Prof. Joel Savarino

We thank Prof. Savarino very much for his careful and thoughtful review of our work. Please see below for point-by-point responses in blue following Prof. Savarino’s comments, 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

The paper needs major revisions before being accepted. The authors should better present their data in light of recent and past publications. Many important works are not referenced and it seems difficult to follow the conclusions (mainly part 4) of the authors based on only snow concentrations when other publications measuring all aspects of atmospheric parameters struggle to conclude on the fate of nitrate, its origin, formation, transport deposition and post deposition.

(1) author's response

We agree with Prof. Savarino. Some recent/past publications were not referenced in previous version. Following the comments from Prof. Savarino and two anonymous referees, the references were updated.

The discussion section (part 4) was substantially revised following the comments/suggestions, with the aid of publications on the atmospheric parameters. In addition, our recent measurements of atmospheric NO_3^- were included in the discussion (see responses to Referee 1).

(1) author's changes in manuscript

The references were updated.

The discussion part was revised

Please see the revision-tracked version of manuscript.

(2) comments from Referees

Reference to work suggesting an extraterrestrial source of nitrate in ice has been repeatedly dismissed (1-3 just for the most recent publications). Clearly state this fact or remove any reference to those works. 1-Wolff, E. W., Jones, A. E., Bauguitte, S. J.-B., and Salmon, R. A.:

Reassessment of the factors controlling temporal profiles of nitrate in polar ice cores using evidence from snow and atmospheric measurements, Atmospheric Chemistry and Physics Discussion, 8, 11039-11062, 2008. 2-Wolff, E. W., Bigler, M., Curran, M. A. J., Dibb, J. E., Frey, M. M., Legrand, M., and McConnell, J. R.: The Carrington event not observed in most ice core nitrate records, Geophys. Res. Lett., 39, L08503, 10.1029/2012gl051603, 2012. 3-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.: Nitrate ion spikes in ice cores not suitable as proxies for solar proton events, Journal of Geophysical Research: Atmospheres, n/a-n/a, 10.1002/2015JD023805, 2016.

(2) author's response

Thanks for this point. The references were now included in the revised manuscript.

(2) author's changes in manuscript

The recent works, both observations and model simulations (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 in the manuscript, as follows,

In addition, while some studies suggested that snow/ice NO_3^- is possibly linked with extraterrestrial fluxes of energetic particles and solar irradiation, with solar flares corresponding to NO_3^- spikes (Zeller et al., 1986; Traversi et al., 2012), other 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).

Please see the revision-tracked version of manuscript, section **1 Introduction**

(3) comments from Referees

Volatilization of nitrate. In Erbland et al. 2013 and Berhanu et al., 2014, 2015 (4-5) isotope fractionations demonstrate that vitalization is not an important loss process in contradiction with the authors statement (line 96). This should be clearly mentioned. What do you call post depositional effects beside photo-dissociation and volatilization? For me they are the post depositional effects. If you think there is more effects to take into accounts please, indicate which ones? 4- Berhanu, T. A., Meusinger, C., Erbland, J., Jost, R., Bhattacharya, S. K., Johnson, M. S., and Savarino, J.: Laboratory study of nitrate photolysis in Antarctic snow. II. Isotopic effects and wavelength dependence, The Journal of Chemical Physics, 140, 244305, 10.1063/1.4882899, 2014. 5- Berhanu, T. A., Savarino, J., Erbland, J., Vicars, W. C., Preunkert, S., Martins, J. F., and Johnson, M. S.: Isotopic effects of nitrate photochemistry in snow: a field study at Dome C, Antarctica, Atmos. Chem. Phys., 15, 11243-11256, 10.5194/acp-15-11243-2015, 2015.

(3) author's response

We agree with Prof. Savarino. The post-depositional effects refer to the two processes, photolysis and volatilization.

In comparison with photolysis, the extent and isotopic effects of NO_3^- volatilization remains poorly understood. Although several laboratory and field experiments have been conducted to examine the volatilization effects, the outcomes seem to vary remarkably among different experiments. Freshly-falling snow in Hanover, New Hampshire was used for examining NO_3^- loss with sublimation at -5 ^oC under controlled laboratory conditions, and NO_3^- loss was found to be negligible after a few days (Cragin and McGilvary, 1995). Similarly, a field experiment conducted on a subtropical glacier also showed that no significant loss of NO_3^- occurs over the course of one month sublimation (with temperature near zero; Ginot et al., 2001). The negligible NO_3^- loss during these experiments could be associated with that deposition of NO_3^- is mainly in particulate form rather than in the form of HNO_3 in the experimental snow. In a laboratory experiment, no detectable NO_3^- loss from the surface of frozen $NaNO_3$ solution acidified to pH=4 at -6^{o}C, and the high dissociation constant of HNO_3 was possibly a main reason (Sato et al., 2008; Riikonen et al., 2014). The wind-blown snow collected from Dome C was exposed to a flow of N_2 for one week in the dark at about -30 ^oC, no NO_3^- loss was detected, consequently the isotopic composition of NO_3^- is relatively constant during the sublimation process (Berhanu et al., 2014). However, the field experiment conducted at Dome C showed 17% (-30 ^oC) to 67% (-10 ^oC) of NO_3^- lost after 14-day sublimation (Erbland et al., 2013). Further investigations are needed to quantify the effects of volatilization for a better understanding of NO_3^- preservation in the snow/ice.

(3) author's changes in manuscript

Following the comments, we re-phrased these sentences, as follows, and also noted that volatilization might be important at warmer temperatures,

The effects of volatilization of NO_3^- are uncertain, given that one field experiment suggests that this process is an active player in NO_3^- loss (17 % (-30 ^oC) to 67 % (-10 ^oC) of NO_3^- lost after two weeks′ physical release experiments; Erbland et al., 2013), while other laboratory and field studies show that volatilization plays a negligible role in NO_3^- loss (Berhanu et al., 2014; Berhanu et al., 2015). Further investigations are needed to quantify the effects of volatilization for a better understanding of NO_3^- preservation in the snow/ice.

Please see the revision-tracked version of manuscript, section **1 Introduction**

(4) comments from Referees

Please also consider this publication for your introduction Bock, J., Savarino, J., and Picard, G.: Air–snow exchange of nitrate: a modelling approach to investigate physicochemical processes in surface snow at Dome C, Antarctica, Atmos. Chem. Phys., 16, 12531-12550, 10.5194/acp-16-12531-2016, 2016

(4) author's response

The very recent modeling work performed by Bock et al. (2016) suggest that co-condensation is the most important process to explain NO_3^- incorporation in snow undergoing temperature gradient metamorphism. The observed summer NO_3^- peaks in surface snow can be explained by this process.

(4) author's changes in manuscript

This reference was included in the revised manuscript, as follows,

However, snow physical characteristics play a crucial role in NO_3^- deposition and preservation. For instance, summertime concentrations in the surface skin layer of snow (the uppermost ~4 mm) can be explained as the result of co-condensation of HNO_3 and water vapour, with little to no photolytic loss in this microlayer (Bock et al., 2016). The combination of concentration and isotopic studies, along with physical aspects of the snow, could lead to the reconstruction and interpretation of atmospheric NO_3^- over time (e.g., Erbland et al., 2015; Bock et al., 2016), if there is detailed understanding of the NO_3^- deposition and preservation in different environments in Antarctica.

Please see the revision-tracked version of manuscript, section 1 Introduction

(5) comments from Referees

Acidity calculation is wrong. H+ = Σanions - Σcations, the equation used is a simplifi- cation and do not for instance takes into account ammonium ions.

(5) author's response

As Prof. Savarino suggested, the formula in the previous version ($[H^+] = [SO_4^{2-}] - 0.12 \times [Na^+] + [NO_3^-] + [Cl^-] - 1.17 \times [Na^+]$, Eq. 1; Legrand and Delmas, 1988) do not consider the effects of ammonium ions. In this case, the calculated H^+ concentrations were potentially over-estimated. In the updated version H^+ concentration is calculated through ion balance, i.e., $[H^+] = [Cl^-] + [NO_3^-] + [SO_4^{2-}] - [Na^+] - [NH_4^+] - [Mg^{2+}] - [Ca^{2+}]$ (Eq. 2), where ion concentrations are in μeq L^{-1}. Concentrations of H^+ calculated from the two methods are as follows,

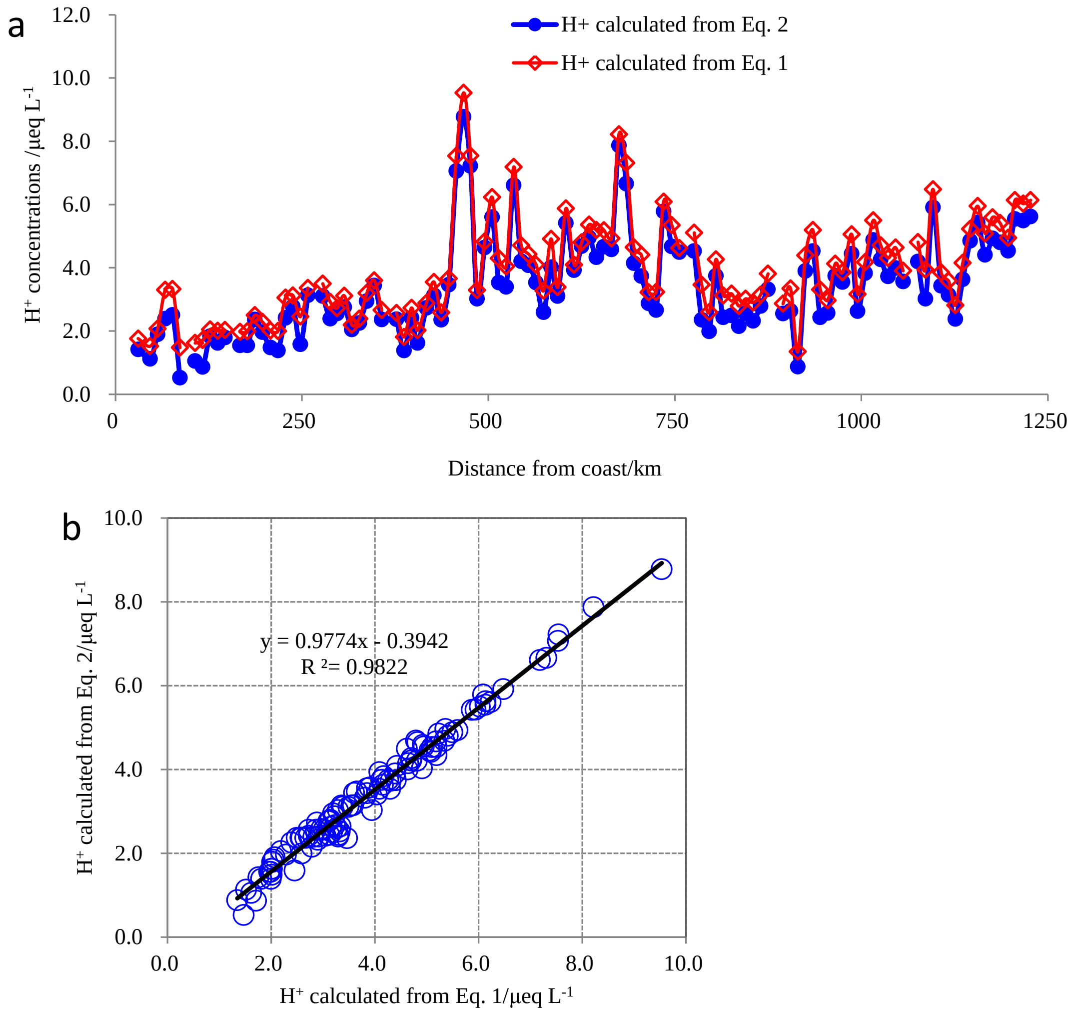

Figure Concentrations of H^+ in surface snow calculated from the two methods (a, upper panel) and the relationship between H^+ levels from the two calculations (b, bottom panel).

In general, H^+ concentrations from the two calculations are generally very close due to the relatively low concentrations of NH_4^+ in Antarctic snow. On average, the difference between the two calculations is <10%. In the revised manuscript, all of the H^+ data was calculated through Eq. 2, following Prof. Savarino' s suggestion.

(5) author's changes in manuscript

Revised, as follows,

For Antarctic snow samples, the concentrations of H^+ are usually not measured directly, but deduced from the ion-balance disequilibrium in the snow. Here, H^+ concentration is calculated through ion balance.

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

where ion concentrations are in μeq L^{-1}.

Please see the revised manuscript, section **2.3 Sample analysis.**

(6) comments from Referees

Cv is not defined (line 206)

(6) author's response

Cv, the Coefficient of Variation.

(6) author's changes in manuscript

Defined in the manuscript.

(7) comments from Referees

Erbland 2013 sampled many snow pits at a higher resolution than Frey 2009 (line 231). It is this reference that should be used and cited here.

(7) author's response

Yes, Erbland et al. (2013) sampled 17 snowpits at a higher resolution on the traverse from DDU to Dome C than Frey et al. (2009). In the work of Frey et al. (2009), only the top ~10 cm of snow (called ‘surface snow’) was sampled on that traverse (in total, 15 samples). In the section of 3.1 NO_3^- concentrations in surface snow, we only compare NO_3^- concentrations in the surface snow. Thus, we cited the work of Frey et al. (2009) in the previous version. We also cited the work of Erbland et al. (2013) in the revised version.

(7) author's changes in manuscript

The work of Erbland et al. (2013) was included in the revised manuscript.

(8) comments from Referees

Line 257 replace "may be" by "as a result of post depositional processing" This is no doubt about that.

(8) author's response

Agree, thanks.

(8) author's changes in manuscript

Replaced.

(9) comments from Referees

Line 288 change proposed by demonstrated - Again isotopes of nitrate have demonstrated the correctness of this assertion.

(9) author's response

Agree. Thanks.

(9) author's changes in manuscript

Changed.

(10) comments from Referees

line 291: Please add France 2011 reference, the first publication to have measured the optical depth of the snow pack in the UV range, years before Zatko France, J. L., King, M. D., Frey, M. M., Erbland, J., Picard, G., Preunkert, S., MacArthur, A., and Savarino, J.: Snow optical properties at Dome C (Concordia), Antarctica; implications for snow emissions and snow chemistry of reactive nitrogen, Atmos. Chem. Phys., 11, 9787-9801, 10.5194/acp-11-9787-2011, 2011.

(10) author's response

Yes, France et al. (2011) reported the *e*-folding depth (z_e), where the actinic flux is reduced to 37 % (i.e. 1/*e*) of the surface value, of 10 to 20 cm at Dome C on the East Antarctic plateau, while Zatko et al. (2013) calculated the *e*-folding depth at different sites in Antarctica.

(10) author's changes in manuscript

The reference France et al. (2011) was added.

(11) comments from Referees

line 293 The idea that below the photic zone, nitrate is archived without further modification is an idea developed in Frey 2009, Erbland 2013 and 2015. This should be recognized.

(11) author's response

Agree.

(11) author's changes in manuscript

The statement was re-phrased, and the references were included in the manuscript, as follows,

In this case, NO_3^- in the bottom snowpit, i.e., below the photic zone, can be taken as the archived fraction without further modification on the basis of previous observations (Frey et al., 2009; Erbland et al., 2013; Erbland et al., 2015).

(12) comments from Referees

line 306: Change dry deposition by apparent dry deposition. See Bock et al. but also the second reviewer's comments.

(12) author's response

Agree. Please also see the responses to the Referee#2.

(12) author's changes in manuscript

Changed in the revised version. This section was substantially revised following the comments from Prof. Savarino and the two anonymous referees, please see the revised manuscript, sections **4.1.1 NO_3^- in coastal snowpack** and **4.1.2 NO_3^- in inland snowpack**

(13) comments from Referees

line 320: it is not the strong correlation between deposition flux and accumulation that makes wet deposition to dominate but the comparison between "dry" and wet fluxes (see your eq 5). The fact that a correlation exists only means that the scavenging ratio of atmospheric nitrate by snowfall is constant or in other words the concentration in snow fall is independent of the snow accumulation (see your equation 5).

(13) author's response

Agree. Thanks for pointing this out.

(13) author's changes in manuscript

This statement was re-phrased in the revised manuscript.

(14) comments from Referees

line 331: K2 is not dimensionless as it allows to convert atmospheric concentration (mass/volume) to snow concentration (mass/mass), it has a unit of m3/g. How K2 is calculated? According to Eq5, K2 x Catm = Cf-snow, so K2 = 43/20 = 2.1 meaning that 1 g of snow scavenged 2 m3 of air. Also note that eq 5 & 6 is nothing else than your eq4. These models are not different models but the same, expressed in different way. It is thus not surprising to find the same dry deposition flux. Comment your dry deposition with respect to previous publication (eg Pasteris 2014)

(14) author's response

Agree, and thanks for this comment. Because the unit of NO_3^- flux, F_{total}, is μeq m^{-2} a^{-1}, the unit of $K_2C_{atm}A$ should be μeq m^{-2} a^{-1}. Considering that the units of C_{atm} and A are μeq m^{-3} and kg m^{-2} a^{-1}, respectively, the unit of K_2 should be m^3 kg^{-1}. Following the linear relationship between NO_3^- flux and snow accumulation rate (A), i.e., $F_{total} = K_1C_{atm} + K_2C_{atm}A$, the slope of the linear fit, K_2C_{atm}, is 0.6 (y = 0.6x + 50.3). The atmospheric NO_3^- concentration on the coast was observed to be 19.4 ng m^{-3} (i.e., 0.3×10^{-3} μeq m^{-3}), then K_2 is estimated to be $\sim 0.2 \times 10^4$ m^3 kg^{-1}, i.e., about 2 m^3 g^{-1}.

Yes, the equations 5 and 6 are the same with equation 4 in previous version, i.e., not different models. This section was substantially revised in the manuscript.

The apparent dry deposition flux is compared to previous observations in Dronning Maud Land (DML) region and at Kohnen Station, where the negative dry deposition flux suggests a net loss of NO_3^- (Weller et al., 2004; Weller and Wagenbach, 2007; Pasteris et al., 2014).

(14) author's changes in manuscript

Following the comments from Prof. Savarino and Referee#1, the models were consolidated. Accordingly, this discussion was revised. Please see the revision-tracked version of the manuscript, sections **4.1.1 NO_3^- in coastal snowpack** and **4.1.2 NO_3^- in inland snowpack**.

(15) comments from Referees

line 342: give the reference for the deposition velocity at South Pole.

(15) author's response

Thanks for this point.

(15) author's changes in manuscript

The following reference was added,
Huey, L.G., Tanner, D.J., Slusher, D.L., Dibb, J.E., Arimoto, R., Chen, G., Davis, D., Buhr, M.P., Nowak, J.B., Mauldin Iii, R.L., Eisele, F.L., and Kosciuch, E.: CIMS measurements of HNO_3 and SO_2 at the South Pole during ISCAT 2000, Atmos. Environ., 38, 5411-5421, doi:10.1016/j.atmosenv.2004.04.037, 2004.

(16) comments from Referees

Line 347: K2 in eq7 cannot be equal to K2 in eq5. K2 in eq5 takes implicitly into account , the density of air, as K2/ = K in eq7, unless I have missed something

(16) author's response

We thank Prof. Savarino very much for pointing this out, and we are sorry for the confusion about the scavenging ratio for NO_3^- in the previous version. K_2 is the scavenging ratio for precipitation (m^3 kg^{-1}), which allows to convert atmospheric concentration to snow concentration of NO_3^- in this study.

If it is assumed that NO_3^- concentration in snow is related to its concentration in the atmosphere, the scavenging ratio for NO_3^- (W) can be calculated on a mass basis from the following expression (Kasper-Giebl et al., 1999),

$$W = \rho_{atm} \times (C_{f\text{-}snow} / C_{atm}) \text{ (Eq. 7)},$$

where ρ_{atm} is air density (g m^{-3}), and $C_{f\text{-}snow}$ and C_{atm} are NO_3^- concentrations in fresh snow (ng g^{-1}) and atmosphere (ng m^{-3}) respectively. If taking $\rho_{atm} \approx 1000$ g m^{-3} (on average, ground surface temperature $t \approx 255$ k, ground pressure $P \approx 0.08$ MPa, in the coastal region), $C_{f\text{-}snow} = 43$ ng g^{-1} (see the main context), and $C_{atm} = 19.4$ ng m^{-3}, W is calculated to be ~2200, generally comparable to previous reports (Barrie, 1985; Kasper-Giebl et al., 1999; Shrestha et al., 2002). It is noted that the calculation here may be subject to uncertainty, due to the complex transfer of atmospheric NO_3^- into the snow. However, the scavenging ratio provides useful insights into the relation between NO_3^- concentrations in the atmosphere and snow and reference values for modeling NO_3^- deposition at large scale in Antarctica.

(16) author's changes in manuscript

K_2, the scavenging ratio for precipitation, and the scavenging ratio for NO_3^- (W) were clarified in the updated version. Please see the revision-tracked version of the manuscript, sections 4.1.1 NO_3^- in coastal snowpack

(17) comments from Referees

line 352: not sure these inferred parameters are better than concentration observations to provide useful reference values for modeling. These are macroscopic, apparent parameters that are unable to describe processes at microscopic scale. See Bock 2016.

(17) author's response

We agree that the calculations here are macroscopic and apparent parameters, which possibly cannot characterize the microscopic processes (e.g., co-condensation; Bock et al., 2016). However, they possibly can provides useful parameter values for modeling NO_3^- deposition at large scale in Antarctica. It is noted that previous modeling work of Zatko et al. (2016) do not include the microscopic processes (i.e., models at the regional to global scale).

(17) author's changes in manuscript

The statement was rephrased in the revised manuscript, as follows,

It is noted that the calculation here may be subject to uncertainty, due to the complex transfer of atmospheric NO_3^- into the snow. However, the scavenging ratio provides useful insights into the relation between NO_3^- concentrations in the atmosphere and snow, which might be useful in modeling NO_3^- deposition at large-scale.

Please see the revision-tracked version of the manuscript, sections **4.1.1 NO_3^- in coastal snowpack**

(18) comments from Referees

Fig5a and fig5b are in contradiction. The same parameter (p-concentration) cannot be linear with respect to a variable A and its reverse 1/A (same for fig5e & fig5f). I also found p-concentration not very expressive. Archived, deep concentration seems more appropriate.

(18) author's response

Thanks for pointing this out. For a direct comparison with previous investigations, only the relationship between archived concentration of NO_3^- and inverse snow accumulation rate was presented, for the observations both at inland and coastal sites.

Agree, and p-concentration was replaced with archived concentration (C_{archived}) in the revised version.

(18) author's changes in manuscript

Figures 5 was re-drawn, and only the correlation between archived concentration of NO_3^- and inverse snow accumulation rate was included, as follows,

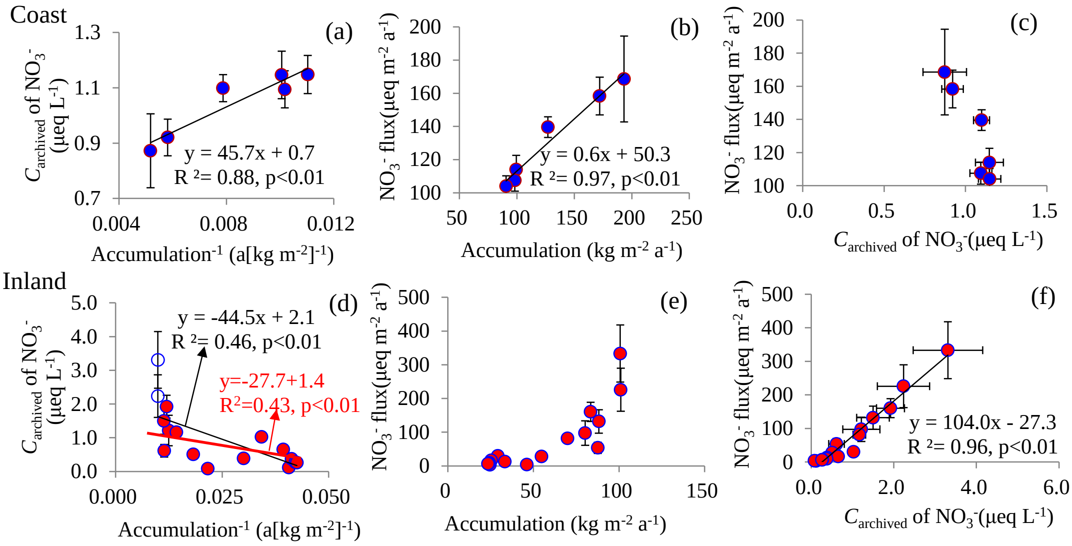

Figure 5. The relationship among snow accumulation rate, the archived concentration ($C_{archived}$), and flux of NO_3^- in coastal (top row, (a), (b) and (c)) and inland (bottom row, (d), (e) and (f)) Antarctica. In panel (d), the linear fit in back line (y = -44.5 + 2.1) include the full date set, while the linear equation in red (y = -7.7 + 1.5) was obtained by excluding two cases (open circles) with snow accumulation rate larger than 100 kg m^{-2} a^{-1} (see the main text). The flux values are the product of $C_{archived}$ of NO_3^- and snow accumulation rate, namely the archived flux. Least squares regressions are noted with solid lines and are significant at $p < 0.01$. Error bars represent one standard deviation (1σ).

The p-concentration was not used throughout the context, please see the revised version of the manuscript.

(19) comments from Referees

Why slope of fig5a & fig6b are so different if no nitrate is lost in coastal region ? In general, Cfirn, Cp-concentration, Cf-snow are poorly labeled on figures (why not using the same as Pasteris 2014), why in fig6 f-snow label is not used, same for fig4? This makes the reading of the figures very confusing.

(19) author's response

Now, we can make a comparison between the two figures (Figures 5a versus 6a), as follows,

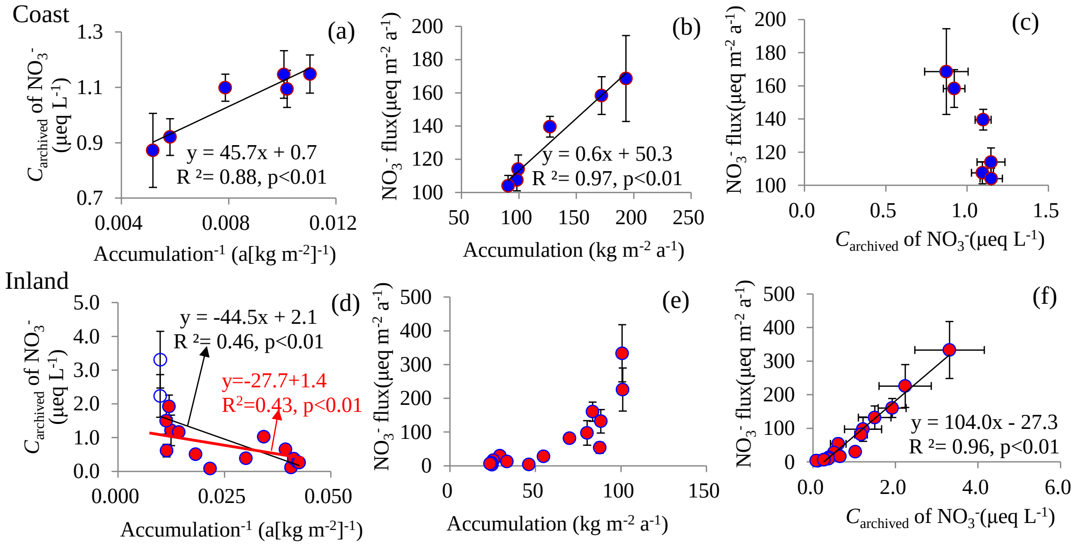

Figure 5. The relationship among snow accumulation rate, the archived concentration ($C_{archived}$), and flux of NO_3^- in coastal (top row, (a), (b) and (c)) and inland (bottom row, (d), (e) and (f)) Antarctica. In panel (d), the linear fit in back line (y = -44.5 + 2.1) include the full date set, while the linear equation in red (y = -7.7 + 1.5) was obtained by excluding two cases (open circles) with snow accumulation rate larger than 100 kg m^{-2} a^{-1} (see the main text). The flux values are the product of $C_{archived}$ of NO_3^- and snow accumulation rate, namely the archived flux. Least squares regressions are noted with solid lines and are significant at $p < 0.01$. Error bars represent one standard deviation (1σ).

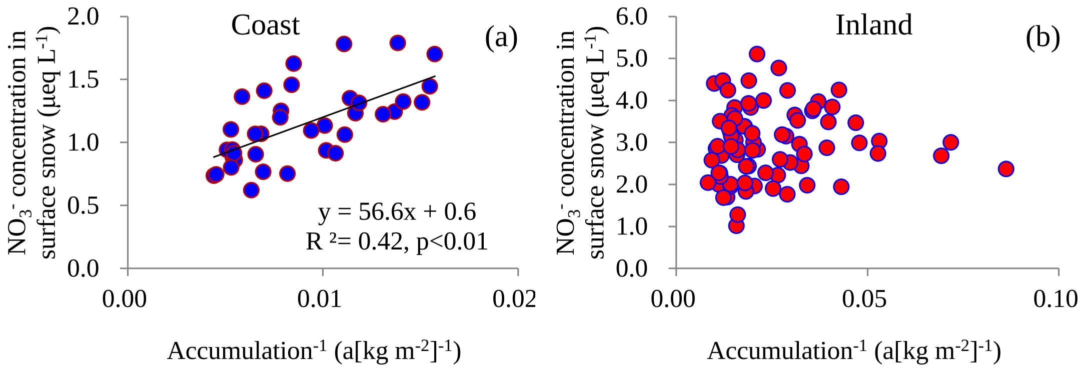

Figure 6. The relationship between NO_3^- concentration and inverse snow accumulation rate in surface snow in coast (panel (a)) and inland (panel (b)) Antarctica. Least squares regressions are noted with solid line and are significant at $p < 0.01$.

In terms of surface snow on the coast, NO_3^- may be disturbed by the katabatic winds and wind convergence located near the Amery Ice Shelf (that is, the snow-sourced NO_x and NO_3^- from Antarctic plateau possibly contribute to coastal snow NO_3^-) (Parish and Bromwich, 2007; Ma et al., 2010). In addition, the sampled ~3 cm surface layer roughly corresponds to the net accumulation in the past 0.5-1.5 months assuming an even distribution of snow accumulation in the course of a single year. This difference in exposure time of the surface snow at different sampling sites, could possibly affect the concentration of NO_3^-, although the post-depositional alteration of NO_3^- was thought to be minor on the coast (Wolff et al., 2008; Erbland et al., 2013; Shi et al., 2015). Taken together, NO_3^- in coastal surface snow might represent some post-depositional alteration. Even so, a negative correlation between NO_3^- concentration and snow accumulation rate was found at the coast (R^2=0.42, p<0.01; Fig. 6a). It is noted that the parameters obtained from Figure 6a (y = 56.6x + 0.6) are generally comparable to those of the coastal snowpits (y = 45.7x + 0.7; Figure 5a), and the small difference could be associated with the influences discussed above. Both the snowpit and surface snow observations suggest that overall the majority of the NO_3^- appears to be preserved and is driven by snow accumulation on the coast.

(19) author's changes in manuscript

All of the figures (Figures 4, 5 and 6) were labeled clearly following the comments, please see the revised manuscript.

(20) comments from Referees

line 381: replace snow accumulation by inverse snow accumulation. Also please comment the difference of nitrate flux loss between you (-73.9 ueq m-2 a-1) and Pasteris 2014 (-22 ueq m-2 a-1), as well as for the slope, 2.7 vs 1.1 when accumulation rates cover the same range.

(20) author's response

Thanks for this comment. A comparison between our observations and previous reports of Pasteris et al. (2014) and Weller and Wagenbach (2007) was made.

(20) author's changes in manuscript

Following Prof. Savarino's comments and suggestion, this paragraph was re-written, as follows,

In comparison with the coast, the correlation between C_{archived} and inverse snow accumulation is relatively weak in inland regions (Fig. 5d), suggesting more variable conditions in ambient concentrations and dry deposition flux of NO_3^-. In addition, the relationship of C_{archived} vs. inverse accumulation in inland is opposite to that of coast. Based on current understanding of the post-depositional processing of NO_3^-, the negative correlation between C_{archived} and inverse snow accumulation (Fig. 5d) suggests losses of NO_3^-.The slope of the linear relationship indicates apparent NO_3^- dry deposition flux of -44.5±13.0 μeq m^{-2} a^{-1}, much larger than that of DML (-22.0±2.8 μeq m^{-2} a^{-1}), where the snow accumulation is generally lower than 100 kg m^{-2} a^{-1} (Pasteris et al., 2014). At Kohnen Station (an inland site in East Antarctica), with snow accumulation of 71 kg m^{-2} a^{-1}, the emission flux of NO_3^- is estimated to be -22.9±13.7 μeq m^{-2} a^{-1} (Weller and Wagenbach, 2007), which is also smaller in comparison with this observation. Weller et al. (2004) proposed that loss rate of NO_3^- does not depend on snow accumulation rate and the losses become insignificant at accumulation rates above 100 kg m^{-2} a^{-1}. Among the inland sites, SP10 and Core2 (~800 km from the coast), featured by high snow accumulation rate (> 100 kg m^{-2} a^{-1}; Table 1 and Fig. 1), exhibit even higher values of C_{archived} and archived fluxes of NO_3^- than those of the coastal sites. It is noted that the two cases influence the linear regression significantly (Fig. 5d). If the two sites are excluded, we can get a linear regression with the slope of -27.7±9.2 μeq m^{-2} a^{-1}, which is comparable to previous reports in DML (Pasteris et al., 2014).

Please see the revision-tracked version of the manuscript, first paragraph in section **4.1.2 NO_3^- in inland snowpack.**

(21) comments from Referees

Figure 6h: There is something difficult to understand and seems to be a circular reasoning in fig6. Since Flux = snow concentration x snow accumulation, and only concentration and accumulation are measured, how fig6g and 6h can produce both a linear trend. In fig6g, slope gives snow concentration, the linear trend then suggests a constant homogeneous snow concentration in fresh snow. Slope of fig6h gives a constant homogeneous accumulation (in clear contradiction with measurements), well if accumulation is constant and snow concentration is constant, how the flux can vary? (same observation for fig5) Your conclusion that accumulation is not the main driver of the preserved nitrate (line 387) contradicts fig6g and the linear trend plotted. I will suggest to remove the linear trend of fig6g, which obviously looks like more exponential than linear.

(21) author's response

Agree, thanks for the comment. In the previous version of the manuscript, data in Figure 6g (snow accumulation rate versus archived NO_3^- flux) can be better depicted by an exponential regression, instead of a linear model, while the strong linear relationship between the archived NO_3^- concentration and flux suggest that accumulation rate is not the main driver of the preserved NO_3^- concentration. In this case, the linear fit was removed and Figure 6 was therefore re-drawn.

(21) author's changes in manuscript

Following the comments, Figure 6 was redrawn, and accordingly the discussion was re-made. Please see the revised manuscript, section **4.1.2 NO_3^- in inland snowpack.**

(22) comments from Referees

line 403: in reference add Erbland 2013, France, 2011

(22) author's response

Agree, added.

(22) author's changes in manuscript

The references were added in the revised manuscript.

(23) comments from Referees

line 405: add Davis et al., 2004 reference
Davis, D., Chen, G., Buhr, M., Crawford, J., Lenschow, D., Lefer, B., Shetter, R., Eisele, F., Mauldin, L., and Hogan, A.: South Pole NOx Chemistry: an assessment of factors controlling variability and absolute levels, Atmos. Environ., 38, 5375-5388, 10.1016/j.atmosenv.2004.04.039, 2004.

(23) author's response

Added.

(23) author's changes in manuscript

Added. Please see the revised manuscript.

(24) comments from Referees

line 413: do you mean fig6e, f instead of 6c & d ?

(24) author's response

In previous version, no significant correlation was found in Figures 6 c and d. Also, the relationship was not so strong (Figure 5f) compared to the correlation for coast (Figure 5b). Figure 6 was redrawn in the revised manuscript.

(24) author's changes in manuscript

Please see the revised manuscript Figure 6 and the main text.

(25) comments from Referees

The part4 needs to be revisited in light of the references given by reviewer 2. There are many misconceptions. The first is that a correlation does not imply a causal effect. nitrate and sulfate summer peaks may have completely unconnected reasons (max photo-denitrification and max marine emission respectively followed by dry and wet depositions). Nitrate aerosols are not on the same aerosols size bin than sulfuric acid (Jourdain and Legrand, 2002). Even in heavily sea salt impacted coastal sites, half of the nitrate is in acid form and rapidly goes to almost 100% inland. There are no reasonable observations to support the conversion of NOx to nitrate by sulfate aerosols (in addition than N2O5 does not exist in summer), neither than nitrate is internally mixed with sulfate aerosols. Jourdain, B., and Legrand, M.: Year-round records of bulk and size-segregated aerosol composition and HCl and HNO3 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, 10.1029/2002jd002471, 2002.

(25) author's response

We agree that a correlation between NO_3^- and the co-existing impurities does not necessarily suggest a causal link. In the surface snow on the traverse from coast to the ice sheet summit, Dome A, non sea salt fraction account for 75 - 99 % of total SO_4^{2-}, with a mean of 95 %, suggesting a dominant source from ocean bioactivities. The percentages are relatively higher in inland regions than at coastal sites. Field observations show 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). But this similar seasonal pattern of the two species is unlikely associated with the 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 addition, 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). Laluraj et al. (2010) found a close correlation between $nssSO_4^{2-}$ vs. NO_3^- in ice ($R^2 = 0.31$, $p<0.01$), and they attributed the relationship to the fine $nssSO_4^{2-}$ aerosols, which have long residence time in the atmosphere (Hara et al., 2014) and could provide nucleation centers forming the multi-ion complexes with HNO_3 in the atmosphere. It is acknowledged that this proposal should be examined further, considering that the complex chemistry of SO_4^{2-} and NO_3^- in the atmospheric is far from understood (e.g., Wolff, 1995; Brown et al., 2006).

In addition, 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; Röthlisberger et al., 2000).

(25) author's changes in manuscript

Following the comments and suggestions from Prof. Savarino and Reviewer#2, this section was re-organized, 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; Röthlisberger 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.

End of responses to Prof. Savarino.

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End of the responses.