

Reviewer #2 (Prof. Michel Legrand)

We are very grateful to Prof. Michel Legrand for his detailed comments and very useful and constructive suggestions. The manuscript has been substantially modified and reformatted based on these comments/suggestions. Below, we give a point-by-point response to the comments and suggestions of Prof. Michel Legrand, in the order of (1) comments from Referees, (2) author's response, and (3) author's changes in manuscript. Prof. Michel Legrand's comments are in black, and the responses are in blue.

(1) comments from Referees

This manuscript reports on analysis of inorganic ions in snow samples collected in the frame of intensive program of snow sampling made along several successive traverses achieved from the coast (Zhongshan Station) to Dome A (East Antarctica). The samplings include 594 surface snow samples (upper 3 cm), and 3 snow-pits (down to 1-2 m depth). It is shown that Cl^- , Na^+ , K^+ , and Mg^{2+} concentrations are high within the narrow coastal region, dropping further inland, while NO_3^- exhibits an opposite trend. No clear spatial trends were found for SO_4^{2-} , NH_4^+ , and Ca^{2+} . Data are discussed with respect to potential origin of ions including for minor ones like calcium, ammonium and potassium.

Overall evaluation: First, the authors have to be congratulated for having successfully conducted such a very large inter-annual snow-sampling program, likely sometimes done under harsh weather conditions.

Whereas these data certainly contain interesting information that would be relevant for the Cryosphere journal, inherent to the poor representativeness of 3 cm snow sampling, an in depth interpretation of data is often very difficult and I find that, in the present version of the manuscript, the authors over-interpret them. The manuscript also reveals several misleading presentations and discussions of data. Finally some key previous works are not adequately referenced in the manuscript.

In conclusion I recommend major revisions and in the following I try to identify what can be removed from the manuscript and reversely what can be developed, in particular (and if possible) taking advantage of more information derived from the snow pit data.

(1) author's response

We appreciate Prof. Michel Legrand taking the time to complete the review and welcome the very helpful and constructive comments.

(1) author's changes in manuscript

We revised the manuscript substantially following the comments and suggestions, see responses below.

(2) comments from Referees

Overall comments:

1. It would be far more logic to first present and discuss the snow pit data showing how large is the seasonal variation of all species including ammonium, nitrate, magnesium, calcium and potassium and discuss the basic causes of that. In the present version, only the well-known species (sodium and sulfate) data in the snow-pit are shown in the main text, whereas ammonium is only seen in the SI. Reporting first (and for all species including ammonium, potassium, calcium, nitrate) on the snow-pit data would permit the readers to appreciate the seasonal variability related to surface snow variability.

(2) author's response

We agree with Prof. Michel Legrand and thanks for the helpful suggestion. Indeed, there is significant seasonal variation of ions at P1 and P2 (Figure 3 in the revised manuscript). The Results section 3 was re-organized and substantially revised as suggested by the reviewer. Section 3.1 now discussed the concentrations of all species in snow pits (including Cl^- , NO_3^- , SO_4^{2-} (nss SO_4^{2-}), Na^+ , NH_4^+ , K^+ , Mg^{2+} and Ca^{2+}). The snow pit data show that there are significant variations in ion concentrations, and even in the same summer or winter 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 are presented, and the spatial variability of ions is 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 roughly represent summertime conditions.

(2) author's changes in manuscript

Section 3 Results was re-organized as suggested by the reviewer, and Figure 3 was reproduced. Please see the revised manuscript.

(3) comments from Referees

2. Instead of using enrichment factors (EFs), more illustrative is the calculation of concentration in excess with respect to the seawater composition done with calculation of error propagation (specially for minor species like potassium, magnesium, and calcium). These data would certainly be more useful than EF values to discuss snow pit data (levels, seasonality, etc).

(3) author's response

We agree with Prof. Michel Legrand and thanks for the comment. The section of EFs calculation in the original version was removed. In the Result and Discussion sections, the ion concentrations in excess with respect to seawater composition (i.e., the non-sea-salt fractions of ions, nssX, with X of an ion) were used to discuss the levels, seasonality, main sources, etc. Accordingly, all the text associated with the EFs was replaced with the excess concentrations (nssX). Also, the error for the calculation of the non-sea-salt fractions were included, also see the response #(18) below.

(3) author's changes in manuscript

For the revisions, please see the revised manuscript sections 2 Methods, 3 Results, and 4 Discussions. Accordingly, the related text in Abstract and Conclusions sections were also revised.

(4) comments from Referees

3. Section 4.4: The value of this discussion is very weak since based on samples of poor seasonal representativeness. Please remove it (see also my comment below for section 4.4).

(4) author's response

We agree with Prof. Michel Legrand. Indeed, the surface samples, which represent different lengths of time at varied locations, are unlikely representative of seasonal (or interannual) variations. Together with the comments of Reviewer#1, this section was removed.

(4) author's changes in manuscript

Section 4.4 (in the original version) was removed. Please see the revised version of the manuscript.

(5) comments from Referees

Comments:

Title: please specify “surface snow” chemistry

(5) author's response

The “surface snow” was specified in the title.

(5) author's changes in manuscript

Now the title reads,

“Spatial and temporal variations in surface snow chemistry along a traverse from coastal East Antarctica to the ice sheet summit (Dome A)”

(6) comments from Referees

Abstract: Please remove the last sentence since, in no way, your data can demonstrate something (statistically thought) on this topic: «The interannual variations in ion concentrations in surface snow on the traverse are likely linked to the changes in the Southern Indian Ocean low (SIOL) from year to year, and the deepening of the SIOL in summer tends to promote the transport of marine aerosols to Princess Elizabeth Land. »

Also your statement «Snow NH_4^+ is mainly associated with marine biological activities »

is not really demonstrated (at least as the manuscript stands, see more comments below).

Finally please reword the confusing sentence: «the negative nssSO₄²⁻ signal in winter snow resulted from inputs of sea salts being completely swamped by the contribution of marine biogenic emissions. I don't understand what you mean here: do you mean that you have never pure winter snow because of wind mixing after deposition ???

(6) author's response

Agreed. Section 4.4 was removed and the abstract as well as conclusions were revised accordingly.

As for the possible sources of NH₄⁺ in surface snow, several sentences were included in the manuscript (see response below).

The sentence “the negative nssSO₄²⁻ signal in winter snow resulted from inputs of sea salts being completely swamped by the contribution of marine biogenic emissions” was rephrased.

(6) author's changes in manuscript

The abstract was revised accordingly, and it 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⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺, and Ca²⁺) 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 Mg²⁺ decreasing rapidly with distance from the coast and NO₃⁻ showing an opposite pattern. No clear spatial trends in SO₄²⁻, NH₄⁺, and Ca²⁺ 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 nssMg²⁺ are mainly associated with terrestrial particle mass. On average, nssCa²⁺ and nssSO₄²⁻ in surface snow account for ~77 and 95 % of total Ca²⁺ and total SO₄²⁻, respectively. The high proportions of the non-sea-salt fractions of Ca²⁺ and SO₄²⁻ are mainly related to terrestrial dust inputs and marine biogenic emissions, respectively. Snow NH₄⁺ 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 nssSO₄²⁻ values, and a significant negative correlation between nssSO₄²⁻ and Na⁺ in wintertime snow was found, suggesting that sea salts originated from the sea ice. In the interior areas, marine biogenic SO₄²⁻ still dominated snow SO₄²⁻ in winter, leading to significant positive nssSO₄²⁻ values. Ion flux assessment suggests an efficient transport of nssSO₄²⁻ to at least as far inland as the ~2800 m contour line.”

(7) comments from Referees

Introduction: Several times the choices of your references are strange:

Line 61-65: You miss to cite here Legrand and Delmas (1985) here for a traverse in Adelie Land. This reference is particularly important since it is one the unique traverse for which acidity had been measured (not calculated), see my comment below.

(7) author's response

Thanks. We are sorry for missing some previous important works in the Introduction section. The reference, Legrand and Delmas (1985) was included in the revised manuscript. Legrand M. and Delmas R.J. (1985) Spatial and temporal variations of snow chemistry in Terre Adélie (East Antarctica). *Ann. Glaciol.* 7, 20-25.

(7) author's changes in manuscript

The sentence now reads,

“Snow chemistry has been broadly investigated along traverses during the International Trans-Antarctic Scientific Expedition (ITASE), e.g., DDU to Dome C, coast-interior traverse in Terre Adelie, Syowa to Dome F, Terra Nova Bay to Dome C, 1990 ITASE, and US ITASE in West Antarctica (Legrand and Delmas, 1985; Qin et al., 1992; Mulvaney and Wolff, 1994; Proposito et al., 2002; Suzuki et al., 2002; Dixon et al., 2013),”

(8) comments from Referees

Line 78: I don't think that Saltzman (1995) for the statement “that sulfate in the snow is mainly from marine biogenic sulphur species » is the adequate reference. Please here cite the review from Legrand (1995) or Legrand (1997).

(8) author's response

Thanks for the point. The references Legrand (1995) and Legrand (1997) were cited here. Legrand M., 1995. Sulphur-derived species in polar ice: a review, in: Delmas, R. (Ed.), *Ice core studies of global biogeochemical cycles*. Springer, pp. 91-119. Legrand M. (1997) Ice-core records of atmospheric sulphur. *Phil. Trans. R. Soc. Lond. B* 352, 241-250.

(8) author's changes in manuscript

The sentence now reads,

“SO₄²⁻ in the snow is mainly from marine biogenic sulfur species, dimethylsulphide (DMS) (Legrand, 1995; 1997),”

(9) comments from Referees

Line 79-80: The two cited references are fine but there are numerous previous works done on that and I would suggest mentioning the article in Nature 1987 (Legrand and Delmas, 1987) for instance.

(9) author's response

Agree that there are previous works done on that large volcanic eruption emissions can episodically contribute to the very high concentrations of SO_4^{2-} in ice core. The important work Legrand and Delmas (1987) was included here.

Legrand M. and Delmas R. (1987) A 220-year continuous record of volcanic H_2SO_4 in the Antarctic ice sheet. Nature 327, 671-676.

(9) author's changes in manuscript

The reference was included, and the sentence now reads,

“with a small proportion from sea salt aerosols, while large volcanic eruption emissions can episodically contribute to spikes in SO_4^{2-} concentration (Legrand and Delmas, 1987; Jiang et al., 2012; Cole-Dai et al., 2013).”

(10) comments from Referees

Line 82-85 : I don't think there is something on the Keene paper on calcium in snow. I suggest citing the study of the Vostok ice core in which the origin (and calculation) of excess potassium, magnesium and calcium were discussed (Legrand et al., 1988).

(10) author's response

Thanks for the point. The reference Keene et al. (2007) was replaced with Legrand et al. (1988).

Legrand M., Lorius C., Barkov N. and Petrov V. (1988) Vostok (Antarctica) ice core: Atmospheric chemistry changes over the last climatic cycle (160,000 years). Atmos. Environ. 22, 317-331.

(10) author's changes in manuscript

The reference was changed, and it now reads,

“Terrestrial sources can also contribute to potassium (K^+) and magnesium (Mg^{2+}) in snow, but the contribution proportion varies significantly among sites (Legrand et al., 1988; Khodzher et al., 2014).”

(11) comments from Referees

Line 92: Please cite the first study of ammonium and discussion of its marine origin in Antarctica by Legrand et al. (2000).

(11) author's response

We agree with that Legrand et al. (1999) (published in 1999 instead of 2000) is the first study of ammonium and discussion of its marine origin in Antarctica. And this work was cited. Since Pasteris et al. (2014) discussed the ammonium source of biomass burning, the reference Pasteris et al. (2014) was kept here.

Legrand M., Wolff E. and Wagenbach D. (1999) Antarctic aerosol and snowfall chemistry: implications for deep Antarctic ice-core chemistry. *Ann. Glaciol.* 29, 66-72.

(11) author's changes in manuscript

The original references were replaced with Legrand et al. (1999), and the sentence now reads, “and biogenic emissions in the Southern Ocean and/or mid-latitude biomass burning were proposed to be the major sources, depending on the investigation sites (Legrand et al., 1999; Pasteris et al., 2014)”

(12) comments from Referees

Lines 100-105: Please cite Weller et al. (2011) (see my comment on section 4.4).

(12) author's response

Thanks for the point, and the work Weller et al. (2011) was included in the section.

Weller R., Wagenbach D., Legrand M., Elsässer C., Tian-kunze X., Königlango G. and niglanglo G. (2011) Continuous 25-yr aerosol records at coastal Antarctica: inter-annual variability of ionic compounds and links to climate indices. *Tellus B* 63, 901-919.

(12) author's changes in manuscript

The section now reads,

“On annual to decadal time scales, ion concentrations in snow and ice tend to be associated with changes in transport from year to year (Severi et al., 2009; Weller et al., 2011), and thus large scale atmospheric and oceanic circulation in the Southern Hemisphere, such as the Southern Annular Mode (SAM), Southern Oscillation (SO) and Southern Indian Ocean Dipole (SIOD), could potentially influence variations in ions in ice (Russell and McGregor, 2010; Weller et al., 2011; Mayewski et al., 2017).”

(13) comments from Referees

Section 2.2:

Line 197-189: This statement “In Antarctic snow, concentrations of H^+ are usually not measured directly, but deduced from the ion-balance disequilibrium in the snow » is wrong and very misleading. In fact more than 1000 Antarctic snow and ice samples covering various time periods (present-climate, last glacial age) and collected at various places were measured for H^+ (Legrand, 1987; Legrand and Delmas 1984) including along a traverse in Adelie Land (Legrand

and Delmas, 1985). From that it was shown that the measurement of chloride, nitrate, sulfate, proton, sodium, ammonium, potassium, magnesium and calcium permit to verify the good balance between measured anions and measured cations. And from these studies that it was postulated that if not available the H⁺ concentration can be derived from the equation $[H^+] = [SO_4^{2-}] + [NO_3^-] + [Cl^-] - [Na^+] - [NH_4^+] - [K^+] - [Mg^{2+}] - [Ca^{2+}]$.
So please modify this section accordingly.

(13) author's response

Agreed and thanks for this comment. Indeed, from a number of observations (e.g., Legrand and Delmas, 1984; Legrand and Delmas, 1985; Legrand, 1987) it is deduced that concentrations of H⁺ can be reasonably deduced from the ion-balance disequilibrium, if the direct measurements of H⁺ are unavailable. This section was revised following the reviewer's suggestion.

Legrand M. (1987) Chemistry of Antarctic snow and ice. *J. de Phys.* 48, 77-86.

Legrand M. and Delmas R.J. (1985) Spatial and temporal variations of snow chemistry in Terre Adélie (East Antarctica). *Ann. Glaciol.* 7, 20-25.

Legrand M.R. and Delmas R.J. (1984) The ionic balance of Antarctic snow: a 10-year detailed record. *Atmos. Environ.* 18, 1867-1874.

(13) author's changes in manuscript

This section was re-written, and it now reads,

“In Antarctic snow, previous observations suggested that concentrations of H⁺ can be reasonably deduced from the ion-balance disequilibrium, if the direct measurements of H⁺ are unavailable (Legrand and Delmas, 1984; Legrand and Delmas, 1985; Legrand, 1987). Here, H⁺ concentration is calculated as follows.

$[H^+] = [SO_4^{2-}] + [NO_3^-] + [Cl^-] - [Na^+] - [NH_4^+] - [K^+] - [Mg^{2+}] - [Ca^{2+}]$ Eq. (1),”

(14) comments from Referees

Section 2.3:

Why do you play with EF instead of the amount of species present in excess with respect to the seawater composition. The calculations of excess are far more useful to discuss data (see further comments). In any case, calculations of error propagation are clearly needed here, especially for potassium, calcium, and magnesium.

(14) author's response

We agree that the calculation of non-sea-salt fractions of ions (nssX) (i.e., the concentrations in excess with respect to the seawater composition) is a more illustrative way. In the previous version, the enrichment factors (EFs) is a measurement of whether or not an ion is present in a relative abundance similar to that of seawater, which is an alternative way of showing the enrichment of an ion with respect to the bulk seawater composition. In the revised manuscript, we only focused on the calculation

and discussion of the non-sea-salt fractions of ions in snow, following the reviewer's suggestion.

(14) author's changes in manuscript

Section 2.3 was removed, and the associated text in the manuscript was revised accordingly. Also, the errors of non-sea-salt fractions of ions are now presented in the revised manuscript ($nssX \pm 1\sigma$).

Please see the revised manuscript, sections 2, 3, and 4.

(15) comments from Referees

Sections 2.4:

In your case (and it is often the case) the PCA approach does not give more information than those that can be simply derived by checking your plots. Checking your Figure 2, it immediately appears that (as expected) you have more sea-salt at the coast than inland (leading to your PC1). Outside of that, other information derived from the PCA analysis are not very powerful (see my further comments).

(15) author's response

We agree that the spatial variations in ions on the traverse and the correlation plots of ions versus Na^+ (Figures 2 and 5 in the revised manuscript) can provide the information on main sources of ions in surface snow. For instance, the sea salt related ions show high concentrations on the coast. In this case, it seems that the PCA approach is redundant. As for the PCA approach, it can offer information on the geochemical behaviors of chemical ions in addition to the sources, i.e., PCA is a powerful tool for identifying the common sources and/or transport process of chemicals in different environments. Sometimes, chemicals with different sources can also be highly loaded in the same principle component, possibly due to the common geochemical behaviors (e.g., transport process). Therefore, the text associated with PCA was kept in the revised manuscript. But we made more use of the ion concentrations in excess with respect to the seawater composition when identifying the main sources of ions in snow, following the reviewer's comments.

(15) author's changes in manuscript

In the methodology section, the subsection on principal component analysis (PCA) of ions was kept in the manuscript. In section 4.2 Groups of ions in surface snow, the PCA results were shortened, and the main source identification of ions in surface snow were performed by making more use of the non-sea-salt fractions of ions.

Please see the revised manuscript, sections 2.3, 4.1, and 4.2.

(16) comments from Referees

Section 3.1:

Line 234-245: It is for a very short time that melted snow is under saturated with respect to

CO₂. In fact after 10 min or so the equilibrium is reached but don't forget that another important factor is the temperature (colder is water more CO₂ is dissolved). Another source of uncertainty here is the PCO₂ in the lab of analysis (related for instance to the number of people). Please report the temperature at which your pH measurements were done. Would be good here to compare your calculated H⁺ not only with previous similar estimates but also with previous actual measurements (Legrand and Delmas, 1985, for instance).

(16) author's response

Thanks for the helpful comments. We agree that temperature is an important parameter influencing the amount of CO₂ dissolved in the water, and the lower temperature is water and the more CO₂ is dissolved. Also, we agree that the number of people in the lab can influence the pH values potentially. In this study, the measurements of pH were performed in a class 100 clean room at room temperature (~20°C)

Following the reviewer's suggestion, the calculated H⁺ concentrations in this study were also compared with previous direct measurements (Legrand and Delmas, 1985).

Legrand M. and Delmas R.J. (1985) Spatial and temporal variations of snow chemistry in Terre Adélie (East Antarctica). *Ann. Glaciol.* 7, 20-25.

(16) author's changes in manuscript

The measurement temperature was added in the revised manuscript, and the sentence reads, "pH values of surface snow sampled in 2013 were measured with a glass pH electrode in a class 100 room at room temperature (~20°C),"

The calculated H⁺ concentrations were compared to the direct measurements in previous investigation, and a paragraph was added in the revised manuscript, as follows,

"Here, the calculated H⁺ concentrations vary in the range of 0.51-10.01 μeq L⁻¹, with a mean of 3.53 ± 1.61 μeq L⁻¹. In general, the calculated H⁺ values of the coastal surface snow are generally comparable to previous direct measurements in Terre Adélie (Legrand and Delmas, 1985).

(17) comments from Referees

Section 3.2:

Line 274: I feel that after having calculated nssSO₄ (excess sulfate) you will identify an increasing trend of excess-sulfate from the coast to inland due to dry deposition (the sulfate one being obscured by the large amount of sea salt at the coast).

(17) author's response

In surface snow on the traverse, nssSO₄²⁻, on average, accounts for more than 90% of the total SO₄²⁻. That is, most of the SO₄²⁻ in surface snow is from the marine biogenic emissions. The concentrations of nssSO₄²⁻ in surface snow depend on the transport efficiency towards inland, the snow accumulation rate at a specific site, etc, and there was no significant correlation between nssSO₄²⁻ concentrations and distance from coast on the traverse (Figure below).

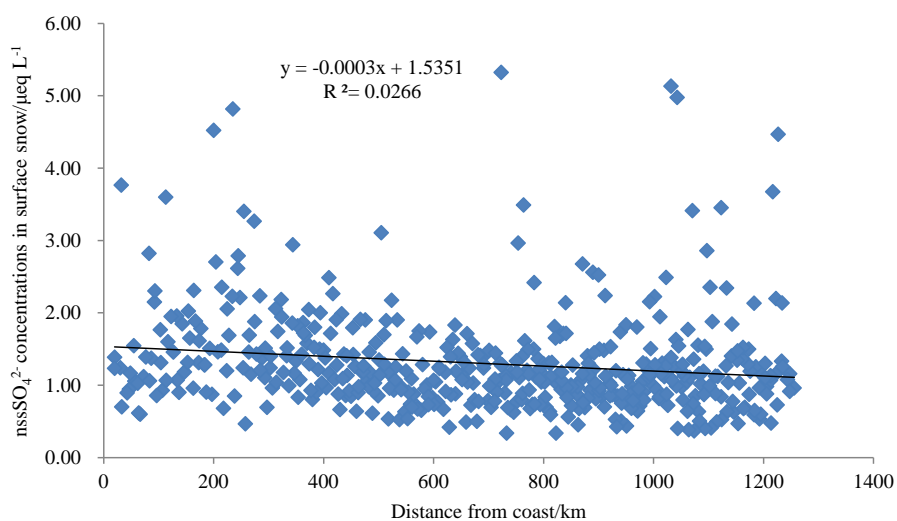


Figure Correlation between nssSO_4^{2-} concentrations in surface snow and distance from coast

(17) author's changes in manuscript

The spatial pattern of nssSO_4^{2-} was included in the revised manuscript, as follows,

“As for SO_4^{2-} (and nssSO_4^{2-}), NH_4^+ , and Ca^{2+} , no clear spatial trend was found on the traverse.”

(18) comments from Referees

Section 3.3:

This section needs to be significantly developed. First please show all species, second calculate excesses and corresponding error bars.

(18) author's response

Thanks for the constructive comment. Together with the comments from Reviewer#1, the Results section was re-organized and substantially revised. Section 3.3 in previous version was changed to section 3.1 (Chemical ion variations in snow pits) in the updated manuscript. In this section, 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+}) are presented. Then the non-sea-salt fractions of K^+ , Mg^{2+} , and Ca^{2+} are calculated (including the errors). Considering figure 3 (in revised manuscript) now contains the profiles of Cl^- , NO_3^- , SO_4^{2-} (nssSO_4^{2-}), Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , and the ratio of $\text{SO}_4^{2-}/\text{Na}^+$, the variations in nssCl^- , nssK^+ , nssMg^{2+} , and nssCa^{2+} were not included in Figure 3. (Note that nssSO_4^{2-} and the ratio of $\text{SO}_4^{2-}/\text{Na}^+$ included in Figure 3 help to date the snow pits). A new paragraph was added in the revised manuscript to show the variations in non-sea-salt fractions of ions in the three snow pits, and the profiles of non-sea-salt fractions of ions (nssCl^- , nssK^+ , nssMg^{2+} , and nssCa^{2+}) in snow pits P1 (a), P2 (b), and P3 (c) are now present in Figure S2 in the supplementary materials.

(18) author's changes in manuscript

This section (section 3.1 in the revised version) was substantially revised, and it now reads,

“Clear seasonal cycles of Na^+ and nssSO_4^{2-} are present in P1 and P2, and thus the two pits can be well dated, spanning ~ 3 years (Figs. 3 (a) and (b)). Based on the snow pit dating, it is estimated that snow accumulation rate is ~ 50 (P1) and ~ 33 cm snow per year (P2), agreeing well with the field measurements (P1: $\sim 150 \text{ kg m}^{-2} \text{ a}^{-1}$; P2: $\sim 100 \text{ kg m}^{-2} \text{ a}^{-1}$; Fig. 2(a)), assuming a snow density of $\sim 0.33 \text{ g cm}^{-3}$. At P1, negative nssSO_4^{2-} values are observed in winter snow, i.e., $\text{SO}_4^{2-}/\text{Na}^+$ ratio below that of bulk seawater, while all of the nssSO_4^{2-} data in P2 pit are positive. It is difficult to assign the samples in the snow pits to the four distinct seasons based on the measured parameters, and thus, in the following discussion, we choose a conservative assignment method, i.e., a summer season featured with higher nssSO_4^{2-} and $\text{SO}_4^{2-}/\text{Na}^+$ ratio (and lower Na^+) and a winter season characterized with the opposite patterns. In addition to SO_4^{2-} and Na^+ , the other species also show seasonal variations, especially in pit P1, where elevated levels of NO_3^- and NH_4^+ are generally present in summer snow, and the values of Cl^- , K^+ , Mg^{2+} , and Ca^{2+} are high in winter. It is noted that even in the same season, ion concentrations could vary among samples at a single site (e.g., shaded areas in Figs. 3(a) and (b)).

As for nssSO_4^{2-} at P3, the very large signal at the depth of ~ 120 cm is most likely the fallout from the massive eruption of Pinatubo in 1991 (Fig. 3(c)), based upon previous observations at Dome A (e.g., Hou et al., 2007). Accordingly, the snow accumulation rate from 1992 to 2010 is $\sim 22 \text{ kg m}^{-2} \text{ a}^{-1}$, in line with previous investigations (Hou et al., 2007; Jiang et al., 2012; Ding et al., 2016). Based on nssSO_4^{2-} signals and the method proposed by Cole-Dai et al. (1997), 19 continuous samples have been identified as influenced by Pinatubo eruption, covering ~ 2.5 years, possibly suggesting that the effects of Pinatubo eruption on atmospheric chemistry lasted at least for 2.5 years over Dome A. Interestingly, only elevated SO_4^{2-} concentrations are present during this period, and anomalous high or low concentrations of other ions are absent. Additionally, no correlation was found between nssSO_4^{2-} and other species during the 2.5 years, possibly suggesting that Pinatubo volcanic emissions contribute less to the ion budgets other than SO_4^{2-} at Dome A.

Previous investigations proposed that Na^+ and nssSO_4^{2-} in surface snow (top ~ 1 cm) collected during a full year at central Antarctica show clear seasonal cycles, with high (low) Na^+ in winter (summer) snow (Udisti et al., 2012). At P3, Na^+ , nssSO_4^{2-} and the ratios of $\text{SO}_4^{2-}/\text{Na}^+$ 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 variability of ions. Following the field measurements of snow accumulation rate at Dome A during 2008-2011 ($\sim 20 \text{ kg m}^{-2} \text{ a}^{-1}$; 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 $\text{SO}_4^{2-}/\text{Na}^+$ 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.

In terms of the non-sea-salt fractions in the snow pits (Fig. S2), nssCl^- is lower at P1 ($0.25 \pm 0.28 \mu\text{eq L}^{-1}$) than at the inland sites P2 and P3 (0.42 ± 0.18 and $0.58 \pm 0.34 \mu\text{eq L}^{-1}$, respectively), while the

concentrations of nssK^+ , nssMg^{2+} , and nssCa^{2+} generally show similar spatial patterns, possibly due to the low snow accumulation rate in interior areas. Different from the sea salt ions and nssSO_4^{2-} , nssCl^- , nssK^+ , nssMg^{2+} , and nssCa^{2+} in pits P1 and P2, do not show significant seasonal patterns. In the coastal pit P1, the non-sea-salt fractions account for less (<~30%) of the total ions, and the contribution percentages of non-sea-salt fractions increased at inland sites P2 and P3, about 30-70 %.”

(19) comments from Referees

Section 4.1:

Please present excess here.

Line 371-373: I disagree with that since only a very small amount of potassium from dust is leachable (and measured with your IC) but it is not at all true for calcium (see Legrand et al., 1988, for instance). Therefore you cannot compare your snow data with the mean crust composition from Bowen that refers to total potassium (insoluble and soluble). Please check in Legrand et al. (1988) or Legrand (1987) information on excess potassium versus excess calcium.

(19) author's response

Thanks for the comments, and the concentrations in excess, instead of the EFs, are now presented.

We agree that the IC can only measure the soluble fractions of the terrestrial particle mass, and the insoluble fraction is largely unknown. The non-sea-salt fractions of ions (e.g., nssMg^{2+} and nssCa^{2+}) can represent, at least partly, the terrestrial crustal materials. For instance, close relationship was found between nssMg^{2+} or nssCa^{2+} and aluminum concentrations at Dome C (Legrand and Delmas, 1988). We admit that possibly different portions of K^+ and Ca^{2+} in the crustal material will be dissolved in the water (snow), and thus the calculation here seems to be uncertain regarding that Bowen's work is for the total crust composition (i.e., including both soluble and insoluble fractions). In this, we cannot get the ratio of $\text{K}^+/\text{Ca}^{2+}$ from the water soluble fractions of terrestrial particle mass, and thus the calculation in this section was removed and additional discussion on the sources of K^+ was included.

Legrand M.R. and Delmas R.J. (1988) Soluble impurities in four Antarctic ice cores over the last 30,000 years. *Ann. Glaciol.* 10, 116-120.

(19) author's changes in manuscript

The calculation of EFs was removed, and all of the EFs in the original version were replaced with the concentrations in excess (i.e., the non-sea-salt fractions).

The calculation of K^+ from the biomass burning emissions was removed, and additional discussion was included in the revised manuscript, as follows,
“Here, no elevated snow nssK^+ peaks were found in austral autumn (i.e., P1 and P2; Fig. S2) when the chemicals emitted from biomass burning (e.g., black carbon) often peaked in Antarctic snow (Sigl et al., 2016). In addition, there is a close relationship between nssK^+ and nssCa^{2+} ($r=0.22$, $p<0.001$), suggesting that snow nssK^+ is unlikely dominated by biomass burning emissions.”

(20) comments from Referees

I find also that your calcium data (Fig 2) are often above 4 ppb (that is also higher than seen in numerous ice cores under present-day climate. Please comment.

(20) author's response

Thanks for the comment. Indeed, values of Ca^{2+} at some sites, especially on the coast and the in the inland regions (about 500-900 km from the coast). In the coastal areas, the high concentrations are likely associated with the marine inputs, while the elevated values at the inland sites could be related to the low and fluctuating snow accumulation rate due to the strong wind scouring (Ding et al., 2011; Das et al., 2013). In fact, similar elevated Ca^{2+} values were found in the glaze/dune regions on the US ITASE traverses across East and West Antarctica (Dixon et al., 2013).

(20) author's changes in manuscript

A paragraph was included in the revised manuscript to explore the possible reasons for the high Ca^{2+} concentrations at some sites, as follows,

“It is noted that some Ca^{2+} concentrations in surface snow (e.g., coastal and some inland sites) are above $0.2 \mu\text{eq L}^{-1}$ (Fig. 2(i)), slightly higher than most reports of snow and ice under present-day climate (e.g., Legrand and Mayewski, 1997). On the coast, the high concentrations could be related to marine inputs (e.g., Bertler et al., 2005), while the elevated values in the inland regions (about 500-900 km from the coast, where the glaze/dune are distributed) are possibly associated with the low and fluctuating snow accumulation rate due to the strong wind scouring (Ding et al., 2011; Das et al., 2013). Similarly, in the glaze/dune regions on the US ITASE traverses across East and West Antarctica, concentrations of Ca^{2+} in snow and ice are also often above $\sim 0.2 \mu\text{eq L}^{-1}$ (Dixon et al., 2013).”

(21) comments from Referees

Line 445-454: I disagree with this discussion. Whereas I agree nitrate is clearly related to atmospheric oxidant, the link to sulfate (its similar presence in PC3) is due to the fact that you concentrate nssSO_4 due to dry deposition at sites with low accumulation rate while nitrate is enhanced for a totally different reason (photochemistry).

(21) author's response

Thanks for the helpful comments. Indeed, high concentrations of both nssSO_4^{2-} and NO_3^- were found at the sites with low snow accumulation rate, while elevated NO_3^- concentrations are on the Dome A plateau and high nssSO_4^{2-} values occurred in the region about 300-800km from the coast (see figure below). Thus, snow accumulation rate is positively correlated with both species (see figure below). The positive loading of SO_4^{2-} in PC3 and the correlation between SO_4^{2-} and NO_3^- in surface snow could be explained, at least in part, by the effects of snow accumulation rate.

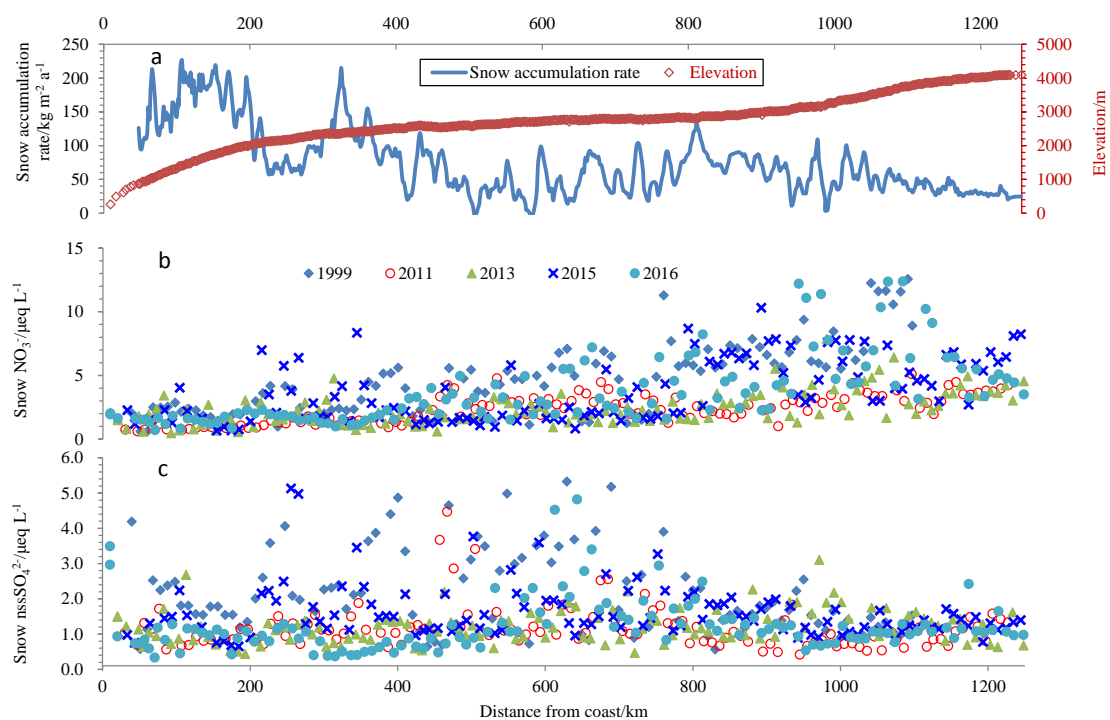


Figure Annual snow accumulation rate, elevation (a) and concentrations of NO_3^- (b) and nssSO_4^{2-} (c) in surface snow collected during five seasons.

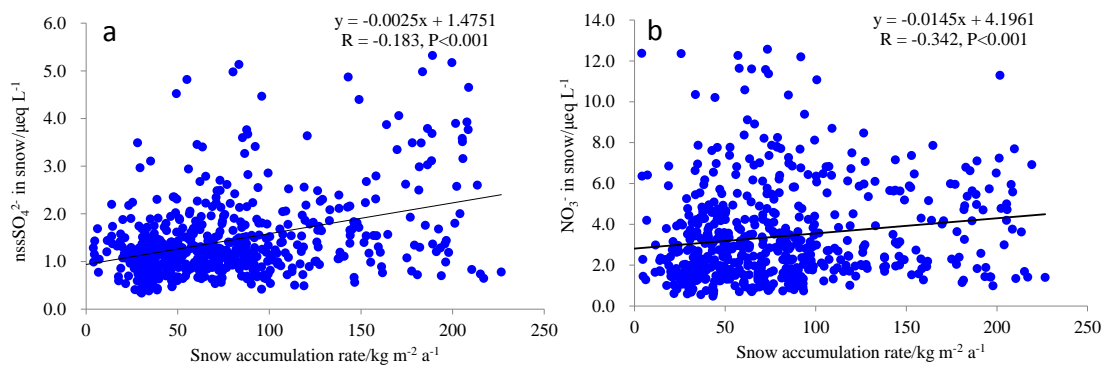


Figure Relationship between snow accumulation rate and nssSO_4^{2-} (a) or NO_3^- (b) in surface snow on the traverse.

In addition, nssSO_4^{2-} and NO_3^- in the atmosphere are mainly associated with secondary aerosols, and the formation of both species is closely related to the oxidants HO_x , RO_x , etc (Ishino et al., 2017; Shi et al., 2018). Therefore, we cannot completely rule out that the correlation between SO_4^{2-} and NO_3^- may be associated with their formations in the atmosphere.

(21) author's changes in manuscript

Following the reviewer's comments, this point was re-discussed in the revised manuscript, as follows, "SO₄²⁻ did not show high loadings in any of the three extracted components. Its positive loading in PC1 (0.55) and weak relationships between SO₄²⁻ 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^- .”

(22) comments from Referees

Section 4.2:

Line 430: Your argument of an absence of correlation between ammonium and organic tracers (Shi et al., 2019) is not correct since the authors invoked a decrease of levoglucosan from the coast to inland due to its photochemical degradation.

(22) author's response

We agree that the levoglucosan could undergo photochemical degradation in the snow because of the 24-h sunlight during summertime in Antarctica. Thus the absence of correlation between NH_4^+ and levoglucosan could be associated with the post-degradation of levoglucosan. A recent observation on the same traverse suggested no correlation between NH_4^+ and black carbon that is mainly from Southern Hemisphere biomass burning emissions (Ma et al., 2020), in surface snow. In addition, no association was observed between NH_4^+ and phenolic compounds (i.e., vallinic and syringic acids, which are derived mostly from the combustion of lignin) in surface snow (Shi et al., 2019). In this case, the observations do not support that NH_4^+ is dominated by biomass burning emissions.

(22) author's changes in manuscript

This section was re-written following the comments from both reviewers, as follows,

“ 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.”

(23) comments from Referees

Line 433: Checking your Fig S1 in fact you have an outstanding value (17 ppb). Discarding this value I have difficulty to identify a seasonal cycle. Also your mean value (removing the outlier) is close to 2 ppb (it is slightly but significantly higher than what was seen in previous ice core studies). Please comment.

(23) author's response

Thanks for the comment. Indeed, there is one sample with very high NH_4^+ concentration, $\sim 0.9 \mu\text{eq L}^{-1}$. If this data was excluded, there is no significant difference between the concentrations in summer and winter (Independent samples t test; $p > 0.05$), although the mean concentration is slightly higher in summer than in winter. The discussion on the seasonal variation in NH_4^+ and Figure S1 (in original version) were removed in the revised manuscript.

Indeed, the mean value of NH_4^+ at snow pit P1 is $0.16 \mu\text{eq L}^{-1}$ (removing the outlier). The NH_4^+ concentrations here are slightly higher than what was observed in previous ice core studies, but generally comparable to some coastal observations (e.g., coastal sites in Terre Adelie) (Legrand and Delmas, 1985; Legrand et al., 1998). At the sites close to coastal line, especially near the penguin colony, snow NH_4^+ could be influenced by the penguin colony emissions (Legrand et al., 1998; Rankin and Wolff, 2000), but this effect is very localized, likely within several km from the colony (Rankin and Wolff, 2000). Considering that site P1 is 46 km from the coast and the ratio of Na^+/K^+ is close to that of bulk seawater (Figure 9 in the revised manuscript), snow NH_4^+ at P1 is unlikely dominated by local penguin colony emissions. Thus, NH_4^+ at the coastal site P1 is more likely influenced by marine biogenic emissions.

Legrand M. and Delmas R.J. (1985) Spatial and temporal variations of snow chemistry in Terre Adélie (East Antarctica). *Ann. Glaciol.* 7, 20-25.

Legrand M., Ducroz F., Wagenbach D., Mulvaney R. and Hall J. (1998) Ammonium in coastal Antarctic aerosol and snow: Role of polar ocean and penguin emissions. *J. Geophys. Res.* 103, 11043-11056.

Rankin A.M. and Wolff E.W. (2000) Ammonium and potassium in snow around an emperor penguin colony. *Antarct. Sci.* 12, 154-159.

(23) author's changes in manuscript

The profile of NH_4^+ in P1 was included in Figure 3 in the revised manuscript, and the outstanding value was excluded. Also the relatively high concentrations of NH_4^+ in P1 was discussed, as follows,

“ 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.”

“In addition, NH_4^+ concentration at P1 ($0.16 \pm 0.05 \mu\text{eq L}^{-1}$) is slightly higher than the previous reports of ice cores but comparable to some coastal observations (e.g., coastal sites in Terre Adelie) (Legrand and Delmas, 1985; Legrand et al., 1998), possibly associated with marine biogenic emissions (i.e., close to the coast) (discussed above).”

(24) comments from Referees

Section 4.4: Given the poor representativeness of snow samples, it is clear that examination with respect to SOI is very difficult. For your information, based on a continuous record of 25 year of aerosol, Weller et al. (2011) examine the inter-annual variability with respect to climate-related indices (SAM, SOI, SIE) and nothing very significance had appear.

Weller R., Wagenbach D., Legrand M., Els ässer C., Tian-kunze X. and Königlanglo G., Continuous 25-years aerosol records at coastal Antarctica: I. Inter-annual variability of ionic compounds and links to climate indices, *Tellus*, 63B, 901-919, DOI:10.1111/j.1600-0889.2011.00542.x, 2011.

(24) author's response

We agree that the surface samples, which represent different lengths of time at varied locations, are unlikely representative of seasonal (or interannual) variations. Together with the comments of Reviewer#1, this section was removed.

Also, thanks for the reference, which provides much longer time of information. Indeed, it is difficult to construct the potential association between chemicals and the climate-related indices.

Weller R., Wagenbach D., Legrand M., Els ässer C., Tian-kunze X. and Königlanglo G. (2011) Continuous 25-yr aerosol records at coastal Antarctica: inter-annual variability of ionic compounds and links to climate indices. *Tellus B* 63, 901-919.

(24) author's changes in manuscript

This section was removed in the revised manuscript.

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End of responses to Prof. Michel Legrand.