

Flux and size fractionation of ^3He 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. ^3He 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 ^3He flux derived from Antarctic ice samples is similar to ^3He 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 $1.25 \pm 0.37 \times 10^{-12} \text{ cm}^3 \text{ STP cm}^{-2} \text{ ka}^{-1}$ (mean $\pm 2\text{se}$). 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 ^3He flux. Size fraction experiments show that the majority of the ^3He flux is delivered by particles in the 5-10 micron size range, consistent with the hypothesis that helium in IDPs is primarily solar helium implanted in particle surfaces.

Keywords: helium isotopes, interplanetary dust, ice cores, Antarctica.

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 ^3He , Ir and/or Pt in marine sediments or ice as tracers of IDP input (Gabrielli et al., 2006; Winckler and Fischer, 2006; Gabrielli et al., 2004; Winckler et al., 2004; Karner et al., 2003; Higgins et al. 2002; Marcantonio et al., 1999; Patterson

and Farley, 1998; Farley and Paterson, 1995). Initial reports of 100 ka cyclicity in ^3He flux (Farley and Patterson, 1995; Paterson 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 and Fischer, 2006; Gabrielli et al., 2004; Winckler et al., 2004). 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 ^3He 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 ^3He in particles extracted from ice cores and showed that the extraterrestrial component dominates ^3He in such samples. ^3He 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 $0.01 \text{ m}^2\text{a}$. 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 ^3He 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 Fischer (2006) reported ^3He 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

^3He 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 ^3He flux. This is important for understanding particle transport properties and the origin of the extraterrestrial component.

2. Methods

To examine the reproducibility of extraterrestrial ^3He in ice core samples we obtained a 3-meter 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 112-115 m depth interval. The ice accumulation rate is $2.0 \text{ g ice cm}^{-2}\text{yr}^{-1}$ 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 $\pm 10\%$; 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-meter 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 $0.04\text{-}0.06 \text{ m}^2\text{a}$. 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 micron 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 microns. Each sieve was then inverted over a smaller stainless steel filtration funnel and particles were rinsed on to 0.45 micron silver filters. All filters were wrapped in aluminum foil boats for analysis. The melt water containing <10 micron particles was filtered sequentially through 5 and 0.2 micron 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 micron 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 hours, and then was rinsed with ethanol in the same fashion as samples. The 12-24 hour 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 hours contained $28\text{-}30 \times 10^{-12} \text{ cm}^3 \text{ STP } ^4\text{He}$ after correction for line blanks of $43\text{-}45 \times 10^{-12} \text{ cm}^3 \text{ STP}$. Hot furnace blanks (corrected for line blanks) were $2\text{-}6 \times 10^{-12} \text{ cm}^3 \text{ STP } ^4\text{He}$ ($n=4$) 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 $^3\text{He}/^4\text{He}$ ratio of $7.8 \pm 3.1 R_a$ (R_a is the atmospheric ratio, 1.38×10^{-6}). Blank tests with water or ice ranged from 57.1 to $337.3 \times 10^{-12} \text{ cm}^3 \text{ STP } ^4\text{He}$. In most cases the isotopic composition could be

determined, and the ^3He content of these blank filters ranged from 0.6 to $8.6 \times 10^{-15} \text{ cm}^3$ STP ^3He (Table 1). Furnace hot blanks run during these measurements were less than $8 \times 10^{-12} \text{ cm}^3$ STP ^4He . Contamination in excess of furnace blanks is presumably due to particulate material present on the filtration apparatus. The high (above atmospheric) $^3\text{He}/^4\text{He}$ ratios for the blank tests employing water or ice suggest contamination with ^3He 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 ^3He relative to the blank, corrections for contamination are relatively small for total ^3He , which is the important measurement for this study. We use the mean of the 10 blank experiments to determine a blank correction of $168.7 \pm 49.5 \times 10^{-12} \text{ cm}^3$ STP ^4He and $3.4 \pm 2.3 \times 10^{-15} \text{ cm}^3$ STP ^3He (mean \pm 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. $^3\text{He}/^4\text{He}$ ratios were calculated from the blank corrected concentrations, and uncertainties in the blank correction were propagated into uncertainties in concentrations and the $^3\text{He}/^4\text{He}$ ratio. The ^4He 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 ^3He correction for these samples is < 2%. For the sequentially filtered samples the ^4He blank ranged from 15-25% of the ^4He in the size fractions. For ^3He 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 ^3He contents.

3. Results and Discussion

3.1. Replicate Samples and the Reproducibility of the Extraterrestrial Signal

The ^4He content of the replicate ice samples ranged from 0.5 to $3.8 \times 10^{-12} \text{ cm}^3$ STP g^{-1} ice, with $^3\text{He}/^4\text{He}$ ratios from 136 to 206 R_a (Table 2). Brook et al (2000) and Winckler and Fischer (2006) found similar high $^3\text{He}/^4\text{He}$ ratios in particles extracted from ice core samples. These high ratios are similar to those measured in individual stratospheric IDPs by Nier and Schlutter (1992), who reported a mean value of 202 R_a , and range of 137-462 R_a . Later work by Pepin et al. (2000; 2001) reported a similar range of ratios for individual IDPs collected in the stratosphere, but discussed puzzling occasional measurements of much higher ratios, in two cases in excess of 2000 R_a (Pepin et al., 2000), which could be a signature of cosmogenic production by high energy cosmic rays. Such high ratios have not, to our knowledge, been observed in ice samples. Although helium deposited in Antarctic snow should be a mixture of terrestrial and extraterrestrial origin, the high $^3\text{He}/^4\text{He}$ ratios for the ice samples demonstrates that ^3He is dominantly extraterrestrial in these samples. Using a two-component mixing model, and extraterrestrial and terrestrial end-member $^3\text{He}/^4\text{He}$ ratios of 290 and 0.015 R_a respectively (Brook et al., 2000), the fraction of extraterrestrial ^3He is greater than 99.99% in all 9 replicates. Therefore, no correction for terrestrial ^3He is made to the total concentrations. The mean ^3He concentration of the ice samples is $625 \pm 174 \times 10^{-18} \text{ cm}^3$ STP g^{-1} ice (mean \pm 2 se). The individual measurements range from 148 to 961

$\times 10^{-18} \text{ cm}^3 \text{ STP g}^{-1} \text{ ice}$, with the wide range presumably reflecting the statistical problems of sampling small numbers of IDPs (Farley et al., 1997).

The mean ^3He flux derived from the mean ^3He concentration and assuming an ice accumulation rate of $2.0 \text{ g cm}^{-2} \text{ yr}^{-1}$ is $1.25 \pm 0.35 \times 10^{-12} \text{ cm}^3 \text{ STP cm}^{-2} \text{ ka}^{-1}$ (mean \pm 2se). Despite the fact that the samples represent deposition over only ~ 140 years, this value is similar to results from marine sediment records (Higgins, 2001; see Figure 1) and to the mean flux for the last 13,000 years based on 14 samples from the EDML ice core (Figure 1). The uncertainty derived from the 9 replicates (28%) is smaller than might be expected from the model results of Farley et al. (1997) for samples with low AT values. Following Brook et al. (2000) the expected statistical distribution for a sample with AT of $0.5 \text{ m}^2\text{a}$ was calculated from the model results of Farley et al. (1997) using a Monte Carlo method, employing the predicted size distribution and flux of IDPs heated to $< 600^\circ\text{C}$ and assuming ^3He content is correlated to surface area (Farley et al., 1997). 2000 realizations were generated, and 1000 random samples of $n=9$ were drawn from these realizations. Uncertainties less than the measured value of 28% occurred in less than 28% of the simulations. Brook et al. (2000) reported similar results for smaller samples, and suggested that the size distribution of incoming particles used in the entry-heating model may be incorrect, though clearly larger data sets are needed.

Insert Figure 1 about here.

The uncertainty bounds for all of the estimates in Figure 1 are relatively large. In the case of the Holocene data compiled by Higgins (2001) and the EDML data (Winckler and Fischer, 2006) part of the variability could be temporal. Although all of these ^3He flux estimates agree within uncertainties, key questions about the nature of this extraterrestrial tracer remain. In particular, the spatial and temporal scales of variability are unknown. Both issues are critical for developing measurements of ^3He in ice cores

as tracers of extraterrestrial input, and will require further work.

Given that the range of ^3He concentrations in the replicate Vostok samples is quite large (Table 1), individual measurements of kilogram-sized ice samples, even from an ice core site with a low accumulation rate, in this case $\sim 2 \text{ cm ice yr}^{-1}$ ($\text{AT} = 0.04\text{-}0.06 \text{ m}^2\text{a}$), cannot provide precise constraints in the ^3He flux. Significantly larger samples are likely necessary for detailed down core work. 10 kg samples from a site like this, with ice accumulation rate of $\sim 2 \text{ g cm}^{-2} \text{ yr}^{-1}$, would have an AT value of $\sim 0.5 \text{ m}^2 \text{ a}$. By comparison, 0.5 g of typical pelagic sediment accumulating at $1 \text{ g m}^{-2} \text{ yr}^{-1}$ (Farley et al., 1997) would yield the same AT value. Based on the model of Farley et al. (1997) samples of this size would yield a statistically representative sample and would avoid the problem of under-sampling the flux, and biasing the results to an inaccurate low flux estimate. However, even with 10 kg samples, variations of order $\pm 30\%$ in replicates would be expected, and replication would be critical for precise work. The “low bias” of small samples may be evident in our data, as the estimate of ^3He flux from samples smaller than 300 g from Vostok ice in the 110-111 m section of the BH-5 core (Brook et al. 2000) is lower than that reported here for $\sim 1 \text{ kg}$ samples from the 112-115 m section of the same core (Figure 1). In traditional ice coring programs 10 kg samples would be hard to obtain. However, “horizontal ice core” sites (Reeh et al., 2001; Petrenko et al., 2005; Dunbar et al., 2008), sampling waste water from continuous melting of ice for other chemical measurements (Winckler and Fischer, 2006) and, potentially, replicate coring (where a second core section can be obtained from sections of interest) can provide sufficient ice.

3.2 Size Distribution of the Extraterrestrial Signal

The size distribution of particles carrying the extraterrestrial ^3He flux to the earth’s surface is important for understanding transport in the ocean and atmosphere,

statistical sampling effects, and for developing appropriate techniques for sampling particles. Heating of IDPs during atmospheric entry causes helium loss, and the importance of this effect varies with particle size, which influences the degree of heating. Farley et al. (1997) used an entry heating model and the initial size distribution of IDPs from the Long Duration Exposure Facility (LDEF) (Love and Brownlee, 1993) to estimate the size distribution carrying the extraterrestrial ^3He signal. They modeled the size distribution of ^3He bearing particles for both mass-correlated and surface-area correlated scenarios. They concluded that the ^3He flux correlates with the flux of surface area of IDPs, and is therefore most likely a signature of implanted solar particles (solar wind and solar energetic particles), and that most of the flux should be carried by particles less than 20 microns in diameter, with a peak in the size distribution at ~ 7 microns. In contrast, Stuart et al. (1999) argued that extraterrestrial ^3He in deep sea sediments resides in particles > 50 microns due to chemical alteration and loss of ^3He from smaller particles in sediments. Lal and Jull (2005) also argued that large particles are important, postulating that a significant number of larger particles are formed from the atmospheric breakup of meteorites that contain ^3He produced by spallation by galactic cosmic rays. However, later work by Jull et al. (2007) showed that ^3He in 50-300 micron particles from the South Pole water well (Taylor et al., 1998) is dominated by a solar wind, rather than galactic cosmic ray produced component, consistent with previous studies of micrometeorites (Stuart et al., 1999; Osawa and Nagao, 2002). Jull et al. also reported cosmogenic radionuclide data for South Pole water well particles and concluded that concentrations were too low for the particles to be derived from the break-up of larger meteors.

Measurements of ^3He in different particle size fractions can help to address these issues. Mukhopadhyay and Farley (2006) demonstrated that most ($>80\%$) of the ^3He in 33 million year old deep-sea sediment samples from the North Pacific Ocean

resides in particles less than 50 microns, and 57% in particles less than 37 microns, which is not consistent with the suggestions of Stuart et al. (1999) and Lal and Jull (2005)

Figure 2 shows the size fraction data from the sequential sieving and filtration of particles from Vostok ice samples, revealing that most of the ^3He in these samples is present in the 5-10 micron size fraction, broadly consistent with the modeling of Farley et al. (1997) and Lal and Jull (2005).

Insert Figure 2 about here.

The results do not support the existence of a substantial component of the ^3He flux in larger particles, although larger ice samples may be needed to confirm this conclusion (Lal and Jull, 2005). The total ^3He content of the sample used for the size fraction experiment, obtained by summing the individual fractions, is $443 \pm 6 \times 10^{-18} \text{ cm}^3 \text{ STP g}^{-1}$ ice. This is within the range determined for the replicate samples (Table 1), and since the size fraction experiment had only a small component of ^3He in particles greater than 20 microns it seems unlikely that there is a very large component present in larger particle sizes in the other samples, though clearly larger particles containing extraterrestrial helium do exist (Jull et al., 2007; Osawa and Nagao, 2002; Stuart et al., 1999).

In Figure 2 we compare a histogram of the size fraction data with the size distribution of both mass flux and surface area flux for particles heated to less than 600 degrees during atmospheric entry, calculated from the results of Farley et al. (1997). As those authors point out, choosing a different cut-off temperature between 500 and 700 degrees does not substantially alter the model result. The peak in the size fraction data in the 5-10 micron size fraction is broadly consistent with the model for surface area flux, supporting the contention that the ^3He flux is a surface area-correlated phenomenon, due to implantation of solar helium in IDPs (Farley et al., 1997).

The concentration of ^3He flux in the 5-10 micron size fraction has implications for transport of ^3He bearing particles in the atmosphere. Particles in this size range have tropospheric residence times from less than 1 to 100s of days and may be small enough to be subject to a variety of processes that affect the deposition of atmospheric aerosol particles beyond simple gravitational settling (Kreidenweis et al., 1999; Hobbs, 2000), including accumulation scavenging and long range transport by atmospheric circulation. Lal and Jull (2005) predicted that the flux of 2-5 micron IDPs should peak at 45° N and S based on the fall-out pattern of ^{90}Sr , and that the Vostok site would see only 20% of the mean IDP flux. Observations (Figure 1) exhibit some scatter, but are consistent with a more uniform deposition of IDP ^3He based on the agreement of ice core and marine sediment data. More data are clearly needed to further evaluate spatial variability of the IDP flux.

4. Conclusions

Helium in dust separated from kilogram sized ice samples from the Vostok ice core site in East Antarctica has an isotopic composition consistent with an extraterrestrial origin. High $^3\text{He}/^4\text{He}$ ratios, from 136 to 206 R_a , are similar to those measured in interplanetary dust particles (IDPs) collected in the stratosphere. The mean ^3He flux to the ice core site, derived from replicate measurements of nine kilogram-sized sub-samples that are ~ 3800 years old, is $1.25 \pm 0.37 \times 10^{-12} \text{ cm}^3 \text{ STP cm}^{-2} \text{ ka}^{-1}$, similar to the flux inferred from previous studies of Holocene ice and sediment samples. The large range of ^3He content for individual sub-samples indicates that very large samples, or multiple replicates, will be needed to adequately characterize the ^3He flux from ice samples. A size fractionation experiment shows that the majority of ^3He in particles in the ice at the Vostok site is present in a restricted size range, 5-10 microns, consistent with the hypothesis that helium in IDPs is implanted solar helium and resides

primarily in particle surfaces. Further measurements will be required to fully understand ^3He as a tracer of extraterrestrial input in ice cores, and to determine the spatial and temporal scales of variability.

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Figure Captions

Figure 1. Comparison of ^3He fluxes for Holocene ice samples, including results from this study, and a compilation of flux data from marine sediments from Higgins (2001).

Figure 2. Size distribution of ^3He in particles separated from the 112-115 m section of the Vostok BH-5 ice core discussed in text, and model calculation (Farley et al., 1997) of surface area and mass flux to the earth surface for interplanetary dust particles heated to $< 600^\circ\text{C}$ in atmospheric entry. The peak in the size fraction data in the 5-10 micron size fraction is broadly consistent with the model for surface area flux, supporting the contention that the ^3He flux is a surface area-correlated phenomenon, due to implantation of solar helium in IDPs (Farley et al., 1997).

Tables

Table 1. Results of test of contamination 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

^aMean ± 2se.

Table 2. Helium isotope data for replicate samples spanning 112-115 m from the Vostok BH-5 ice core. Concentrations expressed in cm³ STP per gram of ice melted.

Sample	Ice Weight (g)	⁴ He (10 ⁻¹² cm ³ STP g ⁻¹)	³ He/ ⁴ He (R/R _a)	³ He (10 ⁻¹⁸ cm ³ STP g ⁻¹)
Rep. 1	1157.57	0.52±0.04	206.1±3.1	148±2
Rep. 2	820.78	3.15±0.06	136.0±1.0	592±4
Rep. 3	1123.10	3.03±0.04	184.1±1.0	771±4
Rep. 4	1090.37	3.06±0.05	186.9±1.0	791±4
Rep. 5	1200.78	2.58±0.04	199.8±1.1	715±4
Rep. 6	1099.23	3.21±0.04	182.8±0.9	961±5
Rep. 7	1128.19	3.80±0.04	190.0±0.9	845±4
Rep. 8	1044.32	1.61±0.04	177.6±1.3	395±3
Rep. 9	833.27	2.15±0.06	137.6±1.2	410±4
				Mean: 625±174^a

^aMean ± 2se.

Table 3. Helium isotope data for size fractions separated from the 115-118 m section of the Vostok BH-5 Core. Total sample weight was 1502.63 g ice. Concentrations expressed in cm³ STP per gram of ice melted. Two filters were used for the 5-10 and 0.45-5 micron fractions because of clogging. The amount of ⁴He released from the filters for the 0.45 micron filtered fraction was measurable, as was the ³He/⁴He ratio, and the total ³He could be determined. However, ⁴He was lower than the assumed procedural blank correction and we therefore do not report ⁴He concentration or isotope ratio for this fraction.

Sample	⁴ He (10 ⁻¹² cm ³ STP g ⁻¹)	³ He/ ⁴ He (R/R _a)	³ He (10 ⁻¹⁸ cm ³ STP g ⁻¹)
> 63 μm	0.34±0.03	47.2±3.2	22.0±1.5
20-63 μm	0.64±0.03	24.7±1.7	21.8±1.5
10-20 μm	0.17±0.03	193.1±6.8	44.4±1.6
5-10 μm part 1	0.63±0.03	204.3±2.0	179.4±1.8
5-10 μm part 2	0.48±0.03	223.4±2.7	147.2±1.7
<i>Total</i> 5-10 μm	<i>1.11±0.05</i>	--	<i>326.6±4.9</i>
0.45-5 μm part 1	NM	NM	10.9±1.5
0.45-5 μm part 2	NM	NM	17.4±1.5
<i>Total</i> 0.45-5 μm	NM	NM	<i>28.2±2.1</i>



