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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Concentrations, Particle-Size Distributions, and Dry Deposition Fluxes of Aerosol Trace Elements over the Antarctic Peninsula" *by* Songyun Fan et al.

Holly Winton (Referee)

holly.winton@vuw.ac.nz

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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.

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. 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 literate on this topic. Currently, there is no discussion on what the size distribution us about the atmospheric transport mechanisms of the individual trace elements. Also, the conclusions don't come through strongly enough in the discussion. 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? 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.

Technical corrections L11 Suggest replacing with "local mineral dust, long-range transported aerosol and sea salt aerosol." What about local P biogenic emissions? L26 Define course mode. L32 Include sea salt as a source of aerosol. L37 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. L47 Include a refence to McMurdo Sound here as the dustiest region in Antarctica. L62 "...new sources of aerosol trace elements by warming which exposes a greater area of ice-free land and by..." L68 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). L100 Please report precision here or in Table 2. L107 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 an K by ICP-MS? L121 Why did you select Al over Ti as the crustal reference? L128 "...was calculated from the concentration of the trace element in the air and dry..." L141 What are the environmental factors? L142 Please report the uncertainty of your dry deposition velocity estimates. Also, where can the reader find the dry deposition velocity values for each element? L160-185 Can you use refences 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. L186 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. L195 Again, please use refences for the Southern Ocean rather than Northern Hemisphere. Some suggestions for Pb: McConnell, J.R., Maselli, O.J., Sigl, M., Vallelonga, P., Neumann, T., Anschütz, 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. L200-201 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. L208-211 Refer the reader to Fig. 4. L215-216 Al concentrations have been measured in local dust in snow on sea ice in McMurdo Sound which is the dustiest location in Antarctica. L232-235 Comparison to trace element concentrations in regions outside of the Southern Ocean or the Southern Hemisphere seems irrelevant. 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. L243-255 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? L258 Please compare to other studies of aerosol Pb in the Southern Ocean. L2643-264 "The low concentration of Pb observed in samples associated with trajectories that did not pass over Southern South America suggest that. . ." L265 ". . .long-range transported aerosol from Southern Hemispheric continental regions containing a mixture of anthropogenic and crustal emissions such a South America." L272 Do you mean K wasn't influenced by biomass burning. This should be mentioned before you use nss-K as a biomass burning tracer. L266-273 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). L277 Consider renaming "elements from the continents" to "mineral dust" L2278 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. L281 This group of elements all seem to have a coarse mode around the same size. Please include. L281-282 Is this reference for long-range transported dust? L280-289 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 airmass. 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 um indicating. V and Mn have a bimodal distribution with a primary mode around XYZ um and secondary mode around ZXY um. 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 model 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. L318-319 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. L321 Dome C is a high elevation site and receives fine dust with a mode around 2-3 um. L305-340 Please add errors on these fluxes. L323-327 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. L332 Add reference for the 40 % assumption. L331-333 What geographic area does the estimate represent? L337-341 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. L343 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? L348-351 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. 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. Fig. 2: State what the label stand for. Please provide more information in the caption. Fig. 3: Please state the time period the samples represent in the 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? 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. 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. Tables 2-3: Could be moved to the supplement.

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