

Author's Response to Review #1:

We are grateful to Dr. Holly Winton for the valuable comments that helped us significantly improve this manuscript. All comments have been addressed on a point-by-point base. Changes we made based on these comments in the revised manuscript are **highlighted in red**. Below we first list the **Comment**, followed with our **Response** and **Modified text** to it.

Review #1:

Interactive comment on “Concentrations, Particle-Size Distributions, and Dry Deposition Fluxes of Aerosol Trace Elements over the Antarctic Peninsula” by Songyun Fan et al. Reviewed by Holly Winton

Major Comments:

Fan et al. report size segregated aerosol concentrations and dry deposition fluxes for a suite of trace elements and cations from a land-based station on the Antarctic Peninsula over the 2016-2017 austral summer. Aerosol iron solubility data from these samples has been previously published by Gao et al. (2020). While the timeseries is short (8 sets of size segregated samples each with a sampling duration of 1 week), the study fills a gap in data availability of aerosol trace element levels in this sector of Antarctica and also in our understanding of the deposition pathways. The authors have estimated dry deposition velocities for each trace element at the sampling site, providing a more reliable estimate of dry deposition fluxes than previous studies which typically use a generic value not necessarily appropriate for the region with a large uncertainty. The study also reports aerosol size distribution of the trace elements which is extremely valuable data, also scarce in this region, providing important insights into the source and transport pathways of aerosols in pristine airmasses. The manuscript is well written, has a logical structure and provides important data to a wide-ranging community of researchers from atmospheric chemists, biogeochemists and ice core scientists. I recommend publication in ACP after a few suggestions for improvement have been addressed. I hope the suggestions below will strengthen the manuscript.

Response: We appreciate Dr. Winton's evaluation of this study and agree that the revision is needed along the lines of her suggestions.

Suggestions for improvement:

Introduction: It would be helpful to add a short section on what is known about aerosol removal processes in the Southern Ocean and where the gaps are in our understanding. I also think it would be worth briefly summarizing the aerosol iron data here e.g. mineral dust source and probable source region.

Response: We agree with Dr. Winton. A brief explanation about aerosol removal mechanisms and current gaps in our understanding about aerosols over the Southern Ocean and Antarctica has been added in **lines 29-39**. A short summary of ice-free areas that may be potential dust source regions on the Antarctic Peninsula have been added in **lines 62-66**. A brief summary about what we found in our previously published Fe-focused study is added in **lines 66-69**.

Modified text:

lines 29-39: “In the atmosphere, the removal of aerosols involves gravitational settling, impaction, diffusion, hygroscopic growth, and scavenging by precipitation, and the rates of all these processes are dependent on the aerosol particle size (Saltzman, 2009). Over the Southern Ocean and Antarctica, aerosol particle size distributions have been studied (Gras, 1995; Järvinen et al., 2013; Xu et al., 2013; Kim et al., 2017; Herenz et al., 2019; Lachlan-Cope et al., 2020). Seasonal variations of the particle number concentrations were observed in the King Sejong Station, Antarctic Peninsula, and Halley Research Station on the Brunt Ice Shelf. The maximum and minimum of the particle number concentrations at two sites were found in the austral summer and austral winter, respectively (Kim et al., 2017; Lachlan-Cope et al., 2020). Due to the low background concentrations of aerosol particles, new particle formation has been suggested to substantially affect the annual aerosol concentration cycles (Lachlan-Cope et al., 2020). However, most of these studies focused on the physical characteristics of aerosol particle size; the size distributions of aerosol trace elements are still poorly understood, and at present only aerosol Fe has been characterized for particle-size distributions around Antarctica (Gao et al., 2013, 2020).”

lines 62-66: “Under conditions of low precipitation (Van Lipzig et al., 2004) and high wind speed (Orr et al., 2008), several ice-free areas on James Ross Island, off the east coast of the northern Peninsula could serve as local dust sources (Kavan et al., 2018), and such sources may contribute to the atmospheric loading of certain trace elements such as Fe (Winton et al., 2014; Gao et al., 2020). Similar ice-free areas were found in King George Island, Livingston Island, Anvers Island etc. in the Antarctic Peninsula region (Bockheim et al., 2013), and may act as potential sources of aeolian dust.”

lines 66-69: “As part of the current study, the particle size distribution of aerosol Fe measured at Palmer Station showed single mode distribution and was dominated by coarse particles, suggesting that local regional dust emission dominated the concentration of Fe in this region (Gao et al., 2020).”

Discussion: The discussion on aerosol sources and, in particular, particle size distribution needs clarification and further investigation. The particle size distribution is a great dataset but at the moment it hasn't been fully utilized. Incomplete referencing and a limited explanation of the particle distribution let the discussion down. The authors could improve this section with a statistical cluster analysis of the physical characteristics of particle size distributions and/or a thorough understanding/comparison of the Antarctic and Southern Ocean literature on this topic. Currently, there is no discussion on what the size distribution tells us about the atmospheric transport mechanisms of the individual trace elements. Also, the conclusions don't come through strongly enough in the discussion.

Response: We agree with Dr. Winton. The discussion on the particle size distribution has been rewritten. We used k-means clustering to classify the normalized particle size distribution of trace element/ionic species into 5 clusters to further discuss the primary

source of each element: (1) mineral dust (Ti, V, Mn, Ce), (2) Al, (3) Pb, (4) sea salt (Ca, Na⁺, K⁺), and (5) P. Trace elements were grouped by their particle size distributions and then discussed with respect to potential sources. Samples that showed unique size distribution pattern were discussed individually. In this part, we also added a comparison of aerosol size distributions with studies focused on the dust particle size distributions in snow, ice core, and aerosols in Antarctica. Please see the [section 3.3, lines 326-395](#). The method of k-means clustering has been added to [section 2.3.2, lines 155-163](#).

Modified text:

lines 326-395: Please see section 3.3.

lines 155-163: Please see section 2.3.2.

Conclusions and implications: It's fantastic to see the size distribution data. Do you have plans to continue aerosol monitoring at this land-based sampling station to make a long-term record of size segregated trace elements? In this section, you could recommendation future aerosol studies investigate the size distribution to provide additional information on the source and atmospheric transport. What are the implications for the underestimation and overestimation of dry deposition velocities in previous aerosol flux estimates and climate modelling for the region? What about the implications of your new particle size distribution data for new particle formation studies?

Response: We have revised the conclusions and implications. Please see the revised [section 4, lines 442-463](#). We are hoping to continue the measurements of aerosol chemistry in this region through future studies, particularly through long-term observations. The errors associated with the dry deposition velocities could cause a biased dry deposition flux estimation and mislead modeling results. The range of particle sizes (from ~0.1 – 10 μm) in our study is much larger than on the sizes critical for new particle formation (in the subnanometer – nanometer range), and thus it is difficult to address the new particle formation processes with our data. We cannot say much about the new particle formation process.

Modified text: lines 442-463: Please see section 4.

Throughout: Please make it clear throughout the study that you are reporting total acid digestible trace element concentrations and water soluble cations. Please report Ca and K as ions Ca²⁺ and K⁺ rather than elements throughout the manuscript.

Response: All elements except for Na⁺ and K⁺ were reported for the total acid digestible concentrations which were analyzed by ICP-MS. To avoid confusion, we have added charge to all water-soluble cations (Na⁺ and K⁺) in the text.

Technical corrections:

Line 11 Suggest replacing with “local mineral dust, long-range transported aerosol and sea salt aerosol.” What about local P biogenic emissions?

Response: We have revised this part by adding “regional crustal emissions, long-range transport and sea salt.” We have briefly mentioned about local P biogenic emissions. Please see [lines 12-13](#).

Modified text: lines 12-13: “...these elements are derived primarily from three sources: (1) regional crustal emissions, including possible resuspension of soils containing biogenic P, (2) long-range transport, and (3) sea-salt.

Line 26 Define course mode.

Response: We have revised this part of the text. Please see lines 28-29.

Modified text: lines 28-29: “..., and such information is critically needed in climate model for better estimating aerosol climate effects (Adebiyi and Kok, 2020).”

Line 32 Include sea salt as a source of aerosol.

Response: We have added sea salt as a source of trace elements (please see [line 43](#)).

Modified text: line 43: “In addition, sea salt emission, ...”

Line 37 A number of local dust sources in Antarctica are increasingly being quantified. See Delmonte et al. (2019) for a recent review. Delmonte, B., Winton, H., Baroni, M., Baccolo, G., Hansson, M., Andersson, P., Baroni, C., Salvatore, M.C., Lanci, L. and Maggi, V., 2020. Holocene dust in East Antarctica: Provenance and variability in time and space. The Holocene, 30(4), pp.546-558.

Response: We have cited Kavan et al. (2018); Delmonte et al. (2020) and added the local dust emission as a source of atmospheric trace elements to the Southern Ocean and Antarctica in addition to long-range transport (please see [lines 48-49](#)).

Modified text: lines 48-49: “Atmospheric trace elements over the Southern Ocean and Antarctica may derive from distant continental sources through long-range transport (Li et al., 2010) and local dust sources in certain areas (Kavan et al., 2018; Delmonte et al., 2020).”

Line 47 Include a reference to McMurdo Sound here as the dustiest region in Antarctica.

Response: We have cited Winton et al. (2014) as a reference measuring dust in McMurdo Sound (please see [line 64](#)).

Line 62 “: : new sources of aerosol trace elements by warming which exposes a greater area of ice-free land and by: : ”.

Response: We have revised this sentence (please see [lines 83-85](#)).

Modified text: lines 83-85: “The new observational data also provide insight into sources of aerosol trace elements, as influenced recently by warming, which exposes a greater area of ice-free land, and by the impact of human activities in this region.”

Line 68 Mention that a detailed description of the aerosol sampling and protocols to mitigate contamination in the pristine environment can be found in Gao et al. (2020).

Response: We have added a sentence to indicate that the detailed sampling protocols can be found in Gao et al. (2020) (please see [lines 91-93](#)).

Modified text: lines 91-93: “A detailed description of the aerosol sampling, including the protocols for mitigating contamination in the pristine environmental can be found in Gao et al. (2020).”

Line 100 Please report precision here or in Table 2.

Response: We have moved Table 2 to Supplement as Table S1. The precisions were estimated by duplicate measurement and reported in [Table S1](#). A sentence refers to Table S1 has been added in [lines 124-125](#).

Modified text: lines 124-125: “During the ICP-MS analysis, duplicate injections of sample solutions were made every ten samples to check the instrument precision (Table S1).”

Line 107 Did you measure other cations and anions on these samples? If you did, will these data be included in your US Antarctic Program Data Centre data publication? I understand they may not be relevant for this manuscript but I’m sure they would be beneficial to other researchers if the data is available. Why did you not measure Ca and K by ICP-MS?

Response: We measured other cations and anions. Those data will be used for another paper and will be uploaded to the US Antarctic Program Data Center shortly. In this study, we measured Ca by ICP-MS; the water-soluble cations Na⁺ and K⁺ were measured by IC. The main reason for measuring those elements by IC is that in most cases, only water-soluble Na⁺ and K⁺ are used as tracers for sea-salt and biomass burning, respectively.

Line 121 Why did you select Al over Ti as the crustal reference?

Response: Al has been used as the crustal reference by other investigators in many locations in Antarctica, such as the Antarctic Peninsula (Dick, 1991), McMurdo (Lowenthal et al., 2000), the South Pole (Zoller et al., 1974). Therefore, our results can be compared directly with others on the same basis. We have added these citations in the revised manuscript (please see [lines 151-153](#)).

Modified text: line 151-153: “The crustal reference element Al has been widely used to calculate crustal enrichment factors in the Southern Ocean and Antarctica (Zoller et al., 1974; Dick, 1991; Lowenthal et al., 2000; Xu and Gao, 2014).”

Line 128 “: : was calculated from the concentration of the trace element in the air and dry: : :”

Response: We have revised this sentence (please see [lines 165-166](#)).

Modified text: lines 165-166: “Dry deposition flux (F_d , $\text{mg m}^{-2} \text{yr}^{-1}$) of each element in aerosols was calculated from the concentration (C_e , ng m^{-3}) of the trace element in the air and dry deposition velocity (V_d , cm s^{-1}):”

Line 141 What are the environmental factors?

Response: For coarse particles, gravitational settling dominates the dry deposition velocity. In contrast, wind speed, relative humidity, air temperature, and sea surface temperature were considered as the environmental factors that control the dry deposition velocity of fine particles, although these factors also affect the dry deposition of coarse-particles. We have added those factors in the revised manuscript (please see [lines 178-179](#)).

Modified text: lines 178-179:”..., including wind speed, relative humidity, air temperature, and sea surface temperature (Figure 2)”

Line 142 Please report the uncertainty of your dry deposition velocity estimates. Also, where can the reader find the dry deposition velocity values for each element?

Response: We have reported the uncertainties in the revised manuscript; please see [lines 179-181](#). The values of dry deposition velocity for each element is reported in [Table 3](#).

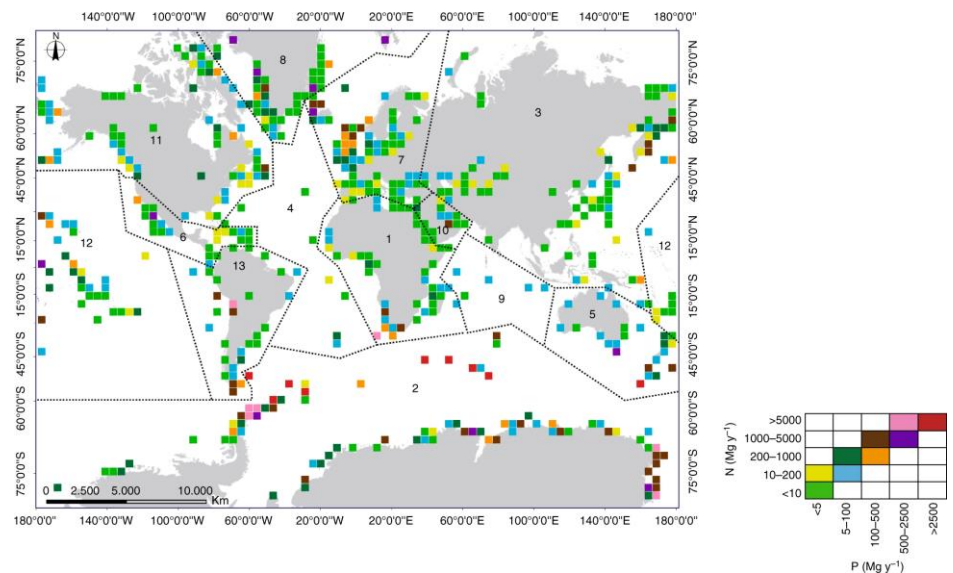
Modified text: lines 179-181: “Therefore, the uncertainties of the dry deposition velocity estimation for the elements that were dominated by coarse particles were about $\pm 30\text{-}60\%$, and the uncertainty of fine particle dominated element (Pb) was about $\pm 100\%$.”

L160-185 Can you use references for P and V sources from the Southern Hemisphere? I’m not aware of any evidence that agricultural and industrial emissions from China are transported to Antarctica. It seems that P at your site is dominantly sourced from local Antarctic emissions

rather than long-range transported agricultural or volcanic emissions. Do airmasses pass over Antarctic soils, or seabird colonies? While ship emissions of V have been detected in aerosols in the Arctic and Atlantic where shipping is more frequent, is there any evidence of this in the Southern Ocean? While it's important to identify all possible sources of these elements, this section could be tightened up to clarify the most likely sources of this group of elements and rule out the unlikely ones to Antarctica.

Response: We have searched the related references for V in the Southern Hemisphere and found only one study conducted in New South Wales, Australia, using V/Ni as a tracer of the use of heavy oils (Keywood et al., 2020). We have replaced the previous references with this one. Please see [line 209](#). In addition, we didn't find any previous studies that reported an elevated EF_{crust} of V due to the ship emissions in the Southern Ocean or coastal Antarctica. It looks like that ship emissions may not be an important source for V in this region, similar to the results found at McMurdo (Lowenthal et al., 2000).

For P, we have removed biomass burning as a source of P since the $nss-K^+$ suggests our samples were barely affected by biomass burning. The 72-hour air mass trajectories show most of the air masses passing over the Antarctic Peninsula were derived from the South Pacific Ocean or South Atlantic Ocean. However, there is a penguin colony on Torgerson Island (~ 1 km from Palmer Station), and it is possible that our samples were affected. In addition, the Antarctic Peninsula has one of the highest P excretions by seabird colonies globally (Otero et al., 2018) (Please see the figure below). We believe that the local crustal emissions from the soil can definitely emit P-enriched dust particles to the atmosphere.



Global distribution of N and P excretion by seabird colonies (Otero et al., 2018).

Modified text: line 209: "...and V in aerosols was found associated with ship emissions due to the use of heavy oil fuel (Keywood et al., 2020)."

Line 186 and 242 Many of these elements occur naturally in the crust but in your samples they

have a non-crustal source. I wonder if you could rename the headings to avoid confusion e.g. rename the group of elements into one of the three categories described in the abstract - local mineral dust, long-range transported aerosol and sea salt aerosol.

Response: We have changed the subtitles in “3.2 Concentrations of trace elements” to “mineral dust”, “anthropogenic aerosol”, and “sea salt aerosol”. Please see [section 3.2](#). However, in the “3.1 Enrichment factors of trace elements” section, the EF_{crust} can only tell us if the elements were derived from crustal or non-crustal sources. We prefer to keep crustal source and non-crustal source.

Line 195 Again, please use references for the Southern Ocean rather than Northern Hemisphere. Some suggestions for Pb: McConnell, J.R., Maselli, O.J., Sigl, M., Vallelonga, P., Neumann, T., Anschutz, H., Bales, R.C., Curran, M.A., Das, S.B., Edwards, R. and Kipfstuhl, S., 2014. Antarctic-wide array of high-resolution ice core records reveals pervasive lead pollution began in 1889 and persists today. *Scientific Reports*, 4(1), pp.1-5. Bollhöfer, A.F., Rosman, K.J.R., Dick, A.L., Chisholm, W., Burton, G.R., Loss, R.D. and Zahorowski, W., 2005. Concentration, isotopic composition, and sources of lead in Southern Ocean air during 1999/2000, measured at the Cape Grim Baseline Air Pollution Station, Tasmania. *Geochimica et Cosmochimica Acta*, 69(20), pp.4747-4757.

Response: We have replaced all the references with Dick (1991), Artaxo et al. (1992), McConnell et al. (2014), and Keywood et al. (2020), which were all conducted in the Antarctica or Southern Hemisphere. Please see [lines 228-229 and line 233](#).

Line 200-201 This sentence isn't clear whether you are suggesting that Ca is derived from seawater or another source? For the modern day, many coastal ice core sites suggest a marine source of Ca, where during glacial periods Ca is dominated by a long-range transported mineral dust.

Response: We have rephrased this sentence. Palmer Station should receive Ca from both seawater and crustal emission. In this study, the high crustal enrichment factor (>10) of Ca suggests the concentration of Ca was dominated by sea-salt aerosol. Please see [line 238](#).

Modified text: line 238: “The high EF_{crust} for Ca at Palmer Station suggests that aerosol Ca was mainly derived from sea-salt.”

Line 208-211 Refer the reader to Fig. 4.

Response: We have referred this sentence to Figure 4. Please see [line 250](#).

Modified text: line 250: “...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)”

Line 215-216 Al concentrations have been measured in local dust in snow on sea ice in McMurdo Sound which is the dustiest location in Antarctica.

Response: We have added a sentence that refers McMurdo Sound is the dustiest location in Antarctica and cite Winton et al. (2016) in the revised manuscript. Please see lines [252-253](#).

Modified text: lines 252-253: “The nearby McMurdo Sound was reported as the dustiest site in Antarctica (Winton et al., 2016).”

Line 232-235 Comparison to trace element concentrations in regions outside of the Southern Ocean or the Southern Hemisphere seems irrelevant.

Response: We have removed the comparison with other studies conducted in the Northern Hemisphere.

L235-237 These pieces of evidence further confirm your EF interpretation that P was mainly locally derived from soil and bird colonies proximal to the station. How did you calculate nss-K? Please state the limitations of using nss-K as a biomass burning tracer in the Southern Ocean as opposed to sites proximal to biomass/fires. See other studies in the Southern Ocean e.g. nss-K at Cape Grim. Is there a better tracer of biomass burning that you have access to over the sampling period? Winton, V.H.L., Bowie, A.R., Edwards, R., Keywood, M., Townsend, A.T., van der Merwe, P. and Bollhöfer, A., 2015. Fractional iron solubility of atmospheric iron inputs to the Southern Ocean. *Marine Chemistry*, 177, pp.20-32.

Response: The nss-K⁺ was calculated using the equation: $[\text{nss-K}^+] = [\text{K}^+] - 0.037[\text{Na}^+]$, where 0.037 is the mass ratio between K and Na in seawater (Millero, 2016). We have added this equation to the methods. Please see [line 138](#).

The main disadvantage of using nss-K⁺ as the tracer of biomass burning is that water soluble K can have multiple sources, which include not only biomass burning and sea-salt, but also mineral dust. During long-range transport, those sources may contribute K to the air mass and cause an increase in nss-K⁺. In our samples, the nss-K⁺ were close to 0. Such a result means K⁺ were dominated by sea-salt aerosol and all the other sources, including biomass burning, played a minor role. We are aware there are some better tracers, such as levoglucosan. However, we do not have enough samples for further analysis. We have revised the sentence and added Winton et al. (2015) as a reference that use nss-K⁺ as a tracer of biomass burning in [lines 272-274](#).

Modified text: lines 272-274: “In this study, nss-K⁺ was used as a tracer of biomass burning (Winton et al., 2015). The calculated nss-K⁺ was indistinguishable from zero, suggesting that K⁺ in aerosol at Palmer Station was primarily derived from sea water, not from biomass burning through long-range transport.”

Line 243-255 Open paragraph with a sentence to let the reader know you are discussing Ni, Zn, Pb and Cu first. It's not clear what caused the large variability in this group of elements (Ni, Zn, Pb and Cu). Can you link these elements with air mass trajectories as you did with Pb? What anthropogenic activities emit Ni, Zn, Pb and Cu in South America?

Response: We have added a sentence in the beginning of this paragraph (please see [line 280](#)). A short discussion about the link between air mass back trajectories and high concentrations of Ni, Cu, and Zn were as added to this part (please see [line 287-291](#)). In South America, the emissions of Ni, Zn, Pb, and Cu were suggested to be primarily associated with vehicle emission, soil dust, and oil combustion (Artaxo et al., 1999; Jasan et al., 2009). In addition, mining activities were suggested as an important anthropogenic source at remote sites in the South America (Carrasco and Préndez, 1991; Klumpp et al., 2000). Please see [line 296-299](#).

Modified text:

line 280: “The concentrations of Ni, Cu, Zn, and Pb in were more variable than the crustal elements.”

lines 287-291: “From the air mass back trajectories, the samples with high Ni (M2, M10), Cu (M1, M2, M4, M10) and Zn (M5) all were impacted by significant amount of air masses from the South Pacific Ocean and South America (Figure 5). The back trajectories of air masses of M7 didn't touch South America but the concentration of Zn in this sample was high. With the fact that aerosol Zn was found in both fine- and coarse-mode fractions (Table 2), both local sources and long-range transport may contribute to this element in the air.”

lines 296-299: “In South America, high enrichment of Ni, Cu, Zn, and Pb in fine mode particles was reported to be primarily associated with vehicle emission, soil dust, and oil combustion (Artaxo et al., 1999; Jasan et al., 2009). Moreover, mining activities were suggested as an important source, especially in remote sites in South America (Carrasco and Préndez, 1991; Klumpp et al., 2000).”

Line 258 Please compare to other studies of aerosol Pb in the Southern Ocean.

Response: We have added in the comparison with Bollhöfer et al. (2005) in Tasmania. Please see [line 305](#).

Modified text: line 305: “...and the annual average concentration of 11 pg m⁻³ in Tasmania (Bollhöfer et al., 2005).”

Line 263-264 “The low concentration of Pb observed in samples associated with trajectories that did not pass over Southern South America suggest that: : :”

Response: We have revised this sentence. Please see [line 311-312](#).

Modified text: lines 311-312: “The low concentrations of Pb observed in samples associated with air masses that did not pass over Southern South America suggest that local anthropogenic emissions were negligible.”

Line 265 “: : long-range transported aerosol from Southern Hemispheric continental regions containing a mixture of anthropogenic and crustal emissions such a South America.”

Response: We have revised this sentence. Please see [lines 312-314](#).

Modified text: lines 312-314: “Thus the major source of non-crustal elements in aerosols over the study region may be long-range transport of aerosols from Southern Hemispheric continental regions containing a mixture of anthropogenic and crustal emissions.”

Line 272 Do you mean K wasn’t influenced by biomass burning. This should be mentioned before you use nss-K as a biomass burning tracer.

Response: At Palmer Station, the nss-K⁺ was close to zero. It means the aerosol samples we collected were barely affected by biomass burning or any other sources, apart from sea spray, contribute K⁺ to the atmosphere. We have added this interpretation in the “Mineral dust” part when we discuss the source of P (please see [line 273-274](#)).

Modified text: lines 273-274: “The calculated nss-K⁺ was indistinguishable from zero, suggesting that K⁺ in aerosol at Palmer Station was primarily derived from sea water, not from biomass burning through long-range transport.”

Line 266-273 See modern day, coastal snow chemistry studies of Ca, Na and K derived from sea salt and sea ice to further back up the high abundance of marine derived elements at your site. Also, open the paragraph with a sentence to let the reader know you are discussing Ca, Na and K. End with a clear sentence stating the source(s).

Response: We have added a sentence in the beginning of this paragraph to show this part is about sea salt species. Please see [line 316](#). We have also added a comparison of the Na/K and Na/Ca ratios with the snow samples collected at James Ross Island (Aristarain et al., 1982). We rephrased the last sentence to clearly suggest Ca and K⁺ were primarily derived from sea salt aerosols. Please see [lines 323-325](#).

Modified text:

line 316: “The concentrations of sea-salt species were much higher than the other trace elements (Table 1).”

lines 323-325: “Such results agree well with the Na/K (29.7) and Na/Ca (39.9) ratios

measured in snow samples collected at James Ross Island (Aristarain et al., 1982). Therefore, the Ca and K⁺ in aerosols were derived primarily from sea salt at Palmer Station.”

Line 277 Consider renaming “elements from the continents” to “mineral dust”

Response: We have revised this section. We used k-means clustering to group all the elements based on their normalized particle size distributions. Please see [lines 327-332](#).

Modified text: lines 327-332: “We applied k-means clustering to the full data set, and the results indicate that aerosol trace elements in the austral summer at Palmer Station can be classified into five groups based on their normalized particle size distributions (Figure 6), with each group showing a unique size distribution pattern: (1) crustal elements from crustal weathering and wind-induced resuspension of soil particles, (2) Al dominated by local minerals, (3) Pb from anthropogenic sources, as a result of long-range transport, (4) sea salt elements from the ocean, through bursting bubbles of seawater, and (5) P from local biogenic and soil resuspension.”

L278 First mention of combustion. You previously ruled out biomass burning. Please state which elements are combustion products in the introduction or discussion section on sources.

Response: We have removed “combustion” and revised this section. Please see [the first paragraph in section 3.3](#).

Modified text: Please see the revised section 3.3, lines 327-334.

Line 281 This group of elements all seem to have a coarse mode around the same size. Please include.

Response: We have revised this section. The elements showing single coarse mode distributions are discussed together in [section 3.3.1, line 335-344](#). “This group of elements all have a coarse mode around the same size” is mentioned in [section 3.3.3, line 375-377](#).

Modified text:
lines 335-344: Please see section 3.3.1.

lines 375-377: “A single-mode Al particle size distribution peaked at around 4.4 μm which shared the same size with the primary mode of Ti, V, Mn, and Ce in sample M8 and M9, indicating the regional crustal emissions.”

Line 281-282 Is this reference for long-range transported dust?

Response: This reference didn’t specify long-range transported dust. We have removed

this citation.

Line 280-289 This section could be considerably improved. Some elements have a secondary mode in some samples but a single mode in other samples e.g. Al, V, Mn, Pb. Please discuss the episodic deposition from multiple sources and include in your discussion that these elements can have more than one source in a particular air mass. Please plot size distributions for all elements in Figure 6. I can't see Na and K. Include in your discussion a comparison with the literature to help attribute individual modes to sources. Some, but not all, references for Antarctic aerosol size distributions are below. I think the elements should be grouped differently according to the particle size distribution rather than the crustal vs non-crustal EF analysis. For example, crustal sources: P, Ti, Ca, and Ce have a single coarse mode distribution around XYZ μm indicating. V and Mn have a bimodal distribution with a primary mode around XYZ μm and secondary mode around ZXY μm . Pb has a bimodal and sometimes trimodal distribution with a fine primary mode that is much finer than and mode in the other elements. Al, again, has a different distribution with a bimodal and sometimes trimodal distribution with mode similar to P, Ti, Ca, and Ce and an additional coarse mode not seen in other elements. Also, Al exhibits a fine mode, similar to Mn and V, but only in one sample. Can you rule out local dust contamination for coarse Al concentrations? Could you compare the Al particle size distribution to mineral dust size distributions in Antarctic snow? A very interesting section could be developed about this comparison. Lachlan-Cope, T., Beddows, D.C., Brough, N., Jones, A.E., Harrison, R.M., Lupi, A., Jun Yoon, Y., Virkkula, A. and Dall'Osto, M., 2020. On the annual variability of Antarctic aerosol size distributions at Halley Research Station. Delmonte, B., Winton, H., Baroni, M., Baccolo, G., Hansson, M., Andersson, P., Baroni, C., Salvatore, M.C., Lanci, L. and Maggi, V., 2020. Holocene dust in East Antarctica: Provenance and variability in time and space. *The Holocene*, 30(4), pp.546-558. Fattori, I., Becagli, S., Bellandi, S., Castellano, E., Innocenti, M., Mannini, A., Severi, M., Vitale, V. and Udisti, R., 2005. Chemical composition and physical features of summer aerosol at Terra Nova Bay and Dome C, Antarctica. *Journal of Environmental Monitoring*, 7(12), pp.1265-1274. Virkkula, A., Teinilä, K., Hillamo, R., Kerminen, V.M., Saarikoski, S., Aurela, M., Koponen, I.K. and Kulmala, M., 2006. Chemical size distributions of boundary layer aerosol over the Atlantic Ocean and at an Antarctic site. *Journal of Geophysical Research: Atmospheres*, 111(D5). Fossum, K.N., Ovadnevaite, J., Ceburnis, D., Dall'Osto, M., Marullo, S., Bellacicco, M., Simó, R., Liu, D., Flynn, M., Zuend, A. and O'dowd, C., 2018. Summertime primary and secondary contributions to Southern Ocean cloud condensation nuclei. *Scientific reports*, 8(1), pp.1-14.

Response: We would like to thank Dr. Winton for sharing these references with us. We have added a discussion about the potential contribution from long-range transport (anthropogenic and crustal) for Al, V, Mn, and Pb in [line 351-359](#). We have added the plots for particle size distribution of Na and K in [Figure 7](#).

We have rewritten this section and grouped the elements by their particle size distributions. At the start of the revised section, we added a result of k-means clustering which classifies the particle size distributions of elements/ionic species into 5 clusters. The particle size distributions of all the elements and ionic species are discussed in 3 sections: single mode distribution, bimodal distribution, and trimodal distribution.

Special samples with unique size distribution are discussed with the air mass back trajectories in this section. At the end of the revised section, we also added a short discussion to compare Al size distribution in this study with the dust particle size distribution measured in snow, ice core, and aerosols in Antarctica. However, we can not rule out local dust contributions to the observed Al concentrations, and we have briefly mentioned this in the revised manuscript. **Please see section 3.3.**

Modified text: Please see all the changes in section 3.3 in lines 326-396.

Line 318-319 Note that estimates of wet deposition fluxes are even scarcer than dry deposition fluxes! Please state the limitations with this assumption but note it is likely the best estimate we have.

Response: We have added an explanation to show this estimate may have large uncertainty. Please see **lines 412-413.**

Modified text: lines 412-413: “Although precipitation varies in different regions, this is the best estimate we have for Antarctic regions.”

Line 321 Dome C is a high elevation site and receives fine dust with a mode around 2-3 μm .

Response: Thanks for sharing that information. We have added the comparison of dust particle size distribution in Dome C ice core samples (Delmonte et al., 2002) in discussion. Please see **lines 392-394.**

Modified text: lines 392-394: “Likewise, the dust particle size distribution in Dome C during the Last Glacial Maximum and the Holocene showed single-mode and peaked at approximately 2 - 3 μm in ice core samples (~3233 m above sea level) (Delmonte et al., 2002; Potenza et al., 2016).”

Line 305-340 Please add errors on these fluxes.

Response: Done. We have added errors in both text (**line 401, 408, and 414**) and revised **Table 3.**

Line 323-327 Suggest removing the comparison to values outside of the Southern Ocean and replacing with a discussion on why the flux is at the lower end of the JRI estimate.

Response: We didn't remove the comparison since it can give the readers a sense that the dust deposition flux at Antarctic Peninsula is extremely low.

There are several active dust sources reported in James Ross Island (Kavan et al., 2018), and we expect higher deposition fluxes close to the source region. In addition, our wet deposition was estimated based on an assumed percentage (40-60%), which could be

underestimated. This could also contribute to a low dust flux. We have briefly discussed this in the revised manuscript. Please see [lines 418-420](#).

Modified text: lines 418-420: “Considering active dust sources were reported at James Ross Island (Kavan et al., 2018), sites close to James Ross Island are likely to receive higher dust deposition fluxes. In addition, without measurements of wet deposition, the total dust deposition flux in our study could be underestimated.”

Line 332 Add reference for the 40 % assumption.

Response: Done. Please see [line 430](#).

Modified text: line 430: “...assumed that dry deposition accounted for 40% of total deposition (Gao et al., 2003)...”

Line 331-333 What geographic area does the estimate represent?

Response: This estimate represents the West Antarctic Peninsula and its marginal seas. We have mentioned this in [line 428](#).

Modified text: line 428: “To examine the potential importance of atmospheric dust input to the particulate elemental concentrations in surface waters of the West Antarctic Peninsula shelf region...”

Line 337-341 Could you estimate the lower bound as well and report both upper and lower bounds for summer? This information would be useful to include in the abstract too.

Response: Our current estimation is based on several assumptions that maximize the impact of atmospheric deposition. It gives us a sense that the atmospheric deposition only plays a minor role for supplying those trace elements to the west Antarctic Peninsula coastal seawater. However, we don't have enough information to provide the upper and lower bounds.

Line 343 This conclusion omits previous discussion of anthropogenic emissions. Please keep a consistent message throughout the manuscript. What about biogenic emissions of P here and in the abstract?

Response: We have rewritten the conclusions and implications section (section 4). We include anthropogenic emission in the conclusion. Please see [lines 444-445](#). A speculation of the P emission is given in [lines 449-450](#).

Modified text:
lines 444-445: “...derived from (1) the regional crustal sources, which includes P-enriched soil resuspension, (2) remote anthropogenic emissions in South America , and

(3) seawater.”

lines 449-450: “We speculate that the modest enrichment of aerosol P over its crustal ratio to Al was caused by the resuspension of regional soils that are P-enriched as a result of the impact of nearby bird colonies.”

Line 348-351 I don't see how you can make this claim without data prior to the commencement of shipping. There are regional differences in aerosol chemistry and dust fluxes around Antarctica. Perhaps rephrase indicating that future impacts of shipping and changes in the ice-free area have an unknown impact on aerosol chemistry in the region.

Response: We have revised this sentence to suggest future study for the unknown impact of shipboard tourism and warming conditions on the aerosol chemistry in Antarctic Peninsula. Please see line 460-463.

Modified text: lines 460-463: “The Antarctic Peninsula is experiencing rapid climate change, with the expansion of ice-free areas and more frequent shipboard tourism with unknown impacts on aerosol chemistry in this region. To quantify future changes in in the atmospheric position and the impact in this region, long-term atmospheric observations of aerosol chemical and physical properties along with coupled studies of the ocean-atmosphere interactions would be needed.”

Fig 1 and Table 1: These look like copies of Fig. 1 and Table 1 in Gao et al. (2020). Please state if the figure is reproduced from this publication.

Response: Yes. The Figure 1 and Table 1 are from Gao et al. (2020). We have referenced this paper.

Fig. 2: State what the label stands for. Please provide more information in the caption.

Response: Done. We have added a description of the label in the figure caption.

Fig. 3: Please state the time period the samples represent in the caption.

Response: Done. We have added the time periods in the figure caption.

Fig. 4: State that these are all land-based sampling sites except one cruise. Are these all total digestible concentrations? Why don't you add other data from cruises in the Southern Ocean?

Response: We have added the information on either land-based or cruise in the figure caption. In this figure, we compared the previous observations of trace element concentrations in the coastal Antarctic region. We included the cruise from Zhongshan Station to Casey Station because it mainly focused on measuring the trace element concentrations in coastal East Antarctica. In this figure, not all the studies used the total

digestion method. Artaxo et al. (1990); Artaxo et al. (1992); Mazzera et al. (2001) and Préndez et al. (2009) used X-ray fluorescence to measure the total concentrations of trace elements. All the other studies applied acid digestion methods. We have added this information in the revised figure caption. Please see the revised [Figure 4](#).

Fig 5: Why are you only showing air mass back trajectories of 4 samples? Please make the continent outline clearer as it is difficult to see under the air mass fetch regions.

Response: We have included the back trajectories for all 8 samples. Unfortunately, these figures were generated on NOAA's website and we cannot change the background. Please see the revised [Figure 5](#).

Fig. 6: Please provide more information in the caption. Group the elements into the size distribution patterns discussed in the text. State what the axis labels stand for and include a reference for the notation in the methods. Are you plotting the number or volume particle size distribution? Please plot size distribution for ALL elements.

Response: In our study, we use the mass size distributions rather than number or volume particle size distributions. We have added a description about how the mass distribution was obtained in method in [section 2.3.2, line 155-163](#). We have revised the figures. Now, the single mode particle size distributions are shown in [Figure 7](#), and the bimodal/trimodal distributions are shown in [Figure 8](#). We have added the figures for size distribution of K^+ and Na^+ . The notation of normalized particles size distribution and reference have been added to Method. Please see [section 2.3.2, line 156-161](#).

Modified text: We have added a new section in method. Please see section 2.3.2.

Tables 2-3: Could be moved to the supplement.

Response: We have moved Table 2 (LOD, %blank ...) to the supplement. However, we believe that keeping Table 1 (sampling information) and Table 3 in the main body of the manuscript (concentrations of trace elements) is convenient to the readers.

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