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Recent increase in Antarctic Peninsula ice core uranium concentrations



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HIGHLIGHTS

- First highly detailed, continuous U record from an Antarctic ice core.
- U concentrations increase up to 10^2 between the 1980s and 2000s.
- Since the 1990s, raw U, excess U, and U Efc values have increased significantly.
- Rise in U primarily attributed to Southern Hemisphere anthropogenic emissions.

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ABSTRACT

Understanding the distribution of airborne uranium is important because it can result in both chemical and radiological toxicity. Ice cores offer the most robust reconstruction of past atmospheric levels of toxic substances. Here we present the first sub-annually dated, continuously sampled ice core documenting change in U levels in the Southern Hemisphere. The ice core was recovered from the Detroit Plateau, northern Antarctic Peninsula, in 2007 by a joint Brazilian-Chilean-US team. It displays a significant increase in U concentration that coincides with reported mining activities in the Southern Hemisphere, notably Australia. Raw U concentrations in the Detroit Plateau ice core increased by as much as 10^2 between the 1980s and 2000s accompanied by increased variability in recent years. Decadal mean U concentrations increased by a factor of ~ 3 from 1980 to 2007, reaching a mean of 205 pg/L from 2000 to 2007. The fact that other terrestrial source dust elements such as Ce, La, Pr, and Ti do not show a similar increase and that the increased U concentrations are enriched above natural crustal levels, supports an anthropogenic source for the U as opposed to a change in atmospheric circulation.

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1. Introduction

Uranium can result in both chemical and radiological toxicity although there are currently no reference concentrations available for evaluating risk assessment for inhaled uranium exposure.

Previous work (Planchon et al., 2002a, 2002b) ascribed recent elevated U concentrations in Antarctica to anthropogenic sources. There are, however, few published records of U concentrations in Antarctic ice and snow and none that offer continuous, sub-annually resolved time series that can be compared and traced directly to modern anthropogenic sources.

Continental dust, volcanism and sea spray are the main natural sources of U to the atmosphere (Planchon et al., 2002a; Vallelonga et al., 2004). While the contribution of natural sources to total concentrations of U can be estimated based on calculation of

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natural abundances, it has thus far been difficult to quantify the contributions of individual natural and anthropogenic sources (Planchon et al., 2002a, 2002b). Until WWII, the vast majority of U input to the atmosphere was from natural sources. Since 1945, increases in Southern Hemisphere U levels have been assumed to be attributed to U mining in South Africa, Namibia and Australia (Planchon et al., 2002b; Brown et al., 2010).

To establish natural, preindustrial background concentrations of U, we examine a suite of Antarctic ice core records shown in Fig. 1. In the Law Dome (East Antarctica) ice core, all of the U is assumed to be of rock dust origin with mean U concentrations of 25.6 pg/L for the period 4500 BCE to 1989 AD based on 31 individual firn- and ice-core samples (Vallelonga et al., 2004). A suite of ice cores and snow pits (Fig. 1) from interior Antarctica demonstrate that mean U concentrations from 1955 to 2006 do not exceed 169 pg/L (Dixon et al., 2013). A snowpit from Coats Land shows elevated levels of U in recent decades relative to the early 1800s, but a source for the elevated U levels is not suggested (Planchon et al., 2002a). Finally, the SPRESSO ice core from South Pole exhibits a mean U concentration of 104.07 pg/L from 1971 to 1999 with no apparent trend.

2. Methodology

2.1. Ice core collection and chemical analysis

In this study we use a 133 m deep ice core (DP-07-1; 64.05°S, 59.39°W) recovered in 2007 from the Detroit Plateau (DP) by the Joint Brazilian–Chilean–USA expedition as part of the Scientific Committee for Antarctic Research CASA (Climate of the Antarctic and South America) and ITASE (International Trans Antarctic Scientific Expedition) research initiatives. DP is located in the northern part of the Antarctic Peninsula (Fig. 1) and has a mean elevation of ~1900 m m.a.s.l. On the basis of borehole measurements, the temperature of the DP ice cap at 10 m depth is approximately –14 °C and very few melt layers were observed in the firn, thus assuring a well-preserved record. Mean annual accumulation is on average 2.5 m w. e. for the last 27 years allowing for sampling resolution sufficient to document seasonal and higher frequency variability. Based on the foregoing, the upper 98 m of this core was processed using ultra-clean handling procedures (Mayewski et al., 1986), yielding a high resolution (average 36 samples/year) U record covering the period 1980–2007 compared to the Coats Land and Law Dome records that have a much lower resolution (several samples per decade).

To prevent the incorporation of secondarily introduced contamination from drilling, ice core packing, and handling the DP ice samples were scraped inside a HEPA clean hood (–20 °C) using a clean ceramic knife. Samples for trace-element analysis were collected directly into acid-cleaned (10% trace metal grade HNO₃) polypropylene Nalgene jars. Before analysis each sample was acidified to 1% with Optima double-distilled HNO₃ under a class-100 HEPA clean bench and left to digest for 60 days. All samples were analyzed for major and trace elements (Sr, Cd, Cs, Ba, La, Ce, Pr, Pb, Bi, U, As, Li, Al, S, Ca, Ti, V, Cr, Mn, Fe, Co, Na, Mg, Cu, Zn, and K) using the Climate Change Institute Thermo Electron Element2 ICP-SFMS. Samples were analyzed using an Elemental Scientific Apex Q with nitrogen addition and a self aspirated ESI PFA-ST nebulizer with a sample uptake of 100 ul/min. U-238 was measured in low resolution and was calibrated before each sample run with a calibration ranging from 10 to 1000 pg/L. The U method detection limit was 6.46 pg/L and was calculated by measuring 7 process blanks and calculated as three times the standard deviation of process blanks (Osterberg et al., 2006).

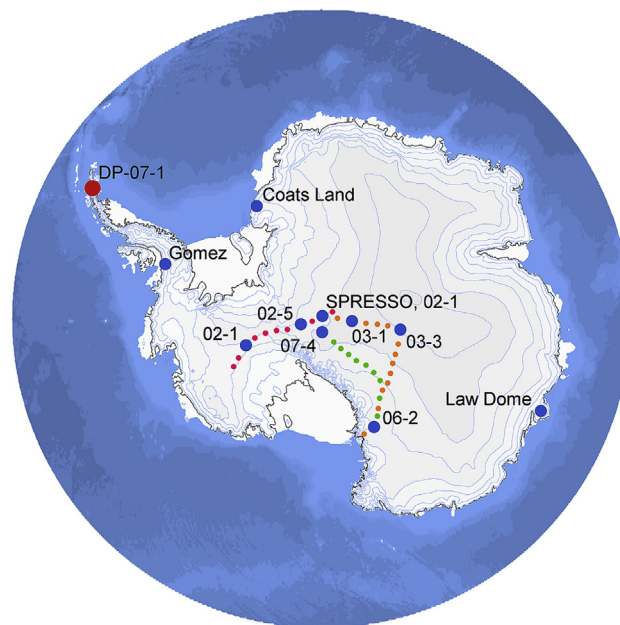


Fig. 1. Location of DP-07-1 ice core drilling site in Antarctica. Also shown are locations of the US ITASE-02-1, 02-5, 02-6, 03-1, 03-3, 06-2, 07-4 (pink dots US ITASE-02 traverse, orange dots US ITASE-03 traverse, green dots US ITASE-07 traverse; Dixon et al., 2013) and Coats Land (Planchon et al., 2002a, 2002b), and Law Dome (Vallelonga et al., 2004) snow pit sampling and firn/ice core drilling sites discussed in this paper.

2.2. Dating of the ice core

The DP-07-1 depth-age scale is based on a well-preserved, well-defined seasonal signature in photochemically-produced hydrogen peroxide (Fig. 2a) with highest and lowest values representing the summer and winter solstice, respectively (Frey et al., 2006). The stable water isotope deuterium (δD) record from DP does not have a clear enough seasonal variability to yield annual signals and as a result it was not useful for ice core time scale development (Fig. A3 in supplemental).

3. Results

3.1. Variation in uranium concentrations for the period 1980–2007

Mean U concentrations in the DP ice core increase from 70 pg/L in the 1980s (Fig. 2b), to 90 pg/L in the 1990s, ultimately reaching 205 pg/L between 2000 and 2007. Overall there is a factor of ~3 increase in mean U concentration during the period 1980–2007. During the 1980s, there is less variability in the U concentrations compared to 1990–2007. Raw U values range from 10 pg/L to 610 pg/L in the 1980s and from 6 pg/L to 1900 pg/L in 1990–2007. To determine if sampling resolution (decreasing with depth) plays a role in the observed differences, we resampled the U data to seasonal resolution (4 samples/year). Seasonal U values range from 23 pg/L to 180 pg/L during the 1980s and from 26 pg/L to 500 pg/L during 1990–2007 maintaining the notable difference in concentration between the two periods.

3.2. Flux values

The Gomez ice core record (Fig. 1) collected 1150 km south of DP-07-01 near the base of the Antarctic Peninsula, reveals an increasing trend (over the last 150 years) in annual snow

accumulation (Thomas et al., 2008, 2009). An increasing trend is also captured in the DP-07-1 snow accumulation record (Fig. 2c). In DP-07-1, the covarying and increasing snow accumulation and U concentrations result in large U annual depositional flux values (flux = concentration \times accumulation rate/per unit area). Since the flux calculation only magnifies the relationship and does not affect the overall trend in the data we use concentration for the remainder of this study.

Net snow accumulation rate changes on the Antarctic Peninsula are attributed to an intensification of the transport strength of the westerlies (Thompson and Solomon, 2002; Turner et al., 2005; Abram et al., 2011). To test whether U concentration increases are caused by intensified atmospheric transport or an increase in U source emission strength, we examine the behavior of other terrestrial and marine source chemical tracers in the DP-07-1 record. Crustal (Ce, La, Pr and Ti) and marine (Na and Sr) tracers do not show a significant background concentration increase since 1980 (Fig. A1 in supplemental). Therefore, the observed rise in U

cannot be attributed to an increase in crustal dust or marine-source aerosols.

3.3. EOF analysis

EOF (empirical orthogonal function) analysis of DP-07-1 chemistry data is used to reveal common associations such as sources, transport, and timing (Table A1 in supplemental). Using this approach, we interpret EOF 1 as a dust signal because it is composed primarily of crustal elements (31% of the Total Variance Explained (TVE)). EOF 1 also accounts for 41% of the total U signal. We interpret EOF 2 as a marine signal because it is composed primarily of marine elements (22% TVE). However, EOF 2 only accounts for 2% of the total U signal (Table A1 in supplemental). Most of the remaining U signal is captured by EOF 9 (2% TVE) and EOF 12 (2% TVE), representing 21% and 20% of the total U, respectively. EOF 9 and EOF 12 are represented almost exclusively by U, and in contrast to EOF 1 and EOF 2, EOF 9 and EOF 12 display significant

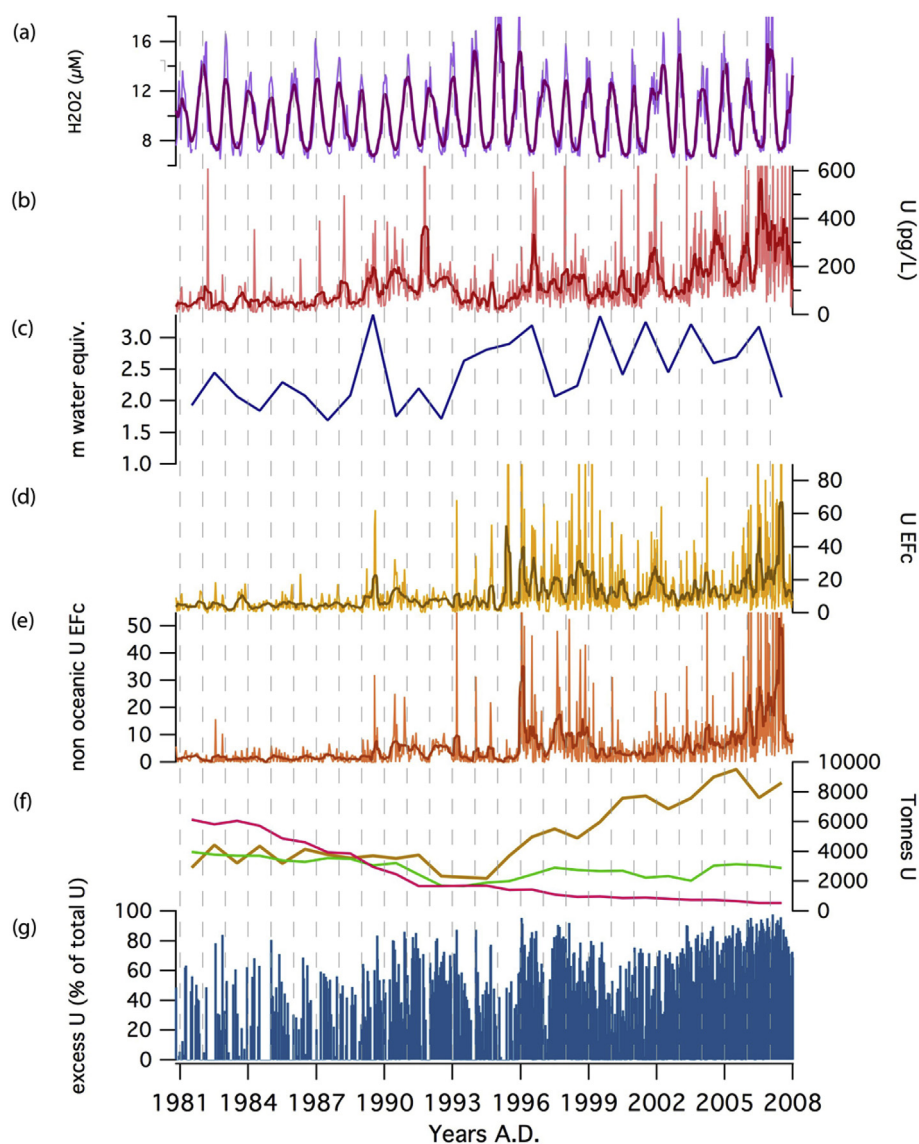


Fig. 2. DP-07-1 (a) H_2O_2 molar concentration (μM); (b) U concentration (pg/L); (c) annual accumulation in water equivalent (m); (d) U mean crustal enrichment factor (EFc) values and (e) mean non-oceanic U crustal enrichment factor values; (f) Australian (brown), Namibian (green) and South African (pink) uranium production (tonnes); (g) excess U (% of total U). The light color lines in concentration and enrichment factor profiles are raw data and the dark color lines are the 9-point smooth data.

increases since 2000 AD (Fig. A2 in supplemental). Therefore, we conclude that EOF 9 and EOF 12 capture the non-marine, non-crustal sources of U.

3.4. Crustal and oceanic enrichment factors

To elucidate potential sources, we calculate a mean Enrichment Factor (EF) for U (Fig. 2d) using methodology similar to Osterberg et al. (Osterberg et al., 2008). Using mean upper crustal (Wedepohl, 1995) and oceanic (Lide, 2005) elemental abundances, we estimate the average crustal EF (E_{Fc}) and average oceanic EF (E_{Fo}) such that: $E_{Fc} = ([U/r]_{\text{sample}}/[U/r]_{\text{upper crust}})$, where r is one of the conservative crustal elements: Ce, La, Pr, and Ti. The final E_{Fc} for U is the mean E_{Fc} from these four elements. The ice core drill was made out of Al, therefore, we do not use Al as a conservative crustal indicator. E_{Fo} for U is calculated using the same method as E_{Fc} using Na and Sr as conservative marine source elements.

Calculated E_{Fc} and E_{Fo} values up to 10 are considered to be almost entirely from crustal and oceanic sources, respectively, and allow for the natural regional fluctuations on Earth (Planchon et al., 2002a; Vallelonga et al., 2004; Duce et al., 1975). EF values greater than 10 are assumed to indicate a strong additional source. The mean E_{Fc} over the past 150 years in the Coats Land study (Planchon et al., 2002a) was 1.8 with a mean E_{Fc} of 4 and a maximum E_{Fc} of 9 over the 1959–1990 period. This is indicative of primarily natural sources of U reaching Coats Land up until 1990. From 1980 to 1990, the DP-07-1 mean E_{Fc} value is 6. From 1990 to 2008 the DP-07-1 mean E_{Fc} is 15, with a maximum value of 370 (Fig. 2d).

Ocean spray can contribute a significant amount of U to Antarctic coastal sites (Vallelonga et al., 2004; Dixon et al., 2013; Toggweiler and Russell, 2008). We calculate the ocean water fraction of U according to the following formula: oceanic fraction for U ($U_o = \text{mean}(r_{\text{sample}} \cdot [U_{\text{ocean}}/r_{\text{ocean}}])$), where r = Na and Sr). Then we subtract U_o from our total U to get non-oceanic U. We then calculate the non-oceanic U E_{Fc} values. Although slightly lower as a consequence of the foregoing, the majority of the non-oceanic U E_{Fc} values are still highly enriched (i.e. E_{Fc} > 10) from 1990 to 2008, and the trend of increasing enrichment is unchanged (Fig. 2e).

3.5. Excess uranium concentration

To partition the natural and anthropogenic sources of U, we calculate the excess (xs) U concentration. To determine xsU (i.e. the U contribution from sources other than crustal dust and ocean spray), we calculated U_o and U_c and subtracted them from the raw U concentrations. The xsU concentrations indicate that significant (in many cases >60%; Fig. 2g) additional U inputs occur after 1990, likely from anthropogenic sources.

4. Discussion

In order to assess the possible contribution by U mining to U loading in the DP ice core, we examine all available historical data on Southern Hemisphere U mining operations. Major above-ground mining operations and U ore processing in the Southern Hemisphere take place in Namibia, South Africa and Australia (World Nuclear Association, 2014). Australian U production (Fig. 2f) matches closely with DP-07-1 U concentrations and xsU values, further validating U mining as a potential primary anthropogenic U source. The other major producers of U in the Southern Hemisphere, Namibia and South Africa (Fig. 2f), show a marked decrease in U production since the early 1980s. Because the measured U concentration trend in DP-07-1 is inverse to the Namibian and South African long-term U production trends, we conclude that these sites likely do not constitute a significant source of U for the

Antarctic Peninsula. Further, high DP-07-1 xsU (Fig. 2g) between 1989 and 1993 are followed by a return to baseline values until ~1996. This return to baseline coincides with a reduction in Australian U production. The increase in Australian U production from 1996 to 2008 also coincides with increasing DP-07-1 xsU. This is in contrast to a weak rise in Namibian U production over the same period and a continued slow decline in South African U production, further highlighting Australia as the most likely anthropogenic U source for the Antarctic Peninsula.

Sub-annual deposition timing of U in DP-07-1 may be related to rainfall seasonality in the vicinity of major Australian U mining centers. To investigate this possibility, the xsU record was resampled to 2-monthly resolution using the summer maxima and winter minima H₂O₂ values for each year and dividing each half-year into 3 equally-spaced intervals. The sub-annual maximum in the DP-07-1 xsU record appears during July–August when mean monthly rainfall over Australian mining regions (e.g. Ranger Mine and Olympic Dam Mine) is at a minimum (Fig. 3). During austral winter, mean rainfall values for the Olympic Dam region in South Australia are often below 3 mm per month. During such dry periods, large volumes of dust can enter the atmosphere and subsequently are transported with the westerlies to the Antarctic Peninsula (Li et al., 2008). This process conceivably occurs as dry soils in these subtropical and dry-season tropical locations are subjected to dry convection resulting in lifting of dust to higher altitudes. In winter (Fig. A4(c) in supplemental) the circumpolar westerlies attain their most equatorward position, particularly over Australia where they comprise the subtropical jet stream (STJ). Previous work suggests that it takes 3–10 days for dust to reach Antarctica from Patagonia or Australia (Krinner and Genthon, 2003; Sudarchikova et al., 2014). Thus, the fast mid-troposphere flow accompanying the STJ over Australia and its associated southern branch (middle-latitude jet) could explain the rapid transport of U to Antarctica. Separating the recent period into two approximately equal-length sub-periods (1981–1994, 1995–2007) suggests that the more recent strong increase in U measured at DP-07-1 is unlikely to be the result of more rapid transport in the mid-to upper-troposphere over Australia since the STJ was weaker in the second sub-period (Fig. A5 in supplemental). It is also suggested that only South America and Australia commonly spawn air masses that reach the Antarctic within 30 days of transport from source (Dixon et al., 2012; Albani et al., 2012). However, there was very little U mining in South America over the period of record in this study.

To further prove that South American air masses seldom reach the study site, we assess air mass origin at DP. We use ERA-Interim Reanalyses data (Dee et al., 2011; The European Centre for M) to plot a wind rose diagram (Fig. A7 in supplemental) of monthly mean wind speed and direction over the 1981 to 2007 period. The wind rose shows westerly winds to be dominant over this time period. Additionally, we perform Hysplit (Stein et al., 2015; Rolph, 2016) back-trajectory cluster analysis for each season during 2004, which is a high U concentration year in the DP chemical record. The cluster analysis results (Fig. A8 in supplemental) show that during the period of maximum U deposition (JJA; Fig. 3), the majority of the air masses reaching DP are coming from the west.

Empirical Orthogonal Function analysis shows that the mean annual rainfall, Australian U production, and DP U Concentration are all positively associated in EOF 1 (total variance explained by EOF 1 = 55%) at 65%, 74%, and 74% respectively (Table A2 in supplemental). This result shows that DP U Concentrations are most strongly associated with Australian U production. However, it is also worth noting that despite an increase in mean annual rainfall (Fig. A6 in supplemental) over the Australian open pit U mining region, DP U concentrations are still increasing.

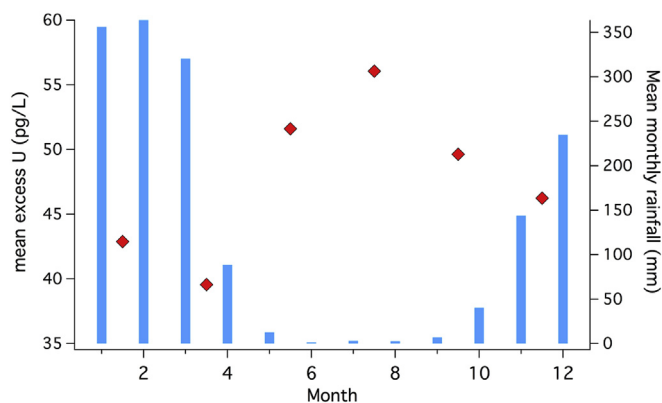


Fig. 3. Comparison of mean excess U concentration (pg/L) resampled to two month intervals with mean (1971–2013) monthly rainfall (mm) in Ranger Mine (Jabriu Airport), Northern Australia.

5. Conclusions

Our study provides the first continuous, high resolution record of U concentrations from an Antarctic Peninsula ice core. We report significant U increases in the atmosphere over the last two decades. Australia is the most likely source of U pollution captured by this 27-year Antarctic Peninsula snow archive. Therefore, we conclude that U deposition at DP is likely related to U mining operations in Australia.

This study demonstrates that dramatic levels of anthropogenic source pollution from Southern Hemisphere industrial sources, in this case open pit mining, are impacting at least the northern fringes of the Antarctic continent. Impacts of this exposure on Antarctic ecosystems are yet to be determined, but they should now be considered. Baseline investigations of the distribution of human source pollutants are essential to understanding changes in human and ecosystem health. Finally, DP offers a good monitoring platform for assessing changes in Southern Hemisphere source pollutants entering Antarctica.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2016.06.010>.

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