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Flux and size fractionation of ^{3}He in interplanetary dust from Antarctic ice core samples

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ABSTRACT

Accretion of extraterrestrial material to earth is of interest for a variety of reasons, including as a possible driver of long or short-term climate change, and as a record of solar system events preserved in the geological record. ^{3}He is highly enriched in extraterrestrial material, and provides a useful tracer of its input into sedimentary archives. Previous work showed that polar ice could be a suitable archive for studying variations in extraterrestrial input. Additional measurements reported here confirm that the late Quaternary ^{3}He flux derived from Antarctic ice samples is similar to ^{3}He fluxes determined from marine sediments. The mean flux from nine replicate ~1 kg ice samples from the Vostok ice core site (112–115 m depth, age of ~3800 years) is $1.25 \pm 0.37 \times 10^{-12}$ cm^3 STP cm^{-2} ka^{-1} (mean ± 2se). The large range for the 9 replicates is probably due to the small number of interplanetary dust particles (IDPs) present, and illustrates that large ice samples are required for precise constraints on temporal variations in the ^{3}He flux. Size fraction experiments show that the majority of the ^{3}He flux is delivered by particles in the 5–10 μm size range, consistent with the hypothesis that helium in IDPs is primarily solar helium implanted in particle surfaces.

1. Introduction

The suggestion that late Quaternary 100,000 year climate cycles might be driven by variations in extraterrestrial dust accretion (Muller and MacDonald, 1995, 1997) stimulated a number of studies of past variations in the interplanetary dust particle (IDP) flux, using ^{3}He, Ir and/or Pt in marine sediments or ice as tracers of IDP input (Farley and Patterson, 1995; Patterson and Farley, 1998; Marcantonio et al., 1999; Higgins et al. 2002; Karner et al., 2003; Winckler et al., 2004; Gabrielli et al., 2004; Gabrielli et al., 2006; Winckler and Fisher, 2006). Initial reports of 100 ka cyclicity in ^{3}He flux (Farley and Patterson, 1995; Patterson and Farley, 1998) were later questioned based on evidence that "sediment focusing" in the ocean can create apparent variations unrelated to the primary IDP flux (Marcantonio et al., 1999; Higgins et al., 2002) and lack of evidence for such cyclicity in some records (Winckler et al., 2004; Gabrielli et al., 2004; Winckler and Fisher, 2006). The history of accretion of extraterrestrial material is also of more general interest as a possible record of solar system events, and as a "constant-flux proxy" that may be useful for constraining sediment accumulation rates.

The ^{3}He content of particles in polar ice cores provides a record of IDP flux that complements the marine record. Brook et al. (2000) reported measurements of ^{3}He in particles extracted from ice cores and showed that the extraterrestrial component dominates ^{3}He in such samples. ^{3}He fluxes derived from late Holocene Greenlandic and Antarctic samples were similar to those previously reported from marine sediments, indicating that ice cores can provide a record of IDP flux. The samples used in that study were relatively small (~200 g of ice), with area–time products (AT) of less than 0.01 m^{2}a. AT is the ratio of the sample mass to accumulation rate; samples with larger values of AT accumulate more IDPs per gram of sediment. The agreement of the results from these small samples with marine sediment data was surprising, given that models of IDP-borne ^{3}He accretion suggest that samples this small would suffer from extreme statistical variability due to small numbers of IDPs present (Farley et al., 1997). More recently, Winckler and Fisher (2006) reported ^{3}He fluxes for the last 30,000 years from samples from the EPICA Drønning Maud Land (EDML) ice core that are consistent with the results from Brook et al. (2000) and show a relatively constant (within a factor of 2–3) flux over that time period. This paper reports additional results from larger samples of Antarctic ice, in an effort to confirm previous estimates of the ^{3}He flux and establish the reproducibility of the measurements. A size fraction experiment was conducted to determine which particle sizes are most important in carrying the ^{3}He flux. This is important for understanding particle transport properties and the origin of the extraterrestrial component.

2. Methods

To examine the reproducibility of extraterrestrial ^{3}He in ice core samples we obtained a 3-m section of the BH-5 ice core from Vostok Station, Antarctica. BH-5 is a shallow core (179 m) drilled with a dry electromechanical system in 1991–1992. The samples were from the

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112–115 m depth interval. The ice accumulation rate is 2.0 g ice cm^{-2} yr^{-1} based on estimates for the 80–130 m depth interval for the main Vostok core site (Sowers et al., 1993). The uncertainty in this value is probably ± 10%. Ideally, future work on young ice should be conducted on cores where more precise accumulation estimates are available through annual layer counting or identification of dated volcanic horizons. We vertically sub-sectioned the 3-m interval with a band saw in a −20 °C freezer to create 9 replicate samples. Sample weights for the subsections were from 821 to 1200 g. The AT value for these samples ranged from 0.04 to 0.06 m^2a. Each sub-section was melted at room temperature in a large stainless steel filtration funnel attached to a vacuum flask. All melting and filtration was conducted in a class 100 clean hood. Particles were filtered on to 0.45 μm silver filters (Osmotics–Poretics). The funnel was rinsed with alcohol to collect particles adhering to the walls and remove water. Each filter was wrapped in an aluminum foil "boat" for helium isotope analysis. Foil boats and filters were melted in an ultra-high vacuum furnace and helium isotopes were measured at WHOI using established techniques (Kurz et al., 1996).

An additional 1503 g subsection from the same 3 m section (112–115 m) of Vostok BH-5 was melted and the melt water was passed through a series of three nylon sieves with openings of 63, 20, and 10 μm. Each sieve was then inverted over a smaller stainless steel filtration funnel and particles were rinsed on to 0.45 μm silver filters. All filters were wrapped in aluminum foil boats for analysis. The melt water containing < 10 μm particles was filtered sequentially through 5 and 0.2 μm silver filters, which were then also wrapped in foil boats.

Because early blanks yielded higher than expected helium levels, filtration procedures were tested for contamination in a variety of ways. Several tests employed deionized water previously filtered through a 0.45 μm cartridge filter. In these tests ~1000 g of water was added through the funnels and passed through a filter. The apparatus remained in place for 12–24 h, and then was rinsed with ethanol in the same fashion as samples. The 12–24 h waiting time was used because filtration of some samples took this long. Tests were also conducted with ice made from filtered deionized water (1.2–1.3 kg), to more closely mimic sample handling. In some cases (Table 1) the outer surfaces of these "blank ice" pieces were shaved with a band saw (steel blade) or steel chisel to test if using those devices caused contamination. One additional blank test exposed filters in the clean hood to lab air, with no liquid filtered through them. Blank results are presented in Table 1.

Two filters exposed only to lab air within the filtration apparatus (with vacuum on) for 24 h contained $28–30\times10^{-12}$ cm^3 STP ^{4}He after correction for line blanks of $43–45\times10^{-12}$ cm^3 STP. Hot furnace blanks (corrected for line blanks) were $2–6\times10^{-12}$ cm^3 STP ^{4}He (n = 4) on the same day. Previous measurements on unused filters revealed minimal contamination (Brook et al., 2000). The isotopic composition of only one of the two filters could be determined (Table 1) and had a ^{3}He/^{4}He ratio of 7.8 ± 3.1 R_a (R_a is the atmospheric ratio, 1.38×10^{-6}). Blank tests with water or ice ranged from 57.1 to 337.3×10^{-12} cm^3 STP ^{4}He. In most cases the isotopic composition could be determined, and the ^{3}He content of these blank filters ranged from 0.6 to 8.6×10^{-15} cm^3 STP ^{3}He (Table 1). Furnace hot blanks run during these measurements were less than 8×10^{-12} cm^3 STP ^{4}He. Contamination in excess of furnace blanks is presumably due to particulate material present on the filtration apparatus. The high (above atmospheric) ^{3}He/^{4}He ratios for the blank tests employing water or ice suggest contamination with ^{3}He enriched particles, possibly from previous samples passed through the filtration apparatus or from other aspects of sample handling in our ice core storage freezer. Numerous hot blank tests, including Aluminum foil "boat" blanks, showed that memory effects in the furnace or mass spectrometer were not responsible for the elevated blanks. Correction for contamination adds uncertainty to the determination of the helium content and isotopic composition of the samples. However, because all samples have high amounts of ^{3}He relative to the blank, corrections for contamination are relatively small for total ^{3}He, which is the important measurement for this study. We use the mean of the 10 blank experiments to determine a blank correction of $168.7\pm49.5\times10^{-12}$ cm^3 STP ^{4}He and $3.4\pm2.3\times10^{-15}$ cm^3 STP ^{3}He (mean ± 2 standard deviation). There are no obvious relationships between the different blank experimental treatments and the measured contamination, with the exception of the filters only exposed to air, which are systematically lower. ^{3}He/^{4}He ratios were calculated from the blank corrected concentrations, and uncertainties in the blank correction were propagated into uncertainties in concentrations and the ^{3}He/^{4}He ratio. The ^{4}He correction is less than 10% for all of the replicate samples from 112–115 m, with the exception of replicate 1, where the correction is 22%. The ^{3}He correction for these samples is <2%. For the sequentially filtered samples the ^{4}He blank ranged from 15–25% of the ^{4}He in the size fractions. For ^{3}He the blank was 1–9% of the total measured amount in the size fractions. Previous work (Brook et al., 2000) did not encounter the elevated blank levels reported here. A smaller, all-glass filtration apparatus was used in that study, which also employed smaller ice samples, and it is possible that particles adhere to the stainless steel surfaces of the large funnels used here and are not completely removed by rinsing and cleaning between samples. Although we cannot conclusively identify the source of the elevated blanks, they are small with respect to the ice-IDP ^{3}He contents.

3. Results and discussion

3.1. Replicate samples and the reproducibility of the extraterrestrial signal

The ^{4}He content of the replicate ice samples ranged from 0.5 to 3.8×10^{-12} cm^3 STP g^{-1} ice, with ^{3}He/^{4}He ratios from 136 to 206 R_a

Table 1
Results of contamination tests during sample processing.

Blank test	Water or ice weight (g)	^{4}He (10^{-12} cm^3 STP)	^{3}He/^{4}He (R/R_a)	^{3}He (10^{-15} cm^3 STP)
DI ice after replicates	1181.2	148.5 ± 0.3	7.4 ± 0.6	1.5 ± 0.1
DI water before filtering/sieving-through sieves	1000.0	149.8 ± 0.3	NM	NM
DI water after filtering/sieving	1000.0	57.1 ± 0.4	92.8 ± 3.0	7.3 ± 0.2
DI ice after filtering/sieving	1276.4	337.3 ± 0.2	18.5 ± 0.4	8.6 ± 0.3
DI ice sawed A	1000	151.3 ± 0.5	11.8 ± 0.6	2.5 ± 0.1
DI ice sawed B	1000	101.1 ± 0.4	5.9 ± 0.9	0.8 ± 0.1
DI ice chiseled A	1000	119.4 ± 0.4	8.7 ± 0.7	1.4 ± 0.1
DI ice chiseled B	1000	234.2 ± 0.5	9.0 ± 0.4	2.9 ± 0.1
DI ice A	1000	178.1 ± 0.3	18.8 ± 0.5	4.6 ± 0.1
DI ice B	1000	209.9 ± 0.3	2.2 ± 0.5	0.6 ± 0.2
Air blank A	–	30.0 ± 0.3	7.8 ± 3.1	0.3 ± 0.1
Air blank B	–	28.3 ± 0.3	NM	NM
Mean procedural blank		168.7 ± 49.5[a]		3.3 ± 2.3[a]

[a] Mean ± 2se.