

Response to Reviewer 1

We sincerely thank the reviewer for constructive criticisms and valuable comments, which were of great help in revising the manuscript. Accordingly, the revised manuscript has been systematically improved with new information and additional interpretations. Our responses (AC) to the referee's comments (RC) are given below.

RC 1. The authors present a record of insoluble dust particles, Cr, Zn, Ba and Cu fluxes for the period 1960-2005, from a firn core drilled at coastal Dronning Maud Land (DML). Fluxes of these impurities were observed to double after 1985, a phenomenon which the authors attribute to changing Southern Annular Mode from negative values in the 1960's to positive values in the 1990's. The manuscript presents a limited data set (In terms of length and the number of analytes) and the interpretation also seems to be limited and somewhat a-priori. The choice of data is quite selective – important data that could be used to support the authors' arguments have been left out, there is an over-reliance on reanalysis data (which must carry large error bars in the pre-satellite period), and the interpretations are poorly supported. The description of the relevance of SAM is cursory (it is present in only one sentence of the introduction) and no evaluation of other climate modes (such as ENSO) are presented.

AC: Changes in atmospheric circulation over the past five decades have greatly enhanced the wind-driven inflow of air mass onto the Antarctic continent, which controls the deposition of various constituents in the Antarctic continent. This increase in deposition of dust associated with trace metals is a major environmental concern in the modern era. Studies were conducted recently about the dust variability during the past several decades, not only because of the anthropogenic warming during this period, but also due to the availability of reliable instrumental and satellite data to validate the proxies. While Antarctic ice core records have been extensively used in the past to understand the large scale variations in climate-influenced parameters like dust during periods of completely different boundary conditions in the past (like the glacial to interglacial fluctuations), only few ice records are available with seasonal/annual resolution that deal with periods before and after the industrialization. Our exceptionally high-accumulation (12-18 samples per year) and well-dated firn core record from coastal East Antarctica offer an unprecedented opportunity to study the interannual to decadal climate variability dust input to the Antarctic continent. Considering that we have analysed large number of samples ($n = 470$) for different size components and important trace metals, we believe that while the time period represented is limited, it offers better insight into the utility of ice records for the study of short-term climatic changes. The fact that significant and systematic changes are observed within the last 50 years and because the supporting climatological data are more reliable during this period, the data offers value to the Antarctic climate scientists.

We agree with reviewer on the importance of the evaluating the major climatic modes – SAM and ENSO – on observed variability. Hence, we conducted a statistical correlation analysis to identify the relation between dust with SAM and ENSO. It showed that while there is a strong positive correlation ($r=0.68$; $p < 0.00000001$) between the dust flux and SAM, similar correlation with Southern

Oscillation Index (SOI) is weak (0.21; $p < 0.09$). Apparently, the relation between dust and SOI is insignificant during the time period of study. As suggested, in the revised text, we have also made detailed discussions on SAM and its influence on the Antarctic climate variability.

RC: The English used in the document needs to be markedly improved; there are many grammatical errors and the flow of the paper is often obstructed by the inclusion of numerous unrelated references. The manuscript shows promise but needs to be improved with regard to a number of points - the authors seem to link references rather than use them appropriately and as support for a well-constructed interpretation. The over reliance on references results in jumps between unrelated topics in consecutive sentences (at one stage every sentence is based on a reference). The brevity of the time series fails to convince that the trend observed in the dataset isn't just noise; the link to SAM is tenuous and unsupported by statistical analysis; papers reporting and interpreting glacial dust fluxes are used to interpret modern dust flux data; and finally, the impact of land use changes and the influence of human activity on both dust and trace element fluxes appear to have not been considered.

AC: We agree with the reviewer that sufficient care was not taken in the original manuscript vis-à-vis the English and interpretations. In the revised manuscript, the English and grammatical errors are corrected and the interpretations and flow have been significantly improved. To evaluate the contribution of dust supply on the geochemical variability within the core during the last 50 years, we have calculated the crustal enrichment factors (E_{FC}) for the trace metals through the core depth. Considering that the source region to the core site is attributed to the Patagonian region and since geochemical composition of the Patagonian top soil and dust (Gaiero et al., 2003 & 2005) are significantly different from the upper crust values of Wedepohl (1995), in the revised manuscript, we have used the trace metal composition of Patagonian dust for normalization and demonstrate that the geochemical record mimics the source characteristics. We have also used statistical techniques in the revised text to support the causal relation between the SAM and dust transport to the core site. Further, we have carefully revised the text to avoid relating the recent changes to large scale glacial-interglacial shifts. Discussions have also made on the possible influence of land use changes and influence of human activities on dust and trace metals in the ice record.

RC: On a more general note, I think that the authors should be cautious about using such a short data set to evaluate changes in climate. The other data sets that the authors cite, to support their argument that dust fluxes to Antarctica have increased in the 20th century, are at least two hundred years in length, and consider dust flux changes over broader periods. Enormous changes in dust flux are observed in Greenland on annual timescales, so determining the signal from the noise may require a longer data set. The authors state that a 65 m core was drilled but only the top 30 m are reported here; the impact of this data set would be greatly improved if the dust record could be extended into the 19th century.

AC: While we agree that a longer record would be more valuable to evaluate the longer climate variability, we are convinced that the distinct changes taken place in dust flux during the last 50 years reveal significant climate relationships. Thus it does make sense to look at the proxy record during the short period. Also, the available climatological data are more reliable during this time. Considering that we have analysed large number of samples (n = 470) for different size components and important trace metals, we believe that despite the fact that the time period represented is limited, it offers better insight into the utility of ice records for the study of short-term climatic changes. Processing of the entire core require longer time period and will be undertaken in future.

Specific comments:

RC: p.1842, line 10. I don't agree that the authors demonstrate a "dominant" role for SAM variability on dust transport.

AC: We have revised the text and have removed such wordings as per the reviewer's suggestion.

RC: p.1842, line 11. "1985s" is incorrect. Either "1980's" or 1985.

AC: We agree.

RC: p.1842, line 21. This sentence contains grammatical errors and should be improved. "Antarctica holding fingerprints" is a particularly awkward metaphor.

AC: The sentence is corrected as well as the whole text is revised to improve the flow.

RC: p.1843, line 3. "Vigorous atmospheric disturbances" is an unusual way of describing the changing conditions at the deglaciation. The role of the hydrological cycle should also be explicitly mentioned. Papers such as Mahowald et al (2005, GBC) and Maher (2010, ESR) are good sources of information regarding the current state of understanding.

AC: Necessary changes have been made and this part is now rewritten.

RC: p.1843, line 7. Generally speaking, insoluble volcanic particles are identified as ash or tephra and are considered distinct from insoluble dusts, which are of mineral dust origin.

AC: We estimated the total dust concentration and number by size fraction. We have revised the sentences to avoid such confusions.

RC: p.1843, line 10. There is more recent research than that of COHMAP. For example, the two papers I mention above. Also the role of glaciation and deflation should be considered in dust production (e.g., Sugden et al., NGeo 2009).

AC: We agree and have made reference to more recent publications, including the paper by Sugden et al. (2009).

RC: p.1843, line 16. Define short-term and long-term.

- AC: In the present context, long term means climate change that taken place over 1000s of years and short-term means interannual to decadal.
- RC: p.1843, line 22. All of the isotopic and elemental evidence points to a dominant source of southern South American (SSA) dust during the glacial. You should specify that Australian is potentially a significant source of dust only during interglacials.
- AC: Necessary correction has been incorporated as per reviewer's suggestion.
- RC: p.1843, line 29. This is the only sentence of the introduction that mentions SAM. I would have expected a much more significant description of this climate mode, upon which the interpretation of the data relies. The assertion that shifts in SAM "might" have changed dust deposition is a very weak argument.
- AC: As per reviewer's comments, we have revised this section and added the causal relationships between SAM and the Antarctic climate variability.
- RC: p.1845, line 29. The detection limits can't be compared to fluxes. Please also provide averages and ranges for the trace element data in ug/L.
- AC: We agree. The averages and ranges for the trace element concentration data is now added to the manuscript.
- RC: p.1846, line 4. I don't have access to Naik et al (2010) but if the results of the NIST1640 standards are not included there, they should be included here.
- AC: The certified values of NIST 1640 standards are included in the revised text.
- RC: p.1847, line 1. I agree that the pre-1979 data is likely to have some bias. It would be good for the authors to indicate some error boundaries or confidence levels.
- AC: Since the NCEP-NCAR reanalysis data was used for comparison purpose only, the detailed error estimation is beyond the scope of our study.
- RC: p.1848. This paragraph is almost unreadable. It contains too many different references and topics, as well as grammatical errors and omissions. It needs to be completely rewritten. Particularly annoying is that the authors refer interchangeably to glacial interglacial and decadal variability in dust deposition. Millennial-scale dust flux variability cannot be treated in the same way as decadal variability.
- AC: We have now completely rewritten this paragraph and have removed the superfluous and irrelevant discussions and related references.
- RC: p.1849, line 9. A more complete evaluation of the trace element data is required. Were the elemental concentrations enriched relative to mean crustal dust concentrations? Are the data affected by pollution and/or contamination? Given that many evaluations of crustal composition (e.g., Wedepohl, GCA 1995 and McLennan, GGG, 2001) have the following order of abundance: Ba >Cr>Zn>Cu, I am inclined to think that the Zn and Cu results are influenced by pollution, contamination or volcanism.
- AC: The entire section is now revised. To evaluate the contribution of dust supply on the geochemical variability within the core during the last 50 years, we have calculated the crustal enrichment factors (EFc) for the trace metals through the core depth. Considering our backtrajectory study as well as various available data, the source

region to the core site is attributed to the Patagonian region. Since the geochemical composition of the Patagonian top soil and dust (Gaiero et al., 2003 & 2005) are significantly different from the upper crust values of Wedepohl (1995), normalization by the geochemical data from Patagonian region would be more reliable for the EFC estimation. We have used Ba, a dominantly crustal element among the geochemical components studied here, for normalization. The enrichment factors thus estimated with reference to both top soil and the aeolian dust (Gaiero et al., 2003 & 2005) reveal significant differences (Table 1). While the ice core data reveal significant enrichment of Zn and Cu (Zn>Cu) compared to the Patagonian top soil, the EFC estimation with respect to aeolian dust from Patagonia (Gaiero et al., 2003 & 2005) reveals no major enrichment for the trace metals. Comparing the EFC of Cu and Zn in the top soil and aeolian dust from the Patagonia indicate that the aeolian dusts may be systematically scavenging Cu and Zn from the South American atmosphere compared to the top soil. Further, it is proposed that the aeolian dust could further accumulate trace metals from the atmosphere before reaching the Antarctic environment, leading to slightly increased concentration of Cu and Zn in the Antarctic ice compared to the Patagonian dust (Table 1). Such an inference is supported by the finding that trace metal pollutants could be scavenged by the mineral dust and transported to long distances in the Southern Hemisphere (Marx et al., 2008).

RC: p.1849, line 25. I would not describe these correlations as strong.

AC: We have undertaken a local significance test for the linear correlations using the technique by Davis (1976) and modified by Yuan and Martinson (2000). These techniques find the effective degrees of freedom. Using this technique, it was found that the one tailed probability between dust and trace metals are $p < 0.00002$, $p < 0.0018$, $p < 0.009$, $p < 0.03$ for Ba, Cr, Zn, and Cu respectively. Thus all the correlations are significant above 99.94%.

RC: p.1805, line 3. As mentioned earlier, the authors should also consider that Zn and Cu are emitted quiescently by local Antarctic volcanoes. Zn and Cu do not have to be attributed to anthropogenic sources, particularly in the case of Antarctica.

AC: Although volcanic contribution was suggested as a source for the increased Zn and Cu in Antarctica (Zreda-Gostynska and Kyle, 1997), such a possibility is not supported by our studies. No SO_4^{2-} anomalies are present in the core except for the Pinatubo (1991) and Agung (1963) volcanic eruptions (Naik et al., 2010). Also, the possibility of local volcanic source within the Antarctic continent is ruled out as there are no such sources exist in the East Antarctica. Zreda-Gostynska and Kyle, (1997) have suggested that fallout from Mount Erebus volcano in West Antarctica is not evenly deposited over the Antarctic continent. Considering that we employed clean processing and analytical protocols and the fact that both the Zn and Cu show good correlation with the dust flux ($r = 0.33$ $p < 0.009$ for Zn and $r = 0.31$ $p < 0.03$ for Cu), we suggest that the high Zn and Cu in the core are indicative of the increased scavenging of these trace metals by the fine dust particles from its source of origin.

RC: p.1851, line 9. This sentence is purely speculative, and is a good indication of the poor data interpretation present in the manuscript. Instead the authors should refer to the abundant data (Rare Earth Elements, Sr/Nd/Pb isotopes, etc) showing that Antarctic dust can originate from SSA and Australia during interglacials. This is also confirmed by various modelling studies.

AC: We have revised this sentence and have also referred to the radiogenic isotope records to support the source contribution. To reconstruct the transport pathways of the dust and trace metals at the present core site, back wind trajectories were constructed for 10 days using the HYSPLIT model for two representative years of low and high dust fluxes. The selected periods were chosen since they are in the same phase of El-Nino Southern Oscillation (ENSO) and since ENSO has a known linkage with southern hemisphere climatic variability (Heureux and Thompson, 2005). Our findings are supported by studies in Patagonia by Li et al. (2010). To evaluate the contribution of dust supply on the geochemical variability within the core during the last 50 years, we also calculated the crustal enrichment factors (EFC) for the trace metals through the core depth. Also, to understand the SSA contribution of trace elements, we have evaluated the published data by Gaiero et al., 2003 & 2005.

RC: p.1851, line 24. The authors offer only a visual comparison between dust and SAM. I would start to be convinced if the authors could quantify such a correlation.

AC: In order to identify the relation between estimated dust and SAM index, we carried out a correlation study between dust and SAM. We found a strong positive correlation ($r= 0.68$ $p<0.00000001$) indicating the role of SAM on dust transportation over East Antarctica.

RC: p.1852, line 21. I think it is safe to say that dust reaching East Antarctica is always wind-borne.

AC: Correction has been incorporated.

RC: p.1853, line 1. How do local conditions relate to the SAM index? Can you be certain that the changes in dust flux aren't just related to local katabatic wind strength and hence do not reflect all of Antarctica? The link to SSA sources would be much more convincing if you could demonstrate the changes (or lack thereof) of dust deflation from SSA sites. In this regard, the authors might look for sediment trap data from coastal SSA, or the extensive publications of Gaiero and coworkers.

AC: We don't have any in-situ observations at the core site to see the local conditions on SAM index. Majority of the trajectories during winter (dominant season of dust input at the site) show that the transport is from the north-western direction, away from the Antarctic coast.

We have verified the metal data published by Gaiero et al. (2003 & 2005) from bed sediments in coastal region, top soil and aeolian dust from Patagonian region, southern South America to identify the background metal values at SSA. We have calculated the metal enrichment with respect to metals from source region to identify the cause for observed high metal concentrations for the present study. We find that trace metal data is similar to the aeolian dust from Patagonia. We have significantly revised the part based on these findings.

RC: pp. 1853-4. The paragraph spanning these pages is again filled with many references and little structure. The authors interchange glacial and interglacial dust transport patterns and conditions, and repeat details that they have previously mentioned, or are not appropriate. Very confusing and frustrating to read.

AC: To improve the discussion and readability, these paragraphs are revised now.

RC: p.1863. The surface winds shown here seem to disagree with the data you present on page 1853.

AC: The Figure 1 represent the wind outside the Antarctic continent and Figure 2e represent the wind speed at the core site taken from the nearest grid point from the NCEP-NCAR reanalysis data.

RC: p. 1865. I don't get a lot of information from this chart. It would be better to relate the different data by charts of [element] flux vs dust flux, and showing associated gradients and correlation coefficients. If this chart is retained, the other Antarctic dust flux records reported in the text (e.g., by McConnell et al.) should also be shown.

AC: We have now discussed the relation between dust and trace metal flux and its associated correlations in the text. Hence, we have removed the chart. McConnell et al. (2007) used aluminosilicate dust flux with aluminium concentration. Thus a direct correlation with our data is not possible.

RC: p.1867. This is not a very useful schematic. Instead it would be better to see an image of the different phases of SAM and how they affect Antarctic climate.

Ac: The figure 5 is now revised as per the suggestion by reviewers. We have plotted a new figure by calculating the wind difference between negative phase of SAM (before 1985) and positive phase (after 1985). This figure clearly demonstrates an increase in speed of westerlies after 1985 compared to the period prior to it.

Table 1: EFC calculated for geochemical components studied here against the concentrations in aeolian dust and top soil published by Gaiero et al. (2003 & 2005).

Enrichment factors with respect to aeolian dust from Patagonia			
Period	Zn	Cu	Cr
Before 1985	3.6	4.7	2.1
After 1985	3.3	6.8	1.7
Enrichment factors with respect to top soil from Patagonia			
Before 1985	39	13	1.9
After 1985	36	18	1.4

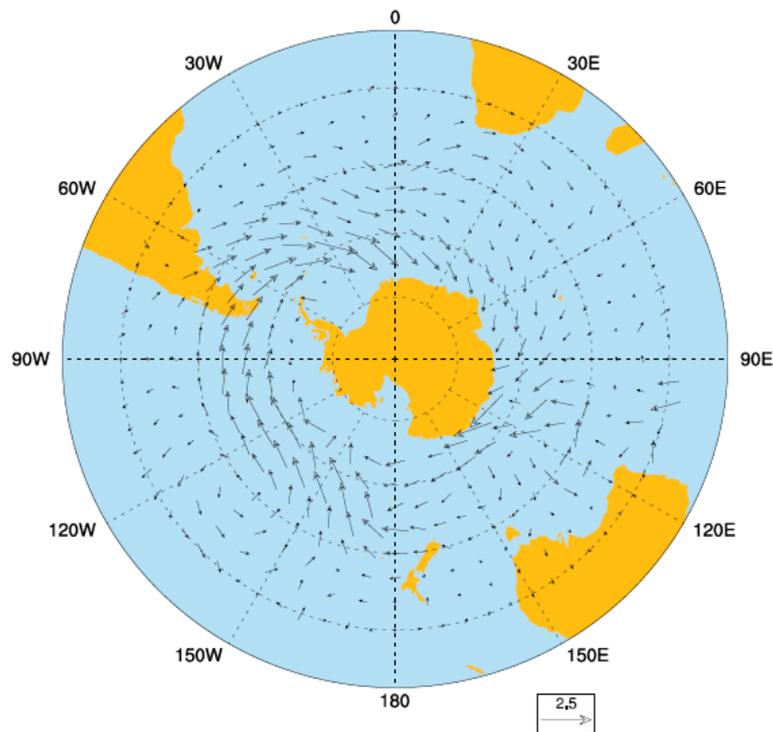


Figure 5. The wind anomalies computed between positive phase of SAM (After 1985) and negative phase of SAM (before 1985). The surface wind data taken from NCEP-NCAR Reanalysis data is used for calculating the difference.

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