

We very much thank the two reviewers for their thorough analysis of our paper and for their valuable comments and suggestions. They had been carefully considered and most of them are accounted in the revised manuscript. Answers and explanations to all detailed questions and annotations raised by the reviewers are provided in the following.  
(RC: Reviewer comments; AC: Author comments).

## Reviewer comments 1

### Specific Comments

10 **RC 1:** The description about the development and validation of the chronology is rather poor, not to say completely lacking. If high resolution meteorological data are compared to record obtained from natural archives, it is necessary for the latter ones to be accurately and precisely dated. The authors just state that the chronology was based on annual layer counting (using Na and S records) with the additional consideration of 4 major volcanic eruptions. No further details are given. A previous work (Schwanck et al., 2016 Atmos. Environ.) is cited as reference for the chronology, but also in this work few details are found. This part needs a substantial extension. A first element would be the comparison between annual layer counting and historical eruptions, which error is found? Is this consistent with a record which is claimed to present a seasonal resolution?

20 **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.*

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**RC 1:** Another important part of the paper is dedicated to the calculation of different contributions for each element, i.e. crustal, volcanic, marine and biogenic. This part is a little bit confused. The authors follow three different approach: the selection of reference elements and reference elemental ratios, the

calculation of enrichment factors and the calculation of Pearson's coefficients. It would be important to  
30 put all this elements together, discussing them in a comprehensive way and not separately. If the  
discussion is kept separated controversial results are found. For example, we can consider Mg.  
According to the use of reference, elements and ratios it has a dominant marine source (30 %, supplementary material) and a secondary associated to crustal material (5 %). But Pearson's coefficients  
35 reveal that Mg is strongly associated to Al, a typical crustal element. Also successive interpretation  
about the comparison with meteorological data point to strong similarities. The application of a multi-  
variate statistical tool as principal component analysis could greatly improve this section of the work.  
PCA could help the authors to identify different contributes and to understand the role played by each  
element in these different contributions. Since its starting point is the calculation of Pearson coefficients  
please consider to make a further step in this sense and complete data treatment with PCA.

40 **AC:** *We agreed that this chapter was confusing, so we decided to take the comment into account and  
redo the analysis using PCA. 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  
45 the total variance, while S and Mn are negatively correlated. We are still working on the interpretation  
of these results.*

**RC 1:** In addition I suggest the authors to improve the method they used to distinguish ss and n-ss Na.  
The assumption that Al is only crustal is justified, but this is not the case for the assumption that Na is  
50 only marine. Please consider to separate the two fractions by using Al as crustal reference and an UCC  
Al/Na ratio.

**AC:** *We revised the whole calculation of ss and nss taking Al into account as crustal reference.  
Non-sea-salt ratios were calculated using the equation reported below (Palmer et al., 2002, Becagli et  
al., 2005):*

55 
$$nssS = S - 0.084 \times ssNa,$$

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:

$$ssNa = Na - nssNa$$

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

60  $nssAl = Al - ssAl$

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

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).

65 **RC 1:** Section 3.2 should be deeply revised. In its current version it seems a review about atmospheric depositional issues in Antarctica, but very poor discussion points are reported. High time resolution data described in this work should be better exploited to understand seasonal dynamics. Are elements presenting parallel seasonal oscillations? If this is not the case and no significative observations are found please consider to dramatically shorten the section and to merge it with section 3.4, so as to have  
70 a single section about temporal variability.

**AC:** *We agree. Sections 3.2 and 3.4 have been merged into a single section (Section 3.2 - Interannual atmospheric variability). We changed the order of discussion, placing the transport session (3.3 - Atmospheric transport to Mount Johns ice core site) to the end.*

*We are also expanded the discussion of results.*

75

**RC 1:** Section 3.4 I suggest to develop the discussion presented here with a comparison with back-trajectories analysis and seasonal trends. Some interesting trends are observed but their interpretation is poor. The authors present a huge amount of observations concerning literature and what was observed in other studies, but the connection between their evidences and literature is lacking. For example  
80 looking at Fig.7 the correlation of  $Al$  and  $Mg$  is completely different with respect to the other elements. This is clearly pointed in the text, but a true interpretation is missing. The phenomenon could be related

to a different seasonality pattern, with dust peaks and marine aerosol peaks occurring in different periods of the year, when SST is different.

85 **AC:** *The section is being rewritten and improved. We agreed that some interpretations were superficial and we are expanding on these issues. The order of the sessions was changed, leaving the discussion of transport to the end. We are also expanding the discussion on how transport and seasonality have affected trace elements concentrations on MJ area.*

## Technical Comments

90 **RC 1:** Line16: insert “of” between “reanalysis” and “trace”

**AC:** *Done*

**RC 1:** Line22: remove “that of the”

**AC:** *Done*

95

**RC 1:** Line27: please consider to add a short passage about the importance of WAIS in relation to climate dynamics and sea level.

**AC:** *The text was changed to: “During recent decades, rapid changes have occurred in the WAIS sector, including flow velocity acceleration, retraction of ice streams, and mass loss (Pritchard et al., 2012). These changes influence the global climate through their contributions to sea level rise (Pritchard et al. 2009, Shepherd et al. 2012) and deep ocean circulations (Holland and Kwok, 2012). WAIS contains sufficient water to raise the global sea level by over 3 m (Bamber et al., 2009, Fretwell et al., 2013)”.*

100

105 **RC 1:** Line40: change “recognize” with “distinguish”

**AC:** *Done*

**RC 1:** Line41-42: remove from “furthermore” to “continent” and replace with “both presenting specific seasonal cycles.”

110 **AC:** *The text was changed to: “To interpret chemistry records from Antarctic ice cores, it is imperative to distinguish the long-range transportation of continental dust and regionally derived sea salt, both presenting specific seasonal cycles.”*

**RC 1:** Line54-56: please reformulate, it is not clear.

115 **AC:** *The text was changed to: “Marine aerosol concentrations are strongly linked to cyclone frequency and intensity that provides high wind speeds over the ocean surface, with the aerosols deposited along the storm track.”*

**RC 1:** Line56: change with “Another primary source of aerosol is mineral dust. It is  
120 transported. . .”

**AC:** *Done*

**RC 1:** Line57: add a further reference. Li et al., 2008 is based only on models, add Revel-Rolland et al., 2006 EPSL which is based on isotopic data from EPICA Dome C. Also the consideration of New  
125 Zealand as dust source for Antarctica is still only an hypothesis based on modeling works, no direct evidences are known.

**AC:** *Done*

**RC 1:** Line69: add “of WAIS” after “systems”

130 **AC:** *Done*

**RC 1:** Line76: please give a reference for modern snow accumulation rates in the considered area

**AC:** *We add Medley et al., 2014*

135 **RC 1:** Line86: In the text, Mount Johns is never described. Is it a topographical height of Pine Glacier? A peripheral area of this glacial system?

**AC:** *Mount Johns is a nunatak in the Pine Island Glacier area.*

**RC 1:** Line116-117: please specify only significant digits

140 **AC:** *We have removed the values. Details about MDL values are contained in the supplement information.*

**RC 1:** Line310: some references concern Talos Dome, which is located in EAIS, not WAIS.

**AC:** *The reference was being used as an example but to avoid misunderstanding we decided to remove.*

145

**RC 1:** Table1: is it possible to add a further column with average uncertainty for each element?

**AC:** *Done*

**RC 1:** Figure2: I guess that y-axis of upper figure is wrong. Al EF should be 1, not 0.1. Is this right?

150 **AC:** *This is correct; some elements presented EFC less than 1.*

**RC 1:** Figure3: here you present some examples to show seasonal variations. You considered Na and Mg. What about considering also Al? Being exclusively crustal it could present a different behavior.

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

**RC 1:** Figure4: specify in the caption that volcanic eruptions were identified using sulfates.

**AC:** *Done*

160 **RC 1:** Figure6-7-8: Why for each figure you report different elements. It would be nice to have three perfectly comparable figures with all the elements you considered in this work. Did you try to apply the same procedure to nss and ss-S. It would be nice to see them.

**AC:** *The graphs that were not shown in the figure did not present very significant results. These have now been added. We did not simulate for ss-S and nss-S only for total S.*

165

## Reviewer comments 2

### 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.

195 **AC:** The chapter has been rewritten and improved. We revised the whole calculation of ss and nss considering AI 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  
200 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*  
205 *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  
210 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*  
215 *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  
220 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).*

225

**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”*

230 **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”*

235 **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.*

- 240
- *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.*

245 **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 it's 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*

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*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.*

255 *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)*

260 **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.*

265 **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.*

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

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**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.*

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**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*  
285 *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.

290 **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):*

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

295 *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:*

$$ssNa = Na - nssNa$$

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

$$nssAl = Al - ssAl$$

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

*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).*

305 **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.*

315 **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.*

320 **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?

325 **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.

330 **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.*

335 **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.*

## 340 **A 125-year record of climate and chemistry variability at the Pine Island Glacier ice divide, Antarctica**

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**Abstract.** The Mount Johns (MJ) ice core (79°55'S; 94°23'W) was drilled near the Pine Island Glacier ice divide on the West Antarctic Ice Sheet during the 2008–2009 austral summer, to a depth of 92.26 m. The upper 45 m of the record covers approximately 125 years (1883–2008) showing marked seasonal variability. Trace element concentrations in 2,137 samples were determined using inductively coupled plasma mass spectrometry. In this study, we reconstruct mineral dust and sea salt aerosol transport and investigate the influence of climate variables on the elemental concentrations to the MJ site. The ice core record reflects changes in emissions as well as atmospheric circulation and transport processes. Our trajectory analysis shows distinct seasonality, with strong westerly transport in the winter months and a secondary northeasterly transport in the summer. During summer months, the trajectories present slow-moving (short) transport and are more locally influenced than in other seasons. Finally, **our reanalysis correlations with trace element** suggest that marine derived trace element concentrations are strongly influenced by sea ice concentration and sea surface temperature anomalies. The results show that seasonal elemental concentration maxima in sea-salt elements correlate well with the sea ice concentration winter maxima in the West Amundsen and Ross Seas. Lastly, we observed an increased concentration of marine aerosols when sea surface temperature decreased.

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355

## 1 Introduction

360 The West Antarctic Ice Sheet (WAIS) is more susceptible to marine influences than East Antarctica Ice Sheet (EAIS). The lower average elevation of the WAIS compared to EAIS, 1100 m and 3000 m, respectively, (Bedmap 2 project data; Fretwell et al., 2013) facilitates the advection of air masses toward the interior of the continent, thereby directly contributing to the ice sheet's surface mass balance through precipitation (Nicolas and Bromwich, 2011). During recent decades, rapid changes have occurred in the WAIS sector, including flow velocity acceleration, retraction of ice streams, and mass loss (Pritchard et al., 2012). These changes influence the global climate through their contributions to sea level rise (Pritchard et al. 2009; Shepherd et al. 2012) and deep ocean circulations (Holland and Kwok, 2012). WAIS contains sufficient water to raise the global sea level by over 3 m (Bamber et al., 2009; Fretwell et al., 2013).

Presently, Pine Island Glacier (PIG) is responsible for 20 % of the total ice discharge from the WAIS (Rignot et al., 2008). Accelerated thinning observed since the 1980s is directly linked to enhance sub-ice-shelf melting, which is induced by the recent alteration of Circumpolar Deep Water (Pritchard et al., 2012; Steig et al., 2012; Favier et al., 2014). Sea level pressure and geopotential height anomalies, associated with increased strength of the circumpolar westerlies (Steig et al., 2012), favor reduced sea ice extent in the Amundsen and Bellingshausen Seas (Schneider et al., 2011) and the advection of warm air onto the continent (Steig et al., 2009; Ding et al., 2011). In this context, both atmospheric and oceanic variability are important for determining the response of the WAIS over long time-scales

Polar ice cores contain important chemical records linked to the climate that extend continuously for the past 800,000 years (Lüthi et al., 2008; Delmonte et al., 2008), making them a valuable tool for interpreting climate trends. Despite the increase in ice core sites in Antarctica, they are spatially distant. In particular, there is a lack of ice core records from the WAIS. Previous studies have emphasized the importance of measuring elemental composition of ice cores in this region in order to understand the recent atmospheric circulation changes (Criscitiello et al., 2014; Pasteris et al., 2014; Tuohy et al., 2015). To interpret chemistry records from Antarctic ice cores, it is imperative to distinguish the long-range transportation of continental dust and regionally derived sea salt, both presenting specific seasonal cycles.

Major and minor impurities in polar ice originate from sources such as oceans, landmasses, volcanism, biogenic activity, biomass burning, and anthropogenic inputs (Legrand and Mayewski, 1997; Planchon et al., 2002; Vallelonga et al., 2004; Weller et al., 2008; Dixon et al., 2013; Schwanck et al., 2016). These impurities are transported over long distances to the polar regions through the troposphere (Petit and Delmonte, 2009) and stratosphere, e. g. volcanic emissions (Krinner et al., 2010). Sea salt and mineral dust can be used to reconstruct climate conditions as well as atmospheric transport patterns (Albani et al., 2012; Chewings et al., 2014). These chemical elements are strongly influenced by the climate conditions in the source region, and the depositional record provides important information about cyclone activity, wind intensity (Koffman et al., 2014a), sea ice conditions (Criscitiello et al., 2014), and aridity and vegetation cover (McConnell et al., 2007).

Sources can be either primary, such as sea salt or mineral dust, or secondary, such as those produced in the atmosphere through oxidation of trace gases (Legrand and Mayewski, 1997). Sea salt is produced by two dominant mechanisms: 1) bubble bursting and sea spray over open water (Leeuw et al., 2011), and 2) by the formation of frost flowers on the surface of young sea ice at high latitudes (Kaspari et al., 2005; Fischer et al., 2007). Marine aerosol concentrations are strongly linked to cyclone frequency and intensity that provides high wind speeds over the ocean surface, with the aerosols deposited along the storm track (Fischer et al., 2004). Another primary source of trace elements is mineral dust. It is transported from arid, continental regions such as Australia (Revel-Rolland et al., 2006; Li et al., 2008), South America (Delmonte et al., 2010; Li et al., 2010), and probably New Zealand (Neff and Bertler, 2015) to the Antarctic. Once entrained into the atmosphere, small grains (0.1 to 5  $\mu\text{m}$ ) can be transported over long distances (Gaiero et al., 2007; Mahowald et al., 2014) through advection before they are deposited on the Antarctic snow surface (Prospero et al., 2002; Delmonte et al., 2013). Here, we present a 125-year record of eleven trace elements (Al, Ba, Ca, Fe, K, Mg, Mn, Na, S, Sr, and Ti) from the Mount Johns (MJ) ice core in the WAIS. We focus on the influence of atmospheric circulation, surface temperature, and sea ice concentration on the transportation of mineral dust and sea salt aerosol to the MJ site. Correlations were made between trace element concentrations and the European Centre for Medium Range Weather Forecasts (ECMWF) Reanalysis Interim (ERA-Interim; Dee et al., 2011) climate variables. Modeled air mass trajectories were also employed to explore atmospheric transport, independent of deposition processes, from 1979 to 2008.

## 2 Methods

### 2.1 Site description and core collection

Pine Island, Thwaites, and Smith Glaciers are the principal drainage systems of WAIS into the Amundsen Sea (Shepherd et al., 2002; Rignot et al., 2002). These glaciers receive a significant amount of precipitation due to their low elevation and coastal locations. The lower elevation allows moisture-rich cyclones to penetrate deeper into the interior (Nicolas and Bromwich, 2011). Kaspari et al. (2004) presented accumulation records from four ice core sites collected within the Pine Island-Thwaites drainage system during the International Trans-Antarctic Scientific Expedition (ITASE). Based on these records, they found that recent snow accumulation (between 1970 and 2000) had increased when compared to the 1922–1991 average. However, radar-derived annual accumulation records show no significant trend over Thwaites Glacier between 1980 and 2009 (Medley et al., 2013).

Satellite observations show ice mass loss along the Bellingshausen and Amundsen Seas has increased over, at least, the past two decades (Rignot et al., 2008; Medley et al., 2014) as a result of ocean-driven basal melt (Pritchard et al., 2009; Pritchard et al., 2012). WAIS is grounded below sea level where warming waters rapidly melt the ice shelves resulting in increased glacial flow (Mercer, 1978; Bamber et al., 2009), further draining the ice sheet (Steig et al., 2012; Dutrieux et al., 2014). 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 (Steig et al., 2009; Pritchard et al., 2012). Recent studies

have indicated that the pronounced warming in the central tropical Pacific Ocean is related to a pressure anomaly north of the Amundsen Sea Embayment, an increase in sea surface temperatures (SST), and negative anomalies in sea ice extent (Steig et al., 2009; Ding et al., 2011; Steig et al., 2012).

In this study, we use an ice core drilled at the nunatak Mount Johns and near the Pine Island Glacier divide (Fig. 1) to reconstruct mineral dust and marine aerosol transport and the influence of climate variables on the elemental concentrations. The MJ ice core (79°55'28" S, 94°23'18" W, and 91.20 m depth) was recovered in the austral summer of 2008/2009. The ice thickness at the ice core site reaches 2,115 m (determined from the Bedmap 2 project data; Fretwell et al., 2013). The average accumulation rate for the period is 0.21 m water equivalent per year (based on calculated field measurements of snow and ice density) and the mean surface temperature (measured at a depth of 12 m) is -33°C, measured using a calibrated platinum probe.

Drilling was performed using the Fast Electromechanical Lightweight Ice Coring System (FELICS) (Ginot et al., 2002). The MJ core (8.5 cm diameter) was cut into sections of approximately 1 m in length, packed in polyethylene bags and then stored in high-density Styrofoam boxes and transported by air to Punta Arenas, Chile. Then, it was sent frozen to the Climate Change Institute (CCI) at the University of Maine, USA, where it was sub-sampled and analyzed using Inductively Coupled Plasma Sector Field Mass Spectrometry (ICP-SFMS).

## 2.2 Laboratory analyses

Decontamination of the MJ ice core was carried out in a certified cold room class 100 with temperature below -20°C. Prior to melting, core ends and breaks were manually scraped using a clean ceramic knife to reduce potential contamination from drilling or other sources (procedure performed according Tao et al., 2001 and described in Schwanck et al., 2016). The core was melted using a continuous ice core melter system developed by CCI researchers (details in Osterberg et al. 2006). This system uses fraction collectors to gather discrete, high resolution, continuous and co-registered meltwater samples. Our samples were collected into acid-cleaned (Optima HNO<sub>3</sub>), low density polyethylene (LDPE) vials and acidified to 1 % with double-distilled HNO<sub>3</sub>. All sample handling was conducted in a class 100 clean room under a laminar flow High Efficiency Particle Air (HEPA) bench. The samples were allowed to react with the acid (at room temperature) at least one month before analysis. This process is important for the dissolution of particulate material (following studies Rhodes et al., 2011 and Koffman et al., 2014b).

Trace element concentrations in 2,137 discrete samples, corresponding to the upper 45 m of the MJ ice core, were determined using the CCI Thermo Electron Element2 ICP-SFMS coupled to an ESI model SC-4 autosampler. Working conditions and measurement parameters are described in Table S1 (Supporting Information). The ICP-SFMS is calibrated daily with five standards that bracket the expected sample concentration range. Although there is no certified reference material for trace elements in polar snow and ice, the analyzed samples were certified with water reference material (SLRS-



455 4, National Research Council Canada, Ottawa, Canada) to ensure the concentrations were within the certification range, confirming the accuracy of this method (details in Osterberg et al., 2006).

Samples of de-ionized water, or “blanks”, were prepared, treated, and analyzed using the same method as snow samples. The Method Detection Limits (MDL) were defined as three times the standard deviation of blank samples (10 blank samples were used). Concentrations below the MDL were replaced with the MDL values, which occurred in a few cases for Al, Ba, 460 Ca, Fe, Mn, Sr, and Ti (less than 1 %). Blank concentrations and MDLs were either similar or less than published values using comparable methods and instruments (Table S2 – Supporting Information).

Water isotope analyses were performed at the Centro Polar e Climatico, Brazil, Isotopes Lab using a Picarro L2130-i wavelength-scanned cavity ring-down spectroscopy (WS-CRDS) instrument (Picarro Inc., USA). The samples collected at the CCI were sent frozen to Brazil and melted on the day of analysis. Aliquots of water were filled in 2 ml glass vials and 465 sealed with polytetrafluorethylene/silicone caps. The vials were then placed in a PAL COMBI-HTCxt autosampler (CTC Analytics AG, Switzerland) connected to the Picarro L2130-i for  $\delta D$  and  $\delta^{18}O$ . Reproducibility of measurements is typically 0.9 ‰ for  $\delta D$  and 0.2 ‰ for  $\delta^{18}O$ .

The aerosol records show strong seasonal variations (chapter 3.2) due to seasonality in aerosol sourced and transport efficiency, and these seasonal signals can be used to detect annual layers (e.g. Sigl et al., 2016). A typical annual layer at MJ 470 site is characterized by a maximum of sea-salt aerosol deposition (e.g. Na, Sr) during austral winter, and a maximum of marine biogenic aerosol emission (e.g. S) in late austral summer. 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. The major historical volcanic events during this 475 period, such as Pinatubo (1991), Agung (1963), Santa Maria (1902), and Krakatoa (1883) are identified by large peaks in S concentration and are used as absolute time horizons during timescale development (Fig. S1 – Supporting Information). On the basis of our dating, the upper 45 m of the MJ ice core covers the period from the 1883 to 2008 A. D. We estimate a maximum dating error of  $\pm 1$  years between the 1991 A.D. Pinatubo and 1963 A.D. Agung eruptions, and  $\pm 2$  years between the 1963 A.D. Agung and 1883 A.D Krakatoa eruptions.

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## 2.3 Meteorological data analyses

To explore possible sources of the observed trace elements in aerosols, air mass backward trajectories were simulated for 1000 m above ground level over the MJ ice core site. Trajectory simulations were made using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HySPLIT) model, developed by the NOAA Air Resources Laboratory (Draxler et al., 485 2010) in conjunction with the global reanalysis datasets from the National Centers for Environmental Prediction (NCEP) and the National Center for Atmospheric Research (NCAR), known as NCEP/NCAR reanalysis model (NCEP1) (Kalnay et al., 1996; Kistler et al., 2001). Despite limitations prior to the satellite era (1979), the NCEP/NCAR reanalysis model represents

a useful tool for understanding the climate of the Southern Hemisphere since 1979 (Bromwich and Fogt, 2004) and has been used with success in Antarctic back-trajectory modeling (e.g. Sinclair et al., 2010; Dixon et al., 2011; Markle et al., 2012).

490 Five-day (120 hr), 3-D back-trajectories were created from the MJ site at 00:00 UTC daily from January 1979 to December 2008 (a total of 10,655 trajectories). We have tested the effect of model initiation heights at 500 m, 1000 m, and 1500 m on our trajectories and confirmed they are spatially consistent. The initial heights of 500 m are affected by surface topography, which is imperfectly represented in the reanalysis model (Dee et al., 2011). For this reason, we have chosen the 1000 m level as the initial height condition for our back trajectories. At this altitude, orographic influences are minimized while the trajectories are sufficiently close to the terrain to be dynamically linked to the surface wind field (Sinclair et al., 2010). The 495 five day simulation is an appropriate time-length when considering the maximum lifetime transport (10 days) of small size (0.1 – 2.5  $\mu\text{m}$ ) fractions of mineral dust and other aerosols, while transport of large particles ( $> 2.5 \mu\text{m}$ ) is likely restricted to the first several days (Albani et al., 2012). Cataldo et al. (2013) measured the size of dust particles in a separate ice core drilled near MJ and found that the mean dust size within the core ranges from 1.2  $\mu\text{m}$  to 2.4  $\mu\text{m}$ .

500 In order to obtain information about airflow patterns at the MJ site, a cluster analysis was applied to a database of individual trajectories (10,655 daily trajectories). The HySPLIT model's cluster analysis algorithm groups trajectories by minimizing the spatial variability between trajectories within some defined number of clusters (Draxler, 1999). For the trajectories presented here, it is determined that five clusters are sufficient to capture seasonal variability during the 1979 to 2008 period. Bracegirdle and Marshall (2012) determined that ERA-Interim was the most accurate of six reanalysis models over 505 Antarctica when compared against surface and midtropospheric pressure and temperature observations. ERA-Interim was thus utilized to provide annual mean 2-m air temperature ( $T_{2m}$ ), sea surface temperature (SST), and sea ice concentration (SIC) from 1979 to 2008. ERA-Interim outputs were obtained from the ECMWF Data Server (<http://apps.ecmwf.int/datasets/>) at a resolution of 1.5°. Spatial correlations were performed between MJ ice core element data and ERA-Interim climate variables.

## 510 **3 Results and discussion**

### **3.1 Glaciochemical records**

Concentrations of 11 trace elements (Al, Ba, Ca, Fe, K, Mg, Mn, Na, S, Sr, and Ti) were measured in 2,137 discrete ice core samples. Table 1 shows a statistical summary of the trace element concentrations measured from the MJ ice core.

The soil and rock dust contribution for the measured trace elements were given by the crustal Enrichment Factor (EF<sub>c</sub>) 515 according to Eq. (1) (Osterberg, 2007):

$$\text{EF}_c = \frac{X_{\text{ice}}/A_{\text{ice}}}{X_{\text{ref}}/A_{\text{ref}}}, \quad (1)$$

where  $X_{ice}$  is the trace element concentration in the sample,  $Al_{ice}$  is the aluminum concentration in the sample, and  $X_{ref}$  and  $Al_{ref}$  are the trace element and the aluminum concentrations in the reference material, respectively. Aluminum was used as the reference element in this work because it is one of the major constituents of the earth's crust (Planchon et al., 2002). The mean elemental concentration used for reference is the average composition of the upper continental crust taken from the literature (Wedepohl, 1995).

Figure 2 shows trace element EFC values during the austral summer and winter months in the MJ ice core. Elements with EFC lower than 10 are considered to be non-enriched and predominantly have a crustal dust origin (Duce et al., 1975). EFC higher than 10 indicates contributions from other sources, such as marine aerosol, volcanism, biogenic activity or anthropogenic emissions. Marine aerosol contributions were estimated using the oceanic enrichment factor (EFO) according to Eq. (2) (Osterberg, 2007):

$$EFO = \frac{X_{ice}/Na_{ice}}{X_{ref}/Na_{ref}}, \quad (2)$$

where  $X_{ice}$  is the trace element concentration in the sample,  $Na_{ice}$  is the Na concentration in the sample, and  $X_{ref}$  and  $Na_{ref}$  are the trace element and Na concentration in the reference material, respectively. Sodium is used as the reference element because it is the main sea salt constituent (Weller et al., 2008; Dixon et al., 2013). We used the average composition of ocean water (Lide, 2005) as a reference for the ocean elemental abundances.

Non-sea-salt sulfur ratios were calculated using Eq. (3) (Palmer et al., 2002; Becagli et al., 2005):

$$nssS = S - 0.084 \times ssNa, \quad (3)$$

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 (4) reported below:

$$ssNa = Na - nssNa$$

$$nssNa = nssAl \times (Na/Al)_{crust} \quad (4)$$

$$nssAl = Al - ssAl$$

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

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). Wagenbach and others (1998) suggested that the seawater ratio at Antarctic coastal sites is variable because of fractionation processes (formation of frost flowers on fresh sea ice and precipitation of mirabilite at temperatures below -8.2°C). It can be recognized by obtaining negative nssS concentrations. The Mount Johns record shows a deficit of nssS during the winter in a few years. However, it corresponds to less than 1% of the samples and we consider that the sulfur fractionation does not affect the ratio in the MJ area.

The sulfur contribution from marine phytoplankton activity (via atmospheric oxidation of dimethylsulfide produced by marine phytoplankton during summer months) was estimated from the methanesulfonate (MSA is a specific marker of

marine biogenic emissions) concentration (following Udisti et al., 2016). About 30% of the nssS concentration in the MJ area was considered from biogenic origin.

Approximately 10 to 15 % of the non-sea-salt sulfate/sulfur concentration in the Antarctic atmosphere originates from volcanic activity (Boutron and Paterson, 1986; Hur et al., 2007). We used the Hinkley et al. (1999) element/S ratios to calculate inputs from the global mean volcanic quiescent degassing background for the element Mn and S (there is not data available for the other elements). Furthermore, we used the metal/S ratios from the Mount Erebus (77°32'S, 167°10'E) plume (Zreda-Gostynska et al., 1997) to represent local source contributions for Al, Ca, Fe, K, Mn, Na, S, and Ti. The first step to calculating the volcanic contribution is to remove the oceanic and crustal fraction of elements. We are left with the excess [excess = total – (oceanic + crustal)] elemental concentrations to calculate the local (3 % - 5 %) and global (10 % - 15 %) volcanic contributions (Table S3 - Supporting Information).

Only S and Mn show significant input of volcanic emissions with contributions ranging from 20 to 33 % from regional volcanic sources and 3 to 5 % from global emissions for mean excess elemental concentrations. The other elements (Al, Ca, Fe, K, Na, and Ti) presented less than 1 % of volcanic input. Due to lack of data in the literature, we did not calculate volcanic increments for the elements Ba, Mg, and Sr.

To evaluate the origin and transport of the trace elements determined in the MJ ice core, we applied the principal component analysis (PCA) and cluster analysis (CA) in order to reveal linear relations existing in the chemical data set. For our study, OriginPro 2017 for Windows (OriginLab Corporation, USA), was utilized for the multivariate statistical analysis, and for descriptive and correlation analyses.

PCA is widely used to reduce data (Loska and Wiechuya, 2003; Gabrielli et al., 2008) and to extract a small number of latent factors (principal components, PCs) for analyzing relationships among the observed variables (Table S4 - Supporting Information). The PCA resulted in four PCs reporting 74.77% of the total variance of 11 chemical variables (Table 2). 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, and with K and Na negatively correlated accounts for 13.27% of the total variance. PC3 is dominated by Fe, accounting for 11.16% of the total variance, while Mg is negatively correlated. Finally, PC4 is dominated by Ca and Sr, accounting for 8.11% of the total variance, while S and Mn are negatively correlated.

Cluster analysis (CA) was performed to further classify elements of different sources based on the similarities of their chemical properties. In this study was used Ward's method, with Euclidean distances as the criterion for forming clusters of elements. Figure 3 displays four clusters: (1) Al–Mg–S; (2) Ba–Mn–Ti; (3) Fe–K; (4) Ca–Na–Sr. It is observed, however, that clusters 2 and 3 join together at a relatively higher level implying perhaps a common source, while the distance between Al–Mg and S in cluster 1 may suggest that this cluster can be further divided into two sub clusters.

Based on the analysis of crustal and marine enrichment factors, PCA and CA, we have classified the concentrations as predominantly crustal for the elements Al and Mg, while Na, Sr, and Ca are primarily sea salt derived elements. The elements Ba, Mn, Ti, Fe, and K show to be from mixed sources (mineral dust and sea salt aerosol). Furthermore, the S record has a considerable volcanic and biogenic input and Mn has an additional volcanic input. We acknowledge that Fe may have

an additional contribution of biomass burning aerosol (Winton et al., 2016), but due to lack of data we will not address this issue here. Based on the low values of E<sub>Fc</sub> and E<sub>Fo</sub>, we consider that the presented concentrations are from natural origin and possible anthropogenic contributions to these elements would be insignificant in this area.

### 585 3.2 Intraseasonal and Interannual atmospheric variability

Generally, trace element concentrations from sea salt aerosol observed in coastal and interior West Antarctic ice cores show a clear seasonal signal, with higher concentrations in austral winter and spring months (June to November) and lower concentrations in austral summer months (December-February) (Legrand and Mayewski, 1997; Wolff et al., 2003; Kaspari et al., 2005; Sigl et al., 2016). Impurities from continental dust can peak in both the austral summer (Weller et al., 2008; Tuohy et al., 2015) or winter months (Hur et al., 2007), depending on site location. Additionally, biogenic aerosols (e.g. sulfur) show peaks in summer months due to an increased phytoplankton activity (Weller et al., 2011). We found high concentrations in austral winter and low concentrations in austral summer for most of the elements analyzed, with the exception of sulfur, which presents peaks in the summer due to the biogenic contribution and of the elements with crustal influence (Al, Ba, Mg, and Ti) that present the highest concentrations in spring/summer. This variability was confirmed by the seasonal cycle of water isotopes. The  $\delta^2\text{H}$  maximum represents annual summer peaks, while the  $\delta^2\text{H}$  minimum marks the lowest annual temperature (associated with the winter). The seasonal variability in the MJ ice core is exhibited in Figure S2 – Supporting Information. According to Tuohy et al. (2015), winter peaks are associated with large precipitation events and/or seasonal wind speeds. West Antarctica is largely influenced by cyclonic activity that penetrates into West Antarctica, delivering heat and moisture (Kaspari et al., 2004; Dixon et al., 2011). Previous research on trace element concentrations has indicated that fresh snowfall in high-accumulation sites is generally related to the highest trace element concentrations (Wolff et al., 1998; Kreutz et al., 2000a). Although, dry deposition can contribute between 10 % and 25 % of chemical impurities in snow, and these mechanisms increase in importance with decreasing annual accumulation (Davidson et al., 1981; Tuohy et al., 2015).

The annual mean concentrations of 11 trace elements in the MJ ice core are shown in Fig. 4. Major, historical volcanic eruptions, such as Pinatubo (1991), Agung (1963), Santa Maria (1902), and Krakatoa (1883), were identified by large sulfur concentration peaks. The concentrations are highly variable down the length of the core. For example, mean Mn concentrations range from  $\sim 0.94 \text{ pg g}^{-1}$  to  $450.88 \text{ pg g}^{-1}$ . The highest concentrations are generally observed before 1930 for all elements. Similar trends are noted for Al, Ba, Fe, Mg, S, and Ti. In contrast, the concentrations of K and Mn remain low (with the exception of a few peaks) and correlated well during the entire period. Ca, Na, and Sr presented low annual variability over the period but marked seasonal variability. In particular, there are three distinct phases in the record: (i) between 1885 and around 1930 concentration values peak for Al ( $12.28 \text{ ng g}^{-1}$ ), Ba ( $63.02 \text{ pg g}^{-1}$ ), Fe ( $2.07 \text{ ng g}^{-1}$ ), Mg ( $26.01 \text{ ng g}^{-1}$ ), S ( $29.66 \text{ ng g}^{-1}$ ), and Ti ( $69.54 \text{ pg g}^{-1}$ ); (ii) between approximately 1930 and 1955 minimum concentrations can be observed for the above trace elements; and (iii) between approximately 1955 and 2008 a second increasing trend is

observed for Al, Ba, Fe, Mg, S, and Ti. The measured trace element concentrations generally agree with previous studies (Kreutz et al., 2000b; Dixon et al., 2013) but show visible variability during the 20<sup>th</sup> century. The ice core record reflects changes in emissions as well as atmospheric circulation and transport processes. The El Niño-Southern Oscillation (ENSO) is the variation in southern Pacific sea surface temperature that induces regional-scale changes in atmospheric circulation in those latitudes. Several researchers have reported links between ENSO and aerosol deposition flux in Antarctica (Vance et al., 2013; Criscitiello et al., 2014). Below we will discuss in detail the influence of climate variables in the concentration of trace elements presented.

Sea ice variability can be influenced by fluctuations in atmospheric circulation patterns associated with intraseasonal (e.g. Renwick et al. 2012), interannual (Kohyama and Hartmann, 2016), and decadal climate variability, such as the Southern Annular Mode (SAM) (Lefebvre et al., 2004; Stammerjohn et al., 2008), the El Niño Southern Oscillation (ENSO) (Simpkins et al., 2012), and the Atlantic Multidecadal Oscillation (AMO) (Li et al., 2014). Sea ice concentration trends are generally consistent with sea surface temperature (SST) trends, such that regions of increasing (decreasing) sea ice are nearly always found in an environment of decreasing (increasing) SST. However, winds can influence sea ice concentration in several ways, including atmospheric thermal advection, oceanic currents, and wind-driven dynamic transport (Holland and Kwok, 2012). Schneider et al. (2011) suggest that atmospheric circulation trends have influenced both the sea ice concentration and the temperature trends in the Pacific sector of the Antarctic.

Correlations were made between annual means of reanalysis variables (SIC, SST, and 2-m air temperature) from the ERA-Interim product and the studied trace element annual average concentrations from 1979–2008. Observed correlations suggest a relationship between sea ice concentration and aerosol transport to the ice core site. Figure 5 shows that correlations between SIC and most of the analyzed elements are positive ( $r > 0.55$ ;  $p < 0.05$ ) in the Ross Sea and negative ( $r > -0.45$ ;  $p < 0.05$ ) in the Bellingshausen Sea, consistent with others authors (Simpkins et al., 2012; Turner et al., 2015a). Previous studies using satellite observations show a dipole structure in SIC with increasing sea ice in the Ross Sea (Stammerjohn et al., 2015) and decreasing in the Amundsen–Bellingshausen Seas (Holland and Kwok, 2012). This dipolar pattern is related to thermodynamic and dynamic forcing associated with variability in the pressure anomalies extending over the Amundsen Sea (Turner et al., 2015b).

The strongest positive correlations ( $r = 0.65$ ;  $p < 0.05$ ) are in the region between 180° and 140° W, suggesting that concentrations at the MJ site increase when SIC is high in the West Amundsen and Ross Seas. This may indicate the dominant source of the marine aerosols in the MJ area. Some studies show that processes associated with sea ice formation (e.g. frost flower formation, brine production, and blowing snow released from sea ice surfaces) are the dominant source of sea salt aerosols for Antarctica (Rankin et al., 2002; Wolff et al., 2003; Kaspari et al., 2005; Fischer et al., 2007; Criscitiello et al., 2013). Aluminum and magnesium exhibit negative correlations ( $r = -0.55$ ;  $p < 0.05$ ) in the Ross Sea region and positive correlations ( $r = 0.50$ ;  $p < 0.05$ ) in the Antarctica Peninsula area. Aluminum in polar ice derives almost exclusively from crustal dust (McConnell et al., 2007), which explains the correlation difference between Al and other trace elements. However, Mg exhibits the same pattern observed for Al, showing a relationship between the two elements. As previously

presented in other studies, the nss fraction of Mg can be linked to insoluble silicate or soluble carbonate or evaporite (e.g., gypsum, halite) minerals derived from terrestrial sites (McConnell et al., 2007; Kreutz and Koffman, 2013), which would explain the strong correlation with Al found. The nssMg fraction measured in this work corresponds to approximately 72% of the total concentration. This negative correlation is also related to a different seasonality pattern, with mineral dust and marine aerosol peaks occurring in different periods of the year.

SST plays an important role in oceanic heat content controlling the interactions between the ocean and atmosphere (Reynolds et al., 2007). SST variability in the Southern Hemisphere (SH), related to ENSO, is associated with changes in the atmospheric circulation (Ding et al., 2012; Fogt et al., 2015), Southern Ocean SST anomalies (Simpkins et al., 2014; Ciasto et al., 2015), Antarctic SIC anomalies (Simpkins et al., 2012; Turner et al., 2015a), and Antarctic surface temperature anomalies (Schneider et al., 2012; Steig et al., 2015). Criscitiello et al. (2014) demonstrated that tropical Pacific SST anomalies have influenced the source and transport of marine aerosols to the WAIS.

Ding and others (2011) show that anomalous SST under areas of strong tropical convection in the central tropical Pacific have generated an atmospheric Rossby wave response that influences atmospheric circulation over the Amundsen Sea, causing increased warm air advection to the WAIS. Other studies suggest that tropical SST forcing has a significant impact on the southern annular mode (SAM) during austral winter (Ding et al., 2012; Li et al., 2014), deepening the Amundsen–Bellingshausen Seas low (Li et al., 2015). Previous studies indicate that wind associated with low pressure systems over the Bellingshausen-Amundsen Sea would facilitate the generation and transport of sea-salt aerosols from either an open ocean or a sea ice source (Bromwich et al., 2013; Criscitiello et al., 2014; Pasteris et al., 2014), as well as influence dust transport from circum-Antarctic continents to the Southern Ocean and WAIS (Neff and Bertler, 2015).

Figure 6 shows associations between SST and annual trace element concentrations. Negative correlations ( $r > -0.70$ ;  $p < 0.05$ ) between SST in the Ross, Amundsen, and Bellingshausen Seas with the trace element concentrations indicate increased transport of marine aerosols when the SST is cooler. Strong negative correlation ( $r > -0.75$ ;  $p < 0.05$ ) is also observed in the Weddell Sea and Indian Ocean. On the contrary, Al and Mg concentrations are positively correlated ( $r > 0.65$ ;  $p < 0.05$ ) with the Ross, Bellingshausen, and Amundsen Seas as well as the southern Indian Ocean SST indicating that Al and Mg concentrations increase when SST is high. Once more, Al and Mg exhibit a very strong relationship with each other and an inverse correlation to that presented by the other elements. In the case of these two elements, transport and deposition processes are associated with mineral dust seasonality pattern and influenced by warmer SST (usually associated with the spring/summer period) between the source and the MJ site.

The 1979–2008 correlations for  $T_{2m}$  are displayed in Fig. 7. Annual  $T_{2m}$  anomaly maps generally reveal weak, negative anomalies ( $r > -0.45$ ;  $p < 0.05$ ) over the Ross and West Amundsen Seas for the measured elements, while showing low, positive anomalies ( $r > 0.45$ ;  $p < 0.05$ ) for Al and Mg specifically.

Ligtenberg et al., (2013) show that snowfall and  $T_{2m}$  in the Antarctic continent appear to be linked: periods with increasing  $T_{2m}$  coincide with periods of increasing snowfall. Changes in  $T_{2m}$  show warmer and moister conditions extending considerably farther inland, consistent with enhanced air intrusions (Nicolas and Bromwich 2011). Changes in snow

accumulation are also linked to the deepening of the Amundsen Sea Low (ASL), tropical SST, and large-scale atmospheric circulation (Thomas et al., 2015).

Our results are consistent with other authors who have identified that seasonal concentration maxima in sea salt elements correlate well with the SIC winter maxima (Rankin et al., 2002; Sneed et al., 2011; Abram et al., 2013; Fan et al., 2014). Seasonal and interannual variability of the trace element concentrations are also likely due to wind speed and transport efficiency at the time of deposition. Hoskins and Hodges (2005) suggest that storminess over the Southern Ocean and the strength of inland transport, both of which are enhanced during winter, can explain the winter maxima deposition in the WAIS. However, the elements of crustal origin presented different variability with maximum concentration in period of lower SIC and higher temperature (usually associated with the spring/summer). Mineral dust deposition at other locations in Antarctica also exhibits seasonal variability with a maximum in summer, e.g. Berkner Island (Bory et al., 2010), Windless Bight, McMurdo Sound (Dunbar et al., 2009), and South Pole (Legrand and Kirchner, 1988).

The strong, negative correlation between trace element concentrations and SST further demonstrate that concentrations contained in the MJ ice core record provide an indication of past variability. Previous studies have also identified a teleconnection between the tropical Pacific Ocean and aerosol deposition in Antarctica (Vance et al., 2013; Criscitiello et al., 2014). SST anomalies under areas of strong tropical convection have a significant influence on the atmospheric circulation in the Bellingshausen-Amundsen Sea area through the generation of a large-scale, atmospheric wave train (Lachlan-Cope and Connolley, 2006; Ding et al., 2011). The wave train pattern is prominent in winter and spring months (Lachlan-Cope and Connolley, 2006). Since sea salt aerosols are deposited throughout the year, notably in winter, it is expected that tropical forcing and atmospheric Rossby waves would influence trace element concentrations.

Based on significant, negative correlations with SST, and supported by annual variability of  $\delta^{18}\text{O}/^{16}\text{O}$  (Fig. S3 – Supporting Information), we suggest that high concentrations observed before 1930 in our ice core record are directly related to cooler SST affecting atmospheric transport in the Amundsen-Ross Sea region. We assume that the low concentrations observed later (1930-1955) are consistent with a warming in the region. Schneider et al. (2008) show extreme positive anomalies (representative of West Antarctic surface temperature) during the 1936-1945 period. The authors interpreted these anomalies as indicative of strong teleconnections in part driven by the ENSO (1939–1942). The second increasing trend observed for Al, Ba, Fe, Mg, S, and Ti (between 1955 and 1992), when compared with  $\delta^{18}\text{O}$  record, is also associated with a period of cooling in the MJ area or in the region where these aerosols are generated, Jones et al. (2016) show a cooling of SST in the Ross sector during the study time (1979-2014), this may be related to the increase of concentrations, since our correlations have pointed this region as a source area of marine aerosols. This cooling is followed by a warming period into recent years in MJ record (associated with decreased concentrations of Al, Mg, and S). Therefore, it seems possible that different trace element concentrations respond to different forcings on different timescales.



### 3.3 Atmospheric transport to Mount Johns ice core site

715 Atmospheric transport to Antarctica is dominated by the circumpolar westerly winds over the Southern Ocean and the permanent cyclone belt over the polar fronts (Hoskins and Hodges, 2005). The baroclinic zone, between 60–70°S, is a very active cyclone generating area due to the interaction of cold, dry air from the continent and relatively warmer, moist air from the Southern Ocean. The prevailing mid-latitude westerlies direct the cyclones, circulating around the Antarctic continent (King and Turner, 1997). The Antarctic plateau is dominated by a high-pressure (anticyclonic, counter-clockwise) and the

720 wind regime is governed by katabatic winds (cold, dense air flowing downhill due to gravity). The Amundsen Sea Low (ASL) is a major driver of West Antarctic climate variability (Turner et al., 2013). It is a mobile, climatological low-pressure located between 170–298° E and 80–60° S, in the Southern Pacific (Kreutz et al., 2000b; Hosking et al., 2013). The depth and location of the low-pressure center affects the climatic conditions and the strength of the westerlies along the coastal regions and the interior of West Antarctica (Ding et al., 2011; Schneider et al., 2012).

725 Back-trajectory modeling have been used to understand the synoptic controls on precipitation and the transport of trace elements to the study site. Here, we calculated 5-day, backward trajectories using the NOAA HySPLIT Model and clustered the trajectories over austral spring (September–November), summer (December–January), autumn (March–May), and winter (June–August) months. Daily simulations were generated during the 1979 to 2008 period. For the simulated trajectories, we have determined that five clusters are sufficient to capture the seasonal trajectory variability.

730 The MJ site receives the majority of air masses from the Amundsen Sea and, secondarily, from across the Antarctic Peninsula and Weddell Sea (Fig. 8). An additional source could be a continental local contribution. It is possible to identify two clusters with dominant westerly flow patterns ranging from fast (long) to slow-moving (short) depending on the season; one cluster includes fast-moving trajectories with strong cyclonic curvature around the Ross Ice Shelf (whole year); a group with direction that varies from westerly flow to northeasterly depending on the season, and a continental grouping that

735 contains mainly katabatic flow paths from the interior. The clusters show that air masses circulate around the Antarctic continent until they are diverted to the interior, as synoptic storms, across the Amundsen and Bellingshausen Seas. In some cases, the air masses arrive on the WAIS after being diverted across the Weddell Sea and travelling over the Filchner-Ronne Ice Shelf.

The trajectories are classified into two groups: 1) oceanic influenced (blue), and 2) continental influenced (red). These

740 classifications are defined by characteristics identified from the mean trajectories of each cluster, speed (proportional to trajectory length), source region, and pathway. The frequency distribution of cluster classes for the 1979–2008 period shows distinct seasonality between the austral summer and other seasons, with comparatively stronger, westerly transport in the cold months and a secondary, northeasterly transport in the warm months. On average, the oceanic group has a maximum, seasonal frequency in winter while the continental group peaks in winter/spring. During the summer, the trajectories

745 generally are slow-moving (short) and are more locally influenced than in others seasons.

The highest concentrations of trace elements in the winter concentrations are associated with air masses clustered within the oceanic trajectories. Low pressure centers over the Bellingshausen-Amundsen Sea are associated with cold temperatures and sea ice formation in the region (Bromwich et al., 2013; Criscitiello et al., 2014). The wind associated with the low pressure systems would presumably aid in the generation and transport of sea-salt aerosols from either an open ocean or a sea ice

750 source. These air masses also have the potential to capture impurities transported over the South Pacific from mid-latitude continental regions such as South America or Australia. Moreover, winds remobilize mineral dust from ice-free areas in the WAIS, for example trajectories crossing Marie Byrd Land, Ellsworth Land, the Antarctic Peninsula, and the Weddell Sea. Li et al. (2008) show that due to the prevailing westerlies, the distribution and deposition of dust has an eastward transport so that half of the Atlantic Ocean and Indian Ocean are influenced by South American dust and the Pacific region is influenced  
755 by Australian dust. Modeling studies of dust transport to Antarctica (Krinner et al., 2010) show that the annual mean concentration of dust in West Antarctica (particularly Marie Byrd Land), is mostly represented by dust originating from Australia. This is in agreement with Neff and Bertler (2015) and Tuohy et al. (2015) who show dust transportation from Australia and New Zealand to the South Pacific. As observed previously in other ice core sites from the WAIS (Dixon et al., 2011; Koffman et al., 2014a; Neff and Bertler, 2015), mineral dust reaching the MJ ice core site is unlikely to be associated  
760 with a single dust source due to mixing along-transport from presumed continental sources. Previous work at the MJ coring site, Cataldo et al., (2013) found particle sizes ranging from 1.1  $\mu\text{m}$  to 2.4  $\mu\text{m}$ . Based on these data we assume that the concentrations are more influenced by remote continental sources than local sources. Although we do not disregard that local contributions can be a secondary source for dust.

The westward extent of trajectories from winter (maximum sea ice area) to spring (maximum sea ice retreat) is between 180  
765 and 140°W, coincident with the area of greatest correlation between ice core trace elements and SIC, SST, and  $T_{2m}$ . The mechanism for deposition of trace elements at MJ site is intrinsically linked with enhanced sea ice production in the Amundsen-Ross Sea. Thus, not only is there a clear mechanism for trace elements transport to the MJ site, but the factors governing this transport (for example, local wind conditions and temperature) are also related to the sea ice concentration itself.

770 The Antarctic climate system is strongly interconnected; regional and seasonal complexity as well as physical relationships between different climate variables must be taken into account to identify the mechanisms driving the production, transport, and deposition of trace elements in WAIS. While these results are promising, further work involving assimilation methods using ice core proxy records and climate simulations in order to best reconstruct the past state of the Antarctic atmospheric circulation is required.

#### 775 4. Conclusions

Using high-resolution ICP-SFMS, several trace elements were measured in an ice core from WAIS and used to evaluate inputs from natural aerosol emission sources. It was found that natural contributions from mineral dust are important sources

for Al and Mg, while marine aerosols from the open sea and sea ice in the southern Pacific are important sources of Na, Sr, and Ca, over the MJ site. However, Ba, Fe, K, Mn, S, and Ti show to be from mixed sources (mineral dust and sea-salt aerosols). Additionally, S and Mn exhibit important volcanic contribution and S has a considerable biogenic input in the summer season. Utilizing back trajectories from HySPLIT, we identify two dominant air-mass trajectory clusters: marine and continental. The analysis shows distinct differences between seasons, relatively stronger, westerly transport during the winter months and a secondary, northeasterly transport during the summer months. During the summer, the trajectories present slow-moving (short) and are locally influenced when compared to other seasons. Based on particle size (1.1  $\mu\text{m}$  to 2.4  $\mu\text{m}$ ) for MJ ice core and the air-mass trajectory, we conclude that the trace element concentrations are influenced by remote continental sources more so than local sources, although we do not ignore that local contributions can be a secondary source for mineral dust.

Our reanalysis-element correlations suggest that marine derived trace element concentrations are strongly influenced by SIC and SST anomalies. The results show that seasonal concentration maxima in sea-salt elements correlate well with the SIC winter maxima. The strongest correlations are in the region between 180° and 140° W, suggesting that elemental concentrations in the MJ site increase when SIC is high in the West Amundsen and Ross Seas. This may indicate the dominant source of the marine aerosols over the MJ site. Correlations between SST in the Ross, Amundsen and Bellingshausen Seas with the MJ site indicate increased transport of marine aerosols when the SST is relatively cooler. Previous studies indicate that wind associated with low pressure systems over the Bellingshausen-Amundsen Sea facilitate the generation and transport of sea-salt aerosols from either an open ocean or a sea ice source (Bromwich *et al.*, 2013; Criscitiello *et al.*, 2014; Pasteris *et al.*, 2014), as well as affect the mineral dust transport from circum-Antarctic continents to the Southern Ocean and WAIS (Neff and Bertler, 2015).

We show that both sources and transport of mineral dust and marine aerosols to West Antarctica are controlled by the climate variables in response to remote atmospheric forcing. While these results are promising, further work is needed to obtain a more detailed picture of past variability and its relationship with regional aerosol transport.

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**Table 1. Statistical summary of trace elements concentrations determined in MJ ice core.**

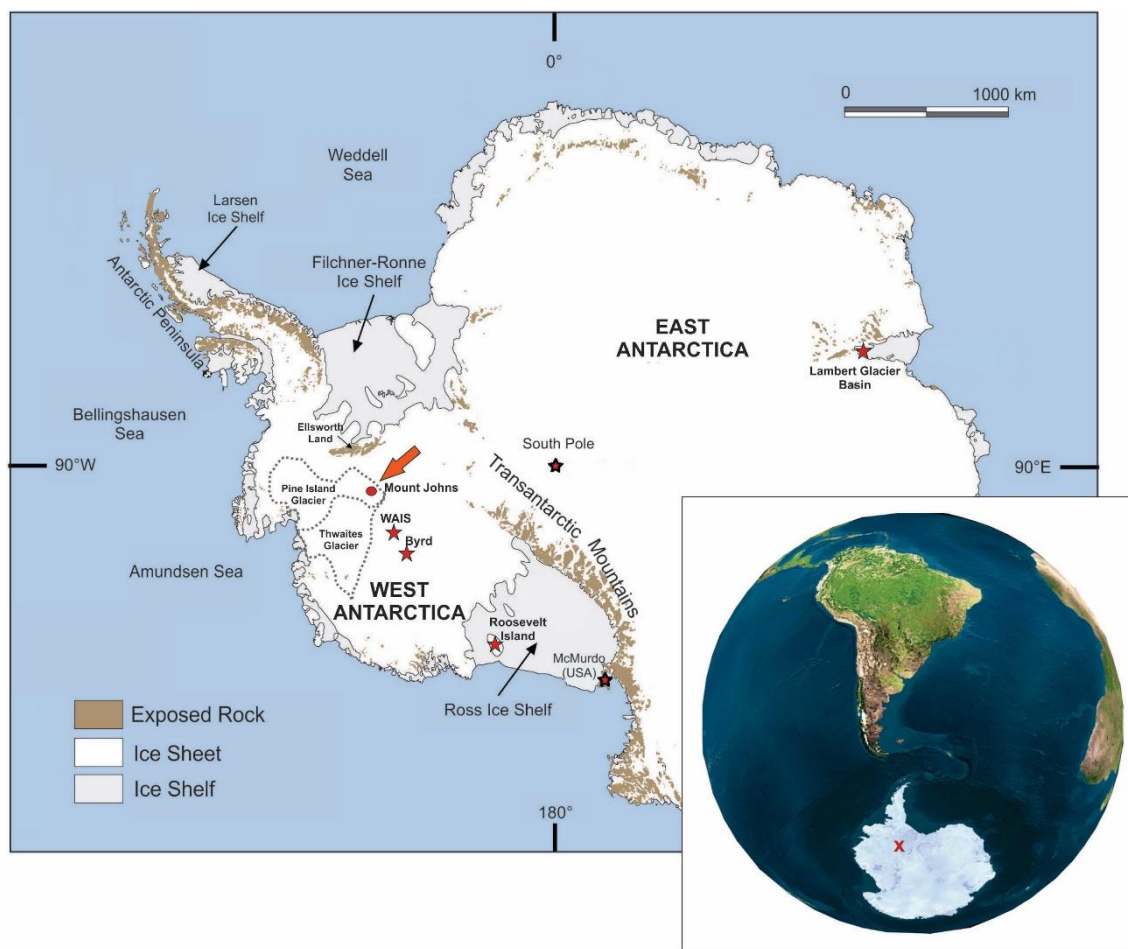
Elements	Mean	Median	SD	Average Uncertainty	Min	Max	MDL*
Al (ng g <sup>-1</sup> )	2.64	2.46	1.65	0.03	0.30	22.77	0.30
Ba (pg g <sup>-1</sup> )	9.31	5.49	11.45	0.24	0.38	93.46	0.38
Ca (ng g <sup>-1</sup> )	3.01	1.88	3.24	0.07	0.09	21.22	0.09
Fe (ng g <sup>-1</sup> )	0.62	0.41	0.68	0.01	0.05	5.58	0.05
K (ng g <sup>-1</sup> )	1.78	0.94	3.39	0.07	0.06	39.54	0.06
Mg (ng g <sup>-1</sup> )	9.27	9.09	4.71	0.10	0.66	45.18	0.38
Mn (pg g <sup>-1</sup> )	28.10	11.99	61.60	1.34	0.94	783.79	0.94
Na (ng g <sup>-1</sup> )	21.91	12.64	33.10	0.69	0.21	381.61	0.21
S (ng g <sup>-1</sup> )	10.24	9.01	6.30	0.14	0.58	62.53	0.06
Sr (pg g <sup>-1</sup> )	23.21	17.74	17.85	0.38	0.74	117.94	0.74
Ti (pg g <sup>-1</sup> )	14.15	9.00	18.76	0.37	0.66	209.35	0.66

\*MDL = Method Detection Limit.

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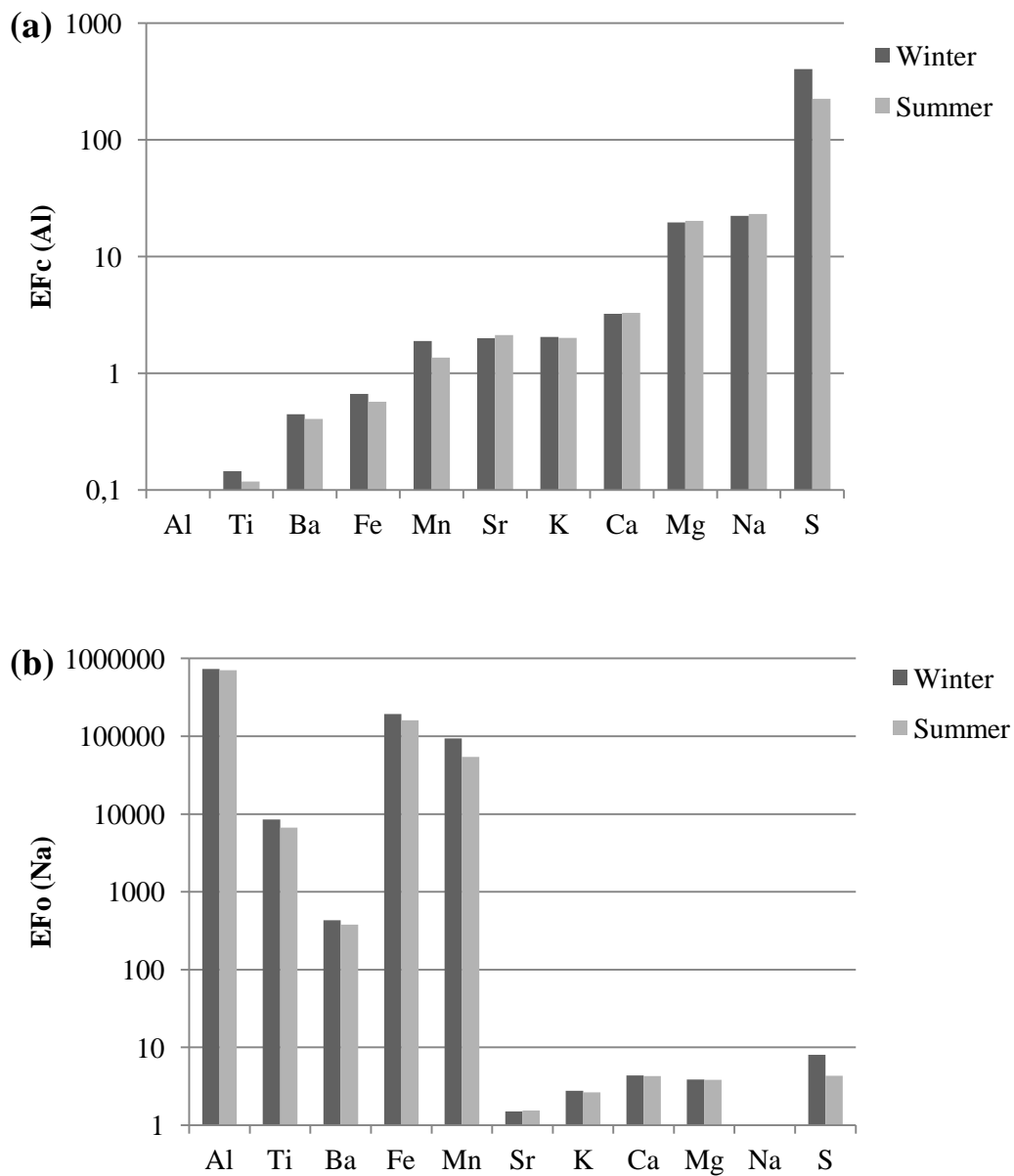
Table 2. Principal component loadings for the Mount Johns trace elements concentrations (PCA loadings > 0.3 are shown in bold).

Element	Component			
	1	2	3	4
Al	0.24	<b>0.59</b>	-0.26	0.05
Ba	<b>0.36</b>	0.01	<b>0.30</b>	-0.07
Ca	0.22	-0.18	0.24	<b>0.66</b>
Fe	0.20	0.19	<b>0.59</b>	-0.02
K	<b>0.32</b>	<b>-0.32</b>	0.06	-0.17
Mg	<b>0.32</b>	0.28	<b>-0.50</b>	0.17
Mn	<b>0.37</b>	-0.18	0.03	<b>-0.36</b>
Na	<b>0.37</b>	<b>-0.34</b>	-0.19	-0.03
S	0.29	0.06	-0.10	<b>-0.44</b>
Sr	<b>0.34</b>	-0.17	-0.17	<b>0.40</b>
Ti	0.21	<b>0.46</b>	<b>0.32</b>	0.03
Percent of variance	42.24 %	13.27 %	11.16 %	8.11 %
Cumulative percent	42.24 %	55.51%	66.67%	74.77%



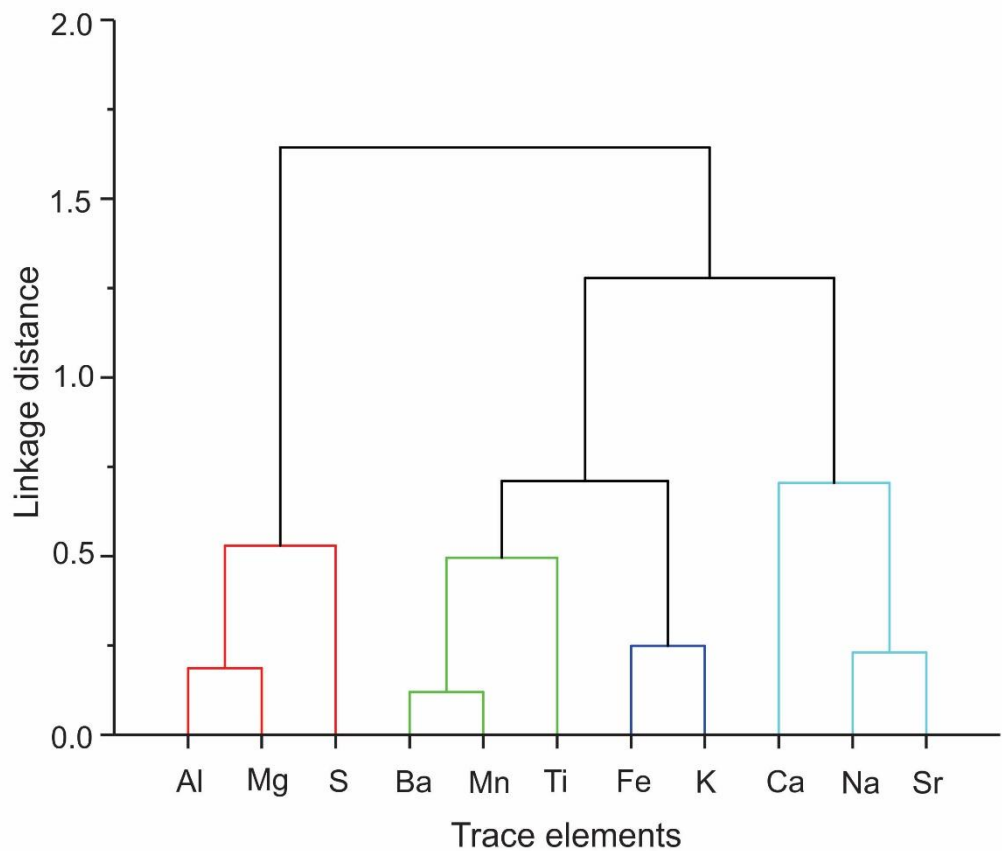
**Figure 1.** Map of Antarctica showing the site of the MJ ice core site (red arrow) and locations of sites discussed in the text (figure adapted from the U.S. Geological Survey, <http://lima.usgs.gov/>).

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1135 **Figure 2.** (a) Mean element enrichment factors in reference to Earth's crust (EFc) and (b) oceanic composition (EFo) at the MJ site for austral summer (December to February) and austral winter (June to August).





**Figure 3. Cluster analysis calculated from the principal components score matrix extracted from the entire data set. Trace elements on PC1 are divided into two subgroups composed of Ba, Mn, Ti, Fe, and K (mixed sources) and Ca, Na, and Sr (marine component). Cluster analysis adds PC2, a subgroup composed of Al and Mg (crustal component) and PC4 composed of S (volcanic influence).**

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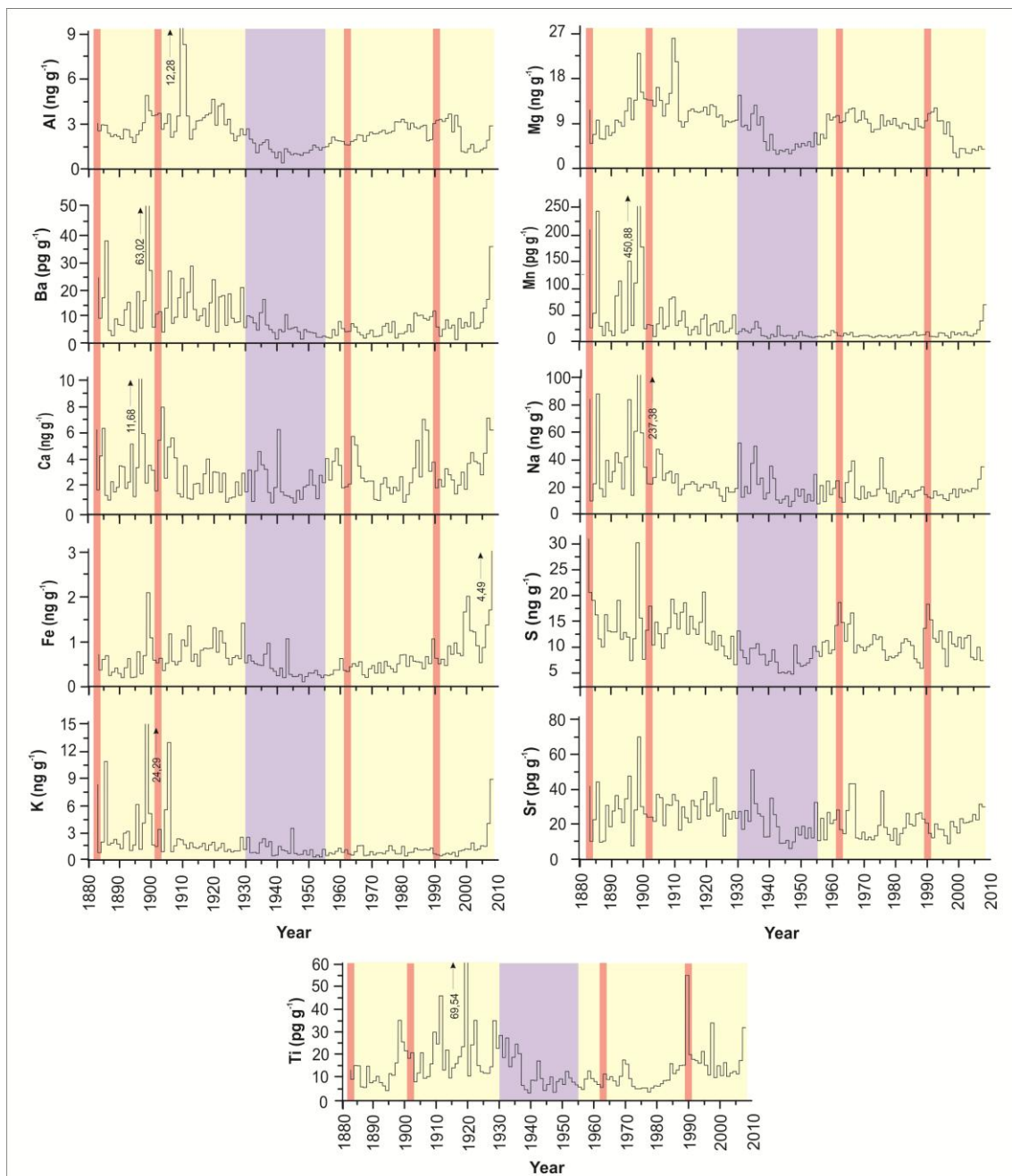
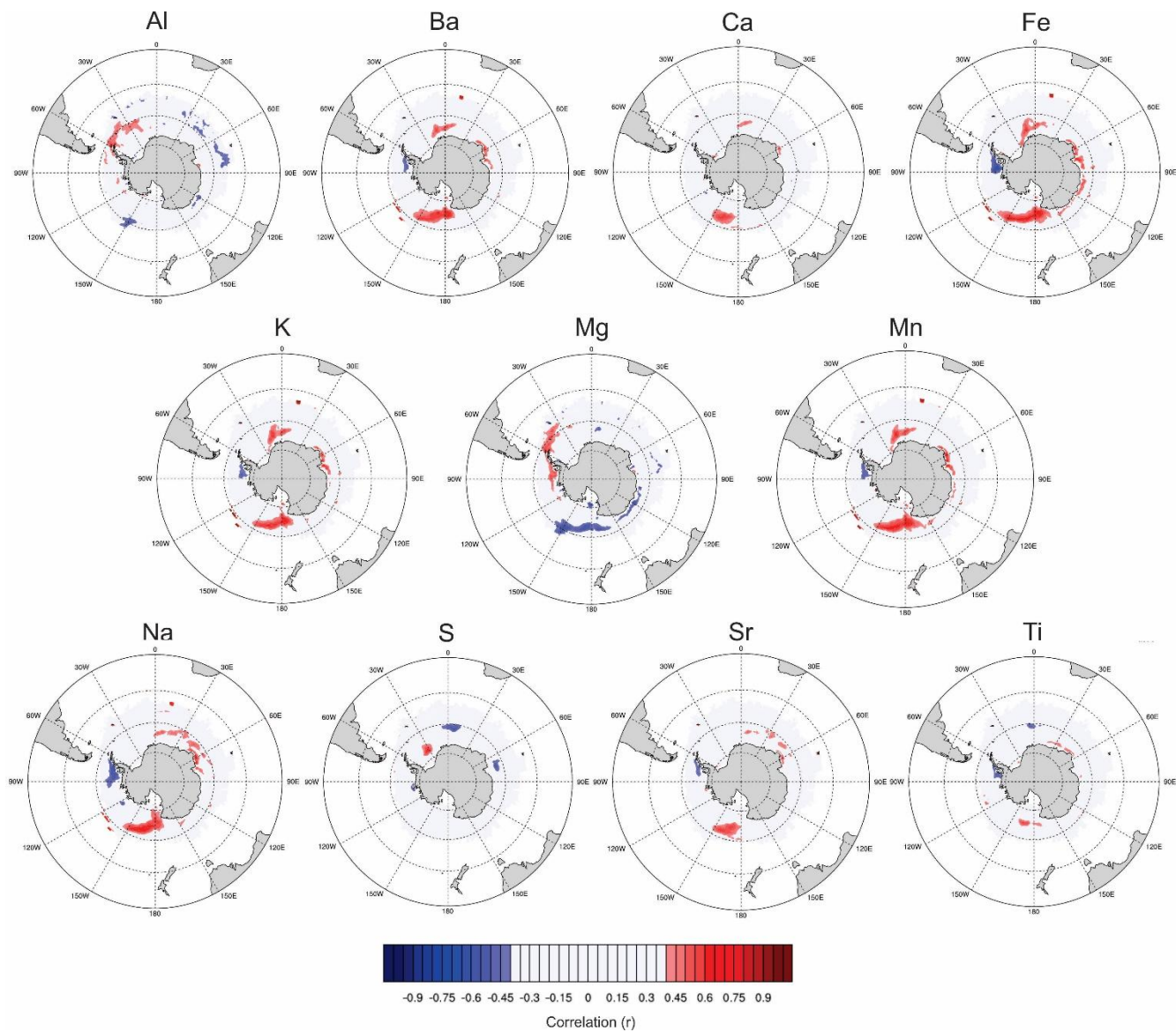
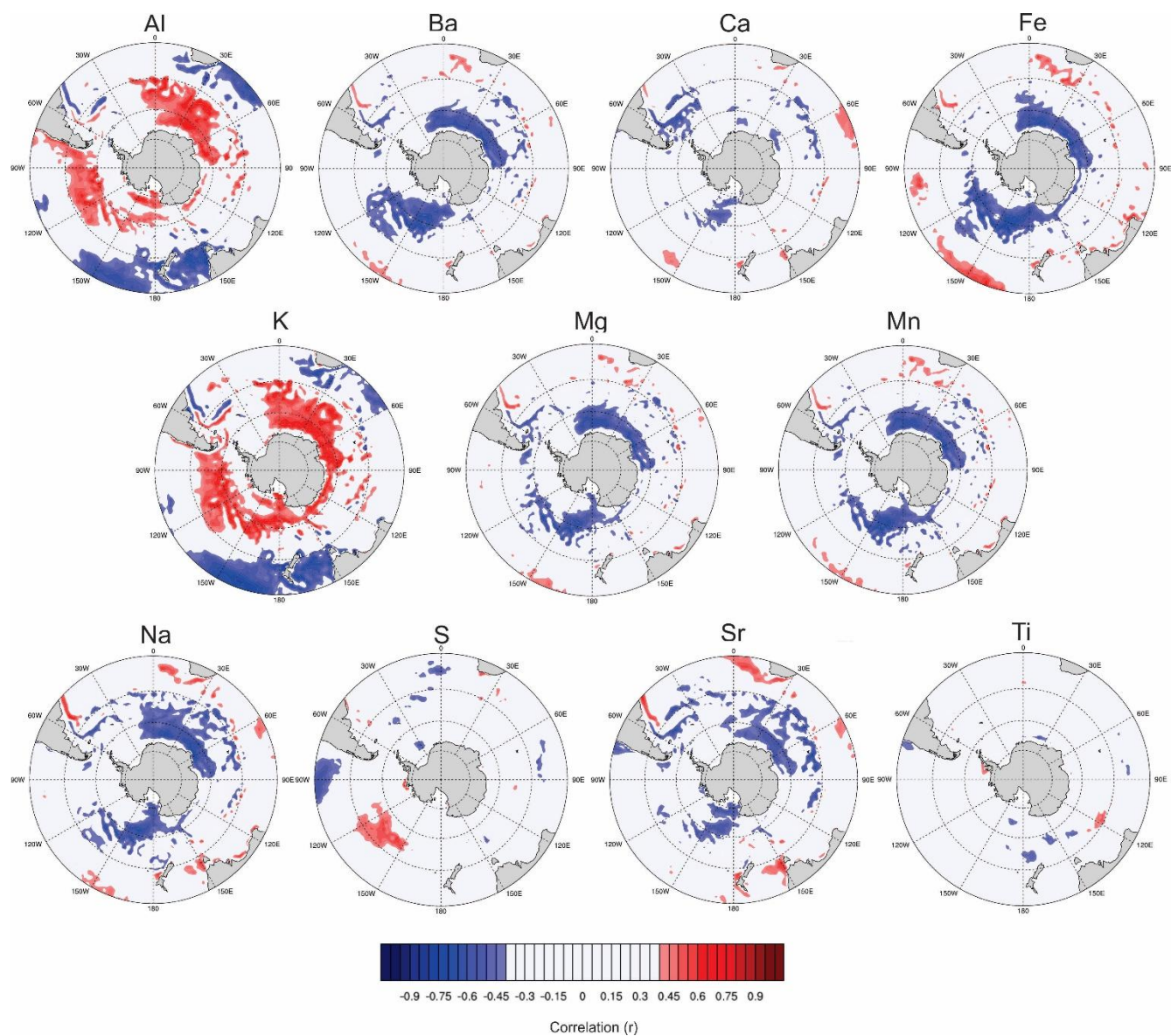


Figure 4. Annual average concentrations of Al, Ba, Ca, Fe, K, Mg, Mn, Na, S, Sr, and Ti measured in the MJ ice core. Colored bands define three distinct phases in the record from 1883 to 2008. The peaks shaded in red were identified using sulfur record and indicate volcanic eruptions corresponding to events: Krakatau (1883), Santa Maria (1902), Agung (1963), and Pinatubo (1991).

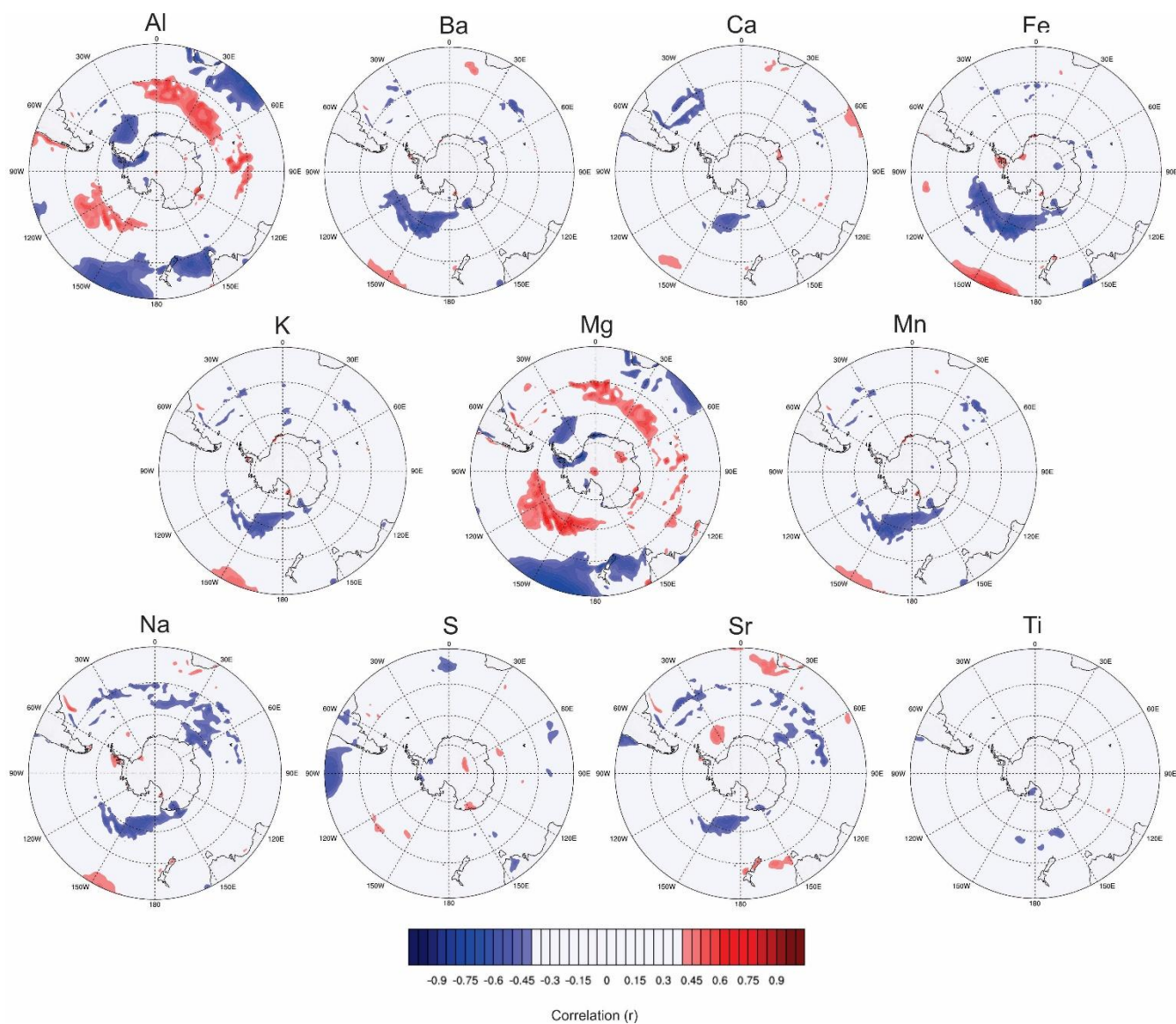


**Figure 5: Correlation of the 1979–2008 ERA-Interim reanalysis sea ice concentration versus annually-averaged trace element time series in the MJ ice core.**



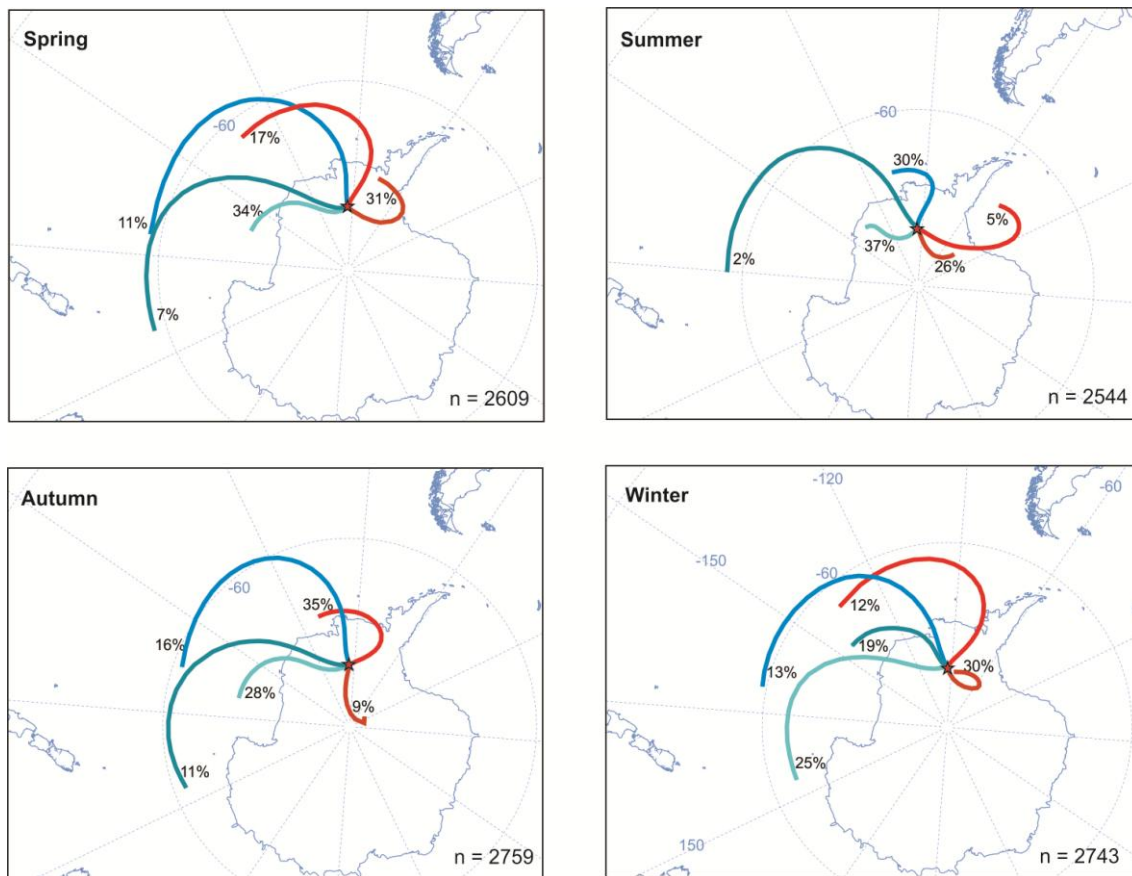
**Figure 6: Correlation of the 1979–2008 ERA-Interim reanalysis sea surface temperature (SST) parameter versus the annually-averaged trace elements time series in the MJ ice core.**





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**Figure 7: Correlation of the 1979–2008 ERA-Interim reanalysis 2-m air temperature parameter versus the annually-averaged trace elements time series in the MJ ice core.**



1165 **Figure 8. HySPLIT seasonal clusters of daily 5 day back trajectories from 1979 to 2008 arriving at the Mount Johns ice core site, West Antarctica. Percentage of daily trajectories included in each cluster is indicated, number of daily trajectories for each season is indicated at the bottom right of each panel. Blue shaded area represent oceanic group clusters, while red shaded area show the continental group clusters. Trajectories calculated using the NOAA Hysplit Model (version 4.9).**

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