the 1950–2000 period in comparison to the 1737–1949 period. 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. However, we cannot assume the same ice velocities for S100 for the pre-1950 and post-1950 periods. By using the S100 core sea-salt results we could 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; and that the calving event at Trolltunga in 1967 (Vinje, 1975), enhanced the input of fractionated sea-salts to the S100 core by modifying the sea-ice conditions around S100. This could be supported by the fact that negative nssSO_4^{2-} values slowly decreased between 1950–1966 showing a marked minimum around 1966 (±3 years), which could have been induced by the calving event.

2. The two firn cores located on the same side of the glacier tongue (KM and KC) show sea-salt concentrations very lower and not characterized by an in-creasing trend (at least for the years covered by the records). In particular, the nearest fin core (KC) show stable (Cl) or slightly decreasing (Na) concentrations about 10 times lower than those measured in the most recent S100 sections (Figure 2b and 2d).

CV. The referee is correct. However, it is difficult to explain, with the current data, the lower sea-salt values found at the ice-rises in comparison to the S100 site after the 1950s. We hypothesize that (section 4): "Considering that we found no correlation between ion concentrations 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 μm) (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, 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."

3. All the firn cores (and, in particular, KC and KM), do not show negative nss-sulfate values (Figure 4c). Authors attribute this different pattern to changes in altitude (lines 25-28, page 16), but KC, for *instance, is only 200 m above the S100 site and particles as small as 1 um (Line 26, page 16) are easily transported to very high altitudes. If the altitude plays a similar dramatic effect, then no contribution of sea salt from frost flowers or salty snow sublimated aerosol should be observed in high-altitude plateau sites. On the contrary, some evidences of ss-sulfate depletion (shown by negative nss-SO4 values) have been found at the sites where Dome Fuji, EPICA-DML and EPICA-DC ice cores were drilled. In conclusion, in my opinion, the Authors hypothesis seems to be not confirmed by the firn cores data and other mechanisms should be investigated.*

CV. This point has now been corrected in view of the high percentage of bio-nssSO_4^{2-} to total SO_4^{2-} found for the ice-rises cores (Table 7). We have now included the following paragraph in section 3.6: “In the KM and BI cores, the estimation of bio-nssSO_4^{2-} surpasses the total SO_4^{2-} observed in these cores, while in the KC core the bio-nssSO_4^{2-} would represent about 50% of total SO_4^{2-}. These high percentages were expected especially in the KC, and BI cores, in which the nssSO_4^{2-} fraction dominates over ssSO_4^{2-} (section 3.5). 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 SO_4^{2-} fractionation in sea-salt found in snow but rather reflect the dominance of bio-nssSO_4^{2-} 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-}.” We also added (in section 4): “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 S4) 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.”

Lines 19-20, page 17. I strong suggest adding at least a composite figure showing the most relevant changes of extension and shape of the glacier tongue since 1967.

CV. Although the data needed to fulfil the referee´s request is available, i.e. front lines for the Trolltunga extent between 1952 and 2014, these data conforms a manuscript by J. van Oostveen (NPI) et al., which is currently in preparation, therefore, we cannot publish the dataset in the present manuscript. We consider that including the Corona Satellite data from 1963 and the present MODIS MOA image (from Quantarctica) is sufficient to show the abrupt change in Trolltunga, that took place within October 1963 and 1973 (J. van Oostveen, personal communication). For more information on the Trolltunga extent over the last decades, please contact J. van Oostveen (jelte.van.oostveen@npolar.no).

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

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Abstract

Major ions were analysed in firn/ice cores~~, and snow pits th~~ ~~ree firn cores from different ice rises~~ 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 ~~firn~~ 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. ~~In addition, Kupol Ciolkovskogo (KC), Kupol Moskovskij (KM), and Blåskimen Island (BI), a 100 m long core drilled near the FIS edge (S100), and five snow pits (M1, M2, G3, G4, and G5~~, 60–90 cm deep) were ~~sampled~~ ~~along Jutulstraumen, the largest outlet glacier in DML~~~~on the ice shelf.~~ 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~~117~~ km) to the sea. Sea-salt species (mainly Na^{+} and Cl^{-}~~$^{-}$~~) generally dominate the precipitation chemistry in the study region. ~~Concentrations of these ions were found to decrease with latitude and distance from the sea.~~ We associate a significant six-fold increase in sea-salts, observed in the S100 core after the 1950s, with ~~a change in deposition regime~~enhanced sea-salt aerosol production from blowing salty snow over sea-ice. 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 ~~by dry deposition~~originating from salty snow found on sea-ice. In contrast, ~~wet deposition of atmospheric sea-salts is dominant in the three ice-rises cores, and no evidence~~ there is no evidence o~~o~~f a significant contribution of fractionated sea-salt to ~~these~~ the ice-rises sites, where the signal~~sites~~ would be most likely masked by the large inputs of biogenic sulfate estimated for these sites~~was found~~. In summary, these results suggest that the S100 core contains a ~~more local~~ sea-salt record dominated by~~ processed~~ ~~signal, dominated by~~ processes ~~during~~ of sea-ice formation in the neighbouring waters. In contrast, the ~~ice~~ ice-rises firn cores register the larger-scale signal of atmospheric flow conditions and a less efficient transport of sea-salt aerosols ~~produced over open water rather than local changes in sea-ice, wind-blown snow accumulated over sea-ice, and frost flower formation~~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 sea-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 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~~2, 2004~~; 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 ~~evidence consists~~that the original seawater SO_4^{2-}/Na$^+$ ratio cannot be used in the non sea-salt sulfate (nssSO_4^{2-}) calculations, leading to ~~of~~ negative ~~non sea-salt sulfate (n~~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~~ Rankin and Wolff,~~ 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~~suggest that the sea-salt source was highly fractionated compared to seawater~~.

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~~ or frost flowers~~, 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~~ the~~ temperature, e.g. sodium sulfate or mirabilite ($Na_2SO_4 \cdot 10\,H_2O$) starts to precipitate at temperatures below −~~-~~8 °C, and 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 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.

2 Methods

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~~50~~ km from the coast) are found at FIS, varying in size from 15 to 1200 km^2, and located approximately 200 km apart.

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 ~~length~~ 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.

~~In addition to the firn and ice cores, five snow pits (60–90 cm deep), (M1, M2, G3, G4, and G5, with additional re-sampling of G4 and G5 as G4a and G5a), were sampled at different sites at FIS during Nov.–Dec. 2009 by a field team from NPI (Figure 1, Table 1). The snow pits were sampled at 20 cm (M1, M2, and G3, G4, and G5) and 4 cm (G4a and G5a) intervals; samples were transported and kept frozen until analysis. The previous summer surface at the snow pit sites was estimated to be located at a depth between 120 and 160 cm (Sinisalo et al., 2013), therefore, snow pit samples covered the last year deposition.~~

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 %.

~~In addition, major ions (Cl^-, Br^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+} and Ca^{2+}) were measured in the FIS snow pits. Triplicate snow samples were taken at each depth interval. Hence, average concentrations of each ion at each depth interval are reported here. Additionally, pits G4 and G5 were re-sampled at 42 cm intervals and denoted as pits G4a and G5a, with one sample per depth interval (pits G4a and G5a). For these snow pits sampled at high resolution only Cl^-, NO_3^-, and SO_4^{2-} are available. All snow pit samples were transported frozen from the sampling site to NPI and then to the Department of Earth Sciences, Uppsala University, where major ions were~~

~~quantified using a Metrohm ProfIC 850 ion chromatograph. The reproducibility of the measurements was within 5 % and detection limits (DL) were below 0.3 μmol L^{-1} for each ion.~~

~~Hereafter, brackets are used to denote ionic concentrations expressed in μmol L^{-1}.~~

Cl^{-}, SO_4^{2-}, K^{+}, and Mg^{2+} n~~N~~on 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:

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

where

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

using the standard mean chemical composition of seawater with ion concentration expressed in μmol L^{-1} (k values are listed in Table S~~1~~2), 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 u~~as:~~

~~$[\mathrm{nssNa}^{+}]=k \times [\mathrm{Ca}^{2+}]_{\mathrm{total}}$,~~

~~and~~

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

~~u~~using Ca^{2+} as reference ion and k= 1.40 for Earth's crust composition (Lutgens and Tarbuck, 2000) (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). ~~Water stable isotope analyses were not performed in the FIS snow pits.~~

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 ($\delta^{18}O$), 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), 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).

~~2.5 Snow pits timescales~~

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

3 Results

3.1 Ion concentrations and sources

Median, mean, maximum, minimum, and standard deviation (σ) of ~~-~~concentration ~~values~~ for all ions measured in the cores ~~and snow pits~~ are shown in Table 2. ~~In~~

~~addition, b~~Box-plots of raw ion concentrations in the different cores ~~and snow pits~~ are shown in Figure~~s~~ S1~~–S3~~ in the S~~s~~upplementary material. In addition, median, mean, maximum, minimum, and σ 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, concentrations in the KM core are higher than in the other ice~~-~~-rises cores, and snow pits, e.g. about eight-fold higher concentrations of Na^+, K^+, Mg^{2+} and Cl~~$^{-}$~~$^{-}$ in KM than in the KC core 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).

Ion concentrations in the snow pits (Table S3) are in reasonable agreement with firn and ice core values ~~reported in Table 2~~ and with ion concentration ranges for snow pits previously sampled at FIS (Mulvaney et al., 1993). 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 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 ~~ion concentrations were logarithmized~~ (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 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. ~~The h~~High loadings of NO_3^- and MSA in PC2, and thus, coherence between both species, ~~was already~~have been 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) suggest that e~~E~~nhanced 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).

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^+~~, SO_4^{2-}~~, and Mg^{2+} median ~~, and Ca^{2+}~~ 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 ~~S1~~ S4 in the

S~~s~~upplementary material). Consequently, 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).

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 S~~1~~4~~2~~ in the S~~s~~upplementary material). Such ~~a~~ significant linear trend was not observed in the KC ion record~~s~~ ~~from the KC core~~ over the same period.

Ions, with the exception of MSA, also show a positive and significant trend between 1737–1949, (Table S4~~1~~), however, the increase is less marked than during the 1950–2000 period.

3.3 Sub-annual variability of ion concentrations

~~3.3~~ The lack of extensive precipitation measurements at sub-annual resolution near the sampling sites at FIS, ~~difficult~~makes a precise reconstruction of the precipitation regime at the area difficult. ~~In order~~ T~~t~~o obtain a time scale for the KC, KM, and BI ice-rises cores, Vega et al. (2016) employed δ^{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 ~~due~~ a manifestation ~~to~~of the semi-annual oscillation of the circumpolar trough (Schlosser el al., 2008). Considering the above,

~~T~~to investigate the ~~seasonality~~ sub-annual variability of the different ion groups in the KM, BI, and S100 cores, we associated ~~used~~ the ~~δ^{18}O~~ winter minima and summer maxima in δ^{18}O determined ~~by Vega et al. (2016)~~ in the KC, KM, and BI cores (Vega et al., 2016), and in the S100 core (Kaczmarska et al. ,~~(~~2004)~~ in the S100 core~~, ~~to~~with~~ define~~ the months of July and January, respectively. The values for April and October were derived by interpolation~~Then we used the intermediate values~~ between January–July, and July–January, ~~to estimate the months of April and October,~~ 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). ~~a composite year of data was produced for each ion species, i.e. for each year, a linear interpolation was done in the concentration domain of each ion at a resolution of 0.01-years (3.65 days). The seasonal~~Summer and winter mean concentrations~~ averages~~ were then calculated ~~for the months MAM (autumn), JJA (winter), SON (spring) and DJF (summer).~~ using~~All logarithmized~~based on logarithms of raw ion concentrations ~~were~~ expressed in µmol L^{-1}~~ and were logarithmized~~. Ion concentrations were not available at the top 2 m (~~snow pit data~~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, ~~seasonalities~~ sub-annual variability were investigated only during the period 1995–2000, where the concentrations have sufficient temporal resolution~~ for subannual analyses~~. The resulting ~~seasonal~~ summer and winter mean ~~average~~ concentrations in the cores are presented in Figure 3~~.~~ .

Sea-salt species (Na^{+} and Cl^{-}, Figure 3a) show ~~minimum~~ lower concentrations during ~~spring~~ summer in the ~~KM~~ BI, and S100 core~~r~~, whereas in the ~~S100~~ KM core ~~minimum~~ summer and winter ~~values are reached during summer~~show similar means. ~~Maximum concentrations are reached during summer (KM) and spring/autumn (S100). In the BI core, sea-salt seasonalities are less pronounced.~~ Both Mg^{2+} and Ca^{2+} (Figure 3b) show ~~high values~~similar means in both summer and ~~in the KM core, and in spring in the S100 core, whereas, again, a less marked seasonality is observed in the BI core~~winter~~.~~ . MSA concentrations (Figure 3c) show ~~consistent and marked spring/summer~~summer maxima in all three cores, ~~and a marked winter minima in the BI core~~with a higher summer to winter difference in the BI core, compared with the KM, and S100 cores.~~,~~ These summer maxima ~~which is~~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 t~~autumn in t~~he S100 core. NO_3^- and SO_4^{2-} concentrations (Figure 3d) show a distinct increase toward the ~~spring/~~summer in the BI core, which is also observed in the KM core, although less defined. 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. ~~In the S100 core, higher NO_3^- and SO_4^{2-} concentrations are found during the spring.~~

3.4 Ions spatial variability

In order to investigate ion spatial variability at FIS, we used median annual ion concentrations in the different ~~snow pits (M1, M2, G3, G4, and G5), and~~ ice-rises cores (KC, KM and BI), and S100 for the ~~year 2009~~~~different~~ overlapping period between 1997 and 2000~~s~~, and compared them with latitude, longitude, site elevation, and distance from the sea (obtained from the GIS package Quantarctica, www.quantarctica.org) (Table 4). ~~Since it has been found that the chemical records in the KM and BI cores appear to be affected by surface topography and local meteorological conditions (Vega et al., 2016), we also calculated the correlation coefficients (R values) between concentration and latitude, longitude, and distance from the sea, only using the snow pit data.~~

~~Concentrations of~~ Only annual SO_4^{2-} and annual MSA concentrations show a significant decrease (at the 95 % confidence level) with latitude, and ~~Br^-, Na^+, K^+, Mg^{2+} and Ca^{2+} show a significant decrease at the 95 % confidence level with increased~~east ~~latitude~~longitude, respectively. ~~i.e. with increasing distance from the coast~~~~lower annual MSA medians towards the eastern sites.~~ ~~Correlation coefficients are high for Cl^- and SO_4^{2-}, however, not significant.~~ No significant relationship is found between the median annual ion concentrations and ~~longitude~~ latitude, site elevation, and distance from the sea for any of the species. ~~When comparing concentrations with the distance from the sea (obtained from the GIS package Quantarctica, www.quantarctica.org), it was found that Br^-, SO_4^{2-}, Na^+, and Ca^{2+} show a significant decrease with increasing distance from the sea when only the snow pits are considered in the analysis; i.e. significance is lost when the KM and BI 2009 median values are included.~~ These findings ~~agree~~ contrast with previous studies from western Dronning Maud Land (WDML) (Stenberg et al., 1998), ~~confirming~~ where a strong correlation between ~~that~~ sea-salt concentrations ~~are~~ and ~~strongly dependent on the~~ distance from the sea was found in this area. ~~This~~ We attribute the lack of significance ~~relationship loses significance when~~for the correlations presented in Table 4 to the local effects on annual SMB due to topography and local meteorology at the ~~these sites~~KM and BI sites, reported by ~~(~~Vega et al. (2~~, 2~~016)~~)~~.

3.5 Sea-salt and non sea-salt fractions

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-salt fractions. Table 5 shows the Cl^-/Na^+ ratio (expressed in µmol L^{-1}) in the KC, KM, BI, and S100 cores. Medians 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 ratio vary between 1.5–3.8, and minima between 0.1–0.9. These results show a clear difference in the Cl^-/Na^+ ratios between the ice-rises cores and the S100 core~~s~~, i.e. a Cl^- to Na^+ unbalance 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~~to the~~ $nssNa^+$ sources (Legrand and Delmas, 1988). T~~, in addition with~~his unbalance can be enhanced ~~a~~by a depletion of Cl^- due to shorter sea-salt atmospheric residence times, 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 ~~sulfur~~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~~, with no evident seasonal differentiation.~~. Since the temporal resolution of the S100 core only allows sub-annual values for the period 1995–2000~~in the S100 core were only available for most recent years of the core time scale (i.e. 1995–2000)~~, is not possible to assess a ~~more conclusive idea on the seasonality~~sub-annual patter on ~~f~~ the Cl^-/Na^+ ratio~~ is not pertinent~~, 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-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 μmol L^{-1}) for Earth's crust (Lutgens and Tarbuck, 2000). This assumption will introduce an overestimation of the $nssNa^+$ fraction proportional to the ratio $Ca^{2+}/Na^+ = 0.02$ (with concentrations expressed in μmol 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 shows the $nssNa^+$, $ssNa^+$, and percentage of $nssNa^+$ to total Na^+ in the different cores. Since some of the calculated $ssNa^+$ values in the KC core were negative (5 %), $ssNa^+$ statistics in Table 5 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 21 % in comparison to the KM, BI, and S100 cores (3 %, 4 %, and 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+} (Table 6). The sea-salt fraction clearly dominates in all ions, with the exception of SO_4^{2-} in the KC, and BI cores, which shows almost three times more $nssSO_4^{2-}$ than $ssSO_4^{-}$. Nss-fractions often have negative values which can be associated to an $ssNa^+$ enrichment or depletion of major ions 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 % for the S100 (1950–2000), and up to 43 % for the KM core.

~~the records from the ice rises KM and BI are included, reflecting the influence of local effects on annual SMB at these sites (Vega et al., 2016).~~

~~Since the elevation differences between the snow pits are only in the order of tens of meters, we did not investigate the elevation dependence of mean ion concentrations.~~ ~~Since it has been found that the chemical records in the KM and BI cores appear to be affected by surface topography and local meteorological conditions (Vega et al., 2016), we also calculated the correlation coefficients (R values) between concentration and latitude, longitude, and distance from the sea, only using the snow pit data.~~

3.6 ~~Deposition regimes~~Evidence for increased fractionated nss-SO_4^{2-} after 1950s

~~3.5~~

~~In order to explain the marked increase in ion concentrations after the 1950s in the S100 ice core, and the negative trend observed in SMB for the S100 and KC cores for the second half of the 20th century (Figure 2e and f) (Vega et al., 2016), we investigated the deposition regimes at FIS. We employed correlation analysis to assess the relationship between the annual average concentrations of all ions and annual SMB at the different core sites for the respective period covered by each core (as noted in Table 2). Correlation coefficients (R_{conc}, Table 75) were < 0.57 and non-significant at the 95 % confidence interval (with the exception of NO_3^- and SO_4^{2-} in the S100 core, which were significant but extremely low as to be considered), which indicates that accumulation rates, and thus, wet deposition is not controlling~~

~~the ion concentration at the different core sites at FIS. We also investigated the relationship between chemical flux, expressed as μmol cm^{-2} yr^{-1} and annual snow accumulation at the different core sites (R_{flux}, Table 75). A significant positive correlation between fluxes and accumulation exists for almost all ion species analysed in the different cores with the exception of sea-salts in the S100 core, with significant R_{flux} values larger than 0.5 for most ions (with the exception of MSA in the KC and S100 cores, and SO_4^{2-}, Mg^{2+} and Ca^{2+} in the S100 core). This indicates that most of the variance in the ion flux can be explained by changing snow accumulation rate at each site for most of ions, with the exception of sea-salts in the S100 core. Therefore, dry deposition of sea-salt aerosols would be the controlling factor in sea-salt fluxes at the S100 site. To confirm that the increase in sea-salt fluxes observed after the 1950s is due to a change in sea-salt deposition mechanism, we calculated the R_{conc} and R_{flux} values for the periods 1737–1949 and 1950–2000 (Table 75). While R_{conc} values for both periods were not significant and low, during 1737–1949, all R_{flux} values were significant (R>0.41). This indicates that during the period 1737–1949, most of the variance in all ion fluxes can be related to SMB at the S100 site. Sea-salt R_{flux} values decreased (with the exception of R_{flux} of NO_3^-) and became non-significant in the case of Cl^-, SO_4^{2-}, and Ca^{2+} during the 1950–2000 period, which points to a change in the deposition regime after the 1950s.~~

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

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-} is strongly depleted in SO_4^{2-} relative to Na^+ (Rankin and Wolff, 2002), it is necessary to account for the biogenic contribution to total nssSO_4^{2-} at each core. 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 μmol 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 ~~8~~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 bio-nssSO_4^{2-} to total SO_4^{2-} (Table ~~8~~7). In the KM and BI cores, the estimation of bio-nssSO_4^{2-} surpasses the total SO_4^{2-} observed in these cores, while in the KC core the bio-nssSO_4^{2-} would represent about 50 % of total SO_4^{2-}. These high percentages were expected especially in the KC, and BI cores, in which the nssSO_4^{2-} fraction dominates over ssSO_4^{2-} (section 3.5). In the S100 core, bio-nssSO_4^{2-} varies according to the time period considered with percentages three~~3~~ 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 SO_4^{2-} fractionation in sea-salt found in snow but rather reflect the dominance of bio-nssSO_4^{2-} 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 ~~calculated~~ used the nssSO_4^{2-} fraction calculated as described in section 2.3, using *k* values of 0.06 (~~see~~ Table S2~~1~~, S~~s~~upplementary material~~ in Vega et al., 2015~~). The percentage of nssSO_4^{2-} relative to total SO_4^{2-} is ~~two~~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~~, and G4~~ (not shown), with~~N~~ negative nssSO_4^{2-} values ~~are~~ being more pronounced after the 1950s~~, which will be discussed in the following sections.~~. These negative values found in the snow, i.e. the sea-salt content in snow is ~~nssSO_4^{2-} is~~ strongly depleted in ssSO_4^{2-} relative to seawater composition~~Na^+ (Rankin and Wolff, 2002)~~, 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 therefore followed the approach by Inoue et al. (2017), to obtain a new value of *k*, *k'*.~~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 ~~fitting~~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) ~~less sensitive to large changes in small sections of the data series (e.g. the ssNa^+ values higher than 200 µmol L^{-1})~~ than the usual least squares method. ~~We used the default robust regression option within of the *fitlm* function found in Matlab (version R2015b).~~ 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 ~~a~~ corrected *k* values (*k'*) ~~This *k'* was calculated~~ by subtracting the absolute value of the linear regression slope ~~of the linear regression (slope= 0.02, in Figure 5) for the nssSO_4^{2-} concentrations vs. Na^+ concentrations (for the whole period, i.e. 1737–2000)~~ from the constant $k = \frac{[SO_4^{2-}]}{Na^+}$ in seawater ~~*k*~~ (~~calculated as described in section 2.3~~Table S~~1~~2), i.e. $k'_{1737-2000} = -0.$~~04~~02~~(in µmol L^{-1}).~~, ~~Similarly, we calculated *k''* for the period 1737–1949 (~~$k'_{1737-1949} = -0.0$~~2~~3~~)~~, and ~~*k'''* for the period 1950–2000 (~~$k'_{1950-2000}$ = ~~k'=~~ $-0.$~~04~~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 ($k = k/k'$~~$k = k/k'$~~) of 5.5 for a firn core drilled at eastern Ronne Ice Shelf. The S100 core presents depletion factors of two~~2~~ for the period 1737–1949, and three~~3~~ for the period 1950–2000.

The annual nssSO$_4^{2-}$ fraction, without the effect of sulfate fractionation, was then recalculated using the values ~~of~~ for *k'* of 0.02 and 0.03 ~~, and *k''*~~ (Table 8~~96~~, and Figure 4a y b, red and blue lines). ~~nssSO$_4^{2-}$ for the KC, KM, and BI cores is shown in Figure 4c (calculated using *k*=0.06). The *k'* value recalculated for the S100 core is similar to the *k'* values described by Palmer et al. (2002), and Plummer et al. (2012) at Law Dome, while *k''* is similar to the value obtained by Inoue et al. (2017) for a Mill Island coastal core.~~ ~~Negative nssSO$_4^{2-}$ values are more pronounced after the 1950s, which will be discussed in the following sections.~~

4 Discussion

From the spatial and temporal variability of sea-salt concentrations in the different FIS cores ~~and snow pits~~ discussed here, it seems that more than one mechanism is 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 S4) 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$_4^{2-}$ ~~fractionation in sea-salts~~depletion in sea-salts measured in the core in comparison to bulk seawater, with ssSO$_4^{2-}$ depletion factors of two~~2~~ for the period 1737–1949, and ~~3~~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 due to cyclonic activity. Trajectory studies of air with high sea-salts concentrations and low ~~sulfate~~SO_4^{2-}/~~sodium~~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 agreement with dry deposition of sea-salts as pointed in section 3.5~~. 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 a much more probable explanation considering the local meteorological conditions in the study area.

Considering that we found no correlation between ion concentrations and site elevation (section 3.4), a decrease in wind transport efficiency of ~~e dominant wind direction at FIS (ENE), and the size of~~ frost flowers (size of 10–20 mm) and aerosol formed via (ii) (size >0.95 ~~-~~μm) (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.~~,~~ ~~wind transport would be more efficient from the neighbouring waters to the low elevation sites (e.g. S100 and the snow pit sites) than to the more elevated ice rises sites.~~ As mentioned in section 3.4, 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.

~~This hypothesis is reinforced by the fact that negative nssSO~~$_4^{2-}$ ~~values are mostly registered in the low elevation sites S100, M1, M2, G3, and G4, but not in the more elevated ice rises sites KC, KM, and BI, where the median nssSO~~$_4^{2-}$ ~~values for the cores are all positive, and sea-salt concentrations considerably lower than at S100 (with the exception of KM, which will be discussed below).~~

The dramatic increase in fractionated sea-salt in the S100 core after 1950s could be associated with an enhanced exposure of the S100 site to primary aerosol, in addition to and enhanced production of fractionated aerosol ~~and frost flowers~~, evidenced by a dominance of negative nssSO$_4^{2-}$~~-~~ values after 1950. Figure 2 a and c show that sea-salts started to increase after 1950 with a marked peak corresponding to 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. 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; and that the calving event occurred at Trolltunga in 1967 (Vinje, 1975) (Figure 1), enhanced the input of fractionated sea-salts to the S100 core by modifying the sea-ice conditions around S100, leading to the marked peak found in sea-salts in 1966 (±3 years). This could be supported 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.

~~in the waters at the eastern flank of FIS as a response to the calving of ice at the tip of FIS (Figure 1), which occurred in 1967 (Vinje, 1975).~~ The longer Trolltunga present before the calving event could have formed a larger bay to the east of it, where compaction of the sea ice occurred due to prevailing easterly winds, resulting in thicker, longer-lasting sea-ice, which limited the sea-spray formation. Such thick sea-ice does not 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 nearby 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 will present a reduced snow salinity in comparison to young sea-ice (Massom et al., 2001). Following the same supposition as Rhodes et al. (2017), i.e. that young sea-ice would be 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 found in S100 after the Trolltunga detachment, could be explained by 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 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 sea-ice sea-salts to oceanic sea-salts (produced by bubble bursting) ratio in Arctic sites, whit higher ratios closer to the sea ice source and when the location is in the path between sea-ice and prevailing winds. To test the~~his~~ hypothesis ~~could be tested~~presented here~~ by~~, a closer analysis of satellite and historical sea-ice data and a model-based study to 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, ~~There~~ any possible ~~is no~~ 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)~~, which suggests that the main source of sea-salts at these sites is wet deposition of sea-spray~~. 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 cores, which could explain the comparatively high sea-salt concentrations (Table 1).

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)~~),~~ ~~and five snow pits sampled along the ice shelf~~. 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~~We found that Na$^+$ concentrations decrease with latitude and distance from the sea when low elevation sites are considered (S100 and the FIS snow pits)~~. 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, and enhanced~~a change~~ input of~~ in~~ fractionated sea-salts~~ sources~~. This increase in sea-salt concentrations was accompanied by a shift in nssSO$_4^{2-}$ toward negative values, suggesting the input of fractionated sea-salts to the ion load in the S100 core most likely by enhancing sea-salts production~~dry deposition~~ by blowing salty snow over sea-ice.~~ Such dry deposition regime is not observed in any of the ice- rises cores.~~ ~~, and e~~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~~vidence of a significant contribution of fractionated sea-salt aerosols to these sites was not found~~. Consequently, the results of this study suggest that the S100 record contains a sea-salt record~~more local sea-salt signal,~~ dominated by processes ~~during ~~of sea-ice formation in the neighbouring waters, whereas the ice-~~-~~rises cores record the signal of larger-scale conditions of atmospheric flow, large inputs of ~~biog~~bio-nssSO$_4^{2-}$~~enic sulfate~~, and ~~transport of sea-salt aerosols produced over open water~~less efficient transport of sea-salts evidenced by lower mean concentrations in comparison 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)~~, and snow pits (M1, M2, G3–G5)~~ locations and sampling details. Distances of the core~~ and pit~~ 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).

Site	**Location**	**Elevation** (m a.s.l.)	**Core length** ***Ice depth*** **Ice temp. at 10 m** (m)	**Distance from the coast** (km)	**Time coverage** (years)	**Average SMB** (m w.e. y^{-1})
KC§	70°31′S, 2°57′E	264	20.0 *460* –~~-~~17.5	42	(1958–2012) ±3	0.24
KM§	70°8′S, 1°12′E	268	19.6 *410* –~~-~~15.9	12	(1995–2014) ±1	0.68
BI§	70°24′S, 3°2′W	394	19.5 *460* –~~-~~16.1	10	(1996–2014) ±1	0.70
S100*	70°14′S, 4°48′E	48	100 - –~~-~~17.5	3	(1737–2000) ±3	0.30

			~~**Snow pit sampling depth** (m)~~			
~~M1~~	~~70°0′S, 1°2′W~~	~~55~~	~~0.6~~	~~21~~	~~Aut.2009–2009/11/28~~	~~0.38 (for the period 1983–2009 inferred from a composite core, see Vega et al. 2016)~~
~~M2~~	~~70°19′S, 0°7′W~~	~~73~~	~~0.7~~	~~64~~	~~Aut.2009–2009/12/17~~	
~~G3~~	~~69°49′S, 0°37′W~~	~~57~~	~~0.9~~	~~27~~	~~Aut.2009–2009/12/21~~	
~~G4(G4a)~~	~~70°54′S, 0°24′W~~	~~60~~	~~0.7~~	~~117~~	~~Aut.2009–2009/12/19~~	

Table 2. Median, mean, maximum, minimum, and standard deviation (σ) of ion concentrations (in µmol L^{-1}) in the KC, KM, BI, and S100 firn/ice cores. ~~and in snow pits M1, M2, G3–G5.~~ 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. ~~Median v~~Values of water stable isotopes and deuterium excess for the KC, KM, and BI are reported by Vega et al. (2016).

~~Site~~	~~Period~~ ~~(years)~~	~~MSA~~	~~Cl^-~~	~~Br^- (×10³)~~	~~$\mathrm{NO_3}^-$~~ ~~$\mathrm{SO_4}^{2-}$~~	~~Median~~ ~~(µmol L^{-1})~~ ~~Na^+~~	~~K^+~~	~~Mg^{2+}~~	~~Ca^{2+}~~
						Median Mean Maximum Minimum σ (µmol L^{-1})			
Site	Period (years)	MSA	Cl^-	~~Br^- ×10³~~ $\mathrm{NO_3}^-$	$\mathrm{SO_4}^{2-}$	Na^+	K^+	Mg^{2+}	Ca^{2+}
KC	1958–2007	0.2 0.2 0.9 0.0 0.2~~0.2~~	10.0~~.0~~ 11.3 59.3 1.7 7.0	~~-~~- 0.6 0.7 1.8 0.2 0.4	1.8 2.1 10.3 0.1 1.5~~1.8~~	9.4 12.1 162.6 1.1 12.2~~9.4~~	0.2 0.2 1.5 0.0 0.2~~0.2~~	0.9 1.0 4.2 0.2 0.6~~0.9~~	0.5 1.8 62.7 0.2 6.5~~0.5~~
KM	1995–2012	0.3 0.5 9.4 0.0 0.6	71.3~~.3~~ 119.7 571.6 6.9 104.4	-~~-~~ 0.4 0.5 5.4 0.1 0.4~~0.4~~	4.5 6.2 84.5 0.5 7.5~~4.5~~	57.7 88.6 654.8 2.9 92.6~~57.7~~	1.5 2.0 16.1 0.1 1.8~~1.5~~	6.3 8.8 45.5 1.0 7.4~~6.3~~	1.6 2.2 10.6 0.4 1.8~~1.6~~
BI	1996–2012	0.4 0.5 2.0 0.0 0.4	23.1~~.1~~ 27.0 185.8 1.8 20.3	-~~-~~ 0.4 0.5 2.3 0.1 0.4	1.9 2.5 11.2 0.3 1.9~~1.9~~	19.0 22.5 161.8 1.7 17.3~~19.0~~	0.5 0.6 5.0 0.0 0.5~~0.5~~	2.0 2.4 15.9 0.3 1.7~~2.0~~	0.6 0.7 3.5 0.3 0.4~~0.6~~
S100	1737–2000	0.1 0.2 5.6 0.0	20.9~~0.9~~ 78.2 2174.1 3.7	-~~-~~ 0.5 0.6 1.8	1.2 2.6 56.0 0.2	20.7 75.5 1315.5 3.7	0.4 1.6 39.6 0.1	2.0 4.4 35.9 0.0	0.7 1.8 40.0 0.1

		0.3	187.7	0.1 0.3	5.2~~1.2~~	149.8~~20.7~~	3.4~~0.4~~	5.4~~2.0~~	3.6~~0.7~~
S100	1995–2000	0.1 0.2 1.0 0.0 0.2	132.4~~.4~~ 220.8 2174.1 11.3 332.1	~~-~~- 0.6 0.6 1.4 0.1 0.3~~0.6~~	3.2 6.0 35.8 0.8 7.4~~3.2~~	144.0 209.0 1315.5 11.0 232.0~~144.0~~	3.3 4.4 39.6 0.3 5.9~~3.3~~	10.7 10.8 35.9 1.5 6.2~~10.7~~	3.0 4.2 40.0 0.5 6.0~~3.0~~
S100	1737–1949	0.1 0.1 0.8 0.0 0.1	16.0~~16.0~~ 18.8 120.9 3.7 12.9	~~-~~- 0.6 0.6 1.8 0.1 0.3~~0.6~~	1.0 1.1 4.8 0.2 0.5~~1.0~~	15.1 18.4 138.8 3.7 14.2~~15.1~~	0.3 0.4 2.1 0.1 0.2~~0.3~~	1.4 1.6 6.3 0.3 0.9~~1.4~~	0.5 0.6 5.7 0.1 0.5~~0.5~~
S100	1950–2000	0.1 0.2 5.6 0.0 0.5	88.5 179.2 2174.1 9.1 280.9	0.5 0.6 1.5 0.1 0.3	2.8 5.2 56.0 0.4 7.9	98.2 172.6 1315.5 8.6 213.2	2.0 3.6 39.6 0.2 5.0	7.9 9.1 35.9 0.0 6.4	1.9 3.7 40.0 0.3 5.4

~~Cont. Table 2~~

~~Median~~ ~~Mean~~ ~~Maximum~~ ~~Minimum~~ ~~σ~~ ~~(µmol L^{-1})~~										
~~Site~~	~~Period (years)~~	~~MSA~~	~~Cl$^-$~~	~~Br$^-$ ×10^2~~	~~NO$_3^-$~~	~~SO$_4^{2-}$~~	~~Na$^+$~~	~~K$^+$~~	~~Mg^{2+}~~	~~Ca^{2+}~~
~~S100~~	~~1950–2000~~	~~0.1~~ ~~0.2~~ ~~5.6~~ ~~0.0~~ ~~0.5~~	~~88.58.5~~ ~~179.2~~ ~~2174.1~~ ~~9.1~~ ~~280.9~~	~~--~~	~~0.50.61.50.10.3~~	~~2.8~~ ~~5.2~~ ~~56.0~~ ~~0.4~~ ~~7.92.8~~	~~98.2~~ ~~172.6~~ ~~1315.5~~ ~~8.6~~ ~~213.298.2~~	~~2.0~~ ~~3.6~~ ~~39.6~~ ~~0.2~~ ~~5.02.0~~	~~7.9~~ ~~9.1~~ ~~35.9~~ ~~0.0~~ ~~6.47.9~~	~~1.9~~ ~~3.7~~ ~~40.0~~ ~~0.3~~ ~~5.41.9~~
~~M1~~	~~aut.2009–sum.2010 for all snow pits~~	~~--~~	~~55.95.9~~ ~~58.9~~ ~~93.7~~ ~~27.1~~ ~~33.4~~	~~0.7.58~~ ~~0.6~~ ~~0.8~~ ~~0.1~~ ~~0.4~~	~~0.50.4~~	~~2.1~~ ~~1.9~~ ~~2.6~~ ~~1.0~~ ~~0.8~~	~~46.4.4~~ ~~44.6~~ ~~68.4~~ ~~19.1~~ ~~24.7~~	~~0.8~~ ~~0.9~~ ~~1.7~~ ~~0.4~~ ~~0.7~~	~~5.65.6~~ ~~5.7~~ ~~8.8~~ ~~2.8~~ ~~3.0~~	~~1.2~~ ~~1.4~~ ~~2.1~~ ~~0.8~~ ~~0.7~~

					~~0.70.20.2~~					
~~M2~~		~~-~~	~~26.2.2~~ ~~65.6~~ ~~199.6~~ ~~10.2~~ ~~89.7~~	~~5.70.~~ ~~6~~ ~~0.5~~ ~~0.7~~ ~~0.3~~ ~~0.2~~	~~0.90.91.40.50.4~~	~~1.2~~ ~~.2~~ ~~2.4~~ ~~6.4~~ ~~0.7~~ ~~2.7~~	~~18.38.3~~ ~~44.4~~ ~~133.3~~ ~~7.7~~ ~~59.5~~	~~0.30.3~~ ~~1.0~~ ~~3.2~~ ~~0.2~~ ~~1.5~~	~~2.72.7~~ ~~7.0~~ ~~21.8~~ ~~0.9~~ ~~9.9~~	~~0.8~~ ~~1.6~~ ~~4.4~~ ~~0.3~~ ~~1.9~~
~~G3~~		~~-~~	~~27.1.1~~ ~~33.5~~ ~~48.0~~ ~~19.4~~ ~~12.8~~	~~7.10.~~ ~~7~~ ~~2.4~~ ~~5.8~~ ~~0.1~~ ~~2.7~~	~~0.50.50.72.50.11.0~~	~~1.4~~ ~~1.5~~ ~~3.0~~ ~~0.5~~ ~~1.0~~	~~61.91.9~~ ~~72.3~~ ~~160.7~~ ~~20.9~~ ~~54.1~~	~~1.51.5~~ ~~1.7~~ ~~3.8~~ ~~0.3~~ ~~1.4~~	~~6.96.9~~ ~~8.8~~ ~~19.2~~ ~~3.1~~ ~~6.4~~	~~2.0~~ ~~2.4~~ ~~5.8~~ ~~0.7~~ ~~2.0~~
~~G4~~		~~-~~	~~7.4.4~~ ~~8.2~~ ~~14.0~~ ~~4.1~~ ~~4.5~~	~~2.90.~~ ~~3~~ ~~0.3~~ ~~0.4~~ ~~0.2~~ ~~0.1~~	~~0.40.50.90.2~~	~~0.4~~ ~~0.5~~ ~~0.9~~ ~~0.2~~ ~~0.3~~	~~5.3.3~~ ~~5.8~~ ~~10.1~~ ~~2.6~~ ~~3.4~~	~~0.1~~ ~~0.1~~ ~~0.2~~ ~~0.1~~ ~~0.1~~	~~0.7~~ ~~0.8~~ ~~1.4~~ ~~0.4~~ ~~0.4~~	~~0.5~~ ~~0.5~~ ~~0.6~~ ~~0.3~~ ~~0.1~~

					~~0.3~~					
~~G5~~		~~-~~	~~7.67.6~~ ~~10.1~~ ~~16.4~~ ~~6.3~~ ~~5.5~~	~~3.90.4~~ ~~0.4~~ ~~0.5~~ ~~0.3~~ ~~0.1~~	~~0.40.40.50.40.1~~	~~0.5~~ ~~0.6~~ ~~0.7~~ ~~0.5~~ ~~0.1~~	~~5.4.4~~ ~~7.4~~ ~~12.4~~ ~~4.5~~ ~~4.3~~	~~0.2~~ ~~0.2~~ ~~0.3~~ ~~0.1~~ ~~0.1~~	~~0.9~~ ~~1.1~~ ~~1.7~~ ~~0.6~~ ~~0.69~~	~~0.5.5~~ ~~0.5~~ ~~0.6~~ ~~0.4~~ ~~0.1~~
~~G4a~~		~~-~~	~~5.65.6~~ ~~7.0~~ ~~23.1~~ ~~2.6~~ ~~4.8~~	~~6.10.6~~ ~~0.6~~ ~~1.1~~ ~~0.0~~ ~~0.3~~	~~0.50.61.90.20.4~~	~~0.3~~ ~~0.4~~ ~~1.4~~ ~~0.1~~ ~~0.3~~	~~-~~	~~-~~	~~-~~	~~-~~
~~G5a~~		~~-~~	~~8.1.1~~ ~~12.9~~ ~~53.6~~ ~~3.3~~ ~~11.7~~	~~8.10.8~~ ~~0.8~~ ~~1.5~~ ~~0.1~~ ~~0.4~~	~~0.30.41.10.20.2~~	~~0.4~~ ~~0.5~~ ~~2.1~~ ~~0.3~~ ~~0.4~~	~~-~~	~~-~~	~~-~~	~~-~~

1

2

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

Table 4. Correlation coefficients (R) for the median annual ion concentrations at the different cores (KC, KM, BI, and S100) vs. latitude, longitude, site elevation, and distance from the ice shelf edge, for the overlapping period 1997–2000. Significant values at the 95 % confidence interval are shown in bold.

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

Table 5. Cl$^-$/Na$^+$ ratio (expressed in μmol 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	Median Mean Maximum Minimum σ (μmol L^{-1})			
Site	Period (years)	Cl$^-$/Na$^+$	nssNa$^+$ (crustal)	ssNa$^+$		nssNa$^+$ to total Na$^+$
				All values	Only positive values	(%)
KC	1958–2007	1.2 1.1 1.9 0.1 0.3	0.7 2.6 87.7 0.3 9.1	8.1 9.6 159.1 -67.3 14.5	(5 %) 8.4 11.2 159.1 0.4 12.1	21
KM	1995–2012	1.3 1.3 3.8 0.8 0.2	2.2 3.0 14.8 0.5 2.5	54.9 85.6 644.4 2.4 90.2	-	3
BI	1996–2012	1.2 1.2 1.5 0.9 0.1	0.8 1.0 4.7 0.4 0.5	18.1 21.5 156.8 1.3 16.8	-	4
S100	1737–2000	1.0 1.0 2.1 0.1 0.2	1.0 2.5 56.0 0.1 5.1	19.5 73.0 1259.4 3.5 145.16	-	5
S100	1995–2000	1.0 1.0 2.1 0.1 0.2	9.1 9.2 30.5 1.3 5.2	135.3 199.8 1285.0 8.6 227.1	-	4
S100	1737–1949	1.0 1.1 1.8 0.6 0.2	1.2 1.4 5.3 0.2 0.8	13.7 17.1 138.0 3.2 13.7	-	8
S100	1950–2000	1.0 1.0 2.1 0.1 0.2	6.7 7.7 30.5 0.0 5.6	90.5 164.9 1285.0 6.4 208.2	-	4

Table 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 (Table 5).

						Median Mean Maximum Minimum σ (μmol L^{-1})			
Core	Period (years)	Cl$^-$		SO$_4^{2-}$		K$^+$		Mg^{2+}	
		ss	nss	ss	nss	ss	nss	ss	nss
KC	1958–2007	9.4 11.1 184.6 −78.6 16.8	(28 %) 0.9 0.2 96.6 −165.6 15.4	0.5 0.6 9.6 −4.0 0.9	(3 %) 1.2 1.6 8.2 −0.5 1.4	0.2 0.2 3.2 −1.4 0.3	(27 %) 0.0 0.1 1.8 −2.0 0.2	0.9 1.1 17.5 −7.4 1.6	(41 %) 0.1 0.0 9.5 −16.8 1.6
KM	1995–2012	63.8 99.3 747.5 2.8 104.7	(5 %) 6.2 10.4 80.5 −200.3 19.4	3.3 5.1 38.7 0.1 5.4	(38 %) 0.9 1.1 45.9 −15.1 4.5	1.1 1.7 12.9 0.1 1.8	(12 %) 0.2 0.3 13.8 −8.4 1.3	6.1 9.4 70.9 0.3 9.9	(43 %) 0.2 −0.7 23.6 −54.1 6.7
BI	1996–2012	21.0 25.0 181.9 1.6 19.5	(4 %) 1.7 2.1 15.3 −1.7 1.8	1.1 1.3 9.4 0.1 1.0	(26 %) 0.8 1.3 7.4 −1.4 1.7	0.4 0.4 3.1 0.0 0.3	(2 %) 0.1 0.1 4.5 0.0 0.3	2.0 2.4 17.3 0.2 1.9	(42 %) 0.0 0.0 2.0 −3.0 0.5
S100	1737–2000	22.7 84.7 1460.9 4.0 168.4	(81 %) −2.1 −6.6 713.2 −583.3 54.2	1.2 4.4 75.6 0.2 8.7	(51 %) 0.0 −1.7 5.2 −44.6 5.0	0.4 1.5 25.2 0.1 2.9	(40 %) 0.0 0.1 14.4 −2.7 0.9	2.2 8.0 138.5 0.4 16.0	(74 %) −0.3 −3.7 3.7 −102.7 11.3
S100	1995–2000	162.0 235.6 1460.9 11.3 259.9	(87 %) −23.7 −14.8 713.2 −583.3 134.6	8.4 12.2 75.6 0.6 13.4	(78 %) −4.5 −6.2 3.2 −39.8 8.2	2.8 4.1 25.2 0.2 4.5	(39 %) 0.1 0.3 14.4 −1.2 2.1	15.4 22.3 138.5 1.1 24.6	(74 %) −5.0 −11.5 3.5 −102.7 18.9
S100	1737–1949	16.6 20.4 159.4 4.0 16.1	(75 %) −1.1 −1.6 10.4 −44.5 4.5	0.9 1.1 8.3 0.2 0.8	(33 %) 0.2 0.1 4.1 −7.2 0.8	0.3 0.4 2.8 0.1 0.3	(41 %) 0.0 0.0 0.6 −0.6 0.1	1.6 1.9 15.1 0.4 1.5	(73 %) −0.2 −0.3 3.7 −14.1 1.2
S100	1950–2000	108.6 194.2 1460.9 9.1 239.3	(90 %) −14.7 −15.0 713.2 −583.3 88.5	5.6 10.1 75.6 0.5 12.4	(81 %) −2.7 −4.8 5.2 −44.6 7.1	1.9 3.4 25.2 0.2 4.1	(38 %) 0.1 0.2 14.4 −2.7 1.4	10.3 18.4 138.5 0.9 22.7	(74 %) −2.3 −9.3 3.5 −102.7 17.0

Table 7. Median, mean, minimum, maximum, and standard deviation (σ) 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 bio-nssSO$_4^{2-}$ to total SO$_4^{2-}$ is shown for all the cores.

Median
Mean
Maximum
Minimum
σ

		MSA/nssSO$_4^{2-}$		**bio-nssSO$_4^{2-}$**	**bio-nssSO$_4^{2-}$ to total SO$_4^{2-}$**
Site	**Period (years)**	**All values**	**Only positive values**		**(%)**
KC	1958–2007	0.1 0.3 −17.2 14.9 1.8	(3 %) 0.1 0.4 14.9 0.0 1.4	1.0 1.2 0.0 5.2 1.0	58
KM	1995–2012	0.1 0.7 −12.9 138.8 9.9	(38 %) 0.3 1.7 138.8 0.0 12.4	1.9 2.9 0.1 52.3 3.5	136
BI	1996–2012	0.3 −0.4 −245.1 30.6 13.1	(25 %) 0.3 0.7 30.6 0.0 2.1	2.1 2.7 0.1 11.3 2.4	108
S100	1737–2000	0.0 0.2 −15.6 11.3 1.8	(51 %) 0.3 1.0 11.3 0.0 1.9	0.7 1.0 0.0 31.4 1.6	37
S100	1995–2000	0.0 0.1 −1.7 9.5 1.4	(78 %) 0.2 1.1 9.5 0.1 2.7	0.5 1.0 0.1 5.4 1.2	17
S100	1737–1949	0.2 0.3 −15.6 10.3 2.0	(33 %) 1.0 1.0 10.3 0.0 1.7	0.7 0.8 0.0 4.4 0.6	72
S100	1950–2000	0.0 0.1 −2.6 11.3 1.4	(81 %) 0.2 1.2 11.3 0.0 2.8	0.7 1.2 0.1 31.4 2.6	24

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

~~**R**~~	~~**Cl^-**~~	~~**NO_3^-**~~	~~**SO_4^{2-}**~~	~~**Na^+**~~	~~**K^+**~~	~~**Mg^{2+}**~~	~~**Ca^{2+}**~~
~~Latitude (˚S)~~	~~-0.74~~	~~-0.10~~	~~**-0.84**~~	~~-0.93~~	~~-0.90~~	~~-0.90~~	~~-0.95~~
	~~-0.62~~	~~-0.07~~	~~**-0.45**~~	~~-0.87~~	~~-0.83~~	~~-0.88~~	~~-0.91~~
~~Longitude (˚W)~~	~~0.81~~	~~-0.40~~	~~0.77~~	~~0.72~~	~~0.59~~	~~0.57~~	~~0.72~~
	~~0.16~~	~~-0.22~~	~~0.30~~	~~0.08~~	~~0.13~~	~~-0.04~~	~~0.02~~
~~Distance from the sea (km)~~	~~-0.84~~	~~-0.07~~	~~-0.91~~	~~-0.90~~	~~-0.83~~	~~-0.83~~	~~-0.93~~
	~~-0.67~~	~~0.05~~	~~-0.64~~	~~-0.70~~	~~-0.70~~	~~-0.61~~	~~-0.70~~

~~Table 5~~7~~. Correlation coefficients (R) for the annual mean chemical concentrations (R_{conc}) and chemical fluxes (R_{flux}) vs. annual snow accumulation in the KC and S100 cores. Significant values at the 95 % confidence interval are shown in bold.~~

~~**R_{conc}**~~	~~**MSA**~~	~~**Cl^-**~~	~~**NO_3^-**~~	~~**SO_4^{2-}**~~	~~**Na^+**~~	~~**K^+**~~	~~**Mg^{2+}**~~	~~**Ca^{2+}**~~
~~KC~~	~~-0.18~~	~~-0.17~~	~~-0.24~~	~~-0.16~~	~~0.05~~	~~-0.08~~	~~-0.21~~	~~-0.12~~
~~KM~~	~~0.14~~	~~-0.06~~	~~-0.02~~	~~0.01~~	~~-0.07~~	~~-0.04~~	~~0.08~~	~~-0.04~~
~~BI~~	~~-0.31~~	~~-0.11~~	~~-0.57~~	~~-0.52~~	~~-0.14~~	~~-0.19~~	~~-0.13~~	~~-0.11~~
~~S100~~	~~0.07~~	~~-0.12~~	~~**-0.04**~~	~~**-0.10**~~	~~-0.12~~	~~-0.10~~	~~**-0.13**~~	~~-0.09~~
~~S100 (1737–1949)~~	~~0.11~~	~~-0.11~~	~~-0.05~~	~~-0.05~~	~~-0.11~~	~~-0.12~~	~~-0.05~~	~~-0.05~~
~~S100 (1950–2000)~~	~~0.16~~	~~-0.09~~	~~-0.06~~	~~-0.10~~	~~-0.08~~	~~-0.03~~	~~-0.15~~	~~-0.03~~
~~**R_{flux}**~~	~~**MSA**~~	~~**Cl^-**~~	~~**NO_3^-**~~	~~**SO_4^{2-}**~~	~~**Na^+**~~	~~**K^+**~~	~~**Mg^{2+}**~~	~~**Ca^{2+}**~~
~~KC~~	~~**0.45**~~	~~**0.76**~~	~~**0.72**~~	~~**0.63**~~	~~**0.59**~~	~~**0.75**~~	~~**0.74**~~	~~0.01~~
~~KM~~	~~**0.59**~~	~~**0.65**~~	~~**0.57**~~	~~**0.71**~~	~~**0.64**~~	~~**0.68**~~	~~**0.74**~~	~~**0.73**~~
~~BI~~	~~0.43~~	~~**0.55**~~	~~0.42~~	~~**0.55**~~	~~**0.53**~~	~~0.49~~	~~**0.56**~~	~~**0.73**~~
~~S100~~	~~**0.18**~~	~~0.07~~	~~**0.63**~~	~~**0.18**~~	~~0.08~~	~~0.10~~	~~**0.16**~~	~~**0.13**~~
~~S100 (1737–1949)~~	~~**0.52**~~	~~**0.45**~~	~~**0.61**~~	~~**0.56**~~	~~**0.41**~~	~~**0.44**~~	~~**0.48**~~	~~**0.43**~~
~~S100 (1950–2000)~~	~~0.22~~	~~0.25~~	~~**0.73**~~	~~0.24~~	~~**0.30**~~	~~**0.36**~~	~~**0.44**~~	~~0.27~~

~~Table 8. Median, mean, minimum, maximum, and standard deviation (σ) 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 bio-nssSO$_4^{2-}$ to total SO$_4^{2-}$ is shown for all the cores.~~

~~Median~~
~~Mean~~
~~Maximum~~
~~Minimum~~
~~σ~~

		~~MSA/nssSO$_4^{2-}$~~		~~bio-nssSO$_4^{2-}$~~	~~bio-nssSO$_4^{2-}$ to total SO$_4^{2-}$~~
~~Site~~	~~Period (years)~~	~~All values~~	~~Only positive values~~		~~(%)~~
~~KC~~	~~1958–2007~~	~~0.1~~ ~~0.3~~ ~~-17.2~~ ~~14.9~~ ~~1.8~~	~~(3 %)~~ ~~0.1~~ ~~0.4~~ ~~14.9~~ ~~0.0~~ ~~1.4~~	~~1.0~~ ~~1.2~~ ~~0.0~~ ~~5.2~~ ~~1.0~~	~~58~~
~~KM~~	~~1995–2012~~	~~0.1~~ ~~0.7~~ ~~-12.9~~ ~~138.8~~ ~~9.9~~	~~(38 %)~~ ~~0.3~~ ~~1.7~~ ~~138.8~~ ~~0.0~~ ~~12.4~~	~~1.9~~ ~~2.9~~ ~~0.1~~ ~~52.3~~ ~~3.5~~	~~136~~
~~BI~~	~~1996–2012~~	~~0.3~~ ~~-0.4~~ ~~-245.1~~ ~~30.6~~ ~~13.1~~	~~(25 %)~~ ~~0.3~~ ~~0.7~~ ~~30.6~~ ~~0.0~~ ~~2.1~~	~~2.1~~ ~~2.7~~ ~~0.1~~ ~~11.3~~ ~~2.4~~	~~108~~
~~S100~~	~~1737–2000~~	~~0.0~~ ~~0.2~~ ~~-15.6~~ ~~11.3~~ ~~1.8~~	~~(51 %)~~ ~~0.3~~ ~~1.0~~ ~~11.3~~ ~~0.0~~ ~~1.9~~	~~0.7~~ ~~1.0~~ ~~0.0~~ ~~31.4~~ ~~1.6~~	~~37~~
~~S100~~	~~1995–2000~~	~~0.0~~ ~~0.1~~ ~~-1.7~~ ~~9.5~~ ~~1.4~~	~~(78 %)~~ ~~0.2~~ ~~1.1~~ ~~9.5~~ ~~0.1~~ ~~2.7~~	~~0.5~~ ~~1.0~~ ~~0.1~~ ~~5.4~~ ~~1.2~~	~~17~~
~~S100~~	~~1737–1949~~	~~0.2~~ ~~0.3~~ ~~-15.6~~ ~~10.3~~ ~~2.0~~	~~(33 %)~~ ~~1.0~~ ~~1.0~~ ~~10.3~~ ~~0.0~~ ~~1.7~~	~~0.7~~ ~~0.8~~ ~~0.0~~ ~~4.4~~ ~~0.6~~	~~72~~
~~S100~~	~~1950–2000~~	~~0.0~~ ~~0.1~~ ~~-2.6~~ ~~11.3~~ ~~1.4~~	~~(81 %)~~ ~~0.2~~ ~~1.2~~ ~~11.3~~ ~~0.0~~ ~~2.8~~	~~0.7~~ ~~1.2~~ ~~0.1~~ ~~31.4~~ ~~2.6~~	~~24~~

Table 8~~96~~. Median annual nssSO_4^{2-} concentrations (in µmol L^{-1}) in the KC, KM, BI, and S100 firn/ice cores~~ and in snow pits M1, M2, G3–G5~~. (-) Not re-calculated.

Site	**Period** (years)	**Median** (µmol L^{-1}) nssSO_4^{2-} k=0.06	**Median** (µmol L^{-1}) nssSO_4^{2-} k'=0.024	**Median** (µmol L^{-1}) nssSO_4^{2-} k''=0.0~~2~~3
KC	1958–2007	1.2~~1~~	-	-
KM	1995–2012	~~0.7~~0.9	-	-
BI	1996–2012	0.8~~7~~	-	-
S100	1737–2000	~~-7×10~~~~$^{-2}$~~0.3	0.9~~.3~~	0.7~~7~~
S100	1995–2000	~~-4.8~~−2.1	~~-0.9~~2.4	~~0.7~~1.3
S100	1737–1949	0.4~~1~~	0.8~~4~~	0.7~~7~~
S100	1950–2000	−1.5~~3.1~~	1.3~~-0.7~~	0.7 ~~0.7~~

Figures

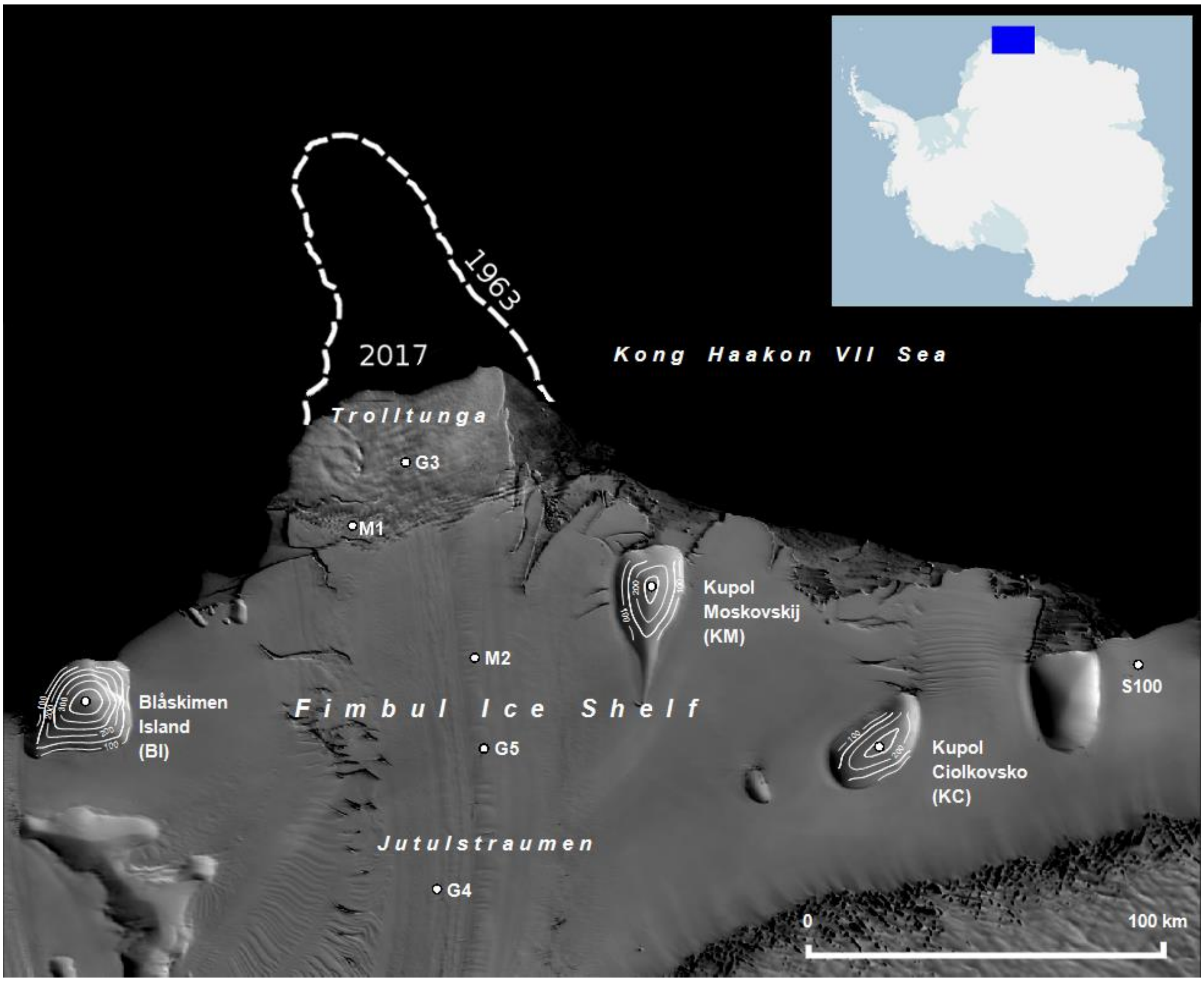


Figure 1. Satellite image of Fimbul Ice Shelf (FIS) showing the KC, KM, BI, and S100 core sites, the M1, M2, G3, G4, and G5 snow pit sites (Supplementary material), Jutulstraumen, and Trolltunga. In addition, 50-m contours are shown at each ice-~~-~~rise, as derived from GPS profiles (V. Goel, personal 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). ~~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).

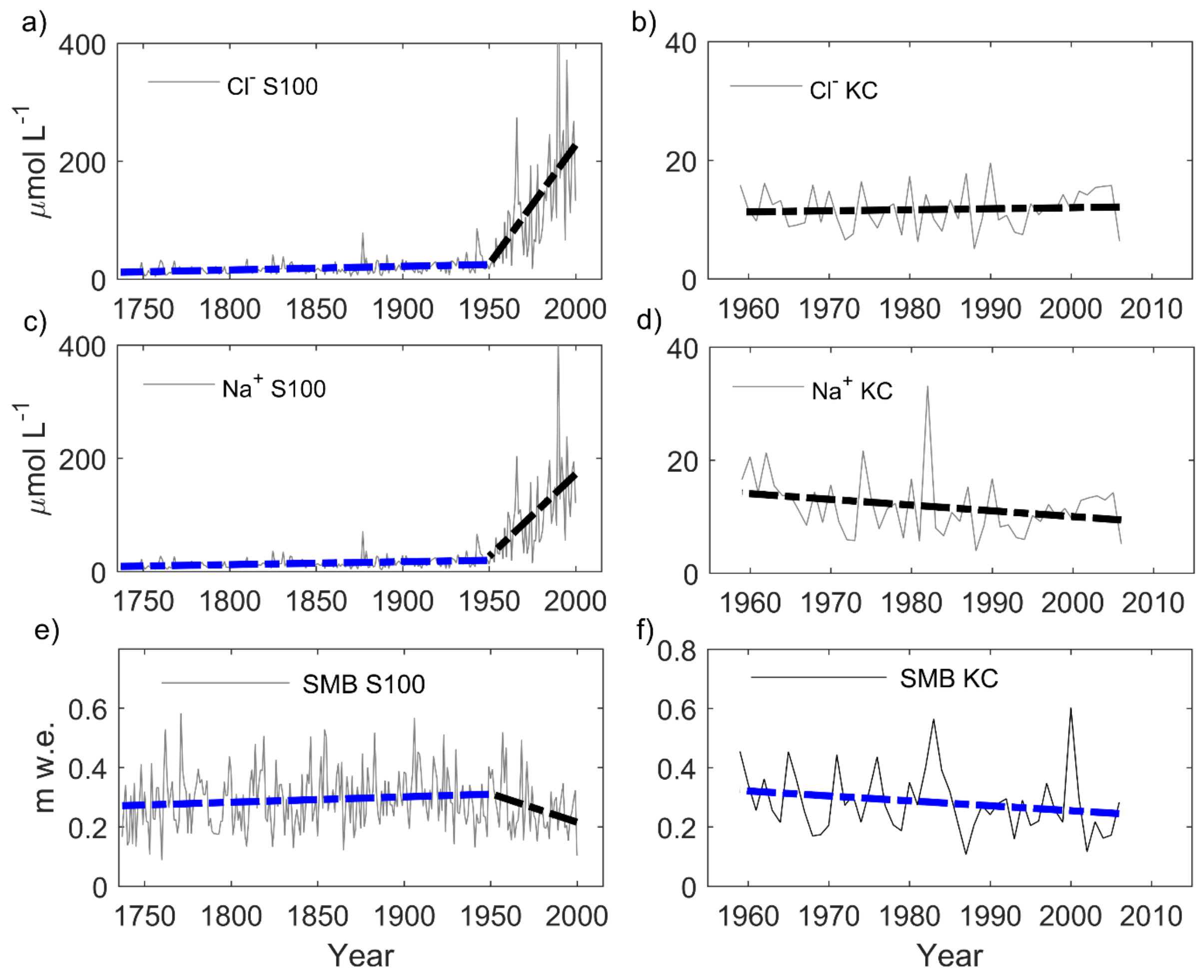
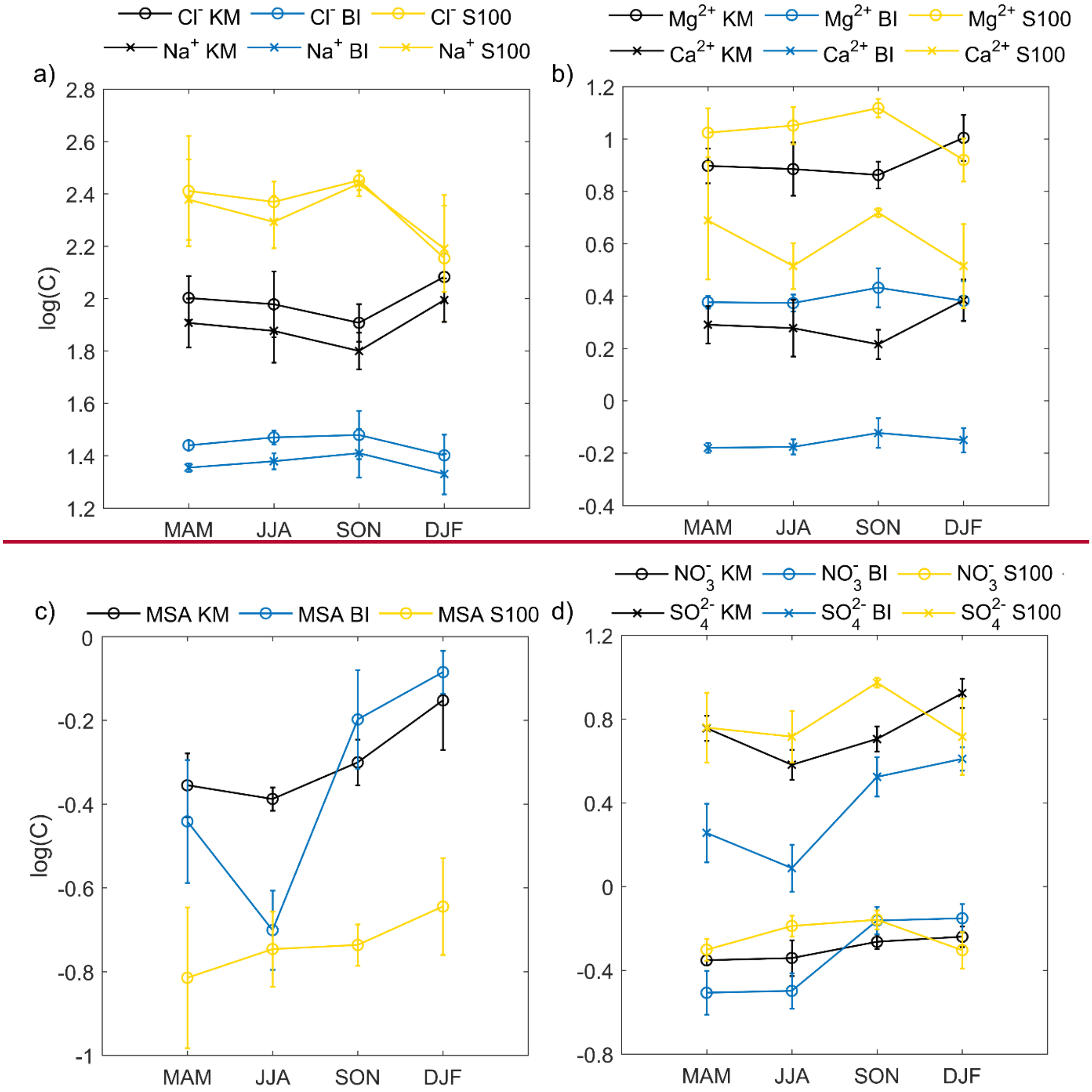


Figure 2. Annual sea-salt (Cl^{-} and Na^{+}) concentrations and surface mass balance (SMB) in the two longest records retrieved at Fimbul Ice shelf, S100, (a), (c), and (e), and KC, (b), (d) and (f). Linear trends in Cl^{-} and Na^{+} concentration, and SMB measured in the S100 core are shown for two 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~~2~~4.


a)
Cl^- KM
Cl^- BI
Cl^- S100
Na^+ KM
Na^+ BI
Na^+ S100
log(C)
2.8
2.6
2.4
2.2
2
1.8
1.6
1.4
1.2
MAM
JJA
SON
DJF
b)
Mg^{2+} KM
Mg^{2+} BI
Mg^{2+} S100
Ca^{2+} KM
Ca^{2+} BI
Ca^{2+} S100
1.2
1
0.8
0.6
0.4
0.2
0
-0.2
-0.4
MAM
JJA
SON
DJF
c)
MSA KM
MSA BI
MSA S100
log(C)
0
-0.2
-0.4
-0.6
-0.8
-1
MAM
JJA
SON
DJF
d)
NO_3^- KM
NO_3^- BI
NO_3^- S100
SO_4^{2-} KM
SO_4^{2-} BI
SO_4^{2-} S100
1.2
0.8
0.4
0
-0.4
-0.8
MAM
JJA
SON
DJF

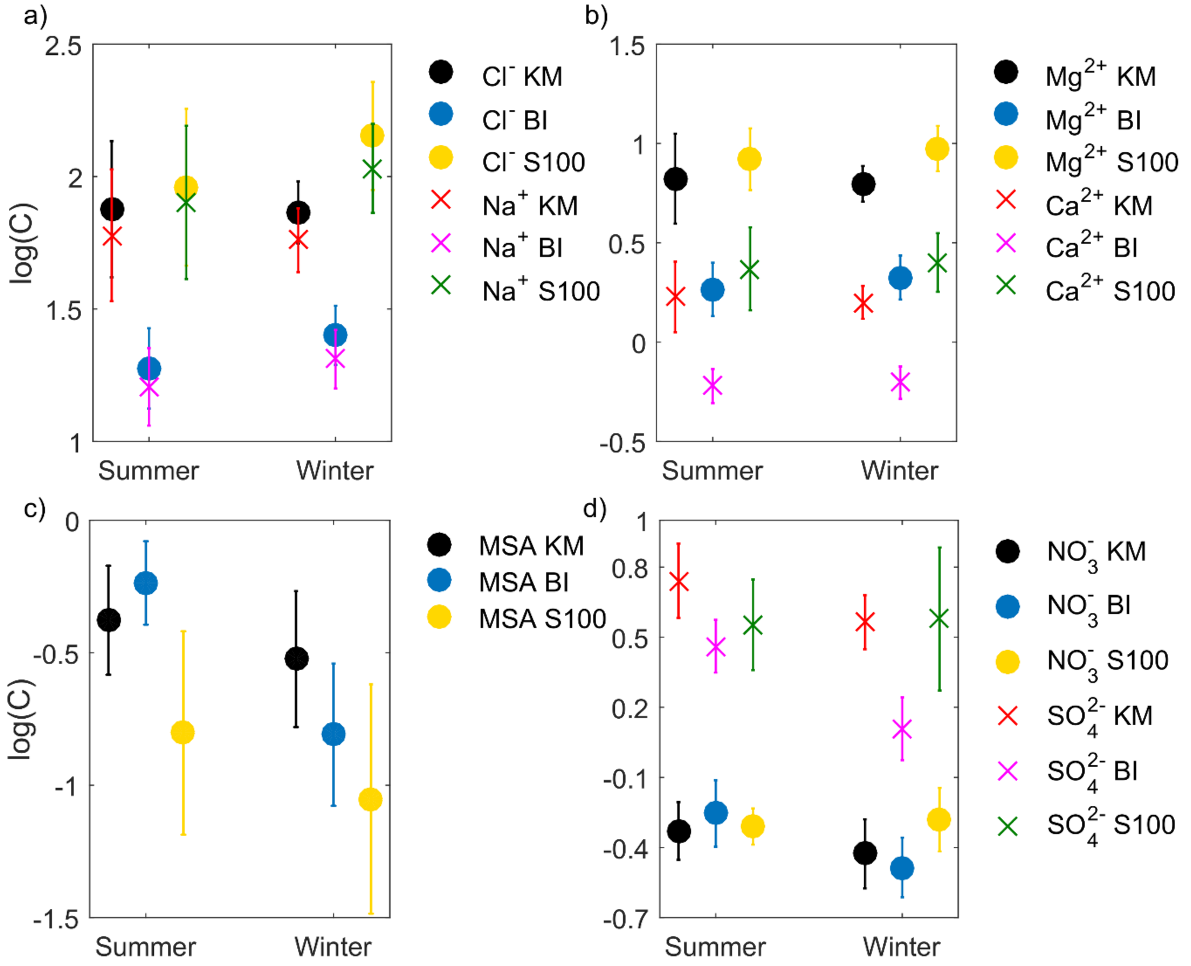
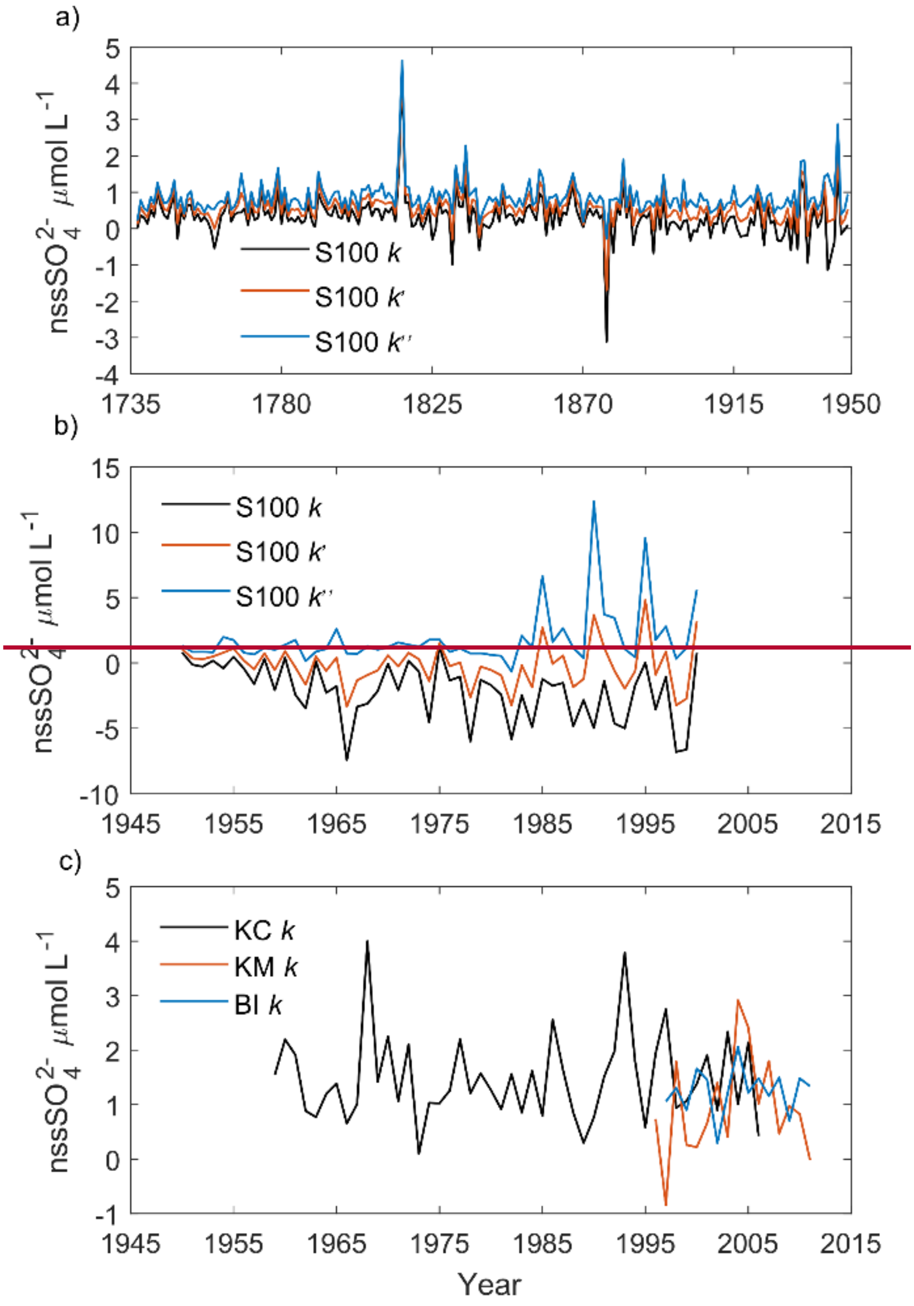


Figure 3. Sub-annual~~easonal~~ variability of selected ions, Cl^{-} and Na^{+} (a), Mg^{2+} and Ca^{2+} (b), MSA (c) and NO$_3^-$ and SO$_4^{2-}$ (d) in cores KM, BI, and S100. ~~Average~~ Mean ~~seasonal values~~summer and winter ~~were~~ concentrations were calculated for the ~~trimesters MAM (autumn), JJA (winter), SON (spring), and DJF (summer)~~months NDJFMA, and MJJASO, for a period ~~from the interpolated composite record (0.01-year resolution)~~ of 16, 15, and 5 years in the KM, BI, and S100 cores, respectively.


a)
nssSO$_4^{2-}$ µmol L^{-1}
5
4
3
2
1
0
-1
-2
-3
-4
1735
1780
1825
1870
1915
1950
S100 k
S100 k'
S100 k''
b)
15
10
5
0
-5
-10
1945
1955
1965
1975
1985
1995
2005
2015
c)
KC k
KM k
BI k
Year

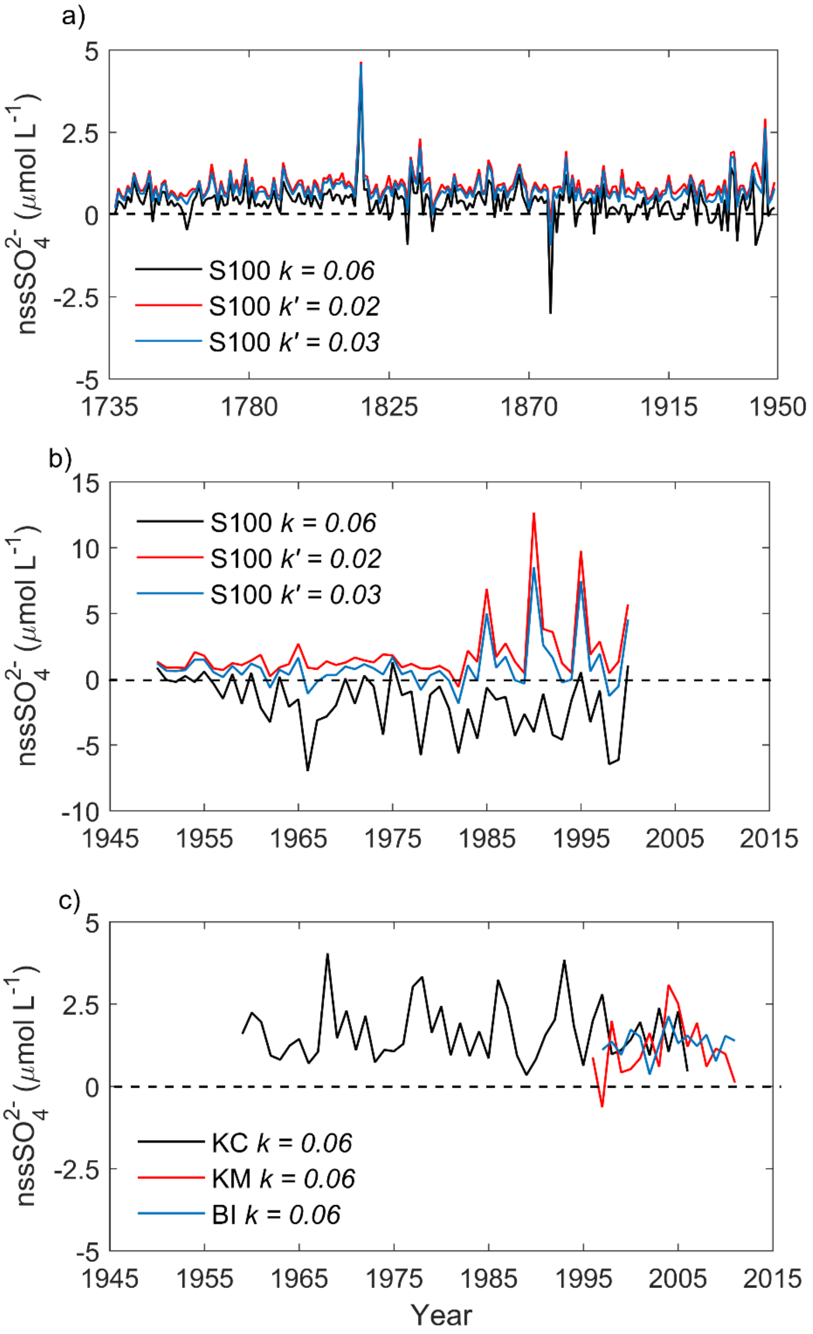
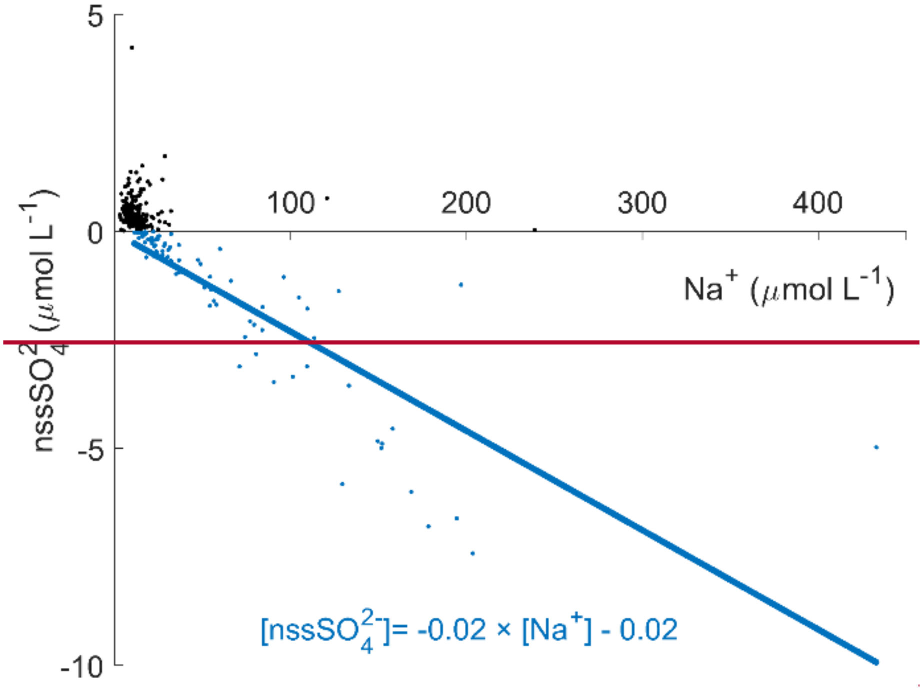


Figure 4. Annual nssSO$_4^{2}$~~–~~- concentrations in the S100 core between a) 1737–1949, b) 1950–2000, and c) in the KC, KM, and BI cores. nssSO$_4^{2}$~~–~~- recalculated using *k*= 0.06, *k'*=0.~~04~~ 02 and *k'*~~'~~'=0.~~02~~ 03 are shown in panels a) and b) with black, red and blue lines, respectively. nssSO$_4^{2}$~~–~~- in the KC, KM, and BI cores was calculated using *k*= 0.06.


5
0
-5
-10
100
200
300
400
Na$^+$ (μmol L^{-1})
nssSO$_4^{2-}$ (μmol L^{-1})
[nssSO$_4^{2-}$]= -0.02 × [Na$^+$] - 0.02

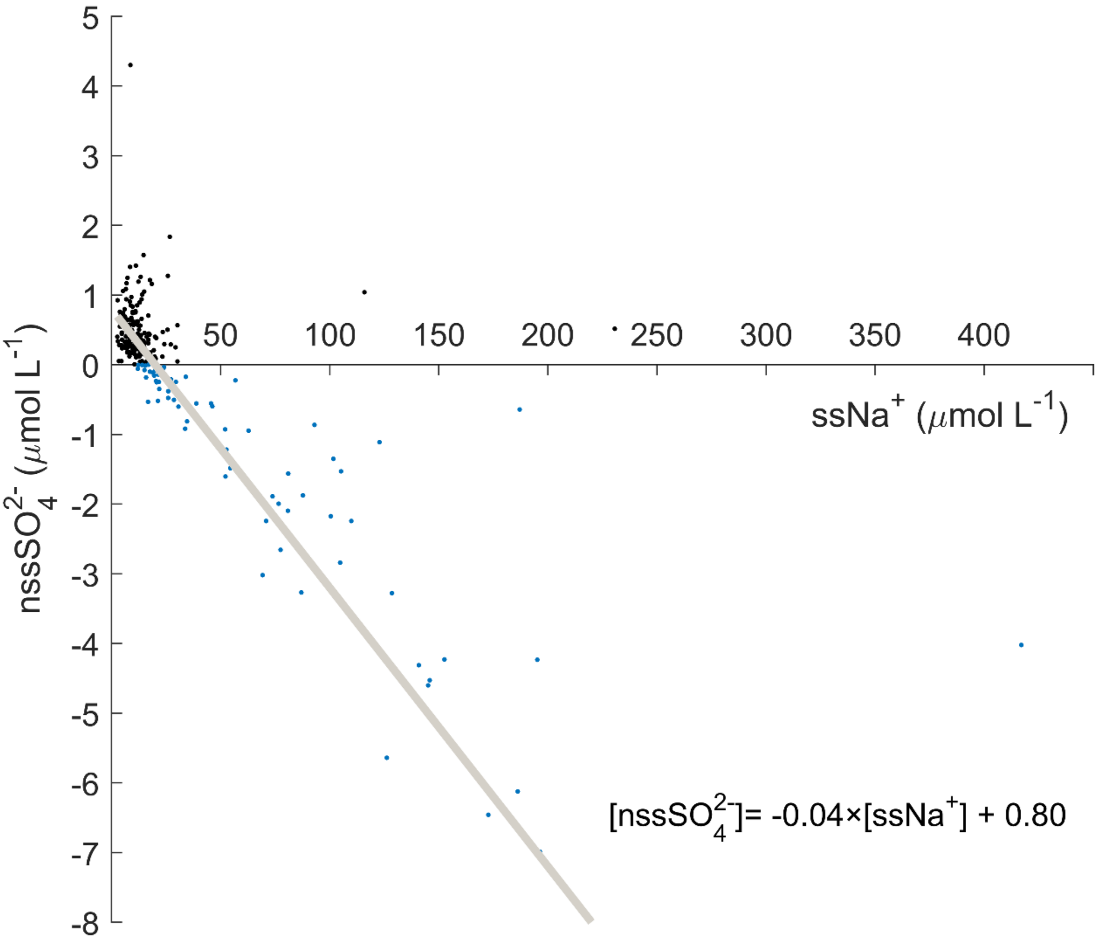

5
4
3
2
1
0
-1
-2
-3
-4
-5
-6
-7
-8
50
100
150
200
250
300
350
400
ssNa$^+$ (μmol L^{-1})
nssSO$_4^{2-}$ (μmol L^{-1})
[nssSO$_4^{2-}$]= -0.04×[ssNa$^+$] + 0.80

Figure 5. Scatter plot of annual n~~on-sea-salt~~ ssSO_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=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 ~~the negative~~all nssSO_4^{2-} ~~values~~ data points to infer ~~a new~~corrected ~~value of~~ *k* value ~~,~~(~~-~~*k'*), following the approach by ~~Inoue~~ Wagenbach et al. (1998~~2017~~).

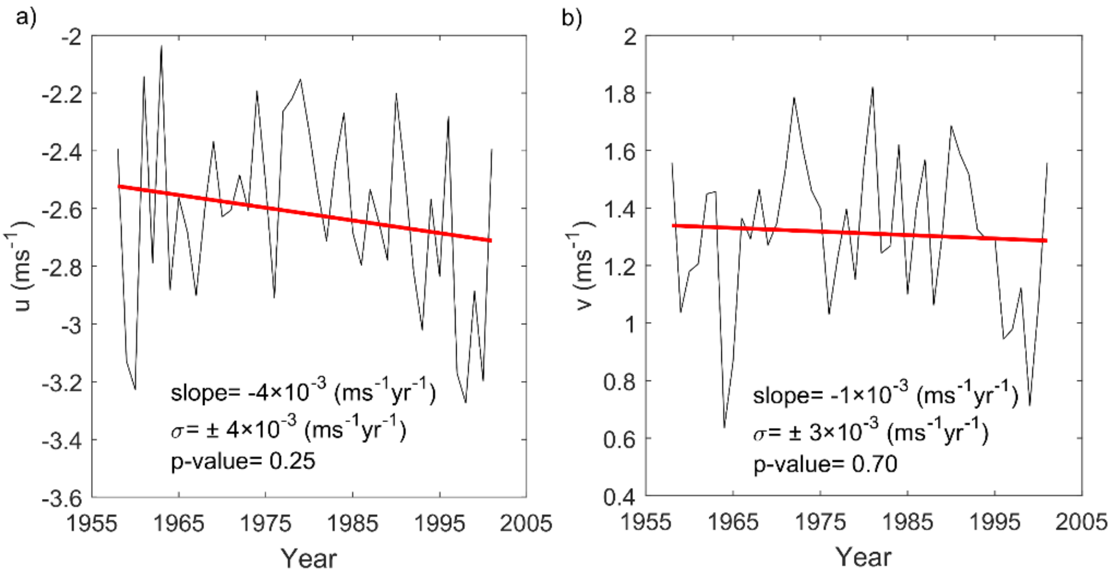


Figure 6. Annual averages of monthly a) zonal, and b) meridional wind speeds (ERA40) for the area (69°S–71°S, 3.5°W–5°E) between 1958–2001. Slope, standard deviation, and *p*-value of the linear regression are shown in the figure.