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Potential genesis and implications of calcium nitrate in Antarctic snow

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Abstract

Among the large variety of particulates in the atmosphere, calcic mineral dust particles have highly reactive surfaces that undergo heterogeneous reactions with nitrogen oxides contiguously. The association between Ca^{2+}, an important proxy indicator of mineral dust and NO_3^-, a dominant anion in the Antarctic snow pack was analysed. A total of 41 snow cores (~ 1 m each) that represent snow deposited during 2008–2009 were studied along coastal–inland transects from two different regions – the Princess Elizabeth Land (PEL) and central Dronning Maud Land (cDML) in East Antarctica. Correlation statistics showed a strong association (at 99 % significance level) between NO_3^- and Ca^{2+} at the near-coastal sections of both PEL ($r = 0.72$) and cDML ($r = 0.76$) transects. Similarly, a strong association between these ions was also observed in snow deposits at the inland sections of PEL ($r = 0.8$) and cDML ($r = 0.85$). Such systematic associations between Ca^{2+} and NO_3^- is attributed to the interaction between calcic mineral dust and nitrogen oxides in the atmosphere, leading to the possible formation of calcium nitrate ($Ca(NO_3)_2$). Forward and back trajectory analyses using HYSPLIT model v. 4 revealed that Southern South America (SSA) was an important dust emitting source to the study region, aided by the westerlies. Particle size distribution showed that over 90 % of the dust was in the range < 4 µm, indicating that these dust particles reached the Antarctic region via long range transport from the SSA region. We propose that the association between Ca^{2+} and NO_3^- occurs during the long range transport due to the formation of $Ca(NO_3)_2$. The $Ca(NO_3)_2$ thus formed in the atmosphere undergo deposition over Antarctica under the influence of anticyclonic polar easterlies. However, influence of local dust sources from the nunataks in cDML evidently mask such association in the mountainous region. The study indicates that the input of dust-bound NO_3^- may contribute a significant fraction of the total NO_3^- deposited in Antarctic snow.

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1 Introduction

Geographically, the Antarctic ice sheet may be remote, its atmosphere however, is linked with the rest of the planet. Fine aerosols from surrounding continents are transported thousands of kilometers and teleconnect with the Antarctic atmosphere. Consequently, the chemistry of the Antarctic atmosphere and its ice sheet is of considerable importance. Apart from sea-salt aerosols, nitrogen oxides (like NO_x, NO_3^-, HNO_3 and N_2O_5) and mineral dust particles make up a significant component in the Antarctic atmosphere (Weller et al., 2002; Albani et al., 2012; Wolff, 2013). Nitrate is ubiquitous in the atmosphere and constitutes a large portion of anions in the Antarctic snow. Several studies have focused on its origin (e.g. Wolff, 1995; Wagenbach et al., 1998; Traversi et al., 2014), seasonal and spatial variations (e.g. Dahe et al., 1992; Lee et al., 2014; Savarino et al., 2006), air–snow interactions (e.g. Antony et al., 2010; Erbland et al., 2013) and its post-depositional processes (e.g. Mulvaney et al., 1998; Wagnon et al., 1999; Weller, 2004). However, there are many uncertainties and speculations over the sources of nitrate, its seasonality and post-depositional losses (e.g. Traversi et al., 2014; Shi et al., 2015).

Natural mineral dust form an active aerosol component in the atmosphere and model simulations show that the mineral dust from surrounding continents are directly linked to the Antarctic climate system (e.g. Li et al., 2008; Gassó et al., 2010). These dust particles are evidently becoming an important factor in which they alter the chemical and optical properties of the atmosphere (Nousiainen and Kandler, 2015). The mineralogy of dust particles is of critical importance since it plays a major role in deciding the manner in which the dust particles alter the chemistry of the atmosphere (Krueger et al., 2004). Previous field studies and model simulations demonstrate that throughout the world, mineral dust is frequently associated with NO_3^- (e.g. Jordan et al., 2003; Fairlie et al., 2010). It was reported that almost all of total NO_3^- in high dust regions and > 40 % of the total NO_3^- throughout most of the global atmosphere is coupled with mineral dust (Usher et al., 2003). This association, depending on the mineralogical composition of

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the dust, yields various nitrate-salt products (Gibson et al., 2006). Studies also show that dust-associated NO_3^- is a large component of total atmospheric NO_3^- due to the presence of Ca^{2+} in mineral dust, which has a high buffering capacity to neutralise atmospheric nitric acid (HNO_3) to yield highly hygroscopic calcium nitrate ($Ca(NO_3)_2$) (Krueger et al., 2003).

In Antarctica, non-sea-salt Ca^{2+} ($nssCa^{2+} = Ca^{2+} - 0.038\ ssNa^+$) has been used as a proxy for mineral dust concentration in ice core records (Ruth et al., 2008; Schüpbach et al., 2013). Röthlisberger et al. (2000) have also documented strong associations between NO_3^- and Ca^{2+} throughout the last glacial maximum during which dust concentrations in the Antarctic atmosphere were up to 20 times that of the present day. However, such associations between NO_3^- and Ca^{2+} have not been widely reported in present day Antarctic snow pack. In this study, we analyse the spatial variability and association of NO_3^- and Ca^{2+} from two different regions in East Antarctica – Princess Elizabeth Land (PEL) and central Dronning Maud Land (cDML). We also discuss the possible formation processes of $Ca(NO_3)_2$ during the long range transport of mineral dust and its significance on Antarctic snow chemistry.

2 Sampling and methods

Forty one snow cores were recovered from the PEL and cDML transects using a KOVACS Mark IV coring system (14 cm diameter) during summer 2008–2009 (Fig. 1). Each snow core was about 1 m deep, enough to cover over a year of snow accumulation. Sampling was carried out in such a way that both transects were perpendicular to the coast and overall slope contours of the region. A total of 21 snow cores were retrieved from coast to inland in PEL covering a distance of 180 km and an elevation of 2100 m a.s.l. Similarly, in cDML, 20 snow cores were retrieved from near-coast to inland region covering a distance of 110–300 km from coast and an elevation up to 2800 m a.s.l. Due to the absence of fresh snow over the Nivlisen ice shelf and the edge of the ice cap, no cores were collected up to 110 km from the coast in cDML (Fig. 1).

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For the sake of clarity, the actual distance from sea (300 km) is mentioned in cDML throughout this paper. Based on the topography, the cDML transect is spatially divided into near-coastal (110–160 km), mountainous (170–240 km) and plateau (> 240 km) sections (Fig. 1).

The snow cores were transferred directly into pre-cleaned high density polyethylene bags and sealed immediately to avoid any contamination during storage and transport. The sample bags were kept in expanded poly propylene (EPP) boxes and were stored at −20 °C throughout the transit to the ice core laboratory of National Centre for Antarctic and Ocean Research in India. Each core was carefully unpacked and sub-sampled at 5 cm resolution under a laminar flow bench housed at −15 °C facility. The outer layers of these sub-samples were removed manually by ceramic knives and the innermost portion of the samples were transferred to pre-cleaned vials. These sub-samples were then melted in a Class 100 clean room prior to the analyses. Major ion concentrations were analysed using Dionex DX2500 with IonPac CS17 column for cations and ICS 2000 with IonPac AS11-HC column for anions. An excellent low detection limit was achieved for both Ca^{2+} (3 µg L^{-1}) and NO_3^- (5 µg L^{-1}) respectively. Reference standards and duplicate samples were analysed in a routine manner to estimate the analytical precision, which was better than 5 % for both the ions. Dust particle concentration and grain size measurements were carried out using a Multisizer 4 Coulter Counter (Beckman), placed in a Class 100 clean room. Size calibration was made using polystyrene latex beads of 5 µm radius and a precision better than 5 % was obtained.

The seasonality from these snow cores are determined as mentioned in Mahalinganathan et al. (2012). High-resolution $\delta^{18}O$ records of each core were studied to differentiate the summer and winter snow deposition. Annual layers were identified based on the seasonal differences of $\delta^{18}O$. A seasonal $\delta^{18}O$ amplitude of 4 ‰ was considered large enough to represent the temperature difference between summer and winter precipitation (Stenberg et al., 1998). A minimum seasonal amplitude of 6 ‰ in $\delta^{18}O$ values were observed in all cores (except cores from 110 km in cDML and from 70 and

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100 km in PEL), which was large enough to differentiate between summer and winter layers.

3 Results

Snow cores from both PEL and cDML clearly showed seasonal variations in $\delta^{18}O$ and the major ions (Fig. 2). The sea-salt (Na^+) ions peaked mostly during winter and were in sync with the $\delta^{18}O$ variations. Though Ca^{2+} and NO_3^- peaks were not in sync with the $\delta^{18}O$ or sea-salt signatures, their seasonal variations were well-marked in the snow cores (Fig. 2). The peak values of NO_3^- and Ca^{2+} were observed during early spring or summer, in agreement with previous studies (Wagenbach et al., 1998; Weller et al., 2011).

Concentrations of NO_3^- and Ca^{2+} ions showed a significant spatial variability along both PEL and cDML transects (Fig. 3). Nitrate concentrations in PEL ranged from 40 to 200 $\mu g\,L^{-1}$ with few samples as high as 350 $\mu g\,L^{-1}$. Similarly, NO_3^- in cDML transect ranged from 15 to 350 $\mu g\,L^{-1}$ with outliers as high as 450 $\mu g\,L^{-1}$. Nitrate did not follow any systematic trend either with changes in distance from the sea or with changes in elevation along both transects. In general, inland sites were characterized by relatively higher values.

Calcium concentrations, on one hand, were significantly lower than that of NO_3^- and ranged from 10 to 70 $\mu g\,L^{-1}$ with few outliers as high as 100 $\mu g\,L^{-1}$ in PEL (Fig. 3). On the other hand, Ca^{2+} concentrations in cDML transect ranged from 5 to 50 $\mu g\,L^{-1}$ except along the mountainous region, where it showed a substantial spread with concentrations up to 150 $\mu g\,L^{-1}$ and outliers as high as 230 $\mu g\,L^{-1}$. The mean Ca^{2+} concentrations were significantly high along the mountainous section in cDML transect. Calcium concentrations were very low in the plateau region of cDML with a narrow range (4–12 $\mu g\,L^{-1}$) irrespective of seasonal variations. Non-sea-salt calculations revealed that the majority of Ca^{2+} was derived from crustal sources in both coastal and

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inland regions of PEL (81 and 94 %, respectively). Similarly, at cDML transect, majority of Ca^{2+} in the mountainous section (96 %), near-coast (92 %) and the plateau (88 %) regions were also from crustal sources.

The relationship between NO_3^- and Ca^{2+} concentrations in snow cores from all sections of PEL and cDML transects were consistent except the mountainous section. Further, to verify the consistency, samples representing one complete year were also evaluated (Fig. 4). Within the near-coastal and the plateau regions of cDML, NO_3^- showed a significant association with Ca^{2+} ($r = 0.76$ and 0.85 respectively). Near the coast, a stronger association ($r = 0.89$) was observed in winter snow deposits while the samples from the plateau section showed a stronger association during the summer ($r = 0.93$). Compared to these, snow cores from the mountainous region in cDML did not reveal any correlation during winter and showed a comparatively weak correlation during summer ($r = 0.49$). In PEL transect, NO_3^- and Ca^{2+} showed a more significant relationship within the inland region ($r = 0.8$) compared to the coastal section ($r = 0.72$). All correlations were significant at 99 % confidence level.

4 Discussion

4.1 Association between Ca^{2+} and NO_3^- in snowpack from distinct transects

Nitrate is one of the major anions with multiple sources in the Antarctic environment. It is predominantly a secondary aerosol produced via oxidation of N_2O and photodissociation of N_2 in the upper atmosphere (Brasseur and Solomon, 1986) and also via sedimentation from polar stratospheric clouds (Legrand and Delmas, 1986; Legrand and Mayewski, 1997). Both stratospheric and lightning sources of NO_x are also thought to contribute to NO_3^- deposited in the Antarctic snow (Legrand and Delmas, 1986; Michalski et al., 2005). Furthermore, it is known that post-depositional processes and photochemical mechanisms also lead to NO_3^- loss in low accumulation sites (Röthlisberger et al., 2002; Erbland et al., 2013), resulting in a complex distribution of NO_3^- in

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the Antarctic snow pack. Due to the variety of its sources, NO_3^- distribution in modern day snow pack is found to be independent of oceanic or topographic influences and is generally expected to have poor relationships with other major ions. Nitrate concentrations in both PEL and cDML did not show any variation with distance from sea or changes in elevation (Fig. 3), indicating a rather mixed input of NO_3^- in the study region. A study of surface snow chemistry across the Antarctic ice sheet by Bertler et al. (2005) also indicated that NO_3^- has no clear associations either with parameters like distance from coast and elevation or with other major ions.

Compared to NO_3^-, sources of Ca^{2+} in the Antarctic environment are relatively well known. A small but significant amount of Ca^{2+} derived from sea spray and sea ice surface is present in the coastal section of Antarctic snow (Sommer et al., 2000). Sea-salt Ca^{2+} may also originate from ikaite crystals ($CaCO_3.6H_2O$) in the Antarctic sea ice during onset of winter (Dieckmann et al., 2008). However, crustal dust from local sources and remote continents are the major sources of $nssCa^{2+}$ in the Antarctic atmosphere and ice sheet (Boutron and Martin, 1980). Many ice core studies have demonstrated that dust flux archived in the Antarctic ice sheet was predominantly transported from Southern South America (SSA) and/or Australia (e.g. Basile-Doelsch et al., 1997; Delmonte et al., 2004; Revel-Rolland et al., 2006). The percentage of estimated $nssCa^{2+}$ component in the total Ca^{2+} in both PEL (> 87 %) and cDML (> 90 %) transects (Fig. 3) support a predominantly crustal origin of Ca^{2+} in the study regions. Furthermore, particle size analyses of dust from snow cores in representative sections of both cDML and PEL transects showed that > 90 % of particles were < 4 µm and the majority of them in the ∼ 1 µm size range, indicating a distant dust source as demonstrated by model studies (e.g. Li et al., 2008, 2010).

Local sources of dust in Antarctica are usually limited to ice-free coastal areas, nunataks and exposed mountainous regions (Tegen and Lacis, 1996; Mahowald et al., 2013). Snow cores from the mountainous section of the cDML transect revealed significant number of coarser particles (> 10 µm) as well as visibly insoluble particles along with a large amount of fine particles. Such coarser particles invariably indicate the in-

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fluence of locally derived dust, especially from Wohlthat mountains, in the prevailing wind direction (Fig. 1).

This study revealed a striking relationship between NO_3^- and Ca^{2+} in snow cores from both PEL and cDML transects in East Antarctica that are > 2000 km apart (Fig. 4). The Ca^{2+} flux calculations ($F_{Ca^{2+}} = C_{Ca^{2+}} \times A$, where $C_{Ca^{2+}}$ = annual mean concentration of Ca^{2+} and A = annual accumulation) (Burkhart et al., 2004) showed an average of 6.1 and 9.1 kg km^{-2} yr^{-1} of Ca^{2+} at PEL and cDML transects respectively, indicating a substantial amount of Ca^{2+} reaching the Antarctic environment throughout the year. Calcium concentrations in the mountainous section were nearly three times higher than the rest of the cDML transect, with concentrations up to 150 to 230 µg L^{-1}, strongly implying additional dust input from the Wohlthat mountains in this sector. Such additional input of Ca^{2+} from local dust sources possibly obscures the Ca^{2+} / NO_3^- association formed during long-range transport, resulting in poor correlation of NO_3^- and Ca^{2+} as observed in the mountainous section of the cDML transect. Moreover, the association between NO_3^- and Ca^{2+} was consistent in snow deposited during both summer and winter throughout the coastal and inland regions of both the PEL and cDML transects (Fig. 4). The excellent correlation between NO_3^- and Ca^{2+} concentrations at all sites (except the mountainous section) strongly suggest that aerosol interactive processes during long range transport are responsible for influencing their relationship.

4.2 Mineral dust reactivity and possible remote sources

While mineral dust aerosol is often discussed as a single entity aerosol, its chemical characteristics depends on the mineralogy of specific source regions. It is a complex mixture with major minerals like quartz, feldspar, hydrous aluminium silicates and carbonates (Pye, 1987). Mineral dust reactivity with trace gases in the atmosphere depends on the composition of the dust itself (Laskin et al., 2005). Carbonates such as calcite ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$) are found to be the most reactive constituents of mineral dust in the atmosphere in the presence of HNO_3 (Usher et al.,

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2003; Krueger et al., 2004). Nitrate-salts such as $Ca(NO_3)_2$ and $Mg(NO_3)_2$ are formed when calcic mineral dust undergo heterogeneous reactions with atmospheric HNO_3 according to the following reactions (Gibson et al., 2006):

$$CaCO_3 + 2HNO_3 \xrightarrow{\%RH} Ca(NO_3)_2 + CO_2 + H_2O \quad \text{(R1)}$$

$$CaMg(CO_3)_2 + 4HNO_3 \xrightarrow{\%RH} Ca(NO_3)_2 + Mg(NO_3)_2 + 2CO_2 + 2H_2O. \quad \text{(R2)}$$

Similarly, there exists a possibility that N_2O_5 present in atmosphere can also react with the calcic mineral dust according to the following reactions:

$$CaCO_3 + N_2O_5 \xrightarrow{\%RH} Ca(NO_3)_2 + CO_2 \quad \text{(R3)}$$

$$CaMg(CO_3)_2 + 2N_2O_5 \xrightarrow{\%RH} Ca(NO_3)_2 + Mg(NO_3)_2 + 2CO_2. \quad \text{(R4)}$$

The reaction between mineral dust and atmospheric HNO_3 or N_2O_5 occurs in two-stages. First, HNO_3 is adsorbed on the dust surface to form thin layers of $Ca(NO_3)_2$, even at very low relative humidity (RH) conditions ($\sim 12\,\%$ RH) (Al-Abadleh et al., 2003). Then, the adsorbed HNO_3 reacts with bulk $CaCO_3$ or $CaMg(CO_3)_2$ to form $Ca(NO_3)_2$. However, this reaction in the atmosphere is limited by the availability of HNO_3 (Laskin et al., 2005). Similarly, laboratory studies have shown that N_2O_5 hydrolysis on mineral dust particles is enhanced in the presence of higher relative humidity (Mogili et al., 2006).

Several ice core and model studies have demonstrated that certain regions in SSA like northern Patagonia, Puna Altiplano (PAP) and San Julian's Great Depression are the main dust emitting sources to East Antarctica (Gaiero, 2007; Lambert et al., 2008; Li et al., 2008, 2010). While a few studies have shown that dust from Australia is important in parts of East Antarctica (e.g. Revel-Rolland et al., 2006), the SSA region is the single-most important dust source to the Atlantic and Indian Ocean sector (e.g. Li et al., 2008, 2010). Studies based on large volumes of surface snow samples at Berkner Island by Bory et al. (2010) also showed that the SSA is the dominant dust source in the Atlantic sector of Antarctica. The mineralogical composition of loess and

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loessoid sediments in SSA reveal the presence of calcic-rich plagioclase and calcite minerals in the dust (Zárate, 2003) that react with HNO_3.

The SSA region is a land mass of significant size extending from 40–56° S, with a typical width varying between 500 and 700 km. It is located inside the westerly wind belt of the Southern Hemisphere and the climate here is controlled mainly by the westerlies from the Pacific Ocean which cross the Andes and continue as dry winds towards the east (Iriondo, 2000). The atmospheric circulation in Patagonia is dominated by strong meridional pressure gradients that promote strong westerlies and high wind speeds throughout the year (Coronato, 1993). These strong westerly winds have a general tendency to form rising air mass capable of lifting dust and aerosols high into the atmosphere (Coronato, 1993). Owing to extreme dryness of the air (with relative humidity as low as 5 %) in the Patagonian region, the westerlies pick up large volumes of dust, which rise to the atmosphere (Iriondo, 1989). Furthermore, meridional surface winds (Muhs and Zárate, 2001) and subtropical jet stream intensify, resulting in very strong winds that lead to sizeable dust storms (Gaiero et al., 2013) moving towards the south and east. The Reactions (R1)–(R4) show that, under humid conditions, HNO_3 strongly promotes the hydrolysis process. Though the extremely low RH conditions in the Patagonian region might not be adequate for hydrolysis, partial solvation could initially provide a reactive site to HNO_3 (Usher et al., 2003).

To further verify the possible dust sources, nine-day forward and back trajectories for everyday at 6 hourly interval for austral winter (JJA) and summer (DJF) during 2008–2009 were reconstructed using the NOAA Hysplit Model v.4 (Draxler and Rolph, 2014) in conjunction with the Global Data Assimilation System datasets (Fig. 5). The forward trajectories were calculated from two prominent dust-emitting hotspots in the SSA region (Li et al., 2010), the North Patagonia (44° S, 67° W, NP in Fig. 5) and San Julian's Great Depression (49° S, 69° W, GD in Fig. 5). The back trajectories were calculated from sampling sites representing coastal, mountainous and inland sections of cDML and PEL. A total of 368 trajectories during winter and 360 trajectories for summer at each location were subjected to a statistical treatment, wherein all trajectories were

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clustered with lower spatial variance. The forward trajectory clusters revealed that both during summer and winter, dust-laden air parcels generally circulate towards south and east from dust sources. The back trajectory clusters revealed, in line with previous studies (Li et al., 2008, 2010), that the majority of air parcels at 1500 m level invariably arrived from regions that are influenced by the air parcels from SSA region. These air parcels ultimately get incorporated into the Antarctic Circumpolar Vortex (ACV) before mass compensation by anticyclonic polar easterlies in the East Antarctic region (Dixon et al., 2012). Both forward and back trajectory clusters show that dust reaching the East Antarctic region is a blend of SSA dust sources that feed into the ACV.

4.3 Formation of calcium nitrate and its significance for the Antarctic snow chemistry

During its transport from SSA over the Southern Ocean, calcic mineral dust particles readily undergo hydrolysis with atmospheric HNO_3 or N_2O_5 as described in the Reactions (R1)–(R4) above. Higher RH levels over the Southern Ocean more likely aid the effortless conversion of $CaCO_3$ to $Ca(NO_3)_2$. Such progressive reactivity of $CaCO_3$ has been observed in laboratory studies (Krueger et al., 2003; Mogili et al., 2006). A simplified schematic diagram depicting the possible mechanism and processes involved in the formation and deposition of $Ca(NO_3)_2$ is shown in Fig. 6. The initial process involves hydrolysis of HNO_3 on calcium-rich dust particles supported by comparatively higher RH over the Southern Ocean. The anticyclonic polar easterlies sink the air masses reaching the Antarctic atmosphere from the westerly wind belt (Dixon et al., 2012). The airmass containing dust particles upon reaching the Antarctic region, undergoes mass compensation within the easterlies leading to the deposition of aerosols and dust particles to the Antarctic surface (Iriondo, 2000). It is therefore proposed, that the availability of calcic mineral dust from SSA under the influence of westerlies and its reaction with nitrogen oxides over the Southern Ocean has facilitated the formation of $Ca(NO_3)_2$ and this would explain the strong association between Ca^{2+} and NO_3^- in Antarctic snow.

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Such a reasoning is also consistent with the poor relationship between Ca^{2+} and NO_3^- reported from sites in proximity to mountainous regions that provide additional local dust which has not undergone any atmospheric chemical processes.

Interestingly, the present findings have important implications for Antarctic snow chemistry. It is considered that NO_3^- in Antarctica is usually deposited in the form of gaseous HNO_3, scavenged by or temporarily adsorbed on snow crystals (Wagnon et al., 1999) with negligible dry deposition (Legrand and Kirchner, 1990). Our study implies that apart from gaseous HNO_3 and NO_3^- from Polar Stratospheric Clouds, dust-bound NO_3^- could also constitute a significant amount of NO_3^- deposited in Antarctic snow. Similar Ca^{2+} / NO_3^- association have been observed in deep ice cores from Vostok (Legrand et al., 1999) and Dome C (Röthlisberger et al., 2002), which was attributed to the high amount of dust in atmosphere during Last Glacial Maximum (LGM). Studies by Lunt and Valdes (2001) have shown that the transport efficiency of dust to East Antarctica is greater at the present day suggesting that an overall increase in dust transport is more likely. Yet, it remained inconclusive if such reactions occurred in the atmosphere or in the Antarctic snowpack. Röthlisberger et al. (2002) also speculated that such reactions in the atmosphere would result in a widespread Ca^{2+} / NO_3^- association throughout Antarctica. Our studies suggest that such association could be widespread, provided the dust-bound nitrate reaches the interior plateau of the continent. However, it is important to consider the possible sources of additional input such as $nssCa^{2+}$ from the mountains or $ssCa^{2+}$ from the oceans along the coast in order to statistically detect the association during data analysis. Further, it is known from several studies that gaseous HNO_3 present in the interstitial snow and NO_3^- adsorbed to snow crystals undergo photolytic loss in the presence of sunlight (e.g. Jones et al., 2000). Volatile loss of HNO_3 have also been documented as a pathway for NO_3^- loss (Röthlisberger et al., 2000; Erbland et al., 2013). However, when NO_3^- is bonded with Ca^{2+} as in $Ca(NO_3)_2$, it could prevent the post-depositional loss of NO_3^- from upper snow layers even in low accumulation regions. Such reactions are of huge significance

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in the Antarctic atmosphere as they have the ability to modify the properties of original aerosols, resulting in modified climate impact, for instance, by variations in light scattering and photochemical activity (Tegen et al., 1997).

5 Conclusions

This study shows a strong and widespread association between Ca^{2+} and NO_3^- in snow from two different regions in cDML and PEL, that are > 2000 km apart in East Antarctica. We hypothesise that such a strong association is due to the interaction between calcium-rich mineral dust and nitrogen oxides in the atmosphere, resulting in the formation of $Ca(NO_3)_2$. The forward and back trajectory analyses suggest, that fine calcic mineral dust from the SSA region has been transported to the East Antarctic region, aided by the westerlies. It is proposed that $Ca(NO_3)_2$ was formed in the atmosphere over the Southern Ocean during the transport of mineral dust from SSA to the East Antarctic region. The $Ca(NO_3)_2$ thus formed, is deposited over Antarctica under the influence of anticyclonic polar easterlies. Our study also shows that local input of dust is more likely to mask the association between Ca^{2+} and NO_3^-. We suggest, that apart from other significant NO_3^- sources, NO_3^- associated with mineral dust could form a significant portion of total NO_3^- deposited in the East Antarctic snow. We also suggest that such association between Ca^{2+} / NO_3^- could be widespread and the $Ca(NO_3)_2$ formation may possibly reduce the NO_3^- post-depositional loss in low accumulation regions.

Author contributions. Both authors contributed equally to the work presented in this paper. K. Mahalinganathan collected samples, analysed data and wrote the paper. M. Thamban analysed data and wrote the paper.

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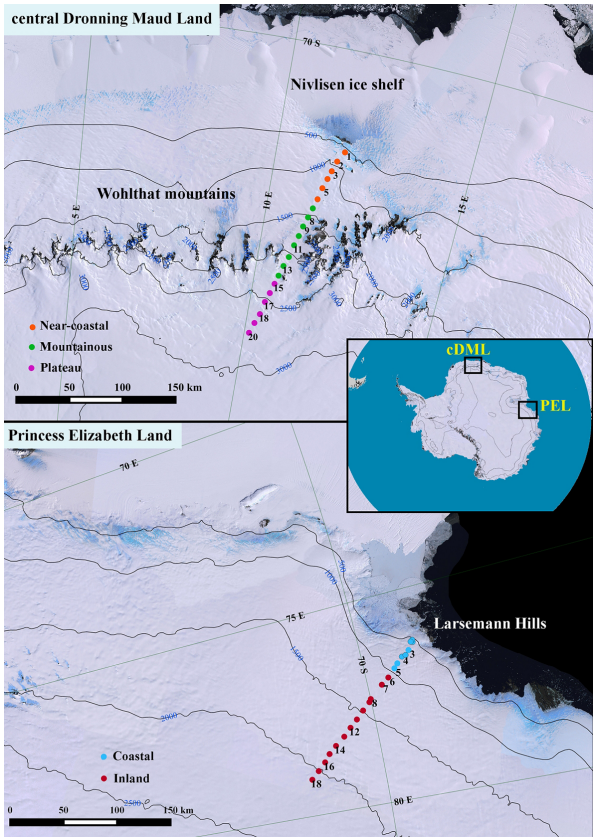

Figure 1. Study region showing sampling locations along transects in central Dronning Maud Land (top panel) and Princess Elizabeth Land (bottom panel), East Antarctica. Colour coded sampling locations indicate different topographic sections. Inset shows the study area highlighted in Antarctica. Maps were created using LANDSAT Mosaic using QGIS Wien (v.2.8) in conjunction with Quantarctica project.

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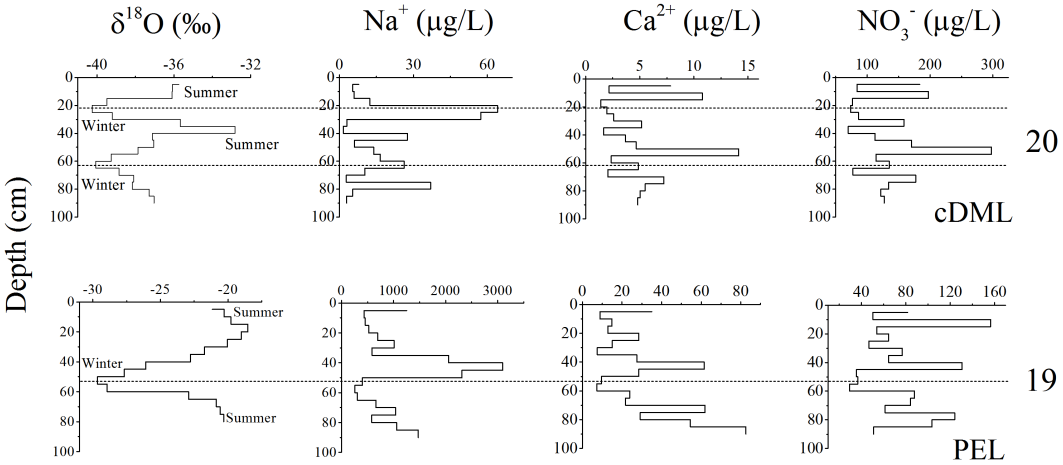

Figure 2. Oxygen isotope ratios and major ions showing seasonality in representative snow cores from inland (cDML, core 20) and coast (PEL, core 19). The dotted lines represent winter maxima in the cores.

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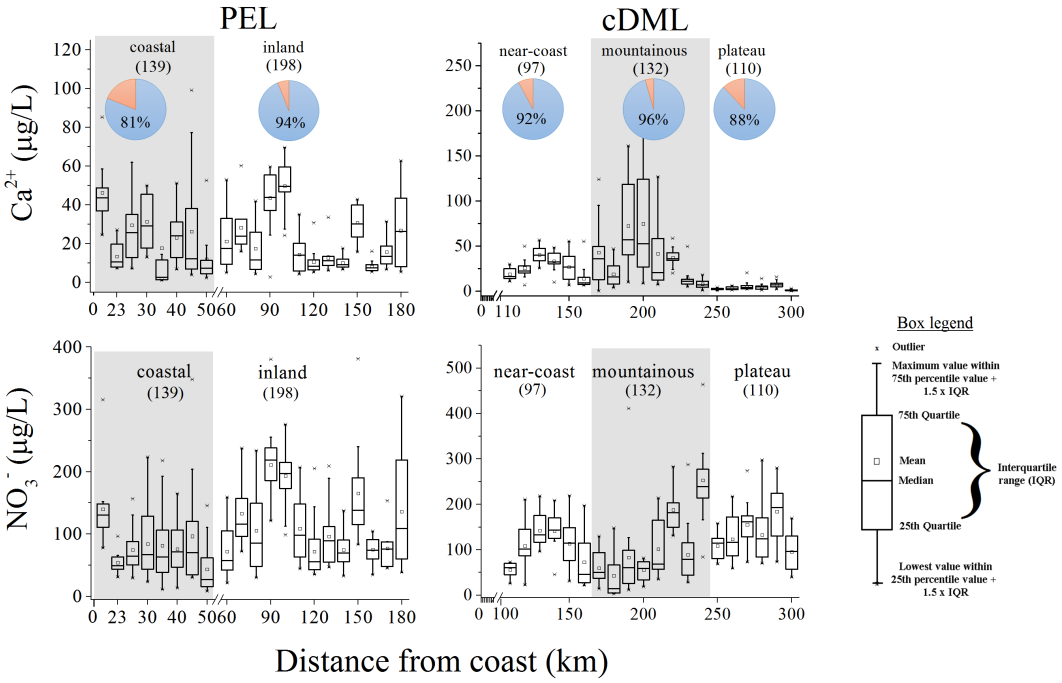

Figure 3. Distribution of Ca^{2+} and NO_3^- concentrations in snow cores along PEL and cDML transects. The % values in circles in Ca^{2+} plots shows $nssCa^{2+}$ %. The numbers inside brackets indicate the total number of samples in respective sections.

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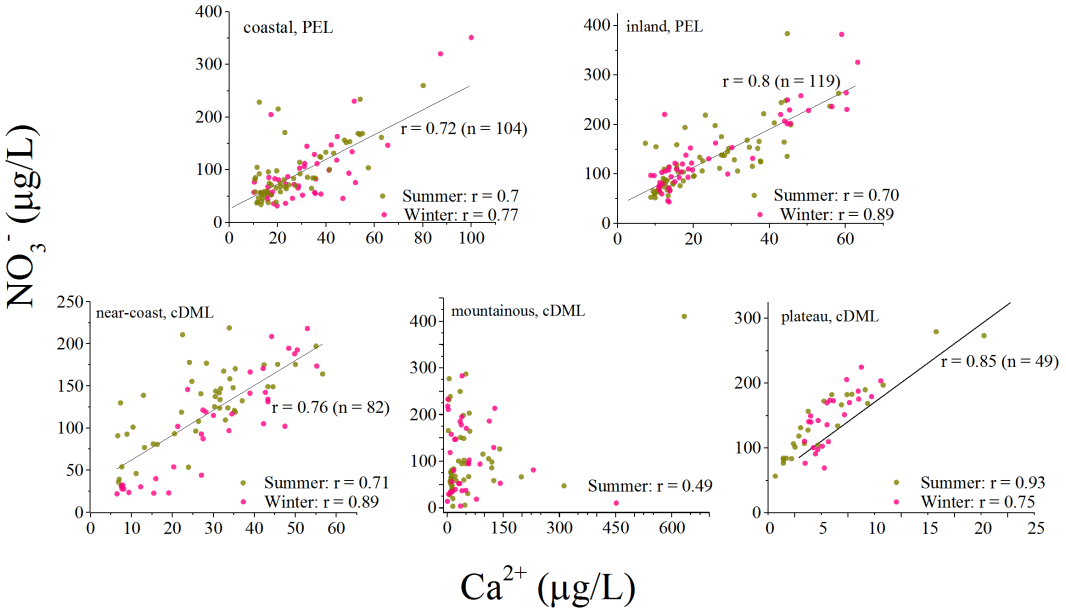

Figure 4. Correlation between Ca^{2+} and NO_3^- during austral summer (green) and winter (pink) in the study regions in cDML and PEL. The samples from coastal and inland sections in both cDML and PEL showed a strong association while the mountainous section in cDML transect showed weak association only during summer.

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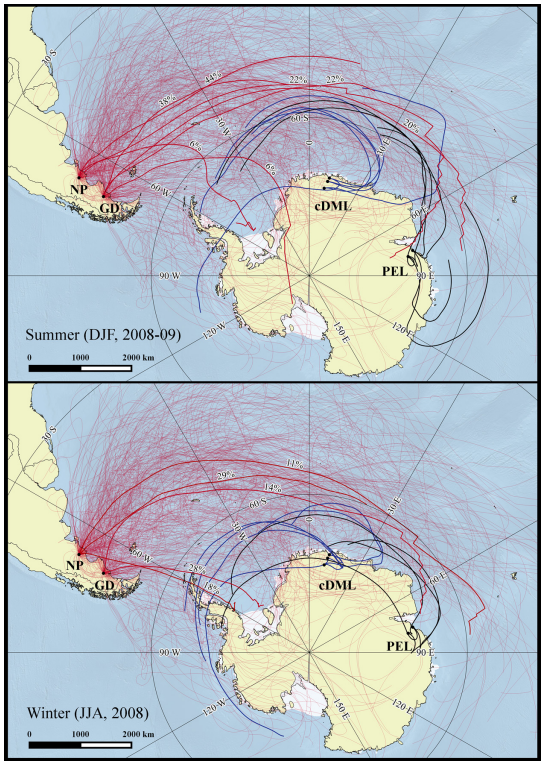

Figure 5. Nine days forward and back trajectory analyses using HYSPLIT Model v.4 in conjunction with GDAS dataset. The forward trajectories (red) were calculated from North Patagonia (NP) and San Julian's Great Depression (GD), and the back trajectories (blue, black) were reconstructed from cDML and PEL. The cloud of thin red lines indicate clusters of 6 hourly forward trajectories while the thick red lines show the cluster average. The thick blue and black lines from cDML and PEL region respectively, indicate the cluster average of back trajectories calculated during the same time period.

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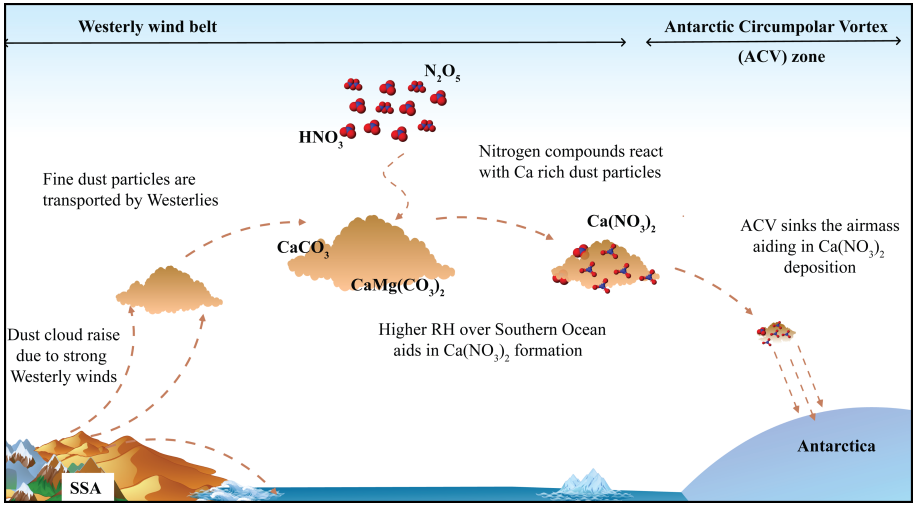

Figure 6. Schematic diagram depicting possible processes and chemical reactions involved during the mineral dust transport from Southern South America (SSA) before depositing over Antarctic ice sheet.

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