Referee #1

We thank the reviewer very much for the thoughtful and thorough review of our manuscript. The very helpful comments and suggestions have greatly improved the quality of this paper. Below, we give a point-by-point response to the comments and suggestions of the reviewer, in the order of (1) comments from Referees, (2) author's response, and (3) author's changes in manuscript (referee comments in black; author's response and changes in manuscript in blue).

(1) comments from Referees

This paper uses surface samples from a series of traverses between the coast and the inland plateau Dome A station, along with 3 pit sequences, to study the geographic pattern of chemical concentrations. At heart it is a very simple study, effectively building on reviews written 15 years ago, but with new data from just a single geographical region. The paper throws a lot of different methods (principal component analysis, enrichment factors, ternary diagrams) at the data. Despite a comparatively long paper, the findings of the study are really nothing new and rather obvious: sea salt ions are closely related and at higher concentrations near the coast; ions such as Ca are mainly terrestrial, sulfate has a marine biogenic source.

I find myself a little torn as to what to recommend for this paper. The data are clearly rare in the sense that there are few data from this sector and from inland sites in general other than Vostok, Dome C and a couple of other sites. The authors do understand previous work and have presented their data in the light of that work. However the whole study is very hampered by the unfortunate fact that surface samples (3 cm surface skims) are just really unsuitable for understanding the chemical climate, and despite the work on the pit samples, it is the surface samples that are the bulk of the paper (7 of 12 data figures plus both tables). Because surface samples inevitably do not cover a full year, it is hard to know what they reveal: at least in some cases they give a misleading impression. For example all the surface samples are collected in the summer, when generally sea salt is low and sulfate is high. Surface samples collected in winter might give a completely different impression, but the paper doesn't make this clear. Surface samples probably represent only a single snowfall, so comparing them from year to year with any seasonal weather statistic (as in Figure 13) is not appropriate.

In the end, I want to be generous and say that the data deserve to be published but a more modest paper is needed, in which the shortcomings of the sample set are more clearly explained, and the paper is stripped down to a shorter length (with perhaps 6 figures and the rest removed or at least moved into the supplement). I don't think the paper brings much insight but it would be a shame if the data were not made available in the peer-reviewed literature so I will recommend major revision, and highlight below where I think the paper can be shortened.

(1) author's response

We greatly appreciate the reviewer for the thoughtful comments of our work.

(2) comments from Referees

Comments on the text:
Abstract is largely OK, but will need to be shortened in line with the text. The section about ternary diagrams is not needed in the abstract as it adds little to the rest of the text, and the part about SIOL should be excluded.

(2) author's response

We agree that the section about the ternary diagrams adds little to the rest of the text, e.g., the section about the excess Cl^- ($nssCl^-$) with respect to the seawater composition. In addition, we agree with both reviewer#1 and reviewer#2 that the section about the factors controlling the interannual variations in chemical ions in surface snow is not robust, considering that the sampled surface ~3cm along the traverse at different locations could represent different periods of snow accumulation, possibly weeks to months or even only a single snowfall event. Thus, the section about the interannual variation and its potential relation to the SIOL was removed in the revised manuscript.

(2) author's changes in manuscript

In the abstract section, the text about ternary diagrams and interannual variations of ions in surface snow (the Southern Indian Ocean low (SIOL)) were excluded. And the abstract was also shortened. Please see the revised manuscript. Abstract now reads,
"There is a large variability in environmental conditions across the Antarctic ice sheet, and it is of significance to investigate the snow chemistry at as many locations as possible and over time, given that the ice sheet itself, and precipitation and deposition patterns and trends are changing. The China inland Antarctic traverse from coastal Zhongshan Station to the ice sheet summit (Dome A) covers a variety of environments, allowing for a vast collection of snow chemistry conditions across East Antarctica. Surface snow (the upper ~3 cm, mainly representing the summertime snow) and snow pit samples were collected on this traverse during five campaigns, to comprehensively investigate the spatial and temporal variations in chemical ions (Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) and the related controlling factors. Results show that spatial patterns of ions in surface snow are consistent among the five campaigns, with Cl^-, Na^+, K^+, and Mg^{2+} decreasing rapidly with distance from the coast and NO_3^- showing an opposite pattern. No clear spatial trends in SO_4^{2-}, NH_4^+, and Ca^{2+} were found. In the interior areas, an enrichment of Cl^- versus Na^+ with respect to seawater composition is ubiquitous as a result of the deposition of HCl, and $nssCl^-$ (nss, non-sea-salt fraction) can account for up to ~40 % of the total Cl^- budget, while $nssK^+$ and $nssMg^{2+}$ are mainly associated with terrestrial particle mass. On average, $nssCa^{2+}$ and $nssSO_4^{2-}$ in surface snow account for ~77 and 95 % of total Ca^{2+} and total

SO_4^{2-}, respectively. The high proportions of the non-sea-salt fractions of Ca^{2+} and SO_4^{2-} are mainly related to terrestrial dust inputs and marine biogenic emissions, respectively. Snow NH_4^+ is mainly associated with marine biological activities, with slightly higher concentrations in summer than in winter. On the coast, parts of the winter snow are characterized with negative $nssSO_4^{2-}$ values, and a significant negative correlation between $nssSO_4^{2-}$ and Na^+ in wintertime snow was found, suggesting that sea salts originated from the sea ice. In the interior areas, marine biogenic SO_4^{2-} still dominated snow SO_4^{2-} in winter, leading to significant positive $nssSO_4^{2-}$ values. Ion flux assessment suggests an efficient transport of $nssSO_4^{2-}$ to at least as far inland as the ~2800 m contour line."

(3) comments from Referees

Line 33/34 "In the interior areas, the negative nssSO42- signal in winter snow resulted from inputs of sea salts being completely swamped by the contribution of marine biogenic emission": this doesn't quite make sense. What you mean is that there are high (positive) nss sulfate in inland snow because of marine biogenic sulfate. You have no data on whether negative values would have been seen, so the current wording serves to confuse the reader.

(3) author's response

Thanks for this comment. The observation suggests that there are high $nssSO_4^{2-}$ values in inland snow during the wintertime, and indeed we have not observed the negative values of $nssSO_4^{2-}$ in interior areas. Following the reviewer's comment, we have reworded this sentence.

(3) author's changes in manuscript

This sentence was reworded, and it now reads,
"In the interior areas, marine biogenic SO_4^{2-} still dominated snow SO_4^{2-} in winter, leading to significant positive $nssSO_4^{2-}$ values. "
For the changes, please see the revision-tracked version of manuscript, section **abstract.**

(4) comments from Referees

The introduction is generally quite good and the English (with a couple of exceptions that will be picked up in proofreading) is fine.

(4) author's response

Thanks for the positive comment.

(4) author's changes in manuscript

Several minor changes were made, including adding some previous important references. Please see

the revised version.

(5) comments from Referees

Methods: After line 154, it should be mentioned that 3 cm at a density of 0.33 as assumed elsewhere is only 1 cm water equivalent (compare snow accumulation in Fig 2a) and therefore the surface samples represent at best a summer sample and in many cases probably a single snowfall.

(5) author's response

   We agree with the reviewer and thanks for the constructive comment. Indeed, since the snow accumulation rate varies, the same 3-cm interval, corresponding to about 1 cm water equivalent assuming snow density of 0.33 g cm^{-3}, would represent different lengths of time at different locations. At locations with high snow accumulation rates (e.g. the coastal region), the upper 3 cm of snow mainly represents deposition from a short period of the summer season (e.g., on the time scale of weeks). In the most inland Dome A region which has the lowest accumulation rate (6-7 cm snow per year), the upper 3 cm of snow may represent deposition over several months. Still, the information contained in the snow roughly indicates summertime conditions, considering the sampling date of late January to February. In this case, it is reasonable to use the sampled surface snow (~3cm) to investigate the spatial patterns and main origins of chemical ions under summertime conditions. While we accept that the sampling protocol could lead to an imperfect interpretation of that data, as both reviewers raised, collection of the samples covering the same time intervals, in practice, is rather challenging considering the significant variability in surface snow accumulation rate, strong snow drift, etc. Accordingly, we use the surface 3 cm of snow to investigate the overall summertime atmospheric conditions.

(5) author's changes in manuscript

In the revised manuscript, we added a paragraph to clarify the representativeness of the surface samples, and it reads,
   "It is noted the surface snow represents different lengths of time at different locations, considering the wide range of snow accumulation rates on the traverse (Fig. 2(a)). At locations with high snow accumulation rate on the coast, the upper 3 cm of snow may represent deposition from a few weeks, while the surface 3 cm of snow could represent deposition over a few months on Dome A plateau. Also, it is possible that the upper 3 cm of snow can be representative of a single snowfall. Still, the information contained in the surface snow generally indicates summertime conditions, as the sampling took place during late January and February in each season. This allows for an investigation of summer snow chemistry patterns on the traverse."

(6) comments from Referees

Fig 2 and section 3.1 already illustrates the problem with the study: there is huge variation at a single

4

site between years, and yet you have no idea whether this variation reflects changes from year to year, or from week to week within a year. Given that the samples are inevitably collected over a period of days to weeks within a year, much of the spatial variability can arise from temporal change in practice. This doesn't completely invalidate the work but it should be explained.

(6) author's response

We agree with the reviewer that the significant variations in ions in surface snow could arise from the temporal changes in chemical ions. Indeed, there is great variation of ions between years at a single site (Figure 3 in the revised manuscript), and the ion concentrations can vary among samples in the same season (shaded areas in Figures 3 (a) and (b) in the revised manuscript).

Together with the comments from Reviewer#2, the Results section was re-organized and substantially revised. At first, concentrations of all species in snow pits (including Cl^-, NO_3^-, SO_4^{2-}($nssSO_4^{2-}$), Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) were presented in section 3.1. The snow pit data show that there are significant variations in ion concentrations, and even in the same season, ion concentrations can also vary among snowfall events at a specific site. In sections 3.2 and 3.3, ion concentrations in surface snow were presented, and the spatial variability of ions was also included. In addition, in the revised version, it was clarified that surface snow mainly represents summertime deposition, and accordingly the spatial patterns of ions on the traverse can generally represent the summertime conditions.

(6) author's changes in manuscript

Following the both reviewers' suggestion, the Results section was re-written, please see the revised manuscript.

(7) comments from Referees

Fig 3b. I don't see the value of packaging all the samples into a single wheel like this. Firstly how is the calculation done: do you add the concentrations from each site (thus giving more weight to the samples with high concentrations) or is each site normalised before averaging? But wouldn't it be more interesting to show this wheel separately for groups of samples, eg <200 km from coast, 200-600 km, and >600 km. Then you could discuss in a holistic way how the composition changes as you go inland.

(7) author's response

Thanks for the suggestion. In the previous version, the contribution percentages of each ion to the total were calculated from the average values (i.e., the percentages) of all sites on the traverse. Following the reviewer's suggestion, the contribution percentages of each ion to the total ion budget were calculated in different regions. In combination of with the spatial patterns of ion concentrations and previous study (Shi et al., 2018), the traverse was divided into three regions, i.e., the coastal 200km, 200-800km, and the Dome A plateau (800km-Dome A), and then the contribution percentages were

calculated in individual regions. Indeed, there are significant variations in the contribution percentages, with increasing values of H^+ and NO_3^- towards inland and an opposite pattern for Cl^-, Na^+, Mg^{2+}, and NH_4^+. On the whole traverse, high contribution percentage of H^+ is observed, agreeing with previous investigations (Udisti et al., 2004; Traversi et al., 2009; Pasteris et al., 2014), and suggesting acidic characteristics of surface snow.

(7) author's changes in manuscript

Following the reviewer's suggestion, this section was re-written and Figure 4 was re-drawn. It now reads,

"The percentages of each constituent to the total ions in surface snow on the traverse are shown in Figs. 4(b)-(d). The most abundant species is H^+, accounting for about 30-40 % of the total ions, followed by NO_3^-, SO_4^{2-}, and Cl^-. In general, ions NH_4^+, K^+, Mg^{2+}, and Ca^{2+} are the smallest component of the ionic composition, with the four cation summing to ~5 % of the total. Spatially, the contribution percentages of H^+ and NO_3^- increase with increasing distance from the coast, with the highest values on Dome plateau (42.3 and 34.5 %, respectively), while Cl^-, Na^+, Mg^{2+}, and NH_4^+ show an opposite pattern and no clear trend was observed for SO_4^{2-}. The high contribution percentage of H^+ is consistent with previous investigations (Udisti et al., 2004; Traversi et al., 2009; Pasteris et al., 2014), and suggests acidic characteristics of summertime surface snow."
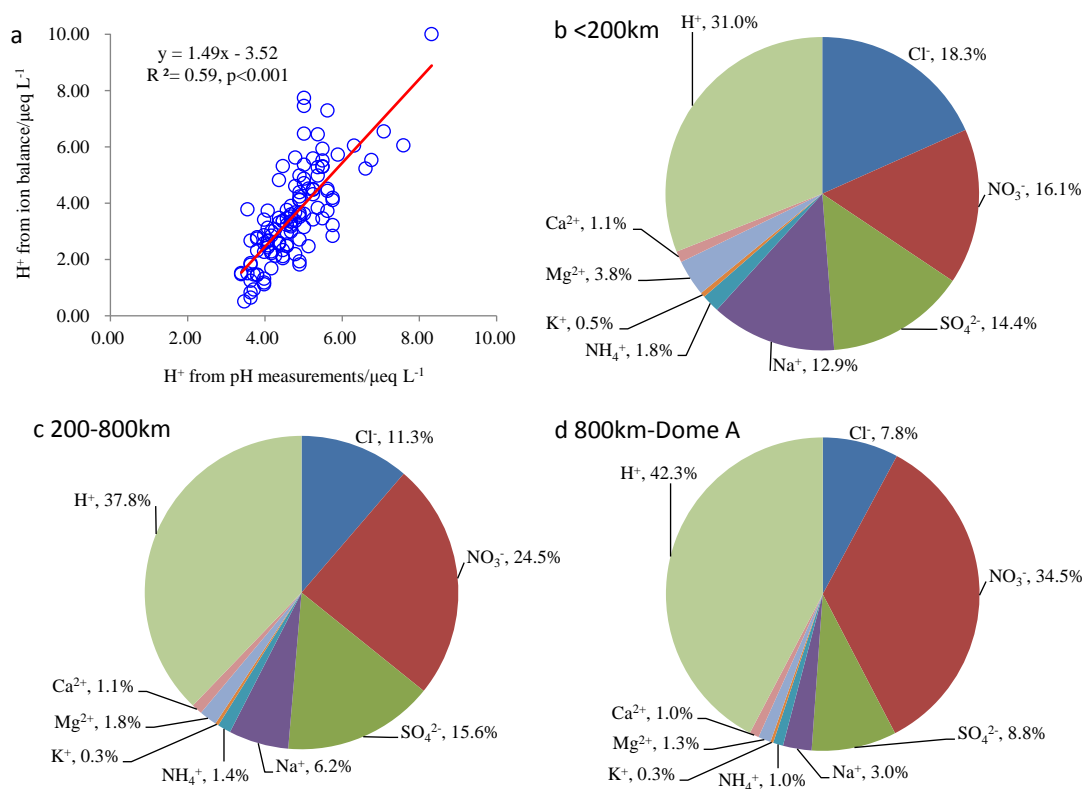

Figure 4. Major ions in surface snow on the Chinese inland Antarctic traverse. Concentrations of H^+ derived from pH versus those from the ion balance method are shown in panel (a), and contribution percentages of each ion to the total in different regions on the traverse are shown in panels (b)-(d), in

6

μeq L^{-1}. The percentages of each ion in individual regions were calculated from the averages of all sites within the region.

(8) comments from Referees

Fig 4. Please state in the caption the year the pits were sampled. I know it's somewhere in the text but it's needed here.

(8) author's response

   Thanks for the helpful comment, and the sampling year of each snow pit was included in the caption of Figure 3 (the revised version).

(8) author's changes in manuscript

The caption of Figure 3 now reads,
"**Figure 3.** Profiles of chemical ions in snow pits P1 (a), P2 (b), and P3 (c). Snow pits P1 and P2 were sampled in the summer season in 2015-2016, and P3 was sampled in January 2010. The ratios of SO_4^{2-}/Na^+ in snow samples were also present. Red arrows in panels (a) and (b) represent the middle of the identified summer, and shaded areas denote summer seasons (see text). The red dashed line in panel (a) represents the ratio of SO_4^{2-}/Na^+ in bulk seawater, while the red dashed line in panel (c) signifies the first snow sample significantly influenced by the Pinatubo eruption. One seasonal cycle generally represents local Na^+ minima and $nssSO_4^{2-}$ and SO_4^{2-}/Na^+ maxima."

(9) comments from Referees

Fig 5 can be removed – it adds nothing, and it is sufficient just to say that the accumulation rate is too low at P3 for seasonal variability to be apparent. If necessary simply point out the relevant section in Fig 4.

(9) author's response

   We agree with the reviewer, and Figure 5 (in original version) was removed. In the revised manuscript, the figure was included in the supplementary materials (Figure S1), to show the variations of ions in the years 2008 and 2009.

(9) author's changes in manuscript

   Figure 5 was removed, and the section was revised accordingly. It now reads,
   "At P3, Na^+, $nssSO_4^{2-}$ and the ratios of SO_4^{2-}/Na^+ fluctuate significantly (Fig. 3(c)), and these contrasts are unlikely indicative of the seasonal cycles as that for P1 and P2. In a full year of snow accumulation at P3, on average, about 7-8 samples were collected, allowing for examining the seasonal

7

variability of ions. Following the field measurements of snow accumulation rate at Dome A during 2008-2011 (~20 kg m^{-2} a^{-1}; Ding et al., 2015), the snow samples covering the years 2008 and 2009 can be roughly identified, assuming an even distribution of snow accumulation throughout the year. In total, there are 7 and 8 samples identified in the years 2008 and 2009, respectively (Fig. S1), and no seasonal cycles in Na$^+$, nssSO$_4^{2-}$, and SO$_4^{2-}$/Na$^+$ ratio were found due to the low snow accumulation rate at P3. In addition, the post-depositional processes (e.g., migration, diffusion, and ventilation processes) and/or wind scouring can obscure the original signal (Cunningham and Waddington, 1993; Albert and Shultz, 2002; Libois et al., 2014; Caiazzo et al., 2016), resulting in the absence of seasonal cycles of ions at P3."

(10) comments from Referees

Fig 6 is unnecessary. Because it pools data from different sites and years the statistics shown really have no meaning. If you really like it, please put it in the supplement.

(10) author's response

Agree with the reviewer. Together with the comments of Reviewer#2, the calculation of Enrichment Factors (EFs) was excluded in the revised manuscript. Instead, the calculation of concentration in excess with respect to the seawater composition was included (i.e., the non-sea-salt fractions of ions). Accordingly, in the discussion section, the EFs of chemical ions were replaced with the excess concentrations.

(10) author's changes in manuscript

Figure 6 in the original version was removed. Please see the revised manuscript.

(11) comments from Referees

Section 4.2: PC1 which is clearly sea salt is fine. However it is then rather obvious that Ca as a terrestrial ion, ammonium and sulfate fall into other PCs, and this can be said much more briefly. Personally I think Table 2 is sufficient and Table 1 adds nothing, but I don't insist on losing it.

(11) author's response

We agree with the reviewer that PC1 is clearly indicative of the sea salt inputs, while SO$_4^{2-}$, NH$_4^+$, and Ca^{2+} fall into other components suggest different main sources. Following the reviewer's suggestion, this section was shortened and the main sources of ions in surface snow were discussed briefly. Table 1 shows the main outcomes of PCA, especially the loadings of individual ions in each PC, and thus this table was still included in the updated manuscript. Then, one can easily read the loadings of individual ions and the contribution percentages of each component to the total variance. Table 2 shows the correlation coefficients of chemical ions in surface snow, together with the PCA results

(Table 1), the main sources of ions can be clearly distinguished.

(11) author's changes in manuscript

In Section 4.2, the different groups of ions and the possible sources were discussed briefly, and the section was shortened. In the revised manuscript, both tables 1 and 2 were included, in order to show PCA and correlation analysis results clearly. The revised text now reads,

"PC1 accounts for 46 % of the variance and is highly loaded by Cl^-, Na^+, K^+, and Mg^{2+}, with the factor loadings higher than 0.7. In addition, the four species are correlated well with each other (Table 2), suggesting the variation of the four species is dominated by sea salt aerosols. PC2 accounts for 17 % of the total variance, and the loading values of NH_4^+ and Ca^{2+} in PC2 are high, ~0.8. Ca^{2+} is mainly from terrestrial particle mass, while NH_4^+ is thought to be mainly associated with biological decomposition of organic matter in the Southern Ocean (Johnson et al., 2007; Kaufmann et al., 2010). In addition, biomass burning from mid-latitudes can contribute to snow NH_4^+ at some sites (Pasteris et al., 2014), and the penguin colony emissions can be important inputs to NH_4^+ in snow several km from the colony (Rankin and Wolff, 2000). On this traverse, no correlation was found between NH_4^+ and biomass burning tracers (e.g., black carbon and phenolic compounds) in surface snow (Shi et al., 2019; Ma et al., 2020), suggesting a minor role of biomass burning emissions. Thus, the high NH_4^+ concentrations on the coast are likely associated with marine biogenic emissions. In this case, it is possible that a similar transport pathway (i.e., preferentially transported long distance in free transport; Krinner and Genthon, 2003; Kaufmann et al., 2010; Krinner et al., 2010) can explain, at least in part, the positive loadings of both NH_4^+ and Ca^{2+} in PC2.

NO_3^- is highly loaded in PC3, which accounts for 13 % of the system variance. On this traverse, NO_3^- in the snow has been extensively investigated, and it is proposed that NO_3^- concentrations were influenced by post-depositional processing which is largely dependent on snow accumulation rate (Shi et al., 2015; Shi et al., 2018a; Shi et al., 2018b). A negative relationship was found between NO_3^- and snow accumulation rate (Fig. 6(b)), suggesting a high degree of NO_3^- cycling driven by photolysis at low snow accumulation sites.

SO_4^{2-} did not show high loadings in any of the three extracted components. Its positive loading in PC1 (0.55) and weak relationships between SO_4^{2-} and sea salts (Cl^- and Na^+) likely supports the contribution of sea salt aerosols, although a minor one. A positive loading of SO_4^{2-} is also present in PC3 (0.42), and a weak correlation was found between SO_4^{2-} and NO_3^-. Both SO_4^{2-} (or $nssSO_4^{2-}$) and NO_3^- are negatively correlated with snow accumulation rate (Fig. 6), but with distinct mechanisms. $nssSO_4^{2-}$ can be concentrated due to dry deposition at sites with low snow accumulation rate, while elevated NO_3^- concentrations are linked to the photochemical cycling and re-deposition (discussed above). In addition, $nssSO_4^2$ and NO_3^- are mainly associated with the secondary aerosols, and the production of both species in summer is closely related to the oxidants HO_x, RO_x, etc (Ishino et al., 2017; Shi et al., 2018a), which may also contribute to the correlation between SO_4^{2-} and NO_3^-."

(12) comments from Referees

Fig 9 and 12 and the associated text seem to add little, and they should go into the supplement of be

deleted.

We agree with the reviewer that the ternary diagram of Cl^-, Na^+, and SO_4^{2-}, and the related text are in part similar with the results of excess concentrations with respect to the seawater composition. The patterns of Cl^-, Na^+, and SO_4^{2-} can more clearly characterize the modification processes to sea salt aerosols, in comparison with the excess concentrations alone. In this case, we keep Figure 9 (in the original version) in the revised manuscript, to show the spatial variation in the degree of sea salt modification in surface snow. Figure 12 (in the original version) shows the seasonal variations in the degree of sea salt modification. Considering that the seasonal variations in ion concentrations and excess concentrations with respect to the seawater composition were discussed extensively in section 4.3, figure 12 (in the original version) was moved to the supplementary materials following the reviewer's suggestion. Accordingly, the associated text was significantly shortened.

(12) author's changes in manuscript

Figure 12 (in the original version) was moved to the supplementary materials, and the associated text was shortened. It now reads,

"The ternary diagram of Cl^-, Na^+, and SO_4^{2-} can well characterize the modification processes to sea salt aerosols, and the ternary plot of the three ions in surface snow is shown in Fig. 7. The values of the ions were normalized via the following equation,

$X=[X]/([Na^+]+[Cl^-]+[SO_4^{2-}])$ Eq. (3),

where [X] is the concentration of ion X in the snow (in μeq L^{-1}). The dashed line between the seawater reference value and the SO_4^{2-} vertex represents the sea salt aerosol composition with additional SO_4^{2-}, i.e., the ratio of Cl^-/Na^+ keeps constant (1.17) with additional SO_4^{2-} along the dashed line. The presence of acids (HNO_3 and H_2SO_4) would result in the liberation of HCl into the atmosphere via reactions R1 and R2, resulting in the changes in Cl/Na ratios, i.e., either Cl^- loss or gain are located right or left of the line, respectively. It is shown that all of the data points are above the seawater plot, suggesting an enrichment of SO_4^{2-} in surface snow. Most of the data points are located left of the line, indicating the general enrichment of Cl^- due to reactions R1 and R2 occurring in the atmosphere and/or in the snowpack. But the coastal data points are generally close to the line, suggesting that the degree of sea salt modification is generally low in the snow.

Similar to the surface snow, the modification processes to sea salt aerosols is negligible in snow pit P1, while the ubiquitous modification process to sea salts throughout the year was found in the interior areas (P2 and P3; Fig. S3). Thus, Cl^- in inland Antarctica, often deviating from the seawater dilution line remarkably in both summer and winter, is not a quantitative indicator of sea salts in snow."

(13) comments from Referees

Fig 13 and the associated text are very misleading. The three traverses show different values for different sampling dates, but this could be day to day, week to week, month to month or year to year

variability and trying to associate with seasonal indices is not relevant. To carry out any such analysis you'd have to calculate the index for the precise dates of the snowfall the surface samples represent. This section should be removed, as should supplement Fig S3.

(13) author's response

We agree with the reviewer and thanks for the comment. Together with the comments of Reviewer#2, section 4.4 was removed in the revised manuscript. In addition, Figure S3, which shows the climatological mean sea level pressure distribution over the southern ocean in the austral summers in different years, was also removed.

(13) author's changes in manuscript

Section 4.4 was removed. Please see the revised manuscript.

(14) comments from Referees

The other supplement Figures (S1 and S2) also don't add to the message and should not be included (S1 is essentially identical to Fig 4a with the addition of ammonium, and Fig S2 shows information that is already visible in Fig 4a.

(14) author's response

We agree with the reviewer. The concentration profile of NH_4^+ in the snow pit is now included in Figure 3a (in the revised version), and thus Figure S1 was removed. The information of Figure S2 is now present in Figure 3a, and Figure S2 was also deleted.

(14) author's changes in manuscript

Figure S1 and S2 were removed

References

Bertler N., Mayewski P.A., Aristarain A., Barrett P., Becagli S., Bernardo R., Bo S., Xiao C., Curran M. and Qin D. (2005) Snow chemistry across Antarctica. *Ann. Glaciol.* 41, 167-179.

Duderstadt K.A., Dibb J.E., Schwadron N.A., Spence H.E., Solomon S.C., Yudin V.A., Jackman C.H. and Randall C.E. (2016) Nitrate ion spikes in ice cores not suitable as proxies for solar proton events. *J. Geophys. Res.* 121, 2994-3016.

Erbland J., Vicars W., Savarino J., Morin S., Frey M., Frosini D., Vince E. and Martins J. (2013) Air-snow transfer of nitrate on the East Antarctic Plateau - Part 1: Isotopic evidence for a photolytically driven dynamic equilibrium in summer. *Atmos. Chem. Phys.* 13, 6403-6419.

France J., King M., Frey M., Erbland J., Picard G., Preunkert S., MacArthur A. and Savarino J. (2011) Snow optical properties at Dome C (Concordia), Antarctica; implications for snow emissions and snow chemistry of reactive nitrogen. *Atmos. Chem. Phys.* 11, 9787-9801.

Frey M.M., Savarino J., Morin S., Erbland J. and Martins J. (2009) Photolysis imprint in the nitrate stable isotope signal in snow and atmosphere of East Antarctica and implications for reactive nitrogen cycling. *Atmos. Chem. Phys.* 9, 8681-8696.

Holland P.R., Bruneau N., Enright C., Losch M., Kurtz N.T. and Kwok R. (2014) Modeled Trends in Antarctic Sea Ice Thickness. *J. Climate* 27, 3784-3801.

Marion G., Farren R. and Komrowski A. (1999) Alternative pathways for seawater freezing. *Cold Reg. Sci. Technol.* 29, 259-266.

Mulvaney R., Wagenbach D. and Wolff E.W. (1998) Postdepositional change in snowpack nitrate from observation of year-round near-surface snow in coastal Antarctica. *J. Geophys. Res.* 103, 11021-11031.

Pasteris D., McConnell J.R., Edwards R., Isaksson E. and Albert M.R. (2014) Acidity decline in Antarctic ice cores during the Little Ice Age linked to changes in atmospheric nitrate and sea salt concentrations. *J. Geophys. Res.* 119, 5640-5652.

Rankin A.M., Wolff E.W. and Martin S. (2002) Frost flowers: Implications for tropospheric chemistry and ice core interpretation. *J. Geophys. Res.* 107, 4683.

Savarino J., Kaiser J., Morin S., Sigman D.M. and Thiemens M.H. (2007) Nitrogen and oxygen isotopic constraints on the origin of atmospheric nitrate in coastal Antarctica. *Atmos. Chem. Phys.* 7, 1925-1945.

Shi G., Buffen A.M., Hastings M.G., Li C., Ma H., Li Y., Sun B., An C. and Jiang S. (2015) Investigation of post-depositional processing of nitrate in East Antarctic snow: isotopic constraints on photolytic loss, re-oxidation, and source inputs. *Atmos. Chem. Phys.* 15, 9435–9453.

Shi G., Buffen A.M., Ma H., Hu Z., Sun B., Li C., Yu J., Ma T., An C., Jiang S., Li Y. and Hastings M.G. (2018) Distinguishing summertime atmospheric production of nitrate across the East Antarctic Ice Sheet. *Geochim. Cosmochim. Acta* 231, 1-14.

Wagenbach D., Ducroz F., Mulvaney R., Keck L., Minikin A., Legrand M., Hall J.S. and Wolff E.W. (1998a) Sea-salt aerosol in coastal Antarctic regions. *J. Geophys. Res.* 103, 10961-10974.

Wagenbach D., Legrand M., Fischer H., Pichlmayer F. and Wolff E.W. (1998b) Atmospheric near-surface nitrate at coastal Antarctic sites. *J. Geophys. Res.* 103, 11007-11020.

Wolff E.W., Bigler M., Curran M., Dibb J., Frey M., Legrand M. and McConnell J. (2012) The Carrington event not observed in most ice core nitrate records. *Geophys. Res. Lett.* 39, L08503.

Wolff E.W., Bigler M., Curran M.A.J., Dibb J.E., Frey M.M., Legrand M. and Mcconnell J.R. (2016) Comment on "Low time resolution analysis of polar ice cores cannot detect impulsive nitrate events" by D.F. Smart et al. *J. Geophys. Res.* 121, 1920-1924.

Wolff E.W., Jones A.E., Bauguitte S.-B. and Salmon R.A. (2008) The interpretation of spikes and trends in concentration of nitrate in polar ice cores, based on evidence from snow and atmospheric measurements. *Atmos. Chem. Phys.* 8, 5627-5634.

Zatko M., Grenfell T., Alexander B., Doherty S., Thomas J. and Yang X. (2013) The influence of snow grain size and impurities on the vertical profiles of actinic flux and associated NO_x emissions on the Antarctic and Greenland ice sheets. *Atmos. Chem. Phys.* 13, 3547-3567.

End of responses to Referee #1.