



Concentrations, Particle-Size Distributions, and Dry Deposition Fluxes of Aerosol Trace Elements over the Antarctic Peninsula

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Abstract. Size-segregated particulate air samples were collected during the austral summer of 2016–2017 at Palmer Station on the Anvers Island, west Antarctic Peninsula, to characterize trace elements in aerosols. Trace elements in aerosol samples, including Al, P, Ca, Ti, V, Mn, Ni, Cu, Zn, Ce, and Pb, were determined by total digestion and sector field inductively coupled plasma mass spectrometer (SF-ICP-MS). The results show that these elements are derived primarily from three sources: (1) regional crustal emissions, (2) long-range transport, and (3) sea-salt aerosols. Elements dominated by a crustal source (Al, P, Ti, V, Mn, Ce) with $EF_{\text{crust}} < 10$ were accumulated mostly in the coarse-mode particles ($> 1 \mu\text{m}$) and peaked at 2.5–7.8 μm in diameter, reflecting the contributions of regional crustal sources. Non-crustal elements (Ca, Ni, Cu, Zn, Pb) showed $EF_{\text{crust}} > 10$. Aerosol Pb was accumulated primarily in fine-mode particles, peaking at 0.078–0.25 μm , and likely was impacted by air masses from South America based on air-mass back trajectories. However, Ni, Cu, and Zn were not detectable in most size fractions and didn't present clear size patterns. Sea-salt elements (Ca, Na, K) showed single mode distribution and peaked at 2.5–7.8 μm . The estimated dry deposition fluxes of dust for the austral summer, based on the particle size distributions of Al measured at Palmer Station, ranged from 0.65 to 28 $\text{mg m}^{-2} \text{yr}^{-1}$ with a mean of 5.5 $\text{mg m}^{-2} \text{yr}^{-1}$. The estimated dry deposition fluxes of the target trace elements in this study were lower than most fluxes reported previously for coastal Antarctica and suggest that atmospheric input of trace elements through dry deposition processes may play a minor role in determining trace element concentrations in surface seawater over the continental shelf of the west Antarctic Peninsula.

1 Introduction

Aerosols affect the climate through direct and indirect radiative forcing (Kaufman et al., 2002). The extent of such forcing depends on both physical and chemical properties of aerosols, including particle size and chemical composition (Piliinis et al., 1995). Size and chemical composition of aerosols influence aerosol optical properties as well as cloud formation and development (Weinzierl et al., 2017). It has been realized that the impact of coarse mineral dust has been underestimated in climate models (Adebisi and Kok, 2020), and this is partly due to the inaccurate understanding of the removal processes of coarse-mode particles.

Atmospheric aerosol deposition delivers nutrient elements to the open ocean, playing an essential role in maintaining marine primary production (Jickells and Moore, 2015; Jickells et al., 2016; Mahowald et al., 2018). A significant source of atmospheric trace elements in the remote oceans is continental dust derived from arid and unvegetated regions (Duce and



Tindale, 1991). In addition, volcanic eruptions, biomass burning, anthropogenic activities and even glacial processes also contribute trace elements to the atmosphere (Pacyna and Pacyna, 2001; Chuang et al., 2005; Guieu et al., 2005; Crusius et al., 2011; Baker et al., 2020). The concentrations of some trace elements in surface seawater of several open oceanic regions, including Al, Fe, Mn, Zn, and Pb, depend strongly on atmospheric inputs (Duce et al., 1991; Prospero et al., 1996; Wu and Boyle, 1997; Measures and Vink, 2000; Moore et al., 2013; Bridgestock et al., 2016).

Atmospheric trace elements in remote regions, such as the Southern Ocean and Antarctica, may derive from various sources, but are generally dominated by long-range transport from distant continental sources (Li et al., 2008). A wide range of aerosol studies have been carried out in Antarctica, with the intention of understanding the processes affecting aerosols and the background level of trace elements in the atmosphere (Zoller et al., 1974; Dick and Peel, 1985; Tuncel et al., 1989; Artaxo et al., 1990; Lambert et al., 1990; Dick, 1991; Artaxo et al., 1992; Loureiro et al., 1992; Mouri et al., 1997; Mishra et al., 2004; Arimoto et al., 2008; Gao et al., 2013; Xu and Gao, 2014; Winton et al., 2016). Over the past several decades, one of the most dramatic warming regions in the Southern Hemisphere has been the Antarctic Peninsula (Vaughan et al., 2003; Bromwich et al., 2013; Turner et al., 2014). A recent study further indicates that slight increases in mean air temperature may cause rapid melting and ice loss in this region (Abram et al., 2013). Under the conditions of low precipitation (Van Lipzig et al., 2004) and high wind speed (Orr et al., 2008), several ice-free areas in the Antarctic Peninsula could serve as local dust sources (Kavan et al., 2018), and such sources could contribute to the atmospheric loading of certain trace elements such as Fe (Gao et al., 2020). Ice core data showed that the dust deposition was more than doubled during the 20th century in the Antarctic Peninsula (McConnell et al., 2007). On the other hand, shipborne tourism in the Antarctic Peninsula has experienced exponential growth since 1990s (Lynch et al., 2010), which might contribute to certain trace elements, such as V and Ni, in the marine atmosphere over this region, possibly similar to the situation at Ny-Alesund in the Arctic (Zhan et al., 2014). As a result, dust emissions induced by regional warming and increased ship emissions may have impacted the concentrations of aerosol trace elements in the marine atmosphere over the Antarctic Peninsula, affecting atmospheric deposition of trace elements to coastal waters off the Antarctic Peninsula and adjacent pelagic waters of the Southern Ocean (Wagener et al., 2008). However, aerosol trace elements are still under-sampled around coastal Antarctica, and thus understanding the chemical and physical properties of aerosols and accurate estimation of the atmospheric deposition of trace elements to the regions are lacking.

This study presents results from an *in-situ* measurement of size-segregated aerosol particles at Palmer Station, Antarctic Peninsula, in the austral summer of 2016–2017. The objectives are to (1) measure the concentrations and size distributions of trace elements in aerosols, (2) determine potential sources of the elements, and (3) estimate dry deposition fluxes of the elements based on the measurement data. Results from this study fill a data gap critically needed for characterization of aerosol properties and for improving quantification of the fluxes that contributes to regional biogeochemical cycles. The new observational data can also provide insight into the new sources of aerosol trace elements by warming and by the impact of human activities in this region. A full discussion of atmospheric Fe in this sample set was published recently (Gao et al., 2020); this paper extends that study by investigating the concentrations, size distributions, and dry deposition fluxes of a suite of additional aerosol trace elements.



2 Methods

2.1 Sampling and sample treatment

Aerosol size-segregated samples were collected during austral summer from November 19, 2016 to January 30, 2017 at Palmer Station (64.77° S, 64.05° W, Figure 1), located on the southwestern coast of Anvers Island off the Antarctica Peninsula.

70 Sampling was conducted using a ten-stage Micro-Orifice Uniform Deposit Impactor™ (MOUDI, MSP Corp., MN, USA) with a 30 L min⁻¹ flow rate. The 50% cut-off aerodynamic diameters of MOUDI are 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, 10, and 18 µm. In this study, size fractions ≤1.0 µm were summed to operationally define fine-mode particles and those ≥1.8 µm were summed to define coarse-mode particles, similar to previous studies which operationally divided aerosol particles into fine and coarse fractions using a cut-off size of 1.0–3.0 µm (Siefert et al., 1999; Chen and Siefert, 2004; Buck et al., 2010; 75 Gao et al., 2019). The aerosol sampler was placed on a sampling platform which was ~300 m east from the station center and ~3 m high above the ground (~20 m above sea-level) in “Palmer’s backyard” (Gao et al., 2020). To avoid local contamination from the research station, a wind control system was set up to pause aerosol sampling when wind direction inside the sector ±60° from the direction of the station and wind speed <2 m s⁻¹. Due to extremely low concentrations of aerosol trace elements over Antarctica, the sampling duration of each sample was approximately one week (Table 1).

80 After each sampling, the MOUDI sampler was carried back to the lab in the research station for sample filter changing and sampler cleaning in a Class 100 cleanroom flow bench. Aerosol samples were stored frozen in pre-cleaned Petri dishes at -20°C before analyses. A total of 8 sets size-segregated aerosol samples were collected on Teflon filters (1 µm pore size, 47 mm diameter, Pall Corp., NY, USA). A full set of blank filters (n = 11) was mounted on the sampler, carried to the sampling platform without running the sampler, and thus defined as field blanks. Meteorological conditions were recorded *in situ* by a 85 weather station (Campbell Scientific, UT, USA) installed on the same platform (Table 1).

2.2 Chemical analyses

2.2.1 Trace elements in aerosols

Aerosol samples were analyzed for the concentrations of trace elements by an Element-1 sector field inductively coupled plasma mass spectrometer (SF-ICP-MS, Thermo-Finnigan, Bremen, Germany) at the Department of Marine and Coastal 90 Sciences of Rutgers University, following a strong-acid digestion method described in Gao et al. (2020). Elemental concentrations were determined for Al, P, Ca, Ti, V, Mn, Ni, Cu, and Zn. Briefly, a quarter of each sample filter was digested in a 15 mL Teflon vial (Savillex, MN, USA) with Optima Grade HF (0.1 mL) and HNO₃ (0.8 mL) (Fisher Scientific, NJ, USA). Sample digestion was performed on a uniform heating HPX-200 (Savillex, MN, USA) hot plate for 4 hours at 165 °C followed by complete evaporation of acids. Then, 2.0 mL 3% HNO₃ with 1 ppb Indium (In) solution was added to re-dissolve 95 the sample, and In was used as an internal standard to correct the instrument drift of ICP-MS. All the digestion processes were carried out in a HEPA filter-controlled Class 100 clean hood in the Atmospheric Chemistry laboratory at Rutgers University. The Teflon vials and test tubes used in this study were thoroughly acid-cleaned. To ensure the data quality, for each batch of



samples, at least two procedural blanks were processed in the same way as the samples to monitor for possible contamination. During the ICP-MS analysis, duplicate injections of sample solutions were made every ten samples to check the instrument precision. The recovery of this analytical protocol was estimated by 7 separate digestions of the Standard Reference Material (SRM) 1648a-urban particulate matter (National Institute of Standard and Technology, MD, USA) (Table 2). The method limits of detection (LOD) were calculated as three times the standard deviation of 11 field blanks and a 200 m³ representative sampling volume (Table 2). The medians of %blank in samples for detectable trace elements were calculated for quality control (Table 2). Elements with concentrations lower than the LOD, including Cr, Co, Cd and Sb, were measured but are not reported or discussed. Aerosol Fe concentrations and Fe solubility were measured in these samples and were reported in Gao et al. (2020). Therefore, they are not included in this paper.

2.2.2 Ionic tracers in aerosols

The concentrations of water-soluble Na and K in aerosols were analyzed by ion chromatography (IC) (ICS-2000, Dionex, CA, USA) with an IonPac CS12A (2 × 250 mm²) analytical column at the Atmospheric Chemistry laboratory at Rutgers University. The cations Na and K were used as tracers to estimate the portion of aerosols derived from seawater and biomass burning, respectively. Sample processing for IC analysis was similar to the method used by Zhao and Gao (2008) and Xu et al. (2013). Briefly, a quarter of the sample filter was transferred to a plastic test tube and leached with 5.0 mL Milli-Q water in an ultrasonic bath for 20 minutes at room temperature. Before being injected into the IC, the leachate was filtered through a PTFE syringe filter (0.45 µm pore size, VWR, PA, USA). The method LODs for Na and K based on 7 blanks and a 200 m³ representative sampling volume were 2 and 1 ng m⁻³, respectively. The precision of the analytical procedures based on seven spiked samples was <±1%.

2.3 Data analyses

2.3.1 Enrichment factors

To achieve an initial estimate of the possible sources for trace elements, enrichment factors relative to upper continental crust (EF_{crust}) were calculated, using the equation:

$$EF_{crust} = \frac{(X_i/X_{Al})_{sample}}{(X_i/X_{Al})_{crust}} \quad (1)$$

where (X_i/X_{Al})_{sample} is the mass concentration ratio of element i to the crustal reference element, Al, in aerosol samples, and (X_i/X_{Al})_{crust} is the abundance ratio of element i to Al in the upper continental crust (Taylor and McLennan, 1995). The crustal reference element Al has been widely used to calculate crustal enrichment factors in the Southern Ocean and Antarctica (Zoller et al., 1974; Lowenthal et al., 2000; Xu and Gao, 2014). When the EF_{crust} is greater than 10, the element likely has additional contributions from other sources (Weller et al., 2008).



2.3.2 Atmospheric dry deposition flux estimation

Dry deposition flux (F_d , $\text{mg m}^{-2} \text{ yr}^{-1}$) of each element in aerosols was calculated from the air concentration (C_e , ng m^{-3}) and dry deposition velocity (V_d , cm s^{-1}):

$$F_d = 0.315 \times V_d \times C_e \quad (2)$$

where 0.315 is a unit conversion factor (Gao et al., 2013). The V_d for each trace element was computed by dry deposition rates (V_{di} , cm s^{-1}) and particle distribution ratios (P_{dri} , %) following the equation:

$$V_d = \sum V_{di} \times P_{dri} \quad (3)$$

The P_{dri} was derived from the concentrations of trace elements in different size fractions and V_{di} was estimated using a combination model of Williams (1982) and Quinn and Ondov (1998). This model includes the effects of wind speed, air/water temperature difference, sea surface roughness, spray formation in high wind speed and relative humidity. Meteorological parameters used for estimating dry deposition rates were measured *in situ* by the weather station with 1 min temporal resolution that was converted to 1 h averages. Sea surface temperature data was obtained from the Palmer Station Long-Term Ecological Research (LTER) study (<https://oceaninformatics.ucsd.edu/datazoo/catalogs/pallter/datasets/28>). Dry deposition rates of coarse-mode particles were dominated by gravitational settling, whereas the dry deposition rates of smaller particles were controlled by environmental factors (Figure 2). The estimation of dry deposition flux carries substantial uncertainty due to the limited sample masses and the assumptions inherent to the V_{di} estimation (Duce et al., 1991; Gao et al., 2020). Dry deposition fluxes were calculated for the trace elements showing clear particle size distribution patterns, including Al, P, Ca, Ti, V, Mn, Ce, and Pb. For Ni, Cu, and Zn that didn't show clear size distributions, however, the ranges of their dry deposition fluxes were also estimated by applying their mean concentrations to the lowest (Pb, $0.11 \pm 0.12 \text{ cm s}^{-1}$) and highest (Al, $0.49 \pm 0.28 \text{ cm s}^{-1}$) dry deposition velocities. The dry deposition fluxes of dust were also estimated based on the concentrations and particle size distributions of Al in aerosols, assuming that Al accounted for 8% of dust mass (Taylor and McLennan, 1995).

2.3.3 Air mass back trajectories

To explore possible source regions of air masses affecting trace elements in aerosols collected at Palmer Station, the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) was used to calculate 72-hour air mass back trajectories for each sampling duration (Rolph et al., 2017). In this study, the HYSPLIT model was driven by the meteorological data from the Global Data Assimilation System (GDAS) with a 0.5-degree resolution. Each air mass back trajectory was calculated at 3-hour intervals and started from one-half mixed boundary layer height. The back trajectories during each sampling period were used to calculate trajectory frequencies which were defined by the following equation:

$$\text{Trajectory frequencies} = 100 \times \frac{\text{number of endpoints per grid square}}{\text{number of trajectories}} \quad (4)$$



3 Results and discussion

3.1 Enrichment factors of trace elements

Crustal enrichment factors of trace elements in aerosols were calculated as the first step of source identification (Figure 3). Two major EF groups were found, representing crustal and non-crustal elements as follows.

3.1.1 Crustal elements (P, Ti, V, Mn, Ce)

The values of EF_{crust} for Ti, V, Mn, and Ce in aerosol samples were less than 10, indicating that a crustal source is the dominant source for those elements (Figure 3). As typical lithogenic elements (Boës et al., 2011), Ti and Mn in aerosols over oceanic regions are usually derived from natural dust emissions (Shelley et al., 2015; Marsay et al., 2018; Buck et al., 2019). Likewise, V and Ce in aerosols over the South Pole were reported dominated by crustal weathering (Zoller et al., 1974). In addition to crustal emissions, long-range transport may deliver some portion of these trace elements from remote sources to Antarctica (Wagener et al., 2008). For example, aerosol Ce and Mn derived from anthropogenic emissions were thought to be contributed by additives in vehicle fuels (Fomba et al., 2013; Gantt et al., 2014), and V in aerosols was found associated with ship emissions due to the use of heavy oil fuel (Chen and Duce, 1983; Rahn and Lowenthal, 1984; Zhan et al., 2014). However, the EF_{crust} results from this study suggest that nearby fuel combustion did not cause significant enrichment of V in aerosols at Palmer Station. A similar phenomenon was observed at McMurdo Station where light-weight fuel oil was used that was not a significant source of V (Lowenthal et al., 2000). We conclude that Ce, Mn, V, and Ti observed at Palmer Station were derived primarily from crustal sources.

The range of EF_{crust} for P was between 2 and 8, relatively higher than that of the other crustal elements. In Antarctic soils, P has been widely studied (Campbell and Claridge, 1987; Blecker et al., 2006; Prietzel et al., 2019). Around the Antarctic coast, including the northern end of the Antarctic Peninsula, high P inputs to the surface soil were found in seabird colonies (Otero et al., 2018), and a high enrichment of P ($EF_{\text{crust}} = 33$) was reported previously at King George Island (Artaxo et al., 1990). The closest potential source, the penguin colony on Torgerson Island, is only about 1 km from Palmer Station. Given that regional wind-induced dust likely affects aerosol composition over the Antarctic Peninsula (Asmi et al., 2018; Gao et al., 2020), soil-derived P is likely to be emitted to the atmosphere. In addition, biogenic activities in Antarctica also produce abundant gaseous P, such as phosphine, through anaerobic microbial processes in soils and animal digestives, and phosphine gas can be transformed to other low-volatile P-containing compounds in the atmosphere or soils (Zhu et al., 2006). A portion of P could derive from anthropogenic emissions, such as agricultural and industrial activities, through long-range transport as observed over the East China sea with EF_{crust} of 35 for P (Hsu et al., 2010). Primary biogenic aerosols, sea-salt aerosols, and volcanic emissions could also contribute P to the atmosphere, causing an elevated EF_{crust} for P (Zhao et al., 2015; Trabelsi et al., 2016).



3.1.2 Non-crustal elements (Ca, Ni, Cu, Zn, Pb)

The enrichment factors of atmospheric Ni, Cu, Zn, and Pb relative to the crustal element Al, were found to be greater than 10 in some samples, suggesting contributions from non-crustal sources during the corresponding sampling periods in this study (Figure 3). High enrichments of these elements have commonly been found in regions where the aerosol composition is largely controlled by long-range transport, including the polar regions (Boutron and Lorius, 1979; Maenhaut et al., 1979; Shevchenko et al., 2003; Xu and Gao, 2014; Zhan et al., 2014; Kadko et al., 2016). Aerosol Cu, Zn, and Pb are primarily associated with non-ferrous metal production (Pacyna and Pacyna, 2001; Shevchenko et al., 2003; Laing et al., 2014). Strong variations in the EF_{crust} of aerosol Cu, Zn, and Pb observed at the Dome C, Antarctica, over the 20th century, were attributed to volcanic activities (Boutron and Lorius, 1979). On the other hand, heavy oil combustion was found to be the major source of aerosol Ni and V, and V/Ni ratios are usually used to identify shipping emissions in Sweden (Isakson et al., 2001). Nevertheless, the V/Ni measured at Palmer Station ranged from 0.01 to 0.2, much lower than the $V/Ni = 3.2 \pm 0.8$ characteristic of the discharge from ship engines (Viana et al., 2009; Viana et al., 2014; Celo et al., 2015). Hence, despite the recent increase in tourist ship traffic, it looks that Palmer Station was barely impacted by ship emissions, which is consistent with the EF_{crust} of V. As a common crustal element, Ca accounts for about 3.5% of the weight of Earth's crust, while Ca is also a conservative major ion in seawater. The EF_{crust} of Ca in aerosol samples collected during this study varied from 11 to 48, indicating an enrichment from other sources.

3.2 Concentrations of trace elements

The concentrations of Ca and Al, the two major elements measured in this study, were one to several orders of magnitude higher than other elements (Table 3). To place all of the results of this study into the context of past investigations, we provide a visual comparison of measured concentrations of aerosol trace elements over Antarctica (Figure 4).

3.2.1 Crustal elements (Al, P, Ti, V, Mn, Ce)

The concentrations of Al in aerosols varied from 1.2 to 7.9 ng m⁻³ with an average of 4.3 ng m⁻³ during the period of this study. These concentrations are lower than the mean Al values of ~13 ng m⁻³ reported previously at King George Island (62.02° S, 58.21° W), the northern end of Antarctic Peninsula (Artaxo et al., 1992), but were slightly higher than the 2-year mean Al concentrations of 1.9 ng m⁻³ observed at King Sejong Station (62.13° S, 58.47° W) (Mishra et al., 2004), ~385 km northeast of Palmer Station, the mean of 0.194 ng m⁻³ observed at Larson Ice Shelf to the southeast of Palmer Station (Dick et al., 1991), the 5-year summer mean of 1.3 ng m⁻³ in East Antarctica (Weller et al., 2008) and the summer average of 0.57 ng m⁻³ measured at the South Pole (Zoller et al., 1974; Maenhaut et al., 1979) (Figure 4b). All these results were much lower than the average Al concentrations of 180 ng m⁻³ and 250 ng m⁻³ observed on the two sites at McMurdo Station (Mazzera et al., 2001) due to the impact of the nearby McMurdo Dry Valleys (Figure 4b). They are also lower than the average Al concentration 190 ng m⁻³ observed at Coastal East Antarctica where the samples were also impacted by air masses passing over McMurdo Dry Valleys



(Xu and Gao, 2014) (Figure 4b). The concentrations of Ti and Mn ranged from 140 to 800 pg m⁻³ with an average of 250 pg m⁻³, and from 17 to 44 pg m⁻³ with an average of 30 pg m⁻³, respectively. Both Ti and Mn concentrations at Palmer Station were lower than at King George Island (average Ti: 1600 pg m⁻³, average Mn: 660 pg m⁻³) (Artaxo et al., 1992) and McMurdo Station (average Ti: 26000 pg m⁻³, average Mn: 3000 pg m⁻³) (Mazzera et al., 2001) but comparable to the concentrations in PM_{2.5} samples collected at 2.5 km southeast of the Chilean Bernardo O'Higgins base located on the northwest coast of the Antarctic Peninsula (Préndez et al., 2009) and the concentrations in high-volume cascade impactor samples collected in coastal East Antarctica (Mn: 450–1200 pg m⁻³ with an average of 700 pg m⁻³) (Xu and Gao, 2014) (Figure 4e and g). The concentrations of V ranged from 2.7 to 6.1 pg m⁻³ with a mean value of 4.2 pg m⁻³, which is higher than the numbers reported at the South Pole (average ~1.5 pg m⁻³) (Zoller et al., 1974; Maenhaut et al., 1979) (Figure 4f). However, the V concentration observed at Palmer Station was much lower than previous observations in North Atlantic Ocean (50–3170 pg m⁻³) (Fomba et al., 2013) and eastern Pacific Ocean (average 150 pg m⁻³) (Buck et al., 2019). On the other hand, Ce demonstrated an average concentration of 1.3 ± 0.69 pg m⁻³, which is consistent with the Ce concentrations reported at Neumayer Station, Antarctica (Weller et al., 2008) (Figure 4k). The low concentrations of aerosol V and Ce suggest that Palmer Station was not significantly influenced by fossil fuel combustion. The P concentrations in aerosols during this study ranged from 85 to 250 pg m⁻³ with an average of 150 ng m⁻³. The concentrations of aerosol P at Palmer Station were lower than that at King George Island (3820 pg m⁻³) (Artaxo et al., 1990). Further, the P values from this study are much lower than the P concentrations previously observed on the east coast of Asia (Cohen et al., 2004), the Australian coast (Maenhaut et al., 2000; Vanderzalm et al., 2003), and Europe (Virkkula et al., 1999) which have been heavily affected by biomass burning, industrial activities and other anthropogenic sources. Comparing global aerosol P concentrations, we find that P concentrations over the Antarctic Peninsula are in the same range as those over the Central Pacific Ocean (Chen, 2004). Confirming that Palmer Station was little influenced by aerosols derived from biomass burning through long-range transport, the calculated non-sea-salt-K was indistinguishable from zero. These results agree well with the air mass back trajectories, which indicate that most samples collected during this study were barely affected by South America (Figure 5). In addition, for most of the 72-hour air-mass back trajectories, the highest frequencies were found around northern Antarctic Peninsula, suggesting that aerosol crustal elements observed at Palmer Station were impacted by sources in that region (Figure 5).

3.2.2 Non-crustal elements (Ca, Ni, Cu, Zn, Pb)

The highest concentration of Ni observed during this study was up to 320 pg m⁻³ with an average of 75 pg m⁻³, while the lowest Ni concentration was below LOD (<20 pg m⁻³). Samples M2 and M10 showed relatively high values of Ni with 320 and 200 pg m⁻³, respectively, while the concentrations of Ni in M1, M5, and M6 were much lower, ranging from 17 to 37 pg m⁻³. The concentrations of Cu varied from <20–480 pg m⁻³ (average 150 pg m⁻³), while Zn ranged from <30–710 pg m⁻³ (average 200 pg m⁻³). These results indicate that the concentrations of these elements in aerosols varied dramatically throughout the study period, likely affected by the source strength and meteorological conditions. Artaxo et al. (1992) collected aerosol samples using a 2-stage size-segregated sampler and observed high concentrations of Ni, Cu, and Zn with averages of 240, 790 and



250 7200 pg m^{-3} at King George Island in summertime (Figure 4h, i and j), which were much higher than the results from this study. Those high concentrations may result from local polluted dust emissions around their sampling site (Hong et al., 1999). In addition, their sampling site was considerably north of Palmer Station, and may be more likely to be impacted by air masses from South America (Chambers et al., 2014). At McMurdo Station, the average concentrations of Cu and Zn in PM_{10} samples were 200 and 1200 pg m^{-3} (Mazzera et al., 2001) (Figure 4i and j). Over coastal East Antarctic, the Ni concentrations in aerosols
 255 ranged from <3 to 2200 pg m^{-3} with an average of 750 pg m^{-3} (Xu and Gao, 2014) (Figure 4h). The concentrations of aerosol Pb observed at Palmer Station ranged from 5.0 to 60 pg m^{-3} with an average of 19 pg m^{-3} , higher than the average concentration of 4.7 pg m^{-3} previously observed on the east coast of the Antarctic Peninsula in austral summer (Dick, 1991) (Figure 4l). However, the average Pb concentration reported at King George Island was about 800 pg m^{-3} (Artaxo et al., 1992), considerably higher than observed in this study (Figure 4l). The two highest Pb concentrations observed during this study were in M4 (60
 260 pg m^{-3}) and M5 (30 pg m^{-3}). The results of air mass back trajectories for M4 and M5 show that part of the air masses was derived from South America, which suggests additional contribution from long-range transport (Figure 5b and c). However, when the 72-hour air mass back trajectories did not intersect the South American continent, the concentrations of Pb in the samples under that condition were significantly lower (Figure 5a and d). The low concentrations of heavy metals observed during this study suggest that local anthropogenic emissions were negligible. Thus the major source of non-crustal elements
 265 in aerosols over the study region may be long-range transport from regions impacted by anthropogenic emissions.

The concentrations of aerosol Ca were the highest among all elements measured in this study (range 16–53 ng m^{-3} and mean value of 30 ng m^{-3}). The mean value of Ca is close to the mean Ca concentrations observed previously in coastal regions of the Antarctic Peninsula (Artaxo et al., 1992; Weller et al., 2008) (Figure 4d). In addition, Na, as a tracer of sea-salt, and K, as a tracer of biomass burning (Zhu et al., 2015), were used to further evaluate the contribution of trace elements from other
 270 sources. The concentrations of Na and K showed average concentrations of 890 ± 310 and 28 ± 11 ng m^{-3} , respectively. The Na/K and Na/Ca ratios were 32 ± 3.5 and 31 ± 5.5 which are close to their average mass ratios, 27 and 26, in seawater (Millero, 2016). The results suggest that Ca was dominated by sea-salt aerosol and Palmer station was barely affected by the biomass burning.

3.3 Aerosol particle-size distributions of trace elements

275 The concentrations of trace elements in aerosols observed during this study varied as a function of particle size (Figure 6). Trace elements in aerosols can be classified into three groups based on their potential dominant sources, with each group showing unique size distribution pattern: (1) elements from the continents, due to crustal weathering and wind-induced resuspension, (2) elements from combustion or other anthropogenic sources, as a result of long-range transport, and (3) elements from the ocean, through bursting bubbles of seawater, by which droplets are ejected into the atmosphere.

280 The first group includes Al, P, Ti, V, Mn, and Ce, which were derived from crustal sources and accumulated in the coarse-mode particles. This group was dominated by particles with diameters larger than $2.5 \mu\text{m}$ (Figure 6), consistent with the coarse-mode dominance of natural dust (Adebiyi and Kok, 2020). For Al, the highest mass concentrations in sample M1, M2, M4 and



M7 were found in the $>18\text{ }\mu\text{m}$ fraction whereas in the other 4 sample sets, Al peaked at around $2.5\text{--}7.8\text{ }\mu\text{m}$. The notably high concentrations of Al in coarse-mode particles found in M1, M2 and M4 may indicate stronger contributions from regional or local crustal materials. In most sample sets, P, Ti, V, Mn and Ce peaked at around $2.5\text{ to }7.8\text{ }\mu\text{m}$. Additionally, M2 and M4 showed bimodal distribution for V which had a small peak in fine-mode at around $0.14\text{--}0.44\text{ }\mu\text{m}$. A similar distribution was found for Mn in sample M4. Such enrichments of V and Mn in fine-mode particles might hint at a minor contribution of aerosols from distinct sources, such as ship emissions (Lowenthal et al., 2000) or long-range transport of dust and biogenic aerosols (Artaxo et al., 1994; Weller et al., 2008).

The mass distributions of Ni, Cu, Zn, and Pb as the second group were different from those of the elements derived from crustal sources (Figure 6). Because of the low concentrations and the resulting high blank corrections, the particle size distribution of Ni, Cu, and Zn was not obtained. The mass distribution of Pb showed either a single mode or bimodal distributions in most samples with Pb being accumulated in the fine-mode particles, peaking at around $0.078\text{--}0.25\text{ }\mu\text{m}$. The enrichment of Pb in fine particles is consistent with relatively long residence time and transport distances in the atmosphere (Seinfeld and Pandis, 1998); this was particularly evident in the size distribution of M4, which was impacted by air masses derived from South America (Figure 5c). In addition, sample M4 showed trimodal distribution for Pb, exhibiting 3 peaks at around $0.078\text{--}0.25\text{ }\mu\text{m}$, $0.78\text{--}1.4\text{ }\mu\text{m}$, and $4.4\text{--}7.8\text{ }\mu\text{m}$, suggesting that additional Pb was contributed by distinct sources. The fine-mode Pb exhibited the highest concentration in most samples. Unlike the mass distributions of Pb, other non-crustal elements, including Ni, Cu, and Zn, did not clearly accumulate on either fine or coarse-mode particles, which may indicate that the dominant source of these elements varied during the sampling period.

The mass distributions of sea-salt elements (Ca, Na and K) as the third group were dominated by coarse-mode particles with diameters $2.5\text{--}7.8\text{ }\mu\text{m}$ (Figure 6), consistent with sea salt aerosols observed elsewhere (Lewis et al., 2004; Zhao and Gao, 2008; Xu et al., 2013). In this study, the correlation between the total concentrations of Ca and Na was strongly positive ($R^2 \pm 0.82$, $p\text{-value} < 0.01$), suggesting a strong contribution of Ca from sea salt aerosols.

3.4 Atmospheric dry deposition fluxes

Aerosol particle size distributions have been used to estimate atmospheric dry deposition fluxes of aerosol Fe to the Southern Ocean and coastal Antarctica (Gao et al., 2013; 2020). Similarly, the dry deposition fluxes of aerosol trace elements were estimated in this study (Table 4). Given that the sea-salt elements were the most abundant, Ca showed the highest dry deposition flux which varied from $0.59\text{ to }6.6\text{ mg m}^{-2}\text{ yr}^{-1}$ with an average of $1.2\text{ mg m}^{-2}\text{ yr}^{-1}$. At the other extreme, due to low concentration and fine-mode particle dominance, aerosol Pb showed ~ 5000 -fold lower dry deposition fluxes with a mean of $0.30 \pm 0.23\text{ }\mu\text{g m}^{-2}\text{ yr}^{-1}$. The rough estimates of the dry deposition fluxes of Ni, Cu, and Zn at Palmer Station are close to the low deposition fluxes found in the North Atlantic Ocean (Shelley et al., 2017) but considerably lower than the other measurements conducted in mid- and low-latitude regions (Arimoto et al., 2003; Buck et al., 2019). The estimated dry deposition fluxes of total continental dust at Palmer station ranged from $0.65\text{ to }28\text{ mg m}^{-2}\text{ yr}^{-1}$ with a mean of $5.5\text{ mg m}^{-2}\text{ yr}^{-1}$, among the lowest globally (Lawrence and Neff, 2009). Previous modelling studies estimated that wet deposition of dust



through precipitation scavenging accounted for about 40~60% of the total deposition in the coastal and open oceans in the mid and low latitude oceans (Gao et al., 2003), and similar wet deposition fraction was also found in the Southern Ocean and Coastal East Antarctica (Gao et al., 2013). Assuming this wet deposition fraction applies to the Antarctic Peninsula region, we approximate roughly a total dust flux of $10 \text{ mg m}^{-2} \text{ yr}^{-1}$. This value is at the lower end of the dust flux range, ~ 5 to $\sim 50 \text{ mg m}^{-2} \text{ yr}^{-1}$, estimated from ice core measurements at James Ross Island on the Antarctic Peninsula over the 20th century (McConnell et al., 2007), but it is higher than dust deposition flux at Dome C ($0.2\text{--}0.6 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Lambert et al., 2012), at Talos Dome ($0.70\text{--}7.24 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Albani et al., 2012) and the model estimate for this region ($1.8\text{--}3.7 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Wagener et al., 2008). However, this result is far lower than the total dust deposition fluxes in the North and South Atlantic Ocean (minimum 270 and $150 \text{ mg m}^{-2} \text{ yr}^{-1}$, respectively) (Barraqueta et al., 2019), the South Pacific Ocean ($230 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Prospero, 1989), South Indian Ocean ($240 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Heimbürger et al., 2012) and McMurdo Dry Valleys ($490 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Lancaster, 2002), while it is close to the estimated total dust deposition fluxes for the 63°S region (between $62^\circ 53'\text{S}$ and $63^\circ 53'\text{S}$, along $170^\circ 6'\text{W}$; $33 \text{ mg m}^{-2} \text{ yr}^{-1}$) (Measures and Vink, 2000). Accordingly, the other crustal elements (P, Ti, V, Mn, Ce) were proportional to the dust dry deposition flux and showed extremely low values.

To examine the potential importance of atmospheric dust input to the particulate elemental concentrations in surface waters of the west Antarctic Peninsula shelf region, the maximum possible suspended particulate Al concentrations contributed by dust were estimated. We used the maximum Al dry deposition flux of $2300 \mu\text{g m}^{-2} \text{ yr}^{-1}$ (Table 4), assumed that dry deposition accounted for 40% of total deposition and assumed no settling loss from the mixed layer of mean depth 24 m (Eveleth et al., 2017) over a 4-month summer season. By this calculation, the accumulated concentration of suspended particulate Al contributed by total atmospheric deposition over this period is 3 nmol kg^{-1} . While suspended Al reaches 135 nmol kg^{-1} in coastal surface waters close to the peninsula, outer shelf and off-shelf surface waters generally have concentrations $< 5 \text{ nmol kg}^{-1}$ (Annett et al., 2017), suggesting that this upper limit estimated dust flux could account for a substantial portion of observed surface ocean lithogenic particle concentration. However, the mean Al flux is just 20% of this maximum, and settling loss from the mixed layer over the course of the summer is likely significant. Still, this rough estimate suggests that atmospheric dust deposition, possibly dominated by regional sources on the Antarctic continent itself, may contribute a significant fraction of suspended particulate concentrations of lithogenic elements in the surface waters of the outer shelf and the proximal pelagic Southern Ocean.

4 Conclusions and Implications

Results from this study indicate that trace elements in aerosols over Palmer Station are derived primarily from crustal and marine sources. Crustal elements (Al, P, Ti, V, Mn, Ce) and sea-salt elements (Ca, Na, K) showed single mode distributions and were accumulated in coarse-mode particles, whereas aerosol Pb that was likely impacted by air masses from South America showed high EF_{crust} and was accumulated in finer mode particles. Most of the samples collected during this study were impacted by air masses originating around or passing over Northern Antarctic Peninsula, suggesting the regional influence on the



concentrations of aerosol trace elements observed at Palmer Station. Although the Antarctic Peninsula has experienced significant changes in the past decade, greater exposure of regional continental dust sources through glacial ice loss and more frequent shipboard tourism do not seem to drive the concentrations of these trace elements over the west Antarctic Peninsula to higher values than observed in other remote Antarctic sites. In addition, the total dust deposition flux ($\sim 10 \text{ mg m}^{-2} \text{ yr}^{-1}$) estimated from the Al concentrations in aerosols obtained from this study and the assumption on the wet deposition suggests that dust deposition plays only a minor role in determining the concentrations of trace elements in coastal seawater around the west Antarctic Peninsula but may be more important in offshore regions. As the role of wet deposition is unquantified at present and remains poorly constrained for this region, however, the total deposition fluxes of trace elements may be underestimated, and the importance of the atmospheric deposition of trace elements to the adjacent seawater may need to be re-evaluated.

Data availability. The data used in this paper has been deposited to the U.S. Antarctic Program Data Center. The DOI will be issued and the data will be freely accessible shortly.

Author contributions. YG conceived the research. YG and SY prepared field sampling and collected aerosol samples. SF and YG digested samples. KB and RMS conducted sample analyses. SF wrote the 1st draft of the manuscript. YG and RMS edited the drafts. All authors contributed to writing and approved the submission.

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

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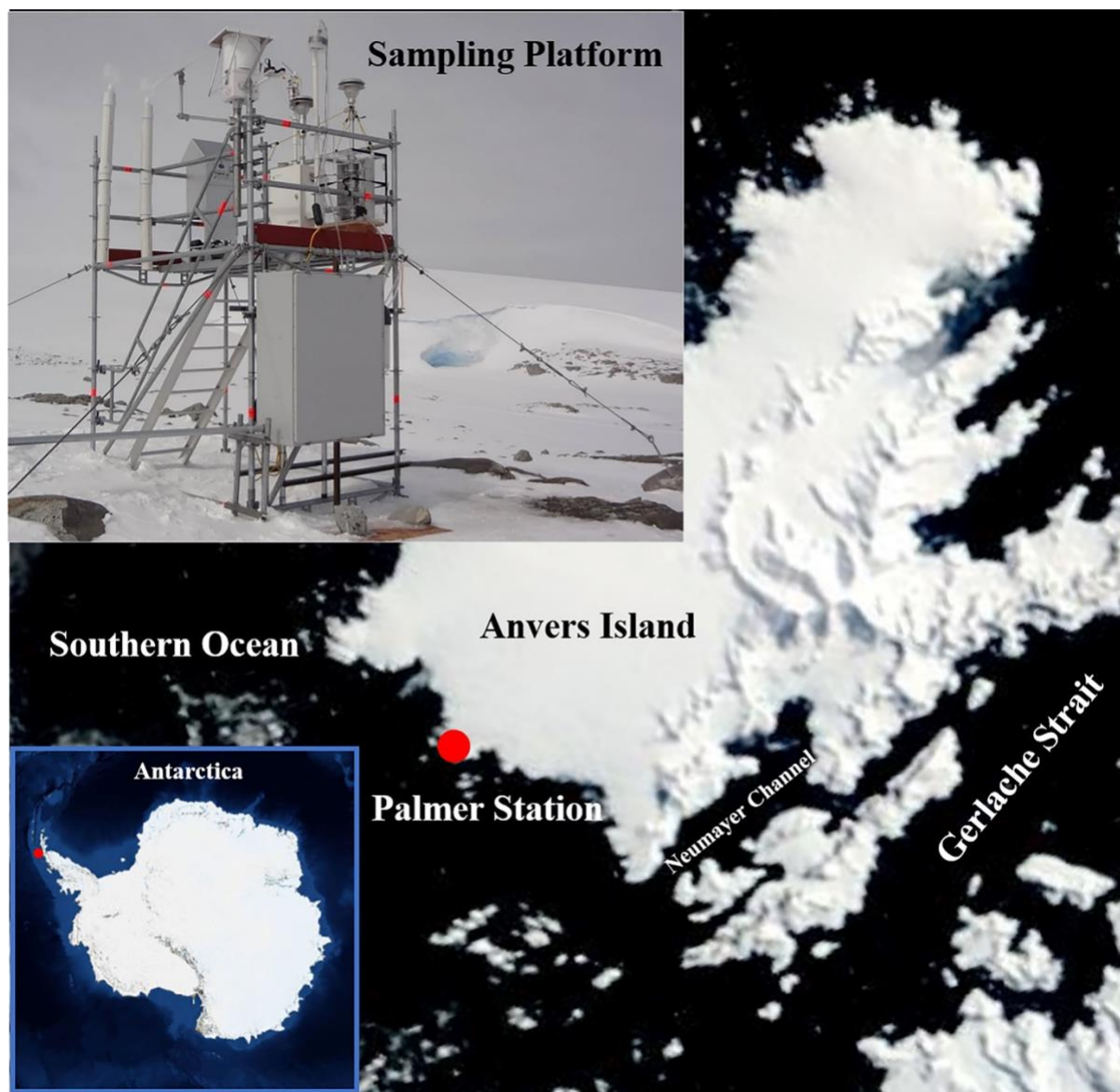
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600 **Figure 1: Sampling site at Palmer Station (red dot) with inset photograph of sampling platform (Gao et al., 2020) (satellite image credits: NASA).**

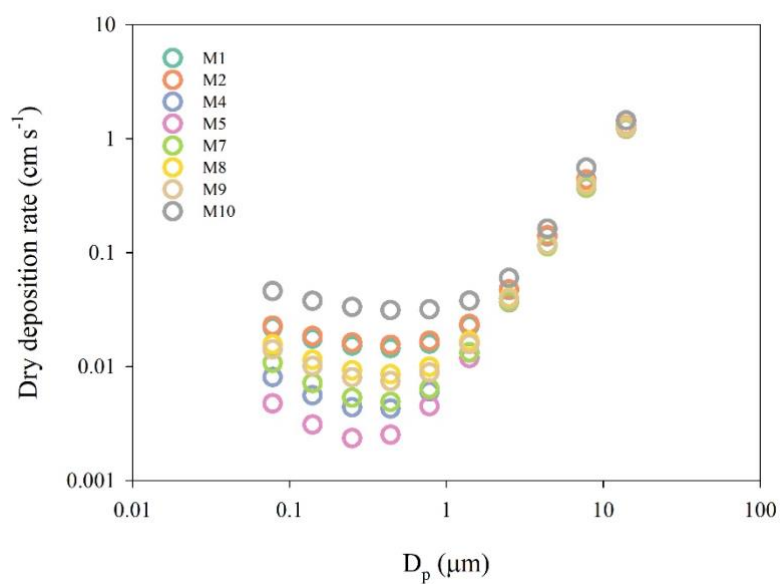
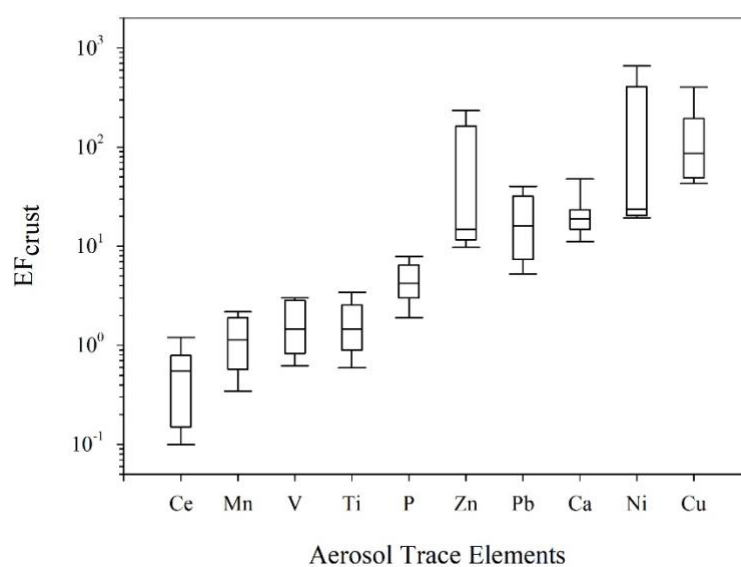


Figure 2: Dry deposition rates of aerosol particles during sampling periods.



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Figure 3: Box plots of EF_{crust} for trace elements in aerosols collected at Palmer Station during this study. The central horizontal line is the mean value whereas the bottom and the top of each box are the 25th and 75th percentiles. The upper and lower horizontal bars indicate the 5th and 95th percentiles of the data

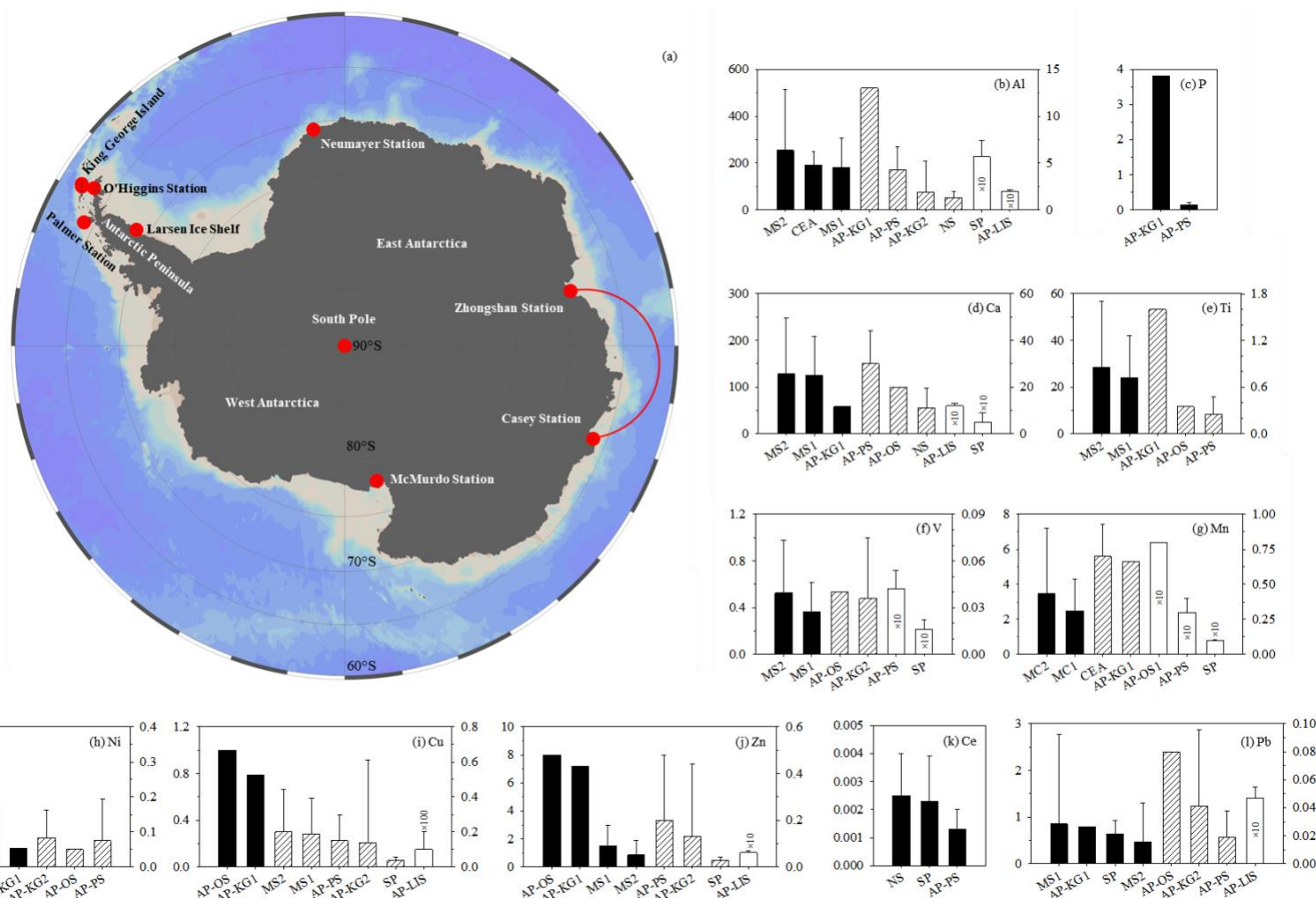


Figure 4: Comparison of the concentrations of aerosol trace elements (ng m^{-3}) over Antarctica. All concentrations are sorted in descending order from left to right. The left y-axis shows concentrations for the black bars, whereas the right y-axis corresponds to the striped and white bars. Some extremely low values are multiplied by 10 or 100 for display purposes, as marked above the corresponding white bars. Data are from observations conducted at South Pole (SP) (Zoller et al., 1974), Hut Point site (MS1) and Radar Sat Dome site (MS2) at McMurdo Station (Mazzera et al., 2001), on a cruise between Zhongshan Station and Casey Station in the Coastal East Antarctica (CEA) (Xu and Gao, 2014), at Neumayer Station (NS) (Weller et al., 2008), and at 5 sites on the Antarctic Peninsula (AP), including Comandante Ferraz Antarctic Station (KG1) (Artaxo et al., 1990; Artaxo et al., 1992) and King Sejong Station (KG2) (Mishra et al., 2004) at King George Island, O'Higgins Station (OS) (Préndez et al., 2009), Larsen Ice Shelf (LIS) (Dick, 1991), and Palmer Station (PS, this study) at Anvers Island.

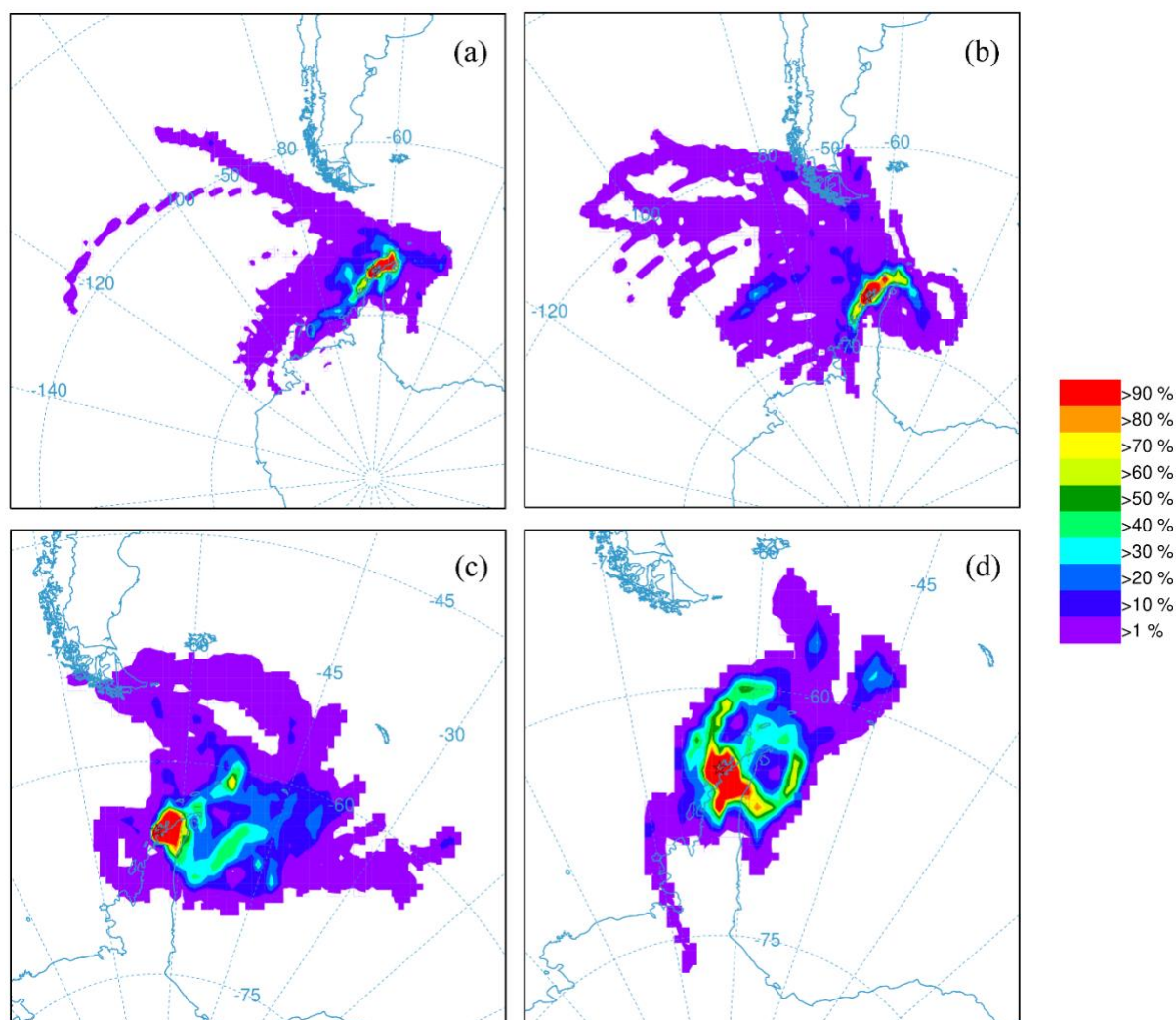


Figure 5: Frequencies of 72-hour air mass back trajectories for samples collected at Palmer Station: (a) sample M2 (Nov 23 – Dec 4, 2016), (b) sample M4 (Dec 6 – Dec 17, 2016), (c) sample M5 (Dec 15 – Dec 24, 2016), and (d) sample M7 (Dec 29, 2016 – Jan 8, 2017).

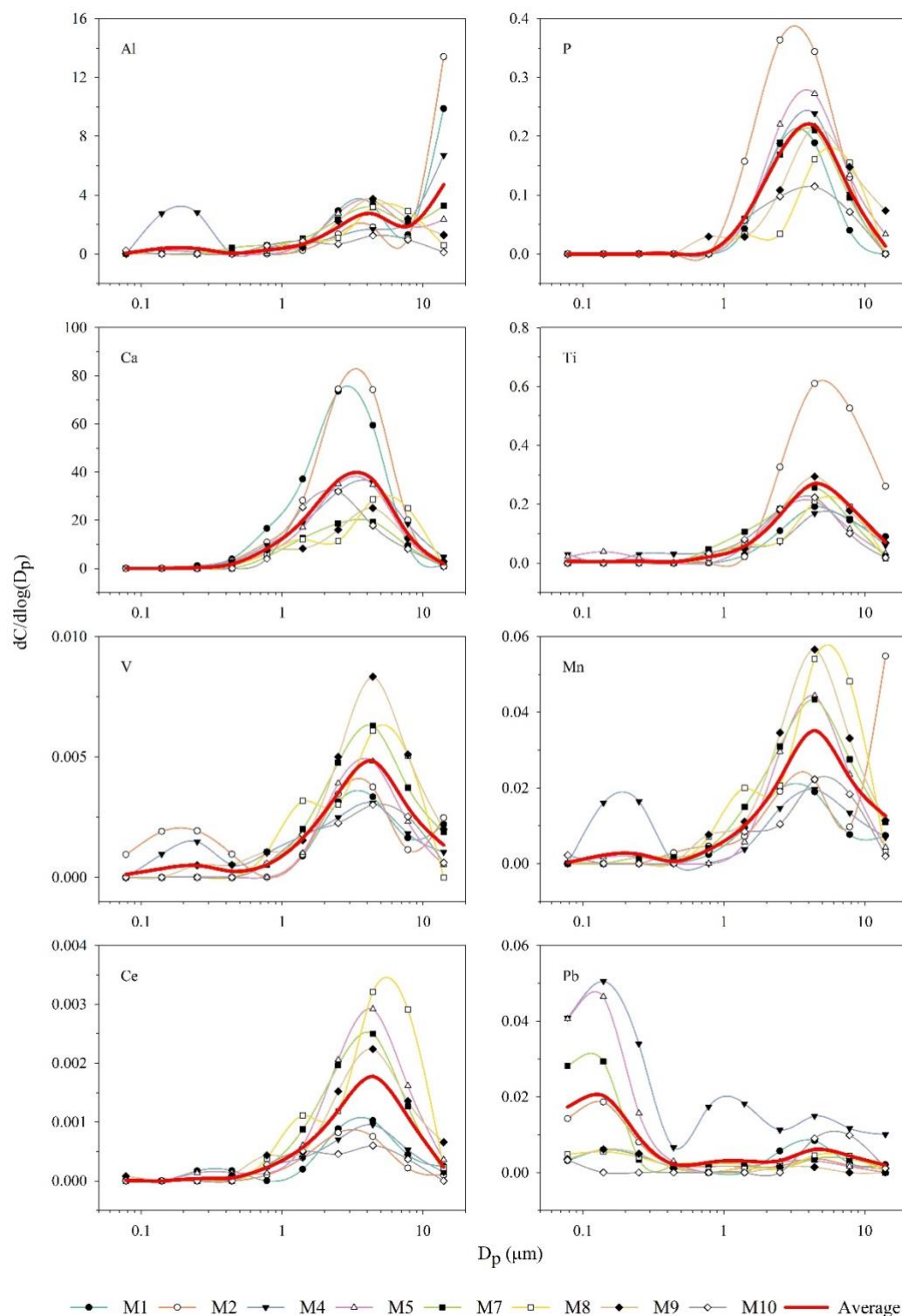


Figure 6: Particle size distributions of selected trace elements in aerosols over Palmer Station.



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Table 1. Sampling periods and meteorological conditions for individual samples.

Sample ID	Sampling Period	Sampling Volume (m ³)	Wind Speed (m s ⁻¹)	Air Temperature (°C)	Relative Humidity (%)	Air Pressure (hPa)	Precip. (mm d ⁻¹)	Solar Intensity (W m ⁻²)
M1	11/19/2016-11/26/2016	225	5.7 (0.7-22)	0.1 (-2.2-4.3)	87 (52-100)	990 (976-1011)	4.19	166 (0-675)
M2	11/26/2016-12/04/2016	233	4.9 (0.4-21)	0.7 (-2.2-5.8)	82 (45-100)	982 (959-997)	1.37	191 (0-847)
M4	12/09/2016-12/17/2016	266	3.5 (0.1-15)	0.7 (-3.3-5.6)	83 (53-100)	985 (969-998)	0.37	207 (1-854)
M5	12/17/2016-12/24/2016	264	2.5 (0.3-9.8)	2.0 (-0.7-5.5)	77 (49-100)	988 (972-996)	0.05	195 (1-723)
M7	01/01/2017-01/08/2017	244	4.1 (0.2-13)	1.8 (-0.7-3.8)	72 (49-99)	987 (972-997)	0.09	272 (0-857)
M8	01/08/2017-01/15/2017	283	5.4 (0.7-16)	2.6 (0.3-5.5)	64 (46-95)	982 (973-991)	0.00	225 (0-792)
M9	01/15/2017-01/23/2017	285	5.0 (0.2-15)	2.1 (-1.0-5.3)	65 (42-86)	987 (981-997)	0.15	260 (0-829)
M10	01/23/2017-01/30/2017	279	8.9 (0.7-21)	4.1 (1.7-7.1)	79 (65-90)	981 (969-997)	3.70	110 (0-594)



Table 2. Limits of Detection (LOD), %blank, and recovery of standard reference material 1648a using acid digestion and ICP-MS procedures.

Elements	LOD (pg m ⁻³)	%Blank (%)	Recov. (%)
	n=11	Median (n)	n=7
Al	80	5.8 (59)	96
P*	20	25 (31)	-
Ca	500	3.5 (57)	93
Ti	10	28 (52)	90
V	0.3	11 (55)	97
Cr	6	ND	38
Mn	1	6.1 (57)	97
Co	2	ND	91
Ni	20	22 (17)	99
Cu	20	19 (22)	93
Zn	30	26 (27)	107
Cd	0.5	ND	100
Sb	3	ND	85
Ce	0.1	10 (48)	86
Pb	0.7	17 (60)	82

*Lack of certified concentration in SRM 1648a prevents calculation of recovery.

ND= not determined in samples because LOD was not exceeded.

The LODs were estimated based on a 200 m³ representative sampling volume.

%Blank is the median field blank/sample concentration×100%, and n is the number of detectable samples.



Table 3. Concentrations of trace elements and ions in aerosols over Palmer Station, the west Antarctic Peninsula.

Element/Ion	Units	Size Fraction	M1	M2	M4	M5	M7	M8	M9	M10	Mean	S.D
Al	ng m ⁻³	Coarse	6.9	7.9	4.4	3.3	3.7	2.2	2.5	0.83	4.0	2.4
		Fine	ND	ND	1.6	0.13	0.30	0.11	0.17	0.41	0.34	0.53
		Sum	6.9	7.9	6.0	3.4	4.0	2.3	2.7	1.2	4.3	2.4
P	pg m ⁻³	Coarse	110	250	150	180	130	95	140	85	140	53
		Fine	ND	ND	ND	ND	ND	ND	15	ND	1.9	5.3
		Sum	110	250	150	180	130	95	160	85	150	54
Ca	ng m ⁻³	Coarse	42	47	25	24	14	18	15	20	26	12
		Fine	8.9	5.4	5.4	3.2	2.4	3.1	3.9	2.0	4.3	2.2
		Sum	50	53	31	27	16	21	19	22	30	14
Ti	pg m ⁻³	Coarse	150	800	120	150	170	130	190	94	230	230
		Fine	ND	ND	48	20	24	9.0	16	65	23	23
		Sum	150	800	170	170	190	140	210	160	250	220
V	pg m ⁻³	Coarse	3.2	3.6	2.4	3.1	4.8	4.1	5.3	2.4	3.6	1.1
		Fine	ND	1.4	1.2	ND	0.27	0.53	0.77	0.28	0.57	0.55
		Sum	3.2	5.0	3.7	3.1	5.0	4.6	6.1	2.7	4.2	1.2
Mn	pg m ⁻³	Coarse	17	42	15	27	32	34	35	14	27	10
		Fine	1.2	2.3	8.2	0.90	2.9	3.6	3.9	2.8	3.2	2.3
		Sum	18	44	23	27	35	38	39	17	30	10
Ni	pg m ⁻³	Coarse	ND	320	ND	17	23	ND	ND	ND	45	110
		Fine	37	ND	ND	ND	ND	ND	ND	200	30	70
		Sum	37	320	ND	17	23	ND	ND	200	75	120
Cu	pg m ⁻³	Coarse	170	200	17	ND	ND	ND	ND	120	63	86
		Fine	69	280	92	52	110	ND	36	34	84	86
		Sum	240	480	110	52	110	ND	36	150	150	150
Zn	pg m ⁻³	Coarse	40	68	32	590	280	25	ND	46	140	200
		Fine	51	ND	30	120	300	ND	ND	56	70	100
		Sum	90	68	61	710	580	25	ND	100	200	280
Ce	pg m ⁻³	Coarse	0.73	0.57	0.62	1.8	1.6	2.1	1.5	0.41	1.2	0.65
		Fine	0.084	0.051	0.12	0.13	0.12	0.19	0.26	0.14	0.14	0.064
		Sum	0.81	0.62	0.74	2.0	1.7	2.2	1.8	0.55	1.3	0.69



Pb	pg m ⁻³	Coarse	5.1	2.4	13.8	2.2	2.4	2.7	0.70	5.0	4.3	4.1
		Fine	3.8	12	46	28	17	4.5	4.3	1.7	15	16
		Sum	9.0	14	60	30	19	7.2	5.0	6.6	19	19
Na	ng m ⁻³	Coarse	1100	1000	900	870	430	450	520	730	749	260
		Fine	220	210	160	120	75	99	89	90	130	57
		Sum	1300	1200	1100	1000	510	550	610	820	890	310
K	ng m ⁻³	Coarse	34	39	34	31	14	12	17	24	26	10
		Fine	4.9	4.5	2.5	3.2	1.6	1.8	1.5	1.4	2.3	1.1
		Sum	39	43	37	34	15	14	19	25	28	11



Table 4. Estimates of dry deposition velocities and dry deposition fluxes of trace elements in aerosols over Palmer Station.

	Element	Velocity (cm s ⁻¹)		Flux (µg m ⁻² yr ⁻¹)	
		Range	Mean	Range	Mean
	Dust			650-28000	5500
EF<10	Al	0.14-1.6	0.49	52-2300	440
	P	0.092-0.52	0.17	3.3-33	7.5
	Ti	0.14-0.92	0.27	7.1-120	18
	V	0.14-0.85	0.26	0.16-0.77	0.32
	Mn	0.18-1.1	0.28	0.94-11	2.5
	Ce	0.10-0.65	0.20	0.017-0.31	0.084
EF>10	Ca	0.073-0.41	0.13	580-6600	1200
	Pb	0.0097-0.83	0.11	0.018-1.5	0.30
	Ni*		0.11-0.49	3.9-17	
	Cu*		0.11-0.49	6.0-25	
	Zn*		0.11-0.49	8.1-36	

*The range of dry deposition fluxes of Ni, Cu, and Zn was estimated based on Pb and Al dry deposition velocities, discussed in the text.