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

Guitao Shi[1,2], Hongmei Ma[2], Zhengyi Hu[2], Zhenlou Chen[1], Chunlei An[2], Su Jiang[2], Yuansheng Li[2], Tianming Ma[2], Jinhai Yu[2], Danhe Wang[1], Siyu Lu[2], Bo Sun[2], and Meredith G. Hastings[3,4]

[1]Key Laboratory of Geographic Information Science (Ministry of Education), School of Geographic Sciences and State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200241, China
[2]Polar Research Institute of China, Shanghai 200062, China
[3]Department of Earth, Environmental and Planetary Sciences, Brown University, Providence, Rhode Island 02912, USA
[4]Institute at Brown for Environment and Society, Brown University, Providence, Rhode Island 02912, USA

Correspondence: Guitao Shi (gtshi@geo.ecnu.edu.cn)

Abstract. To better understand snow chemistry in different environments across the Antarctic ice sheet, we investigated snow ions on a traverse from the coast to Dome A. Results show that the non-sea-salt (nss) fractions of K^+, Mg^{2+}, and Ca^{2+} are mainly from terrestrial particle mass and $nssCl^-$ is associated with HCl. Spatially, the non-sea-salt fractions of ions to the totals are higher in the interior areas than on the coast, and seasonally, the proportions are higher in summer than in winter. Negative $nssSO_4^{2-}$ on the coast indicates sea salts from the sea ice, and marine biogenic emissions dominate snow SO_4^{2-} in interior areas throughout the year.

1 Introduction

Snow chemistry has been broadly investigated along traverses during the International Trans-Antarctic Scientific Expedition (ITASE), e.g., Dumont d'Urville Station (DDU) to Dome C, coast-interior traverse in Adélie Land, Syowa Station to Dome F, Terra Nova Bay to Dome C, 1990 ITASE, and US ITASE in West Antarctica (Legrand and Delmas, 1985; Qin et al., 1992; Mulvaney and Wolff, 1994; Proposito et al., 2002; Suzuki et al., 2002; Dixon et al., 2013), and Bertler et al. (2005) have comprehensively summarized the glaciochemical data across the ice sheet, most of which are for surface snow. Among the major ions, sea-salt-related

ions (e.g., Na^+ and Cl^-), in general, are the most abundant species and typically exhibit a clear spatial trend, with concentrations falling off sharply with distance from the coast.

Temporally, with varied sources and lifetimes, ions in snow often exhibit different seasonal variations; e.g., sea-salt-related ions show high concentrations in winter, while elevated concentrations of SO_4^{2-} and NO_3^- are frequently observed in summer (Neubauer and Heumann, 1988; Gragnani et al., 1998; Traversi et al., 2004; Shi et al., 2015). On annual to decadal timescales, ion concentrations in snow and ice tend to be associated with changes in transport from year to year (Severi et al., 2009; Weller et al., 2011), and thus large-scale atmospheric and oceanic circulation in the Southern Hemisphere could potentially influence variations in snow and ice chemistry (Russell and McGregor, 2010; Weller et al., 2011; Mayewski et al., 2017).

Although investigations of snow chemistry have been carried out along several overland traverses, the investigation of snow chemistry under different environmental conditions and over time is needed, given that the Antarctic ice sheet itself and precipitation and deposition patterns and trends are changing. The Chinese inland Antarctic traverse from the coastal Zhongshan Station to the ice sheet summit (Dome A) covers a range of environments (~ 1250 km); e.g., a high snow accumulation rate is present on the coast and in some interior areas and a low accumulation rate is observed on the Dome A plateau. Several investigations have been carried out

to determine the concentrations of a few ionic species and trace elements on the traverse (e.g., Li et al., 2016; Du et al., 2019), but only limited snow chemistry data were previously available. Therefore, we used surface snow and snow pit samples collected during five Chinese inland Antarctic scientific expedition campaigns to determine the spatial and temporal variations and their controlling factors in a comprehensive set of ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}).

2 Methods

2.1 Sample collection

Snow samples were collected along the traverse from the coast to the ice sheet summit during five Chinese National Antarctic Research Expedition (CHINARE) campaigns (Fig. S1 in the Supplement). In January–February in the years 1999, 2011, 2013, 2015, and 2016, 107, 120, 125, 117, and 125 surface snow samples, respectively, were collected on the traverse. In total, 594 snow samples were collected during the five seasons. For the snow sampling protocols, refer to Shi et al. (2018). It is noted that the collected surface ~ 3 cm of snow represents different lengths of time at different locations, considering the wide range of snow accumulation rates on the traverse (Fig. 1a). At locations with a high snow accumulation rate on the coast, the upper 3 cm of snow may represent deposition from a few weeks or a single snowfall, while the surface 3 cm of snow could represent deposition over a few months on the Dome A plateau. Still, the information contained in the surface snow generally indicates summertime conditions, as the sampling took place during late January and February in each season.

In addition to surface snow, snow pits were sampled in three representative areas on the traverse: P1, located on the coast (69.79° S, 76.49° E; 46 km from the coast), was sampled in December 2015; P2, located in the interior area (76.42° S, 77.03° E; 800 km from the coast), was sampled in January 2016; and P3, located on the Dome A plateau (80.42° S, 77.11° E; 1256 km from the coast), was sampled in January 2010. Sites P1 and P2 are characterized by a high snow accumulation rate (> 100 kg m^{-2} a^{-1}), while the snow accumulation rate at P3 is ~ 25 kg m^{-2} a^{-1}. The depths of P1, P2, and P3 are 180, 100, and 150 cm, respectively, with a respective sampling resolution of 5, 3, and 1 cm. Details on the snow pit sampling are described in Shi et al. (2015). All snow samples were transported and stored under freezing conditions (~ -20 °C).

2.2 Sample analysis

In a class 100 clean room, about 5 mL of the melted sample was transferred to pre-cleaned 8 mL ion chromatography (IC) autosampler vials, and then the lid was tightly screwed on to the vials. The samples were analyzed by an ICS-3000 IC system (Dionex, USA) for the concentrations of ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) in a class 1000 clean room. More details on ion determination are described in Shi et al. (2012). During sample analysis, replicate determinations ($n = 5$) were performed, and 1 relative standard deviation (1σ) for all eight ions was generally < 5 %.

In Antarctic snow, previous observations have suggested that concentrations of H^+ can be reasonably deduced from the ion-balance disequilibrium (Legrand and Delmas, 1985; Legrand, 1987):

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

where ion concentrations are in µeq L^{-1}. In addition, the non-sea-salt fractions of ions (nssX), including nssCl$^-$, nssSO$_4^{2-}$, nssK$^+$, nssMg^{2+}, and nssCa^{2+}, can be calculated from the following expression,

$$[nssX] = [X]_{snow} - ([X]/[Na^+])_{seawater} \times [Na^+]_{snow}, \qquad (2)$$

where [X] is the concentration of ion X and [X]/[Na$^+$] ratios in seawater are 1.17 (Cl$^-$), 0.12 (SO$_4^{2-}$), 0.022 (K$^+$), 0.23 (Mg^{2+}), and 0.044 (Ca^{2+}) (in µeq L^{-1}).

3 Results

3.1 Ion variations in snow pits

Clear seasonal cycles of Na^+ and nssSO$_4^{2-}$ are present in P1 and P2, and thus the two pits can be well dated, spanning ~ 3 years (Fig. S2). In addition to SO$_4^{2-}$ and Na$^+$, the other species also show seasonal variations, especially in P1, where elevated levels of NO$_3^-$ and NH$_4^+$ are generally present in summer and the concentrations of Cl$^-$, K$^+$, Mg^{2+}, and Ca^{2+} are high in winter. As for nssSO$_4^{2-}$ at P3, the very large signal at the depth of ~ 120 cm is most likely the fallout from the massive eruption of Pinatubo in 1991 (Fig. S2), based upon previous observations at Dome A (e.g., Hou et al., 2007). It is noted that only elevated SO$_4^{2-}$ concentrations are present during this period, possibly suggesting that Pinatubo volcanic emissions contribute less to the ion budgets other than to SO$_4^{2-}$ at Dome A.

In terms of the non-sea-salt fractions, nssCl$^-$ is lower at P1 (0.25 ± 0.28 µeq L^{-1}) than at the inland sites P2 and P3 (0.42 ± 0.18 and 0.58 ± 0.34 µeq L^{-1}, respectively), while the concentrations of nssK$^+$, nssMg^{2+}, and nssCa^{2+} generally show a similar spatial pattern. In general, nssCl$^-$, nssK$^+$, nssMg^{2+}, and nssCa^{2+} in snow pits do not show clear seasonal cycles.
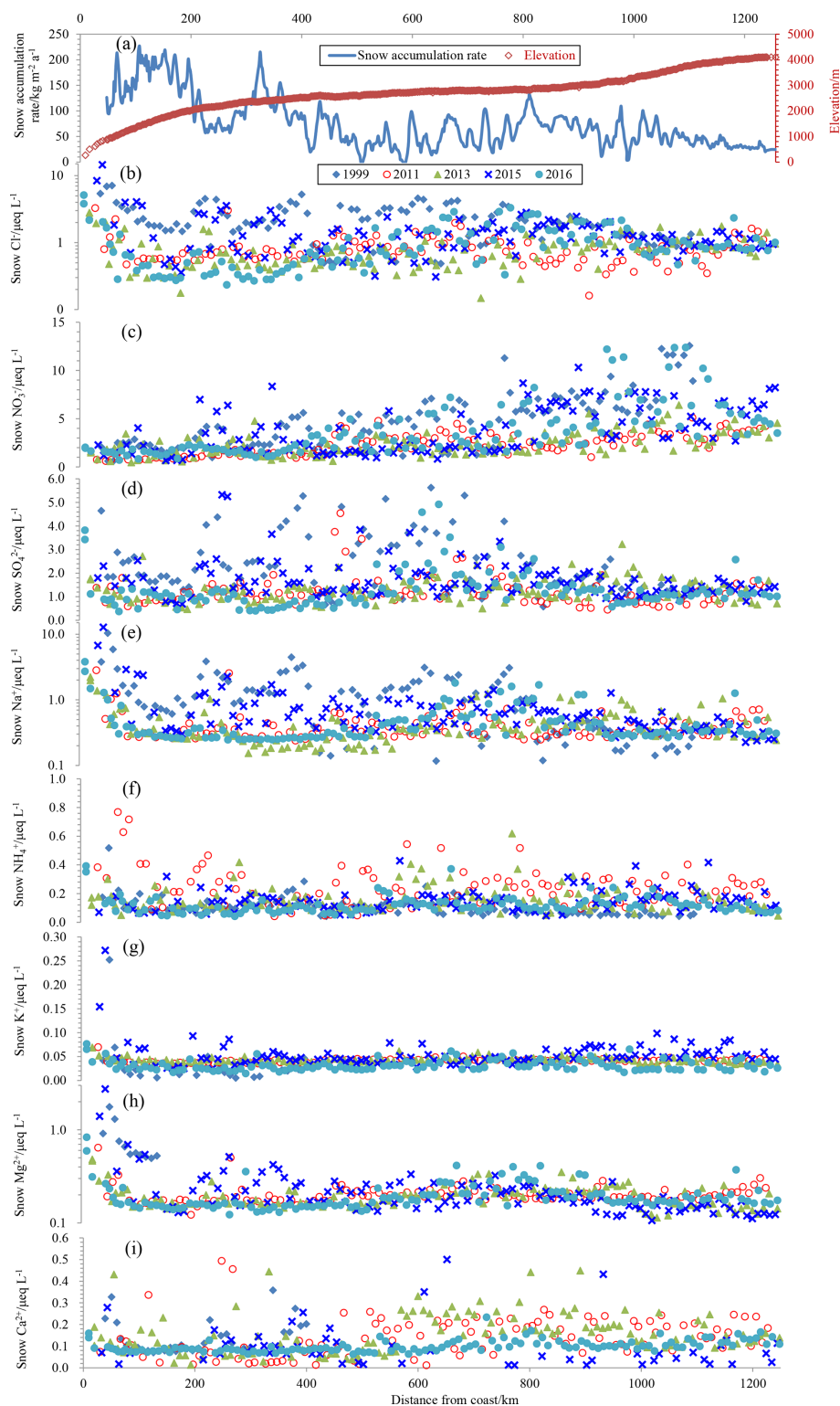
Figure 1. Annual snow accumulation rate and elevation **(a)** and ion concentrations in surface snow collected during five seasons **(b–i)**. Annual snow accumulation rate is obtained from field bamboo stick measurements, updated to 2016 from Ding et al. (2011). The closed diamond, open circle, closed triangle, cross, and closed circle denote ion concentrations in the years 1999, 2011, 2013, 2015, and 2016, respectively. Note that a base-10 log scale is used for the y axis of Cl^- **(b)**, Na^+ **(e)**, and Mg^{2+} **(h)**.

3.2   Ion concentrations in surface snow

Concentrations of ions in surface snow are shown in Fig. 1, and the values generally fall within the reported ranges of the ITASE program sampling (Bertler et al., 2005). Spatially, Cl^-, Na^+, K^+, and Mg^{2+} show very high concentrations within the narrow coastal region and decrease sharply further inland, with minimum values on the Dome A plateau ($> \sim 1000$ km from the coast). It is noted that some samples on the coast also show elevated Ca^{2+} concentrations. The high ion concentrations near the coast may be associated with the strong marine air mass intrusions (Hara et al., 2014). NO_3^- shows an opposite spatial trend, with increasing values towards the inland. As for SO_4^{2-} (and $nssSO_4^{2-}$), NH_4^+, and Ca^{2+}, no clear spatial trend was found.

Among the ions in surface snow, the most abundant species is H^+, accounting for 30 %–40 % of the total ions, followed by NO_3^-, SO_4^{2-}, and Cl^-. In general, NH_4^+, K^+, Mg^{2+}, and Ca^{2+} are the smallest components of the ionic composition, with the four cations summing to (6.0 ± 3.4) % of the total (Fig. S3).

4   Discussions

4.1   Non-sea-salt fractions of ions in surface snow

Correlation plots of ions versus Na^+ in surface snow are shown in Fig. 2. On the coast, most of the Cl^-/Na^+ data are distributed close to the seawater dilution line (Fig. 2a), while most of the plots in the interior areas are above the seawater line, suggesting an enrichment of Cl^-. The $nssCl^-$ accounted for (39 ± 24) % of Cl^- on the traverse, with higher values in the interior areas. The elevated fractions of $nssCl^-$ are likely associated with the "secondary" HCl which is produced by the reactions between sea salts and acids (e.g., HNO_3 and H_2SO_4) (Finlayson-Pitts, 2003).

Mg^{2+} is irreversibly deposited into the snow, and the fraction of $nssMg^{2+}$, on average, represents (44 ± 19) % of Mg^{2+} in snow, with lower (higher) values on the coast (plateau) (Fig. 2d). The enrichment of Mg^{2+} has not been observed in sea salt particles produced by bubble bursting (Keene et al., 2007), and thus enriched Mg^{2+} in the snow is unlikely to be associated with sea salt spray. In the atmosphere, sea salt aerosols can be modified at low temperatures via the formation of mirabilite, thus leading to an elevated ratio of Mg^{2+}/Na^+ if mirabilite precipitates from the aerosols. However, the solid–liquid separation of mirabilite in the aerosol droplet was not observed in the experiments (Wagenbach et al., 1998). Thus, the enrichment of Mg^{2+} in surface snow is unlikely to be associated with sea salt fractionation. Although it is proposed that Mg^{2+} separation in sea salts can occur in surface snow due to the re-freezing process on surface snow (i.e., the quasi-liquid layers on the crystal surface can act like seawater freezing; Hara et al., 2014), our mea-

surement of Mg^{2+} in bulk snow is unlikely to support this process responsible for Mg^{2+} enrichment. A previous observation conducted near this traverse showed a moderate correlation of Mg^{2+} with the element Al in the surface snowpack ($r = 0.53$; $p < 0.05$), indicating a contribution of continental dust (Khodzher et al., 2014). Thus, the most plausible interpretation of $nssMg^{2+}$ is the contribution of terrestrial aerosols. Similarly to Mg^{2+}, most of K^+/Na^+ data points are close to the seawater dilution line on the coast, suggesting a primary contribution of sea salt spray (Fig. 2c). Enriched K^+ is ubiquitous in interior areas, possibly associated with mineral transport and combustion emissions in the Southern Hemisphere (Virkkula et al., 2006; Hara et al., 2013). Note that all sampling sites are at least several tens of kilometers away from the coast, so the contribution of biological activity to snow K^+ would be rather minor (Rankin and Wolff, 2000). A lack of correlation between K^+ (or $nssK^+$) and refractory black carbon (rBC; unpublished data; Fig. S4), which mainly represents the biomass burning emissions in the Southern Hemisphere (Sigl et al., 2016), suggests that K^+ in surface snow is unlikely dominated by biomass burning emissions.

The fraction of $nssCa^{2+}$, on average, accounts for (73 ± 26) % of total Ca^{2+} in surface snow, with high percentages in the interior areas. In Antarctica, snow $nssCa^{2+}$ was thought to be mainly associated with terrestrial inputs, possibly from both South America and Australia (Bertler et al., 2005; Wolff et al., 2010; Du et al., 2018). The $nssSO_4^{2-}$ represents (94 ± 5) % of total SO_4^{2-} in surface snow, with lower (higher) proportions on the coast (plateau) (Fig. 2b), suggesting a dominant role of marine bioactivities. Differently from the coarse-sea-salt aerosols, $nssSO_4^{2-}$ originating from marine biogenic production of dimethylsulfide can form fine aerosol particles in the atmosphere (Legrand et al., 2017a), resulting in a long atmospheric residence time (> 10 d to weeks) and consequently efficient transport (Bondietti and Papastefanou, 1993; Hara et al., 2014). This can help explain the elevated deposition flux of $nssSO_4^{2-}$ frequently found at inland Antarctic sites, e.g., site P2 (discussed below).

4.2   Non-sea-salt fractions and fluxes of ions in snow pits

At P1, the slope values of the linear regression between Na^+ and the four ions are close to those of seawater (Fig. 3), suggesting a dominant source of sea salt aerosols. The proportions of the non-sea-salt fractions of K^+, Mg^{2+}, and Ca^{2+} to the ions in snow are much lower in winter than in summer, as a result of the high sea salt inputs in winter. Negative $nssCl^-$ is present in summer snow, indicating the modification to sea salts (i.e., formation of mirabilite in the atmosphere) in summer when the acid levels (e.g., HNO_3) are relatively high (Savarino et al., 2007). Some winter snow samples featured negative $nssSO_4^{2-}$, i.e., an SO_4^{2-}/Na^+ ratio below the value of seawater (Fig. S2), suggesting sea salt aerosols originating from the sea ice (Marion et al., 1999). In the winter snow, if
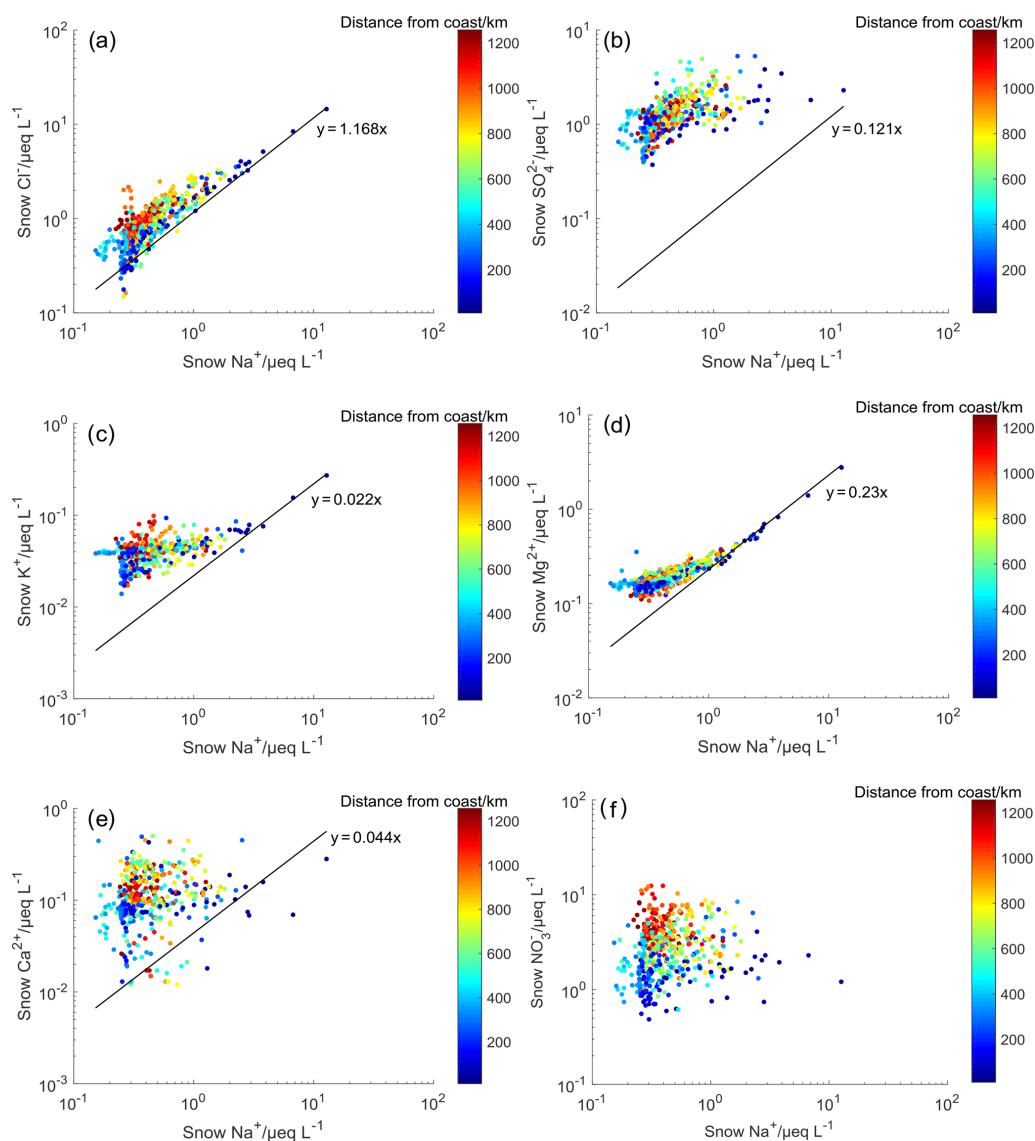
Figure 2. Correlation plots of Cl^-, SO_4^{2-}, K^+, Mg^{2+}, Ca^{2+}, and NO_3^- versus Na^+ in surface snow. The solid black line represents the seawater dilution line, with slopes of typical ions versus Na^+ ratios in seawater (in $\mu eq\,L^{-1}$). The concentration of NO_3^- in seawater is too variable among the seas, and a representative ratio of NO_3^-/Na^+ cannot be presented. Note that a base-10 log scale is used for ion concentrations.

all SO_4^{2-} is from sea salt aerosols, $nssSO_4^{2-}$ is expected to be lower than or close to zero. However, 13 out of the 17 samples classified as winter snow at P1 were characterized by positive $nssSO_4^{2-}$, suggesting a significant contribution from marine biogenic emissions. It is interesting that $nssSO_4^{2-}$ has a strong negative correlation with Na^+ in winter ($r = 0.82$; $p < 0.001$), raising two potential cases: (1) stronger winds transport more sea salt aerosols to P1 which feature depleted SO_4^{2-} from sea ice, thereby resulting in low concentrations of $nssSO_4^{2-}$ and assuming a stable SO_4^{2-} input flux from marine biogenic emissions, and/or (2) with a larger extent of sea ice and strong transport, a large sea salt flux would still

occur but carry less $nssSO_4^{2-}$ from marine biogenic emissions due to the longer transport distance (Wolff et al., 2006, and references therein). If case (2) dominated $nssSO_4^{2-}$ variations in the winter snow, lower $nssSO_4^{2-}$ would be expected at the end than at the beginning of winter when a sea ice coverage minimum is present. The observation at P1, however, does not support this expected seasonal trend (Fig. S2). It is most likely, then, that sea salt aerosol inputs instead of marine biogenic emissions dominate $nssSO_4^{2-}$ variations in winter snow.

The patterns of relationships between ions and Na^+ at P2 are similar to those of P1 except for Ca^{2+} (Fig. 3). Non-sea-
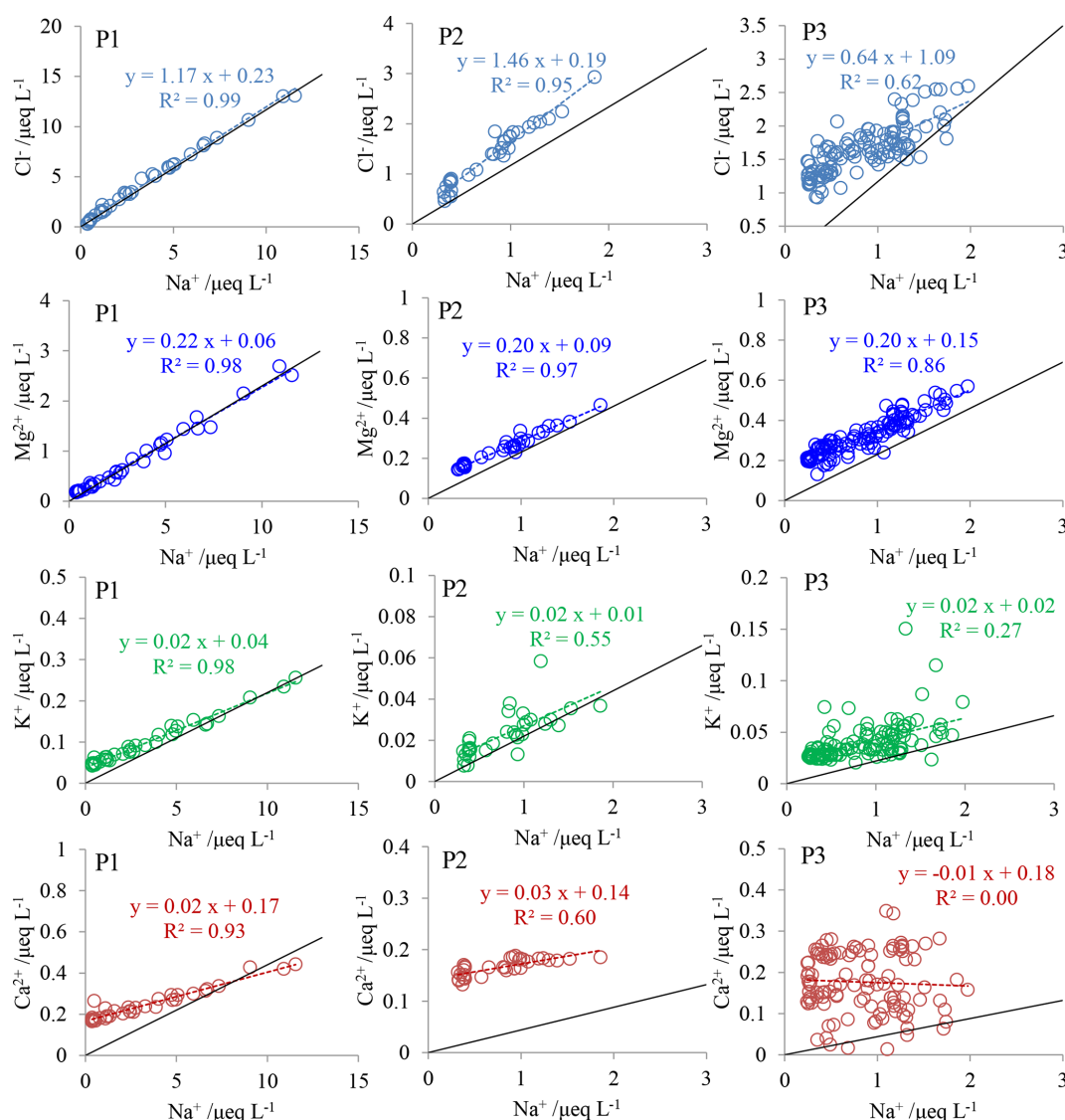
Figure 3. Relationships between Na^+ and Cl^-, K^+, Mg^{2+}, and Ca^{2+} in the three snow pits (P1, P2, and P3). Also shown are the linear regressions between them (dashed line), with all of the linear correlations significant at $p < 0.001$ except Ca^{2+}/Na^+ at P3. The solid black line represents the seawater dilution line. Note that the data of the bottom $\sim 30\,cm$ layer of P3 were excluded in the plots, since they represent a snow layer clearly impacted by volcanic (Pinatubo) eruption emissions.

salt fractions of Ca^{2+} account for $(79 \pm 9)\,\%$ of the total, suggesting a dominant role of the terrestrial source. It is noted that Ca^{2+} remains relatively constant with increasing Na^+ (Fig. 3), possibly suggesting insignificant seasonal variations in terrestrial dust inputs. The fractions of $nssSO_4^{2-}$ to SO_4^{2-} in summer and winter snow are $(94 \pm 4)\,\%$ and $(88 \pm 4)\,\%$, respectively, suggesting a dominant role of marine biogenic emissions, differently from those at P1. Previous investigations proposed that sea salt aerosols emitted from sea ice are an important contribution to the sea salt budget in central Antarctica in winter (Legrand et al., 2016, 2017b). Here, the high $nssSO_4^{2-}$ concentrations indicate that marine emissions could also be an important source of ions in winter.

At P3, Cl^-, K^+, and Mg^{2+} are also correlated well with Na^+ (Fig. 3). The non-sea-salt fractions of Cl^- make up $(38 \pm 24)\,\%$ of the total, similarly to those at P2, indicating the importance of HCl deposition, and consequently result in Cl^- not being a quantitative indicator of sea salts in the interior areas. The $nssSO_4^{2-}$ at P3 accounts for $(95 \pm 2)\,\%$ of SO_4^{2-}. Together with the observations at P2, it can be inferred that SO_4^{2-} in the interior areas is dominated by marine biogenic emissions throughout the year, generally in line with the observation at Dome C (Udisti et al., 2012).

Ion fluxes in the three snow pits can be determined by multiplying the concentrations by the snow accumulation rate, and the highest fluxes of ions except for NO_3^- were gener-

ally present at P1, followed by P2 and P3 (Fig. S5). It is noted that $nssSO_4^{2-}$ fluxes at P1 ($99.4 \pm 46.7 \, \mu eq \, m^{-2} \, a^{-1}$) and P2 ($109.2 \pm 21.6 \, \mu eq \, m^{-2} \, a^{-1}$) are comparable, although P1 is located on the coast and P2 is located further inland (~ 800 km from the coast). In addition, the ratio of $nssSO_4^{2-}$ flux at P1 to that at P3 is 2.2, the lowest among the ratios for the observed ions (17.2, 7.5, 26.7, 8.5, 17.4, 17.0, and 10.0 for Cl^-, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}, respectively), suggesting more efficient transport of $nssSO_4^{2-}$. In other words, atmospheric $nssSO_4^{2-}$ from the open ocean can be efficiently transported to at least as far inland as ~ 800 km from the coast (~ 2800 m above sea level; site P2).

5 Conclusions

Snow chemistry on a traverse from the coastal Zhongshan Station to Dome A was investigated. It is shown that the non-sea-salt fractions of K^+, Mg^{2+}, and Ca^{2+} are mainly associated with terrestrial particle mass, while $nssCl^-$ is linked to the deposition of HCl. Spatially, the proportions of non-sea-salt fractions of ions to the totals are higher in the interior areas than on the coast, and seasonally, the proportions are generally higher in summer than in winter, due to the high sea salt inputs during wintertime. Negative $nssSO_4^{2-}$ observed on the coast indicates sea salts mainly originating from the sea ice in winter, while positive $nssSO_4^{2-}$ is present throughout the year in the interior areas, suggesting the dominating role of marine biogenic emissions. The $nssSO_4^{2-}$ can be transported efficiently to at least as far inland as the ~ 2800 m contour line.

Data availability. This dataset, chemical data on ion concentrations in snow on the traverse from the coast (Zhongshan Station) to Dome A, is available from the Chinese National Arctic and Antarctic Data Center: https://doi.org/10.11856/SNS.D.2021.003.v0 (Shi, 2021).

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Competing interests. The authors declare that they have no conflict of interest.

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