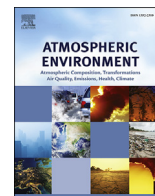




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Recent increase in Antarctic Peninsula ice core uranium concentrations



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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).

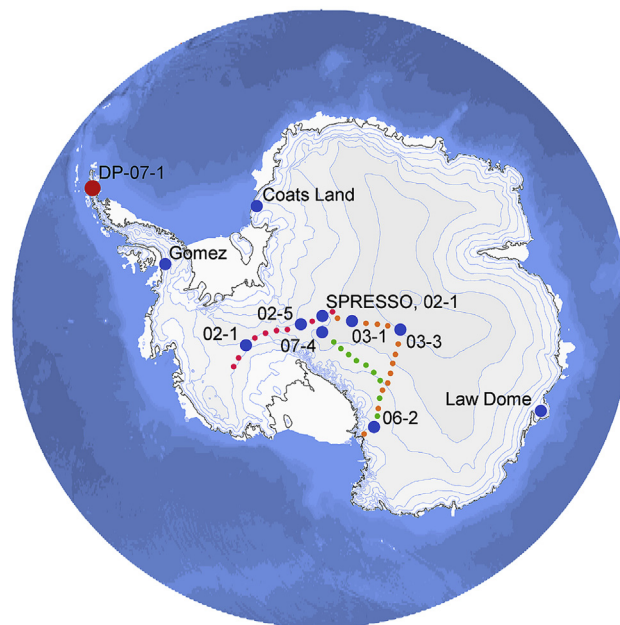


Fig. 1. Location of DP-07-1 ice core drilling site in Antarctica. Also shown are locations of the US ITASE-02-1, 02-5, 02-6, 03-1, 03-3, 06-2, 07-4 (pink dots US ITASE-02 traverse, orange dots US ITASE-03 traverse, green dots US ITASE-07 traverse; Dixon et al., 2013) and Coats Land (Planchon et al., 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

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