

Answers and explanations to all detailed questions and annotations raised by the reviewers are provided in the following. (RC: Reviewer comments; AC: Author comments).

Major comments:

RC2: ERA and NCEP data are notoriously unreliable over Antarctica, with huge biases compared to measurements. Especially the 1000m winds rely mostly on simulated model values, which are also notoriously wrong in Antarctica. Bracegirdle and Marshall (2012) may have determined that ERA-Interim data are the most accurate of the 6 reanalysis models, but that doesn't make them correct or even close to reality. What's the sensitivity of your results when using the other reanalysis datasets? How do the ERA data compare with climatology time series of monitoring stations close to MJ?

AC: *The observations in Polar Regions, such as Antarctica, are extremely scarce making reanalysis heavily simulated in these regions. Biases are to be expected where observations are scarce. Since the closest weather observation site is Byrd Station (over 550 km away and about 550 m lower in elevation), even a comparison between reanalysis models and Byrd Station would not give an accurate estimate of how reanalysis compare to atmospheric conditions at Mount Johns. We understand that biases, whether large or small, are to be expected over Mount Johns due to a lack of weather observations; however, reanalysis models are the best estimate of atmospheric states in this region and is necessary for studies such as this. Because there are several climate reanalysis datasets available for investigating climatological behavior we inter-compare our results between ERA-Interim and an ensemble average of the four leading third-generation reanalyzes models (Gen 3) (Auger et al., in review). The models within Gen 3 are CFSR (Climate Forecast System Reanalysis), MERRA (Modern Era Retrospective Reanalysis for Research and Applications), JRA55 (Japanese 55-year Reanalysis), and ERAI. The variables behavior are captured equally well in both ERAI and Gen 3 ensemble representations. Therefore, we considered that the sensitivity of the correlation results on this work using different datasets is low.*

- Auger, J.D., Birkel, S.D., Maasch, K.A., Mayewski, P.A., Schuenemann, K.C., 2017. *An ensemble average and evaluation of third generation global climate reanalysis models. J. Geophys. Res. (in review).*

RC2: The whole chapter 3.1 is methodically flawed (see minor comments below). This

puts into doubt most of the interpretation based on these data.

AC: The chapter has been rewritten and improved. We revised the whole calculation of ss and nss considering Al as crustal reference (details below). In addition, we replaced the Pearson's correlation by Principal Component Analysis.

RC2: In chapter 3.3, you mention that modeling studies suggest Australia as the main source for the Antarctic Peninsula, but that ice core studies mostly identify a mix of sources. What about your results of non-marine tracers, you don't mention these in the paper. If you decide to concentrate on marine tracers, then the calculation of nss concentrations is not necessary.

AC: *In this chapter we focus on atmospheric transport through trajectory simulations, the model does not allow us to directly use the measured concentrations. However, we made associations between the trajectories and the studied elements looking for marine and continental influences on the concentrations. Some modifications were made in the text, making clearer the discussion about nss-elements.*

RC2: You mention that in the Antarctic Peninsula, wet deposition dominates and the concentration of trace elements depends on cyclonic activity, which is episodic and seasonally variable. However, you do all the correlation analysis using annual means and I don't think that's representative.

AC: *It is difficult to separate the element concentrations into seasons. Although we discuss seasonality of elemental concentrations, it is mainly summer and winter, or low and high, respectively. Therefore, we decided that the best way to make these correlations is using annual concentrations to correlate with annual means of atmospheric variables.*

RC2: One of your conclusions is that "marine derived trace element concentrations are strongly influenced by sea ice concentration and sea surface temperature anomalies". That is a wrong conclusion; all you have is a correlation analysis, no dynamical or physical explanation to imply causation.

AC: *We do not consider this erroneous conclusion, since we use various tools (trajectory simulations, correlations with atmospheric variables, and statistical analyzes) that show us the relations between elements of marine origin and sea ice and*

also the influence of temperature on concentrations (which is clear in winter concentrations).

Minor comments:

RC2: Line 56: Mineral dust is not a source of aerosols.

AC: *The sentence was rewritten to: "Another primary source of trace elements is mineral dust"*

RC2: Line 84: Location of SST and sea ice changes?

AC: *The sentence was rewritten to: "Regional changes in atmospheric circulation and associated changes in tropical Pacific sea surface temperature and sea ice extent also directly influence the warming trend in West Antarctica"*

RC2: Line 98: Please always use SI units. In the case of clean rooms that would be the ISO 14644-1 standard. A class 100 room is equivalent to an ISO 5 level (10^5 particles per cubic meter). The class 100, 1000, etc. standard has been obsolete for over 15 years, it's time people move on.

AC: *We have researched recent references and all refer to class-100 or class-1000. We decided to keep it as in the original.*

- *Uglietti et al. 2015. Widespread pollution of the South American atmosphere predates the industrial revolution by 240 y. PNAS, v. 112(8), p. 2349-2354.*
- *Tuohy, A. et al. 2015. Transport and deposition of heavy metals in the Ross Sea Region, Antarctica, Journal of Geophysical Research: Atmospheres, v. 120 (20), p. 10996-11011.*

RC2: Line 115-119: That method makes no sense to me, although I may just be ignorant of this matter. The standard deviation of the measurements should have no relevance for the detection limit? The instrument could be very precise at medium range, but have a detection limit greater than its precision. Or did you mean the average of the blanks plus 3 times the std?

AC: *The instrument is very precise. Blanks are made to control possible contaminations during the melting process and the acidification. Considering that the blanks (ultrapure water) are coming from different steps of the analytical procedure, it can be considered the real limiting factor for the determination of trace elements at the low and sub pg/g*

level. The detection limits were defined as three times the standard deviation of blank samples (10 blank samples were used). Concentrations below the detection limits were disregarded. This occurred in less than 1% of the samples.

Reference used:

- Barbante *et al.* 1997. Direct determination of heavy metals at pictogram per gram levels in Greenland and Antarctic snow by double focusing inductively coupled plasma mass spectrometry. *Journal of analytical atomic spectrometry*, v.12, p. 925-931)

RC2: Line 120-125: Where did the samples come from? Did you send frozen pieces of the ice core to Brazil? If so how were they treated in Brazil? Or did you send aliquots from the fraction collector? If so how did you send them? Frozen?

AC: *The samples collected at the CCI were sent frozen to Brazil and melted on the day of analysis.*

RC2: Line 127-129: Briefly mention here why S can be used to count layers. Why did you not use other measurements, such as Ca or Al, for the layer counting?

AC: *The dating was improved with the use of stable isotope data (these data were not available until then). More details about dating will be added to the text and to the supplement information. Manual interpretation of the data was done by multiple individuals to identify the individual layers. The CCI software package (Kurbatov *et al.*, 2005) was also used to identify matching seasonal peaks from Ca, Na and Sr and the major historical volcanic eruptions. In this study, water isotopes were used to confirm the dating previously performed in Schwanck *et al.*, 2016.*

RC2: Line 148 – 152: Where is your dust source? If it's Oceania are 5 days enough to transport the particles across the Pacific?

AC: *We believe MJ presents a mixture of sources with main contribution from Australia followed by South America. The five day simulation is an appropriate time-length when considering the maximum lifetime transport (10 days) of small size (0.1 – 2.5 μm) fractions of mineral dust and other aerosols, while transport of large particles (> 2.5 μm) is likely restricted to the first few days.*

RC2: Line 173: The regression line in Figure S1 is just ridiculous. Obviously there is no linear relationship between Na and nssS. Please use common sense and don't blindly apply methods found in other papers.

AC: *This part has been removed. Due to less than 1% of the samples being affected by sulfur fractionation we decided not to apply the correction.*

RC2: Line 202-209: How can you distinguish your calculated excess from the error introduced by crustal Na and oceanic Al? I doubt anything below 10% contribution is significant, once you calculate the calculation uncertainty due to these effects.

AC: *We only use ssNa for the calculation and the oceanic contribution for the aluminum is so low that we consider insignificant. See calculations of nss and ss added. Non-sea-salt ratios were calculated using the equation reported below (Palmer et al., 2002, Becagli et al., 2005):*

$$nssS = S - 0.084 \times ssNa,$$

where S is the total sulfur concentration on the sample, 0.084 is the mean S/Na ratio in seawater (Lide, 2005) and ssNa is the Na actually derived from sea spray. Since some Na derives from continental dust, ssNa was calculated using the four-equation system reported below:

$$ssNa = Na - nssNa$$

$$nssNa = nssAl \times (Na/Al)_{crust}$$

$$nssAl = Al - ssAl$$

$$ssAl = ssNa \times (Al/Na)_{seawater},$$

where the mean Na/Al ratio is 0.3315 in the crust (Wedepohl, 1995) and the mean Al/Na ratio is 0.000000185 in seawater (Lide, 2005).

RC2: Line 214-220: Have you looked at the distributions? Are the elements normally distributed? I doubt it and you cannot use Pearson's correlation then. Try the Spearman or Kendall correlation instead. And redo the classification of crustal and marine elements.

AC: *No, the elements do not present normal distribution. We improved the analysis using Principal Component Analysis. The PCA resulted in four PCs. PC1 is dominated by Ba, K, Mg, Mn, Na, and Sr, accounting for 42.24% of the total variance. PC2, dominated by Al and Ti, accounts for 13.27% of the total variance, while K and Na are*

negatively correlated. PC3 is dominated by Ba, Fe, and Ti, accounting for 11.16% of the total variance. PC4 is dominated by Ca and Sr, accounting for 8.11% of the total variance, while S and Mn are negatively correlated. We are still working on the classification of these results.

RC2: Line 226-227: It may be best to remove table S3 unless you can address all the comments above.

AC: *Due to the changes we made in the text, we decided that Table S3 was no longer needed and was removed.*

RC2: Line 253: Mean of what? And do you really have a 0.01 pg/g measuring accuracy? Please go through all the text and remove all those decimals.

AC: *Yes. Our analyses have this accuracy.*

RC2: Line 325-327: How exactly were these classification defined? It sound rather subjective to me, was there an objective criteria? What about South American influence?

AC: *These classifications are defined by characteristics identified from the mean trajectories of each cluster, speed (proportional to trajectory length), source region, and pathway.*

RC2: Line 333-334: You don't need to cross the Pacific from South America. The South American contribution would come through the South Atlantic cluster.

AC: *Yes. We agreed.*

RC2: Figure 2: Have the same sequence of elements in both (a) and (b) plots.

AC: *The elements are now in the same sequence.*

RC2: Figure 3: remove “concentrations” after variability

AC: *Figure 3 was removed from the text and added to the supplement information. Mg was replaced by Al in the graph.*