Author's Response to Review #2:
We are grateful to the anonymous reviewer for the valuable comments that helped us significantly improve this manuscript. All comments have been addressed on a point-by-point base. Changes made based on the reviewer's 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 #2:
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 anonymous reviewer

Major Comments:
1. The total sampling period of the study is rather short (2 months). Authors need to discuss the possible uncertainties/seasonal influence affecting the element concentrations, especially in the context of comparison with other studies.

> ***Response:*** This study focused on atmospheric trace elements during the austral summer in Antarctic Peninsula. Given that the snow/ice cover could limit local dust emissions, we expect lower concentrations of crustal elements in austral winter. However, it is difficult to evaluate how the other trace elements may vary seasonally without seasonal observations. In the text, we added the detailed study periods (e.g. summer mean concentration or yearly mean concentration) for previous studies of crustal element concentrations, for comparison with our results. In addition, we added "during the austral summer" in the title of this paper to emphases the study season.

2. The study used Al concentration (8% of the dust concentration) to calculate the dry and total deposition of dust in the region. A few major conclusions of the study were based on this assumption. The assumption could lead to uncertainties in the results as it is based on the Al concentration only and may not represent the crustal matter in the study region. Therefore, the authors should provide sufficient evidence to establish that this is a solid assumption. Mass reconstruction of soil/dust based on all available crustal elements concentration (rather than just the Al concentration) can also be considered.

> ***Response:*** The concentrations of Al have been used to represent the contribution from crustal emissions at many sites in Antarctica, such as Antarctic Peninsula (Dick, 1991), McMurdo (Lowenthal et al., 2000), and the South Pole (Zoller et al., 1974). We searched for previous studies that reported rock or soil composition in Antarctica and learned that West Antarctica has a complicated geological history and many different rock types (Pereira et al., 2018). Nelson (1966) reported the percentage of Al ranged from 7.83 % to 10.42 % in 16 rock samples collected at James Ross Island, Antarctic Peninsula. On average, Al accounted for 8.86 ± 0.79 % in the total mass of rock. This number is about 10% different from the average upper crustal abundance of Al, 8.04% (Taylor and McLennan, 1995). In addition, there is a portion of crustal material

contributed by long-range transport, for which we don't know the source region. In this case, we decided to use the average crustal abundance of Al to estimate the dry deposition flux. Such uncertainty is small compared with the uncertainty of dry deposition estimation (a factor of 2 or 3). Given the undetermined mineralogy of the dust we collected, we estimate that dust mass determined by our measurements of aerosol Al and its assumed stoichiometry, as the reviewer suggests, would be at least as uncertain as our approach. Therefore we did not revise our methodology in this case.

3. Some figures and parts of the text should be revised to improve the clarity (e.g., Figures 4, Section 3.2.1, etc.). Specific line-by-line comments are as follows.

Response: We have revised Figure 4, highlighted our study site at Palmer Station in red to improve the clarity. We have also added more detailed information in the figure caption.

Specific comments:

Line 10: "The results show. . ." The reader would be wondering about the study methods/data analysis approach that leads to these results (i.e., enrichment calculation or statistical analysis/ source apportionment tool, etc.?).

Response: We have added the crustal enrichment factor and k-means clustering here as the main approach that to get these results. Please see lines 11-12.

Modified text: lines 11-12: "The crustal enrichment factors (EF_{crust}) and k-means clustering results of particle size distributions show…"

Lines 11-13: "Elements dominated by a crustal source. . . reflecting the contributions of regional crustal sources" repetitive.

Response: We agree with the reviewer and have revised the part of the text. Please see lines 13-15.

Modified text: lines 13-15: "Elements derived from crustal sources (Al, P, Ti, V, Mn, Ce) with EF_{crust}<10 were dominated by the coarse-mode particles (>1.8 μm) and peaked around 4.4 μm in diameter, reflecting the regional contributions."

Line 13: The term "EFcrust" may not be familiar to all readers.

Response: We have added "crustal enrichment factor (EF_{crust})" to define this term in line 11.

Modified text: line 11: "The crustal enrichment factors (EF_{crust}) and…"

Line 13: ". . .coarse-mode particles (>1 μm). . ." contradicts with lines 72-73 that states

‘’. . .those >=1.8 μm were summed to define coarse-mode particles. . .”.

Response: This typo has been corrected. Please see line 14.

Modified text: line 14: “…the coarse-mode particles (>1.8 μm)…”

Lines 26-27: “It has been realized that the impact of coarse mineral dust has been underestimated. . .” I am not sure what argument is presented here.

Response: We have revised this sentence. 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).”

Lines 54-56: “However,. . .are lacking.” Is this the first study on elements sampled in Antarctica? If yes, state clearly, if not, briefly mention about the prior studies that sampled aerosol in this region and what this study introduces. The paragraph that follows (lines 57-65) also does not give much idea about the knowledge gap that this study fills in.

Response: We agree. We have changed the word “lacking” to “inadequate”. Please see line 77. We also added a short summary on what have been done in Antarctic Peninsula and what has not been measured in lines 53-58.

Modified text:
line 77: “…and accurate estimation of the atmospheric deposition of trace elements to the region are inadequate.”

lines 53-58: “In the Antarctic Peninsula, the concentrations of aerosol trace elements were measured at several sites (Dick, 1991; Artaxo et al., 1992; Mishra et al., 2004; Préndez et al., 2009). Total dust deposition in this region was also estimated based on the ice-core record (McConnell et al., 2007). However, the measurement of particle size distribution of aerosol trace elements in Antarctic Peninsula is missing and there is no direct measurement that evaluated the importance of atmospheric deposition as a source of nutrients for primary producers in West Antarctic Peninsula shelf waters.”

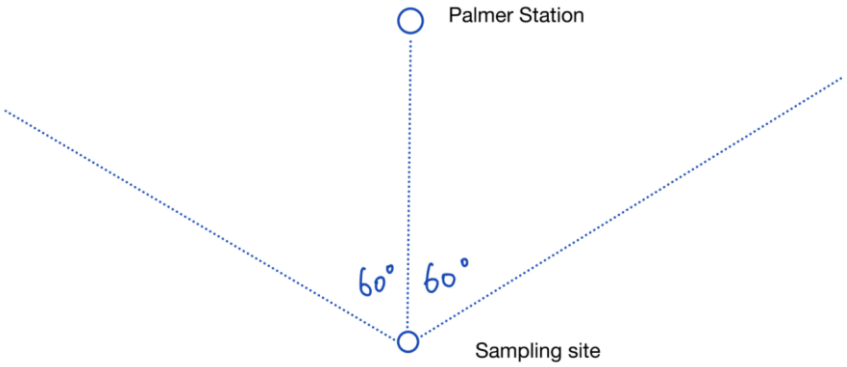
Section 2, Line 66 onward: No mention of how the samples below “limits of detection (LOD)” were treated.

Response: The concentrations below LOD were given a concentration of zero for the purposes of this study. We have added this description in the manuscript. Please see lines 129-130.

Modified text: lines 129-130: “The concentrations below LOD were given a concentration of zero for the purposes of this study.”

Lines 77-78: ". . .wind direction inside the sector ±60° from the direction of the station and wind speed <2 m s-1." Unclear to me.

Response: Please see the plot below. When the wind came from the ±60° from the direction of Palmer Station or wind speed < 2 m s^{-1}, we paused the sampling to avoid local contamination from activities at the station. We have changed "and" to "or" in this sentence. Please see line 101.



Modified text: line 101: "…inside the sector ±60° from the direction of the station buildings or when wind speed <2 m s^{-1}.

Line 91: ". . .Al, P, Ca, Ti, V, Mn, Ni, Cu, and Zn" this list looks different from the one mentioned in the abstract (line 9) that states ". . .Al, P, Ca, Ti, V, Mn, Ni, Cu, Zn, Ce, and Pb...".

Response: This error has been corrected. Please see line 116.

Modified text: line 116: "Elemental concentrations were determined for Al, P, Ca, Ti, V, Mn, Ni, Cu, Zn, Ce, and Pb."

Line 133: The summation is over what variable?

Response: The summation refers to the sum of the concentrations determined for the 10 stages. We have revised the equation to mark i ranges from 1 to 10. Please see the equation in line 170.

Line 135: So, Vd was calculated from the combination of two models?

Response: Yes.

Line 161: "The values of EFcrust for Ti, V, Mn, and Ce in aerosol samples were less than 10. . ." Why P is missing from this list where it has EFcrust<10 (Figure 3)?

Response: The EF_{crust} of P ranged from 2 to 8, slightly different from other crustal

elements. In addition, P possibly has a biogenic source that is also different from Ti, V, Mn, and Ce. Thus, we decided to discuss the P concentration and its source separately in the second paragraph in section 3.1.1.

Lines 170-171: "A similar phenomenon was observed at McMurdo Station where lightweight fuel oil was used that was not a significant source of V" not clear what was referred here.

Response: We refer to the fact that light oil used at McMurdo Station was not a significant source of aerosol V at that location (Lowenthal et al., 2000); such situation could be true for aerosol V at Palmer Station, and our EF_{crust} of V suggests that crustal source is the dominate source for V. Please see lines 210-211.

Modified text: lines 210-211: "Similarly, unenriched V was observed at McMurdo Station where light-weight fuel oil was used that was not a significant source of V (Lowenthal et al., 2000)."

Lines 173-185: My understanding is that the purpose of this paragraph is to establish that the enrichment of P is higher than other crustal elements. But based on the results (Figure 3), it appears that P enrichment is not significantly high in this study, compared to the other relevant studies.

Response: Yes, P was not enriched in our samples. We think P is special in this region since Antarctic Peninsula is one of the places that have the highest P excretion by seabird colonies (Otero et al., 2018). It's worthy to discuss the potential source of aerosol P and explain the reason why EF_{crust} was relatively high in a separate paragraph.

Lines 197-198: "Hence, despite the recent increase in tourist ship traffic, it looks that Palmer Station was barely impacted by ship emissions" assuming this is correct, what is the reason for large variations of Ni (e.g., Figure 3)?

Response: The significant variation of Ni from sample to sample might be attributed to receiving the long-range transport from South America. Please see line 287-289. In addition, the Ni has relatively high percentage of blank as shown in Table S1, and the variation in samples could also be contributed by the variation in the blank.

Modified text: lines 287-289: "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)."

Lines 199-200: "Ca accounts for about 3.5% of the weight of Earth's crust, while Ca is also a conservative major ion in seawater" provide citation.

Response: Done. We have added Taylor and McLennan (1995) as the reference for the

Earth's crustal abundance and Millero (2016) as the reference for the major ions in seawater. Please see lines 237-238.

Modified text: lines 237-238: "Ca accounts for about 3.5% of the weight of Earth's crust (Taylor and McLennan, 1995), while Ca is also a conservative major ion in seawater (Millero, 2016)…"

Line 215: ". . .the impact of the nearby McMurdo Dry Valleys" I could not understand what impact was referred here.

Response: McMurdo Dry Valleys (MDVs) are dry lands in Antarctica. Consequently, the average Al concentration in PM_{10} samples collected at McMurdo Station impacted by the air from MDVs was more than an order of magnitude higher than Antarctic Peninsula region (Mazzera et al., 2001). We have added a sentence to show this information for clarity. 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)."

Lines 217-218: "The concentrations of Ti and Mn ranged from. . .respectively." I am a little confused if the ranges are for individual elements or both elements combined.

Response: We have rephrased this sentence. Please see line 255-256.

Modified text: lines 255-256: "The concentrations of Ti ranged from 140 to 800 pg m^{-3} with an average of 250 pg m^{-3}, while the concentration of Mn ranged from 17 to 44 pg m^{-3} with an average of 30 pg m^{-3}."

Lines 220-223: ". . .but comparable to the concentrations. . . (Figure 4e and g)" is it applicable to both Ti and Mn? Figures 4e and 4g do not support this claim. Mn concentration at AP-OS is more than double of AP-PS (Figure 4g).

Response: We have revised this part of the text. Please see line 259.

Modified text: line 259: "However, these Ti and Mn values observed at Palmer Station were in the same magnitude with the…"

Line 232: ". . .the P values. . ." P concentrations?

Response: It should be P concentrations. We have removed this sentence for clarification.

Line 235-236: "Comparing global aerosol P concentrations. . .as those over the Central Pacific Ocean (Chen, 2004)" provide the concentration values from the referred study.

Response: We realized this comparison is not necessary and have decided to remove it.

Lines 236-237: "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." I could not understand what this means.

Response: We revised this sentence. Please see lines 272-274.

Modified text: line 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."

Lines 239-241: ". . .suggesting that aerosol crustal elements observed at Palmer Station were impacted by sources in that region (Figure 5)." Why this argument applies to only crustal elements?

Response: In this section, we focused on discussing crustal elements. We want to address that the regional crustal sources play more important roles than the sources in distance.

Lines 263-264: "The low concentrations of heavy metals observed during this study suggest that local anthropogenic emissions were negligible." Which metals you are referring to?

Response: It refers to aerosol Pb. We have revised this sentence. Please see line 311-312.

Modified text: line 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."

Lines 264-265: "Thus the major source of non-crustal elements in aerosols over the study region may be long-range transport from regions impacted by anthropogenic emissions" very weak conclusion as it is only based on Pb variation.

Response: We agree. We have added a short discussion to show that the high concentrations of Ni, Cu, and Zn are also associated with air masses derived from coastal South America (lines 287-291). We also include a short summary for the potential sources of the anthropogenic elements in South America (lines 296-299).

Modified text:
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, miming activities were suggested as an important source, especially in remote sites in South America (Carrasco and Préndez, 1991; Klumpp et al., 2000)."

Line 271: ". . .27 and 26, in seawater" respectively?

Response: Yes. We have repaired this sentence. Please see lines 321-323.

Modified text: lines 321-323: "Thus, the Na^+/K^+ (32 ± 3.5) ratios was close to the average Na/K mass ratio in seawater (27) and the Na^+/Ca ratios (31 ± 5.5) were close to the average Na^+/Ca ratio in seawater (26) as well (Millero, 2016)."

Lines 272-273: "The results suggest that Ca was dominated by sea-salt aerosol. . ." what about K?

Response: We have added that K^+ was primarily derived from sea-salt aerosol as well, in the Discussion. Please see lines 324-325.

Modified text: lines 324-325: "Therefore, the Ca and K+ in aerosols were derived primarily from sea salt at Palmer Station."

Line 276: "classified into three groups based on their potential dominant sources" is this classification is in the context of section 3.1? If yes, it should be stated clearly. If that's not the case, provide justification of the grouping.

Response: We have revised the manuscript and added k-means clustering to classify the size distributions into 5 clusters: (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. Please see lines 330-332. We also add the clustering method in "Method". Please see section 2.3.2, lines 161-163.

Modified text:
lines 330-332: "…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."

lines 161-163: "The k-means clustering algorithm was used to cluster the average particle size distribution of each trace element. The optimal number of clusters (k) was selected by choosing the k with the highest Calinski-Harabasz index (Caliński and Harabasz, 1974)."

Lines 301-302: "The mass distributions of sea-salt elements (Ca, Na and K) as the third group were dominated by coarse-mode particles with diameters 2.5–7.8 μm (Figure 6)" I could not find the size distributions of Na and K in Figure 6!

Response: We added a new figure, the original Figure 6 now become Figure 7 and Figure 8. We have added the particle size distribution of Na^+ and K^+ in Figure 7.

Line 303: ". . .the correlation between the total concentrations of Ca and Na was strongly positive (R2±0.82, p-value < 0.01). . ." was it based on the 8 pairs of samples presented in Table 3? What about the correlations of other elements (such as K) with Na? Like Ca, If K is also associated with seasalt (as suggested in lines 270-272) one would expect a good K-Na correlation.

Response: Yes, the correlation between total concentrations of Ca and Na was based on 8 pairs of samples in Table 3. As the reviewer predicted, the correlation between Na and K was even better with a $R^2 = 0.96$. We have revised this section and removed this part.

Lines 311-313: "The rough estimates of the dry deposition fluxes of Ni, Cu, and Zn at Palmer Station. . ." should mention few values from the literature so the readers get an idea of how large or small the values are.

Response: We have showed the dry deposition fluxes of Cu and Zn measured in the North Atlantic Ocean for comparison. Please see lines 403-406.

Modified text: lines 403-406: "The rough estimates of the dry deposition fluxes of Ni and Zn at Palmer Station are close to the median deposition fluxes found in the western North Atlantic Ocean (Ni: 18 μg m^{-2} yr^{-1}, Zn: 16 μg m^{-2} yr^{-1}), whereas the dry deposition flux of Cu is slightly higher than the median Zn dry deposition flux (2.8 μg m^{-2} yr^{-1}) in the western North Atlantic Ocean (Shelley et al., 2017)."

Lines 313-315: "The estimated dry deposition fluxes of total continental dust. . . among the lowest globally (Lawrence and Neff, 2009)" should mention a few global values.

Response: Lawrence and Neff, 2009 provides an average dust deposition fluxes among

the areas receiving dust from local (0–10 km), regional (10–1000 km), and global (>1000 km) scales. For global scale at remote sites, the average dust deposition flux is 0.4 g m^{-3} yr^{-1}.We have revised this sentence in lines 408-409.

Modified text: lines 408-409: "…, and this fluxes is only around 10% of the mean global dust deposition flux at remote sites (Lawrence and Neff, 2009)."

Lines 316-319: ". . .precipitation scavenging accounted for about 40-60% of the total deposition. . ." are these fractions yearly average? Contributions of wet deposition to the total flux is a strong function of season. This study is limited to only two months of sampling. Authors need to discuss the likely uncertainties involved with extrapolating the short-term dry or wet deposition flux to yearly contributions.

Response: The range of 40-60% is a result from a modelling study. We are aware that the precipitation conditions and the proportion of wet deposition in total deposition could differ significantly in different seasons. However, due to the lack of previous measurement of dry and wet deposition fluxes in this region, it's hard to evaluate the uncertainties.

Lines 318-319: "Assuming this wet deposition fraction applies to the Antarctic Peninsula region, we approximate roughly a total dust flux of 10 mg m-2 yr-1" I understand that this is an estimation, but how was it obtained? Assuming dry/total as 0.4? or 0.6?

Response: Yes. We applied the ratio of dry deposition/total deposition (0.4 and 0.6) to the dry deposition fluxes of 5.5 mg m^{-2} yr^{-1}. The result ranged from 9.2 to 14 mg m^{-2} yr^{-1}, which is around 10 mg m^{-2} yr^{-1}. We have added the ratio (0.4 and 0.6) in this sentence to make it clear. Please see line 413.

Modified text: line 411: "Assuming this wet deposition fraction (0.4-0.6) applies…"

Lines 329-341: Authors need to carefully revise this entire paragraph to ensure it is readable.

Response: We have revised and added more information in this paragraph, and please see lines 427-441.

Modified text: Please see the paragraph discussing the importance of atmospheric input to the particulate element concentrations in surface seawater of the West Antarctic Peninsula in lines 427-441.

Lines 346-348: "Most of the samples collected during this study were impacted by air masses originating around or passing over Northern Antarctic Peninsula. . ." This claim is not supported by any strong evidence. Airmass trajectories associated with 4 of the total samples were presented in Figure 5.

Response: We have rewritten the Conclusions and Implications section. This statement has been changed to a conclusion that local/regional sources contributed to the concentrations of crustal elements. Please see lines 446-448. We have also added air mass trajectories for all samples.

Modified text: lines 446-448: "The particle size distributions of crustal elements, including Al, P, Ti, V, Mn, and Ce, were all concentrated in coarse mode, suggesting strong regional emissions likely from ice-free areas on the Antarctic Peninsula and its associated islands."

Lines 354-357: "As the role of wet deposition is unquantified at. . .may need to be reevaluated." I could not understand what this sentence is referring to.

Response: In this study, we only obtained the estimation of dry deposition fluxes, while wet deposition remains unknown. If the future measurements show that wet deposition flux in this region accounts for far more or less than the assumption we made, the total deposition flux of dust need to be reevaluated. We have revised this sentence to make it clear. Please see lines 456-460.

Modified text: lines 456-460: "As the role of wet deposition is unquantified at present and remains poorly constrained for this region, the total deposition fluxes of trace elements during the austral summer could exceed the dry deposition fluxes reported here. Therefore, the importance of atmospheric deposition of trace elements to coastal West Antarctic Peninsula may need to be re-evaluated with additional observations of wet deposition.

Figure 2: Missing proper x-axis label. In addition, the figure caption should mention what the legends (M1. . .M10) are referring to.

Response: We have revised the x-axis label and figure caption. Please see revised Figure 2.

Figure 4: This figure should be revised.
1) What is AP-OS1 shown in Figure 4(g)?
2) What are the vertical lines representing?
3) The acronyms of the sites mentioned in the caption should be consistent with the ones shown on figures (e.g., AP-PS or PS).
4) As mentioned in the text (e.g., line 211), air mass samples from many of the previous studies used for the comparison correspond to PM10 or even PM2.5. The figures should clearly indicate this (e.g., AP-OS(PM2.5)).

Response: (1) We have corrected this typo. It should be "AP-OS". (2) The vertical lines represent the standard deviation of the trace element concentrations in each study. We have included this information in the revised figure caption. (3) We have checked and

revised the incorrect acronyms. We also mark our study in red in the revised Figure 4. (4) We have marked $PM_{2.5}$ and PM_{10} samples in the figure caption. Please see the revised Figure 4.

Figure 5: Why is this figure showing 4 samples only?

Response: We have included the back trajectories for all 8 samples. Please see the revised Figure 5.

Table 4: Should include a few extra columns showing literature values.

Response: Our estimated dry deposition fluxes of trace elements are much lower than the other sites, and we have briefly compared our estimates with literature values in the text. Please see lines 403-406.

Modified text: lines 403-406: "The rough estimates of the dry deposition fluxes of Ni and Zn at Palmer Station are close to the median deposition fluxes found in the western North Atlantic Ocean (Ni: 18 μg m^{-2} yr^{-1}, Zn: 16 μg m^{-2} yr^{-1}), whereas the dry deposition flux of Cu is slightly higher than the median Zn dry deposition flux (2.8 μg m^{-2} yr^{-1}) in the western North Atlantic Ocean (Shelley et al., 2017)."

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