

Reviewer #2

We are very grateful to reviewer#2 for his/her detailed comments and very useful 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 the reviewer, in the order of (1) comments from Referees, (2) author's response, and (3) author's changes in manuscript.

Reviewer comments are in black, and the responses are in blue.

(1) comments from Referees

This manuscript reports on nitrate in samples collected in the frame of an intensive program of snow sampling made along a traverse from the coast to Dome A (East Antarctica). The samplings include 120 surface snow samples (upper 3 cm), 20 snowpits (down to 1.5-3.0 m depth), and a few crystal ice samples. From the coast to the inner plateau, an increasing trend of nitrate present in surface snow is observed whereas the content of deeper snow pit layers are lower at inland sites than at the coast. Extremely high concentrations are found in crystal ice (reaching almost 1 ppmw). Data are discussed with respect to occurrence of post-depositional remobilization of nitrate, wet and dry deposition, and possible role of other ions (sodium and sulfate).

Overall evaluation:

First, the authors have to be congratulated for having successfully conducted such a very large snow-sampling program, likely sometimes done under harsh weather conditions. The data certainly contain valuable information in view to better understand incorporation, remobilisation and partial preservation of nitrate atmospheric signal in cold archives. This topic is clearly relevant for the Cryosphere journal.

(1) author's response

We thank the reviewer very much for reviewing our manuscript and the positive comments. As the reviewer mentioned, the snowpit sampling is usually made under the very harsh weather conditions, e.g., extremely low temperature and heavy blowing snow. We appreciate the Chinese inland Antarctic expedition team members for providing help during sampling.

(1) author's changes in manuscript

We will revise the manuscript following the reviewer's comments and suggestions, see below.

(2) comments from Referees

As it stands the manuscript however requires major revisions and a reevaluation prior to publication. Indeed, at several places in the manuscript data discussions are incorrect, and generally do not enough consider atmospheric information available for the Antarctic atmosphere. Given the scarcity of data presented in this work, I strongly encourage the authors to reformulate the manuscript and in the following I try to identify what would be addressed in an in depth reformulated version of this manuscript.

(2) author's response

We thank the reviewer for pointing out the shortcomings of the manuscript. We agree that the atmospheric information was not considered enough. We will improve the work following the reviewer's suggestions/comments.

(2) author's changes in manuscript

The manuscript was modified according to the comments from the reviewer, see below and section 4 in the revised manuscript.

(3) comments from Referees

Introduction.

This paragraph has to be reworded on several aspects:

Lines 54-86: You missed here several important papers that have discussed in details the origins of nitrate in Antarctica. For instance, Legrand and Kirchner (1990) extensively discussed (1) the absence of link between solar activity and nitrate in snow, (2) what are the main possible sources of nitrate for Antarctica (stratospheric reservoir and long-range transport in the upper troposphere of lightning production, etc). Also model simulations from Legrand et al. (1989) discussed the source of nitrate for Antarctic regions.

Legrand, M., and Kirchner, S.: Origins and variations of nitrate in South Polar precipitation, J. Geophys. Res., 95, 3493-3507 1990.

Legrand, M. R., F. Stordal, I. S. A. Isaksen, and B. Rognerud (1989), A model study of the stratospheric budget of odd nitrogen, including effects of solar cycle variations, Tellus B, 41(B4), 413–426, doi:10.1111/j.1600- 0889.1989.tb00318.x.

(3) author's response

We agree with the reviewer and are sorry for missing the two important references concerning Antarctic NO_3^- budget. In terms of the Antarctic NO_3^- budget, lightning and NO_x produced in the lower stratosphere were thought to play a major role (Legrand et al., 1989; Legrand and Kirchner, 1990). Also, it is suggested that there is not necessarily a connection between solar variability and NO_3^- concentrations (Legrand and Kirchner, 1990).

(3) author's changes in manuscript

The two references were included. The major contribution by lightning and by NO_x produced in the lower stratosphere to Antarctic NO_3^- budget was clarified. In addition, the investigation made by Legrand and Kirchner (1990) suggesting no correlation between solar activity (11-year solar cycle, low solar activity time periods, and solar proton events) and the NO_3^- content of south polar snow was added to the manuscript. Please see the revision-tracked version of manuscript.

(4) comments from Referees

Lines 80-83: You missed here to report two recent papers from Wolf et al. that strongly question the assumption that solar flares and SPE are recorded in ice. Also model simulations do not support at all such an assumption (Legrand et al., 1989; Duderstadt et al., 2014).

Wolff, E. W., M. Bigler, M. A. J. Curran, J. E. Dibb, M. M. Frey, M. Legrand, and J. R. McConnell (2012), The Carrington Event not observed in most ice core nitrate records, *Geophys. Res. Lett.*, 39, L08503, doi:10.1029/2012GL051603.

Wolff, E. W., M. Bigler, M. A. J. Curran, J. E. Dibb, M. M. Frey, M. Legrand, and J. R. McConnell (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. Space Physics*, 121, doi:10.1002/2015JA021570.

Legrand, M. R., F. Stordal, I. S. A. Isaksen, and B. Rognerud (1989), A model study of the stratospheric budget of odd nitrogen, including effects of solar cycle variations, *Tellus B*, 41(B4), 413–426, doi:10.1111/j.1600- 0889.1989.tb00318.x.

Duderstadt, K. A., J. E. Dibb, C. H. Jackman, C. E. Randall, S. C. Solomon, M. J. Mills, N. A. Schwadron, and H. E. Spence (2014), Nitrate deposition to surface snow at Summit, Greenland, following the 9 November 2000 solar proton event, *J. Geophys. Res. Atmos.*, 119, 6938–6957, doi:10.1002/2013JD021389.

(4) author's response

Thanks for pointing this out. The observations and modeling works by (Legrand et al., 1989; Legrand and Kirchner, 1990; Wolff et al., 2008; Wolff et al., 2012; Duderstadt et al., 2014;

Duderstadt et al., 2016; Wolff et al., 2016) were included. Indeed, most of observations and recent modeling studies have established that there is not a clear connection between solar variability and NO_3^- concentrations (Legrand et al., 1989; Legrand and Kirchner, 1990; Wolff et al., 2008; Wolff et al., 2012; Duderstadt et al., 2014; Duderstadt et al., 2016; Wolff et al., 2016).

(4) author's changes in manuscript

The works made by Wolff et al., Legrand et al., and Duderstadt et al., are included in the revised manuscript. Please see the revised version of the manuscript.

(5) comments from Referees

A few sentences on the physical form of nitrate (partitioning between the gas phase, and particulate phase) would be welcome (see my next comment) to better introduce the data discussion with respect to deposition, remobilization, etc.

(5) author's response

A good point, thanks. A summary of the observations on partitioning of NO_3^- between the gaseous phase and particulate phase will be helpful to a better understanding of the deposition and re-emission of NO_3^- . At Dome C on the East Antarctic plateau, observations on the atmospheric NO_3^- have been carried out during the years from 2006 to 2016 (Traversi et al., 2014; Legrand et al., 2016; Legrand et al., 2017b; Traversi et al., 2017), which are important works towards a quantitative understanding of NO_3^- partitioning in the atmosphere.

(5) author's changes in manuscript

Following the reviewer's suggestion, a paragraph summarizing the partitioning between the gas phase, and particulate phase on NO_3^- was included in the revised manuscript, as follows,

In the atmosphere in Antarctica, particularly during spring and summer, NO_3^- is found to be mainly in the form of gas phase HNO_3 , with NO_3^- concentration several times higher in gas phase than in the particulate phase (Piel et al., 2006; Legrand et al., 2017b; Traversi et al., 2017). During the post-depositional processes, the uptake of gaseous HNO_3 is thought to be important in NO_3^- concentrations in surface snow layers (Udisti et al., 2004; Traversi et al., 2014; Traversi et al., 2017). Due to the high concentration in summer, HNO_3 appears to play an important role in acidifying sea-salt particles, possibly accounting for the presence of NO_3^- in the particulate phase in summer (Jourdain and Legrand, 2002; Legrand et al., 2017b; Traversi et al., 2017). It is noted that the significant increase of NO_3^- during the cold periods (e.g., Last Glacial Maximum) could be associated with its attachment to dust aerosol, instead of the gas phase HNO_3 (Legrand et al., 1999; Wolff et al., 2010).

Please see the revised version of the manuscript, section 1 Introduction.

(6) comments from Referees

Data discussion (Section 3): Please reconsider your data in the light of recent papers dealing with nitric acid gas phase and nitrate in the aerosol phase and their changes over the year in Antarctica.

For instance, check the following recent paper and references therein:

Legrand, M., Preunkert, S., Wolff, E., Weller, R., Jourdain, B., and Wagenbach, D. : Year-round records of bulk and size-segregated aerosol composition in central Antarctica (Concordia site) - Part 1 : Fractionation of sea-salt particles, *Atmos. Chem. Phys.*, 17, 14039-14054, <https://doi.org/10.5194/acp-17-14039-2017>, 2017.

(6) author's response

We thank the reviewer for the very constructive suggestion. The partitioning of NO_3^- between gas-phase and particulate phase will be of importance to NO_3^- levels in the snowpack, especially the topmost crystal ice layers. The observed high levels of gas phase HNO_3 in central Antarctica during summer support the importance of the re-emission from snow through the photolysis of NO_3^- in affecting atmospheric $\text{NO}_x/\text{NO}_3^-$ budget (e.g., Erbland et al., 2013). The atmospheric gaseous HNO_3 likely co-condenses with water vapor (Bock et al., 2016), especially on the extensively developed crystal ice layers on Antarctic plateaus (discussed in the main text), leading to an enrichment of NO_3^- in surface snow. In addition, a large concentration of HNO_3 would enhance its reaction with sea-salt, leading to elevated particulate NO_3^- concentrations (Legrand et al., 2017b). The significant correlation between NO_3^- and H^+ ($R^2 = 0.65$, $p < 0.01$) and lack of correlation between NO_3^- and sea salt Na^+ in inland Antarctic surface snow seems to suggest the importance of atmospheric gas phase HNO_3 in affecting surface snow NO_3^- concentrations, in particular NO_3^- levels in the crystal ice samples (correlation between NO_3^- and H^+ , $R^2 = 0.99$, $p < 0.01$).

(6) author's changes in manuscript

The physical form of NO_3^- affecting NO_3^- concentrations in snow was discussed and included in the revised manuscript, as follows,

In inland Antarctica, the dominant NO_3^- species in the atmosphere is gaseous HNO_3 during summertime, while particulate NO_3^- is more important in winter (Legrand et al., 2017b; Traversi et al., 2017). The high levels of gas phase HNO_3 in summer support the importance of the re-emission from snow through the photolysis of NO_3^- in affecting the atmospheric $\text{NO}_x/\text{NO}_3^-$ budget (Erbland et al., 2013). On the one hand, the gaseous HNO_3 can be efficiently co-condensed with water vapour onto the extensively developed crystal ice layers on Antarctic plateaus

(discussed above), leading to an enrichment of NO_3^- in surface snow (Bock et al., 2016). On the other hand, a large concentration of HNO_3 would enhance its reaction with sea-salt, leading to elevated particulate NO_3^- concentrations (Legrand et al., 2017b). The significant correlation between NO_3^- and H^+ in inland Antarctic surface snow ($R^2 = 0.65$, $p < 0.01$) seems to support the importance of atmospheric gas phase HNO_3 in affecting surface snow NO_3^- concentrations, in particular NO_3^- levels in the crystal ice samples (Fig. 1).

Please see the revised version of the manuscript, section 4.1.2 NO_3^- in inland snowpack.

(7) comments from Referees

Two overall comments:

The idea that nitrate is trapped on coarse sea-salt particles is incorrect (or not enough precise): Atmospheric data show that nitrate stays on the intermediate size particles (1-2 micron range) and not on the coarse ones like sea-salt (even at the coast): Jourdain and Legrand (2002); Teilina et al. (2000), Rankin et al. (2003), and Legrand et al. (2017).

Teinila, K., Kerminen, V.-M., and Hillamo, R. (2000), A study of sizesegregated aerosol chemistry in the Antarctic atmosphere, J. G. R. 105, 3893- 3904.

Rankin, A. M. and Wolff, E. W.: A year-long record of size- segregated aerosol composition at Halley, Antarctica, J. Geophys. Res., 108, 4775, <https://doi.org/10.1029/2003JD003993>, 2003.

Jourdain, B. and Legrand, M.: Year-round records of bulk and size-segregated aerosol composition and HCl and HNO_3 levels in the Dumont d'Urville (coastal Antarctica) atmosphere: Implications for sea-salt aerosol fractionation in the winter and summer, J. Geophys. Res., 107, 4645, <https://doi.org/10.1029/2002JD002471>, 2002.

(7) author's response

Atmospheric NO_3^- in Antarctica is mainly in the gas phase (HNO_3), while the particulate phase represents less, particularly in inland Antarctica. As for the particulate phase (also called “aerosol” in previous observations), most of the NO_3^- is found on the intermediate size particles (1 – 2 μm) (Jourdain and Legrand, 2002; Rankin and Wolff, 2003; Legrand et al., 2017b). As the reviewer mentioned, the NO_3^- is not trapped on the coarse sea-salt particles. But the presence of sea salt aerosol can influence atmospheric NO_3^- in two ways. Firstly, higher atmospheric sea salt aerosol concentrations are expected to promote the conversion of gaseous HNO_3 to particulate phase, allowing for the efficient deposition of NO_3^- via the aerosol mechanisms. On the other hand, the saline ice in the atmosphere favors the direct uptake of gaseous HNO_3 on ice surface. Thus, sea salt aerosols play an important role in the scavenging of gaseous HNO_3 from the atmosphere (Hara et al., 2005), and elevated NO_3^- concentrations are usually accompanied by Na^+ spikes in

snowpack (e.g., at Halley station, a coastal location; Wolff et al., 2008).

(7) author's changes in manuscript

Following the reviewer's comments, the relationship between NO_3^- and sea salt in the snowpack was re-discussed, as follows,

In comparison with nssSO_4^{2-} aerosols, the sea-salt aerosols (Na^+) are coarser and can be removed preferentially from the atmosphere due to a larger dry deposition velocity. High atmospheric sea salt aerosol concentrations are expected to promote the conversion of gaseous HNO_3 to particulate phase, considering that most of the NO_3^- in the atmosphere is in the gas phase (HNO_3). In this case, particulate NO_3^- can be efficiently lost via aerosol mechanisms. In addition, the saline ice also favors the direct uptake of gaseous HNO_3 to the ice surface. Changes in partitioning between gas phase (HNO_3) and particulate phase will affect NO_3^- levels due to the different wet and dry deposition rates of the two species (Aw and Kleeman, 2003). Thus, sea salt aerosols play an important role in the scavenging of gaseous HNO_3 from the atmosphere (Hara et al., 2005), and elevated NO_3^- concentrations are usually accompanied by Na^+ spikes in snowpack (e.g., at Halley station, a coastal site; Wolff et al., 2008). Here, no significant correlation was found between Na^+ and NO_3^- in coastal snow (Fig. 7b). The concentration profiles of NO_3^- and Na^+ in coastal surface snow are shown in Fig. 8, and NO_3^- roughly corresponds to Na^+ in some areas, e.g., 50-150 km and 300-450 km distance inland, although in general they are not very coherent. It is noted that amongst the 4 snow samples with $\text{Na}^+ > 1.5 \mu\text{eq L}^{-1}$ (open circles in Fig. 8), only one sample co-exhibits a NO_3^- spike. This is different from observations at Halley station, where Na^+ peaks usually led to elevated NO_3^- levels in surface snow in summer (Wolff et al., 2008). Of the 4 largest Na^+ spikes, one is a fresh snowfall sample (dashed ellipse in Fig. 8), and this sample shows the highest Na^+ concentration ($2.8 \mu\text{eq L}^{-1}$) and low NO_3^- ($0.75 \mu\text{eq L}^{-1}$). It is noted that NO_3^- concentration in this fresh snowfall is close to the model predictions ($0.7 \pm 0.07 \mu\text{eq L}^{-1}$; section 4.1.1), validating that the simple linear deposition model (i.e., the Eq. 6) can well depict the deposition and preservation of NO_3^- in coastal snowpack. At inland sites, no correlation was found between NO_3^- and Na^+ (Fig. 7e), likely explained by the alteration of NO_3^- concentration by post-depositional processing (discussed above).

Please see the revised version of the manuscript, section 4.2 Effects of coexisting ions on NO_3^- .

(8) comments from Referees

The relationship between NssSO_4 and nitrate: The interpretation of the correlation between nitrate and sulphuric acid referring to Brown et al. (2006) is misleading. Indeed this study discussed of the reaction of N_2O_5 on acidic sulphate promoting the formation of HNO_3 in a polluted atmosphere at night. Whatever the Antarctic site, the acidic sulphate is maximum in summer whereas, if present, N_2O_5 can only exist in the Antarctic atmosphere in winter (due to photolysis of the NO_3 radical in summer, N_2O_5 does not exist in summer). So the correlation seen in snow

cannot be explained like that.

(8) author's response

We agree with the referee that the conversion of N_2O_5 to HNO_3 during austral summer could be rather negligible due to the photolysis of NO_3 radical in summertime ($\text{NO}_3 + \text{NO}_2 + \text{M} \rightarrow \text{N}_2\text{O}_5 + \text{M}$). This point was clarified in the revised manuscript. Following previous investigations, the high concentrations of nssSO_4^{2-} aerosols could provide nucleation centers forming the multi-ion complexes with HNO_3 in the atmosphere, possibly leading to elevated NO_3^- concentrations in the snow (Laluraj et al., 2010). On the other hand, the presence of fine nssSO_4^{2-} aerosol may also enhance the direct uptake of gas phase HNO_3 onto the surface, resulting in NO_3^- deposition via aerosol mechanisms. It is acknowledged that these are the plausible explanation of the association between the two anions, and it cannot be ruled out that other processes and/or chemistry would influence the relationship of the parameters. Further works are needed to characterize the formation of SO_4^{2-} and NO_3^- and their potential association in Antarctic atmosphere.

(8) author's changes in manuscript

Following the comments from reviewer#2 and Prof. Savarino, the correlation between SO_4^{2-} and NO_3^- was re-discussed in the revised manuscript, as follows,

In surface snow, the non-sea salt fraction of SO_4^{2-} accounts for 75 - 99 % of its total budget, with a mean of 95 %. The percentages are relatively higher in inland regions than at coastal sites. On the coast, a positive relationship was found between nssSO_4^{2-} and NO_3^- ($R^2 = 0.32$, $p < 0.01$; Fig. 7a). Previous observations suggest that NO_3^- and nssSO_4^{2-} peaks in the atmosphere and snow are usually present in summer (Jourdain and Legrand, 2002; Wolff et al., 2008; Sigl et al., 2016; Legrand et al., 2017a; Legrand et al., 2017b). However, the similar seasonal pattern of the two species is associated with distinct sources, i.e., SO_4^{2-} is mainly derived from marine biogenic emissions while NO_3^- is influenced by photolysis and tropospheric transport (Savarino et al., 2007; Lee et al., 2014; Zatko et al., 2016). In the atmosphere, most of SO_4^{2-} is on the submicron particles, while most of NO_3^- is gaseous HNO_3 and the particulate NO_3^- is mainly on the intermediate size particles (Jourdain and Legrand, 2002; Rankin and Wolff, 2003; Legrand et al., 2017a; Legrand et al., 2017b). Thus, the correlation between NO_3^- and SO_4^{2-} is unlikely explained by the sources or their occurrence state in the atmosphere (i.e., gaseous and particulate phases). Laluraj et al. (2010) proposed that the correlation between nssSO_4^{2-} vs. NO_3^- in ice ($R^2 = 0.31$, $p < 0.01$) could be associated with the fine nssSO_4^{2-} aerosols, which could provide nucleation centers forming the multi-ion complexes with HNO_3 in the atmosphere. This assertion, however, should be examined further, considering that the complex chemistry of SO_4^{2-} and NO_3^- in the atmosphere is far from understood (e.g., Wolff, 1995; Brown et al., 2006). Thus far, the mechanism of nssSO_4^{2-} influencing NO_3^- in the snowpack, however, is still debated, and it cannot be ruled out that nssSO_4^{2-} further affects mobilization of NO_3^- during and/or after crystallization (Legrand and Kirchner, 1990; Wolff, 1995; R  hlisberger et al., 2000). It is noted that no relationship was found between nssSO_4^{2-} and NO_3^- in inland snow (Fig. 7d), possibly due to the strong alteration of NO_3^-

during post-depositional processes, as discussed in section 4.1.2.

Please see the revised version of the manuscript, section 4.2 Effects of coexisting ions on NO_3^- .

(9) comments from Referees

Other comments:

Information on the chemistry of ice crystal are rather rare, so may important to develop this aspect in the revised manuscript (showing the full chemical composition and its comparison with snow).

Did you have measured MSA ?

I think you can say that nssCl^- is HCl and it can be interesting to compare with gas phase HNO_3 .

End of the review.

(9) author's response

Thanks to the reviewer for this suggestion. As the reviewer mentioned, the information on the crystal ice samples on Antarctic plateaus remain limited. So, showing the full chemical composition of the crystal ice can provide important information on snow chemistry in Antarctica. In addition, a comparison of chemical ion concentrations between surface snow and crystal ice was made in the revised manuscript.

Unfortunately, we did not measure the concentrations of MSA now. But we will measure the MSA concentrations in the samples of surface snow/snowpits. Possibly it will be another paper focusing on the biogenic sulfur (nssSO_4^{2-} and MSA).

Yes, the nssCl^- can be taken as HCl. This point was re-discussed in the manuscript.

(9) author's changes in manuscript

Following the reviewer's comment, a figure was included in the supporting information, as follows,

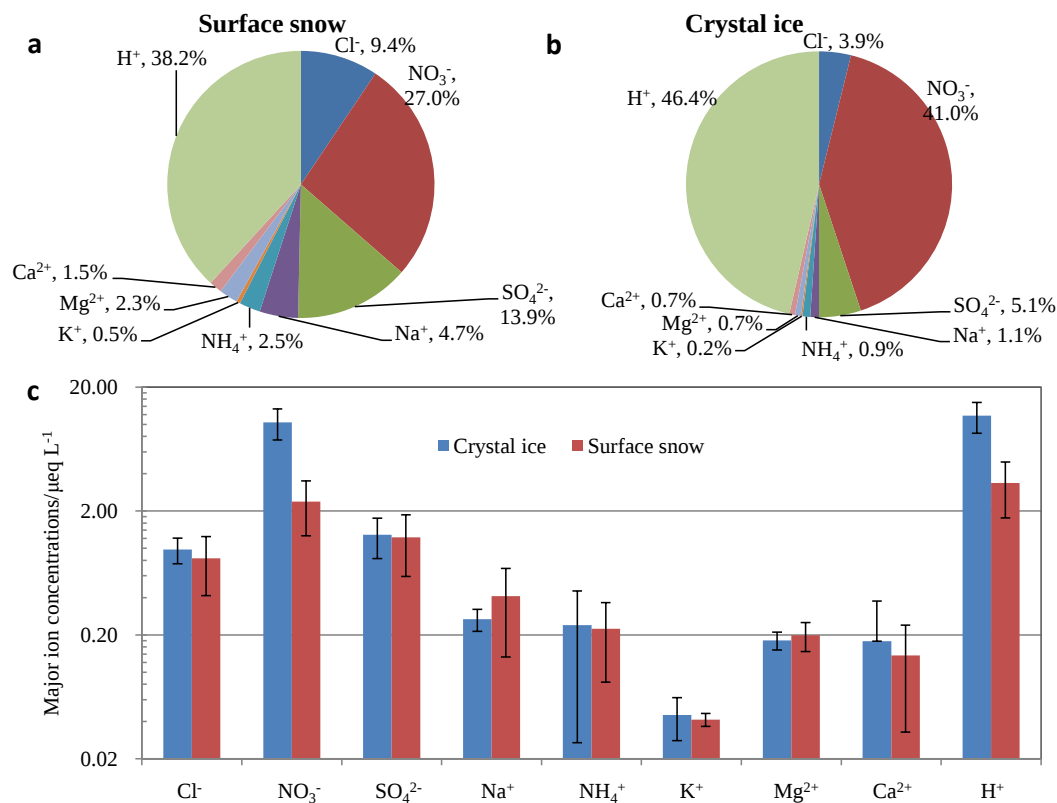


Figure S2 Major chemical ions in surface snow and crystal ice samples on the traverse from coast to the ice sheet summit (Dome A) in East Antarctica. Contribution percentages of each ion to total ion concentrations are shown in (a) and (b), respectively. Concentrations of ions in surface snow and crystal ice are shown in (c), with error bars of one standard deviation (1σ). The concentration of H⁺ is calculated from the difference between sum anions and sum cations. Note that a base-10 log scale is used for ion concentrations in (c).

In addition, the major chemical ion concentrations and a comparison between surface snow and crystal ice was included in the updated version of the manuscript (**3.1 NO₃⁻ concentration in surface snow**), as follows,

In the crystal ice, the means (ranges) of Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺, Ca²⁺ and H⁺ concentrations are 0.98 (0.62 – 1.27), 10.40 (8.35 – 16.06), 1.29 (0.87 – 2.13), 0.27 (0.21 – 0.33), 0.24 (0.03 – 0.56), 0.05 (0.03 – 0.08), 0.18 (0.15 – 0.22), 0.18 (0.05 – 0.57) and 11.75 (9.56 – 18.12) μeq L⁻¹, respectively. H⁺ and NO₃⁻ are the most abundant species, accounting for 46.4 and 41.0 % of the total ions, followed by SO₄²⁻ (5.1 %) and Cl⁻ (3.9 %). The other 5 cations, Na⁺, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺, only represent 3.6 % of the total ion budget. A significant linear relationship was found between NO₃⁻ and the total ionic strength ($R^2 = 0.99$, $p < 0.01$), possibly suggesting that NO₃⁻ is the species controlling ion abundance by influencing acidity of the crystal ice (i.e., H⁺ levels). In comparison with surface snow, concentrations of H⁺ and NO₃⁻ are significantly higher in crystal ice (Independent Samples T Test, $p < 0.01$), while concentrations of Cl⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺ are comparable in the two types of snow samples (Fig. S2 in supporting

information). To date, the information on the chemistry of ice crystal is rather limited but data from the so-called skin layer at Dome C, where NO_3^- concentrations in the top 0.4 cm snow layer are in the range of 9 – 22 $\mu\text{eq L}^{-1}$ in summertime (Erbland et al., 2013), generally comparable to our observations.

In addition, the association between NO_3^- and the major chemical ions in crystal ice was re-discussed (**4.2 Effects of coexisting ions on NO_3^-**), as follows,

With regard to the crystal ice, no significant correlation was found between NO_3^- and the coexisting ions (e.g., Cl^- , Na^+ and SO_4^{2-}), possibly suggesting that these ions are generally less influential on NO_3^- in this uppermost thin layer, compared to the strong air-snow transfer process of NO_3^- (Erbland et al., 2013). It is noted that NO_3^- accounts for most of the calculated H^+ concentrations (81 - 97 %, mean = 89 %), and a strong linear relationship was found between them ($R^2 = 0.96$), suggesting that NO_3^- is mainly deposited as acid, HNO_3 , rather than in particulate form as salts (e.g., NaNO_3 and $\text{Ca}(\text{NO}_3)_2$). This deduction is in line with the observations at Dome C, where atmospheric NO_3^- was found to be mainly in gaseous phase (HNO_3) in summer (Legrand et al., 2017b). On average, the deposition of HNO_3 contribute >91% of NO_3^- in the crystal ice (the lower limit, 91 %, calculated simply by assuming all of the alkaline species (Na^+ , NH_4^+ , K^+ , Mg^{2+} and Ca^{2+}) neutralized by HNO_3 in the atmosphere), suggesting a dominant role of HNO_3 deposition in snow NO_3^- levels. The elevated high atmospheric NO_3^- concentrations observed at Dome A (>100 ng m^{-3} ; 77.12°E, 80.42°S; Table S1 in supporting information) possibly indicate oxidation of gaseous NO_x to HNO_3 , suggesting that NO_3^- recycling driven by photolysis plays an important role in its abundance in snowpack on East Antarctic plateaus.

The relationship between nssCl^- (i.e., the HCl) and NO_3^- in snow was re-discussed in the revised manuscript, please see the revised manuscript, **4.2 Effects of coexisting ions on NO_3^-**

End of responses to Referee #2.

References

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End of the responses.