Geochemical Tracers of Extraterrestrial Matter in Sediments

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ABSTRACT

Every year, tens of thousands of tons of cosmic dust accumulate at the Earth's surface, representing a continuation of the accretion process that started 4.57 billion years ago. The unique geochemical properties of these materials, compared to the Earth's surface, render them excellent tracers of Solar System, atmospheric, oceanographic, and geologic processes. These processes can be recovered from the records preserved in marine and terrestrial sediments, including snow and ice. We review evidence from these natural archives to illuminate temporal and spatial variations in the flux and composition of extraterrestrial material to Earth, as well as the terrestrial processes that affect the distribution of extraterrestrial tracers in sediments.

INTRODUCTION

Imagine if this issue of *Elements* had a sticky front cover capable of holding on to the extraterrestrial particles that are continuously arriving at the Earth's surface. Within one year, assuming a global average annual accretion rate of ~40,000 tons of cosmic dust, this front cover (~0.06 m^2 in surface area) would capture nearly 5 μg of extraterrestrial matter. If you were then to analyze that matter, you would find that the composition of the individual particles would show great diversity, which would reflect the many different sources of those particles.

The most suitable tracers for detecting and quantifying extraterrestrial matter incorporated into Earth surface materials are the platinum group metals, specifically osmium and iridium, and the noble gases, particularly helium. Relative to average Earth surface materials, these elements are most strongly enriched in extraterrestrial matter (TABLE 1), and we use their concentrations and isotope compositions in sedimentary archives to showcase the kind of information that can be reconstructed from such archives. We will only briefly mention chemical indicators of large asteroid and comet impacts

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because they represent the large end of the extraterrestrial size spectrum: though these rare events do contribute significantly to the flux of extraterrestrial matter if that flux is averaged over long enough time periods (FIG. 1; Wetherill and Shoemaker 1982).

SIZE DISTRIBUTION AND FLUX OF COSMIC DUST

Qualitatively, the frequency with which cosmic dust particles reach the Earth decreases with increasing particle size and, by extension, particle mass (FIG. 1). Quantitatively, the size distribution of cosmic dust particles is described by Poisson statistics. This allows us to estimate the number of cosmic dust particles of any given size range that impinge on a given area of the Earth over a given time interval. Sampling a well-defined area for a sufficiently long time is, therefore, essential for capturing a statistically valid sample of the population of small extraterrestrial particles. The particle size spectrum is dominated by sizes in the range ~1–2,000 μm. The area–time product (m^2 y) is a convenient indicator for identifying a statistically valid sample. However, while the surface area of such a sample can be measured easily, the time interval over which a sample accumulated in geologic strata is more challenging to determine accurately.

The size distribution of small extraterrestrial particles in space has been determined experimentally by, for example, the Long Duration Exposure Facility - a NASA-operated cylindrical installation that remained in low-Earth orbit (331 to 480 km) for about 69 months (FIG. 2, LEFT). In one of this Facility's 57 experiments, a 5.6 m^2 aluminum panel was deployed facing space that then accumulated hundreds of hypervelocity impact craters (FIG. 2, CENTER). These allowed Love and Brownlee (1993) to quantify the cosmic dust flux at 40 ± 20 Gg y^{-1}, 30 Gg y^{-1} of which accounts for the accretion rate of 20–400-μm micrometeoroids. This estimate is based on assuming an average impact velocity of 16.9 km s^{-1} (cf. Plane 2012; Huang et al. 2015). The Long Duration Exposure Facility crater record has been used to determine the size distribution of impacting micrometeoroids smaller than about 500 μm in diameter. This size distribution also constrains the minimum area–time products required to accurately estimate the accumulation rates of the chemical tracers of extraterrestrial origin in Earth surface materials: in this case, 0.1 to ~2 m^2 y (FIG. 3), depending on the tracer (Farley et al. 1997; Peucker-Ehrenbrink 2001). An important caveat associated with these estimates lies in the assumption that the particle size distribution in space, such as estimated from the Long Duration Exposure Facility crater distribution, faithfully represents that of extraterrestrial material accumulating in snow, ice, or marine sediments. However, evidence suggests that ablation, melting, or breakup of extraterrestrial particles with diameters larger than 500 μm creates additional small particles—meteoritic smoke particles—during atmospheric entry. If true, the extraterrestrial particle size distribution relevant to Earth's surface environments will differ from that observed in space (Lal and Jull 2003), which would reduce the area–time product required for obtaining statistically valid flux estimates from sediment samples. Ground-based observations with radar at the Arecibo Observatory (FIG. 2, RIGHT) (Mathews et al. 2001) or with LIDAR[4] at the Table Mountain Lidar Facility in Colorado (USA) (Huang et al. 2015) have been used to quantify velocities and size distributions of extraterrestrial particles impinging at the top layers of Earth's atmosphere. The upper end of these flux estimates agrees with that determined using the Long Duration Exposure Facility cratering record. However, the lower-end flux estimates are one to two orders of magnitude smaller. This may be because radar or LIDAR detection requires ionization or vaporization of particles, but the smaller dust particles are less severely heated and many survive atmospheric entry without signs of melting. Such particles are, therefore, not detected by radar or LIDAR observations.

TRACERS OF EXTRATERRESTRIAL MATTER

The simple expression

$$C_{ET} = ETAR/MAR \qquad (1)$$

clearly illustrates the two different ways tracers of extraterrestrial (ET) matter in snow, ice, and marine sediments are typically applied, with C_{ET} = concentration of ET matter in bulk sediment or ice (i.e. the mass ratio of ET particles to total sample); $ETAR$ = particulate ET accumulation rate (g ET particles cm^{-2} ky^{-1}); and MAR = sediment burial flux or sediment mass accumulation rate (g sediment cm^{-2} ky^{-1}).

When combined with independent data constraining bulk mass accumulation rate, measurements of ET matter (C_{ET}) can be used to quantify ET flux at a study site (Kyte and Wasson 1986; Esser and Turekian 1988; Farley 1995; Winckler and Fischer 2006). Alternatively, if one assumes that the flux of extraterrestrial matter is known, then measurements of extraterrestrial matter in climate archives can be used as a means of constraining bulk mass accumulation rate. The former approach is typically applied in recently deposited records where accurate ways of determining mass accumulation rate are readily available (e.g. ice cores; Winckler and Fischer 2006). In marine sediment records, which reach deeper into Earth's past, the latter approach is a potentially very powerful means of reconstructing mass accumulation rate over time intervals that are

[4] LIDAR (light detection and ranging) technology remotely analyzes objects by illuminating the target with pulsed laser light.

otherwise inadequately constrained by conventional magneto- or biostratigraphy (e.g., McGee and Mukhopadhyay 2013). However, using ET flux to reconstruct sediment mass accumulation rate requires a good understanding of how this background flux of extraterrestrial matter varies through time and around the globe. Furthermore, it requires an approach that differentiates between episodic influx, such as large impacts, and the more constant flux of cosmic dust.

In practice, the nearly overwhelming accumulation rate of terrestrial sediment that accumulates with cosmic dust and its alteration products is a great impediment to studying long-term records of cosmic dust accumulation. Even in the remote Pacific Ocean, terrestrial dust flux is on the order of 100 mg per square meter per year (Huneeus et al. 2011), roughly 1,500 times greater than the global average background flux of extraterrestrial matter. Models that account for the focused wet deposition of ablated and recondensed extraterrestrial matter at high southern latitudes (~60°S; Dhomse et al. 2013) yield the highest estimated ratios of extraterrestrial to terrestrial accumulation rates. In these instances, extraterrestrial matter is thought to account for, at most, 10–25% of total Fe deposition.

Elemental and isotopic tracers can be used to "see through" the ubiquitous terrestrial contamination and identify the influence of extraterrestrial matter in snow, ice, and pelagic sediments. The actual concentration of extraterrestrial particles in sediment is very rarely directly measured. Instead geochemical (isotope) tracers are used to infer the presence of extraterrestrial matter. If the average concentration of any given tracer in cosmic dust is known, or assumed, then the nominal concentration of extraterrestrial particles (C_{ET}, see above) can be calculated. This approach is used to translate measured tracer concentration into estimates of total mass influx.

The most effective tracers of cosmic dust are those that exhibit the largest enrichment in extraterrestrial matter relative to common terrestrial materials (TABLE 1). The element iridium is the most widely recognized among the geochemical tracers of extraterrestrial matter. This is because Ir has played an essential role in inferring that a large impact event occurred at the Cretaceous–Paleogene (formerly known as Cretaceous–Tertiary, or K–T) boundary (Alvarez et al. 1981), and because it can be detected at low concentrations by instrumental neutron activation analysis, without the need for chemical separation. Less widely recognized tracers are He and Os. Differences in the He and Os isotope composition of extraterrestrial matter compared to average crustal material provide means of differentiating between terrestrial and extraterrestrial matter that is more robust than can be achieved for Ir.

Iridium (Ir) as a Tracer of Extraterrestrial Matter

In spite of the early importance of Ir in detecting the influence of cosmic dust in terrestrial materials (Barker and Anders 1968) and its use in differentiating the signature of large impact events in the geologic record from the background flux of cosmic dust (Kyte and Wasson 1986), Ir is, nevertheless, an imprecise monitor of cosmic dust flux. The first factor that limits the precision with which the extraterrestrial Ir flux can be quantified is the need to differentiate between terrestrial and extraterrestrial Ir. The second limiting factor is the heterogeneous accumulation rate of Ir around the globe. This is particularly important because it requires either averaging of a great many records to obtain a meaningful estimate of the extraterrestrial Ir flux, or a detailed understanding of the transport processes that deliver Ir to sedimentary archives (FIG. 4). These processes include partial vaporization of Ir during atmospheric entry, and atmospheric removal via wet and dry deposition, which varies regionally. Iridium accumulation in marine sediments is subject to additional complexity related to the chemical form of Ir arriving at the sea surface, as well as differential transport and removal of dissolved and particulate Ir to the seafloor. The variability of these transport and removal processes in time and space contributes significant uncertainty to estimates of extraterrestrial flux based on any single record.

Measurements of Ir in snow and ice reveal unexpected complexity in quantifying ET flux using Ir data (Gabrielli et al. 2004). First, the relative proportions of terrestrial and extraterrestrial Ir can vary greatly with time. Corrections for terrestrial Ir rely upon paired measurements of major elements (Al or Fe). This approach is predicated on the assumption that all terrestrial Ir is carried by detrital material with a relatively constant Ir/Al or Ir/Fe ratio. In some instances, this is explicitly assumed to be an average crustal ratio. The magnitude of this correction varies greatly, for example, in Greenland ice between "dusty" glacial intervals, which largely obscure the extraterrestrial contributions, and interglacial intervals, where 98% of the Ir is estimated to be of extraterrestrial origin. A more surprising result is that the vast majority of Ir in the ice seems to be hosted in a very small size fraction (< 0.45 μm). This feature is interpreted to mean that most of the Ir arriving at the Earth's surface by cosmic dust has been ablated from incoming primary ET particles and recondensed as meteoritic smoke particles. The very small size of meteoritic smoke particles, on the order of nanometers, results in very slow gravitational settling, and this amplifies the importance of atmospheric transport of meteoritic smoke particles and their removal by wet deposition (Plane 2012). Detecting meteoritic smoke particles in ice cores using magnetic methods provides empirical evidence of geographic focusing associated with wet deposition (Lanci et al. 2007).

While strong Ir enrichment relative to average crustal material in slowly accumulating marine sediments is well documented, confidently partitioning this Ir into terrestrial and extraterrestrial fractions is problematic. In their classic study of Ir in North Pacific pelagic clays, Kyte and Wasson (1986) estimated that approximately half of the total Ir burial flux of ~15 pg cm^{-2} ky^{-1} is derived from terrestrial sources. Subsequent work (Dalai and Ravizza 2006) explicitly estimated the terrestrial and extraterrestrial fractions of the total sediment Ir inventory using Th and ^{3}He as proxies for terrestrial and extraterrestrial matter, respectively. Hydrogenous (seawater-derived) Ir was calculated by difference, leading to the conclusion that 70–85% of the Ir in pelagic clays, and in the more rapidly accumulating pelagic carbonates, is scavenged from seawater. Thus, the hydrogenous (seawater-derived) Ir burial flux need not be constant in time or space. Iridium analyses of snow and ice described in the preceding paragraph imply a significant fraction of the hydrogenous Ir may be derived from soluble meteoritic smoke particles deposited on the sea surface. However, the few available analyses of dissolved Ir in rivers suggest that this source of terrestrial Ir is sufficiently large to balance the hydrogenous Ir burial flux. Ultimately, the source of hydrogenous Ir is impossible to discern, and the rate of Ir scavenging from seawater can vary regionally. Consequently, a precise estimate of the background flux of extraterrestrial Ir is beyond our reach. This means that the available Ir data are best regarded as an upper bound on the flux of extraterrestrial matter.

Tracing ET Matter with Os Isotopes

Osmium isotope variations, caused by the decay of ^{187}Re to ^{187}Os, provide a more robust means of correcting for terrestrial background than is possible using Ir. Specifically, slowly accumulating pelagic clays have distinctly lower $^{187}Os/^{188}Os$ ratios and higher Os concentrations than typical marine sediments. These characteristics are attributed to the influence of particulate extraterrestrial matter with high Os concentrations and low $^{187}Os/^{188}Os$ ratios compared to other common surficial sources of Os. Quantifying the fraction of extraterrestrial Os in bulk sediments using Os isotopes is achieved using physical mixing models. These models can account for the influence of both detrital minerals and hydrogenous Os on marine sediments (Fig. 4) and are used to calculate the concentration of Os in the bulk sediment that is associated with extraterrestrial particles (Esser and Turekian 1988; Dalai and Ravizza 2006). The mass of particulate extraterrestrial matter is calculated by assuming a certain chondritic Os concentration for extraterrestrial particles.

Nevertheless, the mixing models used to quantify the amount of Os carried by cosmic dust require assumptions analogous to those used to correct for terrestrial Ir contributions in marine sediments. Consequently, Os isotope-based estimates of cosmic dust flux display a significant range in magnitude (Peucker-Ehrenbrink 2001). This range is probably better interpreted as an uncertainty in the calculations themselves rather than as clear evidence of temporal or geographic variations in cosmic dust flux. Consequently, recent work has used available He isotope data to constrain the application of Os as cosmic dust tracer. For example, the late Eocene increase in cosmic dust flux recognized using ^{3}He (Farley et al. 1998) is also associated with a decline in the $^{187}Os/^{188}Os$ of seawater (Paquay et al. 2014). This finding suggests that Os ablated from cosmic dust during transit through the atmosphere is soluble in seawater and can influence the Os isotopic composition of the global ocean.

Helium and its Isotopes as Tracer of ET Matter

Like the platinum group elements, extraterrestrial matter is highly enriched in helium, a rare gas on Earth but the second-most abundant element in the observable universe. Of the two helium isotopes (^{3}He and ^{4}He), ^{3}He is the proxy of choice for tracing cosmic dust (TABLE 1). The helium isotope ratio ($^{3}He/^{4}He$) of cosmic dust is about four orders of magnitude higher than that of terrestrial sources, which are dominated by ^{4}He from the decay of long-lived radionuclides such as U and Th. As helium in sediments is a mixture of just these two sources, the extreme isotopic contrast allows for a straightforward differentiation of cosmic dust from terrestrial helium carried by, for instance, aeolian dust. In many sedimentary archives, such as marine sediment cores from the remote Pacific Ocean and from ice cores from Greenland or Antarctica, virtually all of the ^{3}He (> 99.5%) is of extraterrestrial origin. In environments that have a higher terrestrial input, the relative contribution of terrestrial ^{3}He can be higher and can generally be well constrained from a simple two-component mixing model (Marcantonio et al. 1995, McGee and Mukhopadhyay 2013).

The behavior of He in extraterrestrial matter differs from that of Os and Ir in important ways (FIG. 4). First, extraterrestrial helium in cosmic dust particles is derived from implantation of solar wind during the dust particles' transit to Earth. Therefore, the extraterrestrial helium signal is correlatable to surface area, as opposed to being volume-related, and this requires smaller area–time products to accurately determine the fluxes of ET matter (FIG. 3). Second, the atmosphere effectively acts as a "helium filter" because the vast majority of cosmic dust particles lose He during entry into Earth's atmosphere as a result of frictional heating to temperatures above 600 °C. Only small cosmic dust particles

(<35 μm), which experience less intense frictional heating, retain their extraterrestrial helium (Farley et al. 1997). As a result, unlike Ir and Os, ^{3}He is uniquely associated with the smallest cosmic dust particles accumulating on Earth, corresponding to only about 0.5% of the total mass flux (FIG. 4).

The accretion history of ^{3}He, as recorded in sedimentary sections in the ocean and on land, can be used to track Solar System events associated with increased cosmic dust flux in the absence of large impactors. During the Cenozoic (a period covering the last 66 My of Earth's history), two such events have been documented: a comet or asteroid shower in the Late Eocene (Farley et al. 1998), and a major asteroid collision in the late Miocene (Farley et al. 2006).

Apart from these events, the extraterrestrial ^{3}He-flux has remained roughly constant over 10 ky to million-year timescales during the Cenozoic. Most observational evidence for a spatially and temporarily constant ^{3}He flux (FIG. 5) comes from Quaternary marine sediment cores (Farley 1995; Marcantonio et al. 1995, 1999; Winckler et al. 2004) and high-latitude ice cores (Brook et al. 2000; Winckler and Fischer 2006). This opens up the opportunity to use cosmic dust–derived ^{3}He as a constant flux proxy to determine rates of sediment accumulation in various geological archives and to constrain paleofluxes, an essential parameter when interpreting the dynamics of past climate systems.

The fact that the flux of ^{3}He is constant means that it can be used as a "clock" to establish the duration of events in Earth's sedimentary record. The most prominent example is the determination of the duration of the Cretaceous–Paleogene event by using ^{3}He as a constant flux tracer. While the asteroid causing the Cretaceous–Paleogene event created the renowned Ir signal (Alvarez et al. 1981), it was—as would be expected for a large impactor—completely degassed of helium. This then allowed the constant background cosmic dust ^{3}He-signal to be utilized to determine the duration of this event, which has now been estimated to be ~ 10 ky (e.g. McGee and Mukhopadhyay 2013).

SUMMARY

Strong enrichment of Ir, Os, and He in extraterrestrial matter compared to typical upper crustal materials renders these elements sensitive tracers of the presence of extraterrestrial matter in ice, snow, and marine sediments. Natural variations in the isotopic composition of Os and He enhance the utility of these tracers for differentiating between the terrestrial and extraterrestrial contributions to element budgets. The volatility of He compared to Os and Ir, and its concentration on the surface of incoming extraterrestrial particles, cause He to trace a very different, much smaller, size fraction of incoming extraterrestrial particles than do Os and Ir. In contrast to He, where vaporized atoms are lost from the Earth system, ablated Ir (and presumably Os) condense into extremely small (nm-sized) meteoritic smoke particles. The behavior of these small particles in the atmosphere and ocean is not well understood, and this makes using Ir and Os as tracers for quantifying total extraterrestrial flux to various sedimentary archives preserved on Earth's surface more complicated. Nevertheless, measuring a variety of tracers of extraterrestrial matter in the same sample will help us gain a comprehensive understanding of the processes that affect the flux of extraterrestrial matter to points on the Earth's surface in the geologic past.

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FIGURE CAPTIONS

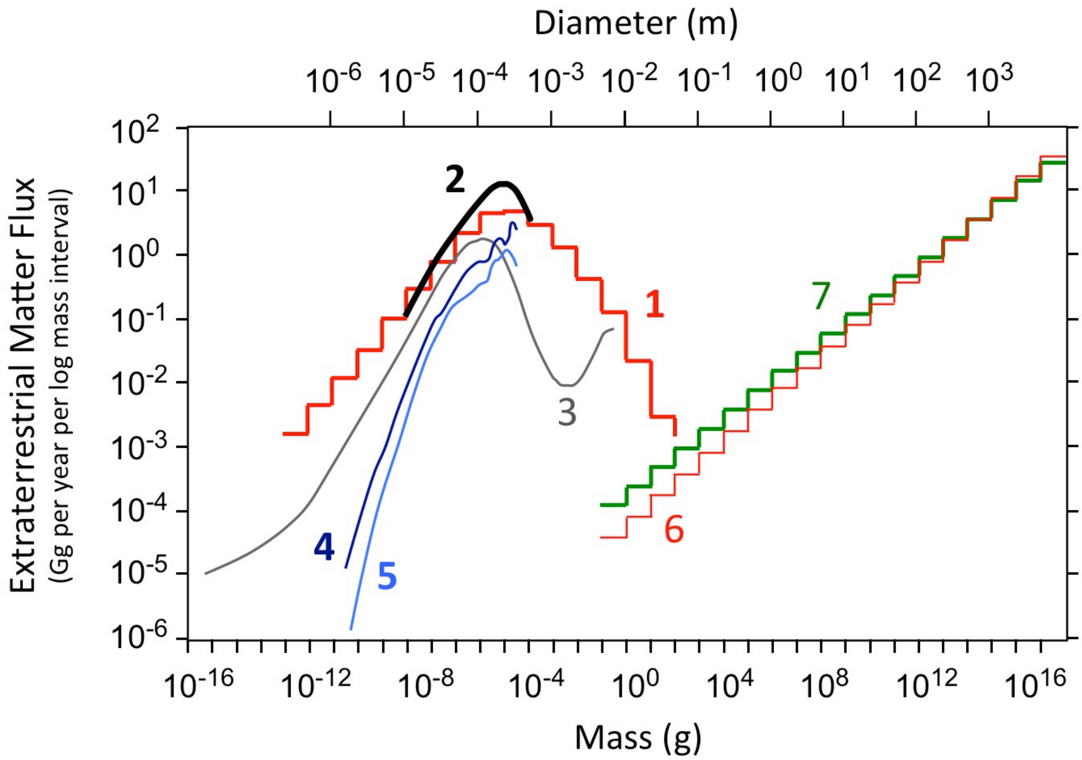

FIGURE 1 Estimated accretion rates of extraterrestrial matter on Earth are dominated by two peaks. The peak at small masses is caused by cosmic dust, micrometeorites and debris ablated from a larger object during atmospheric entry (2.5 g cm^{-3} density; Love and Brownlee 1993). Estimates for different extraterrestrial accretion rates are shown by numbered colored lines: (**1**) cosmic dust flux (Hughes 1978); (**2**) Long Duration Exposure Facility impact record (Love and Brownlee 1993); (**3**) radar micrometeor observations of annual whole-Earth mass fluxes at the Arecibo Observatory in Puerto Rico (Ceplecha et al. 1998); (**4**) same as 3 but for 1998 only; (**5**) same as 3, but for 1997 only; (**6**) the increase in mass flux at large masses reflects large impacts (3 g cm^{-3} density; Kyte and Wasson 1986), quantified using cratering records (Wetherill and Shoemaker 1982); (**7**) same as 6 but recalculated using a 5% larger exponent in the exponential relationship between the number of extraterrestrial objects and the object's radius.

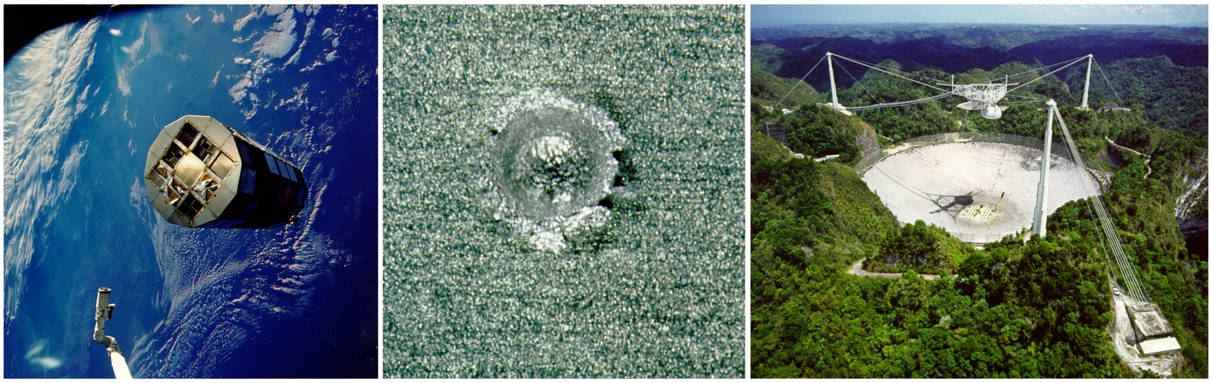

Figure 2 (**left**) Space-facing end of the Long Duration Exposure Facility in orbit. Photo courtesy of NASA. (**center**) Submillimeter impact crater in an aluminum panel from the Long Duration Exposure Facility. Photo courtesy of NASA. (**right**) The Arecibo Observatory, a radio telescope constructed within a karst sinkhole in Puerto Rico. Photo courtesy of NAIC Arecibo Observatory, a Facility of the United States National Science Foundation.

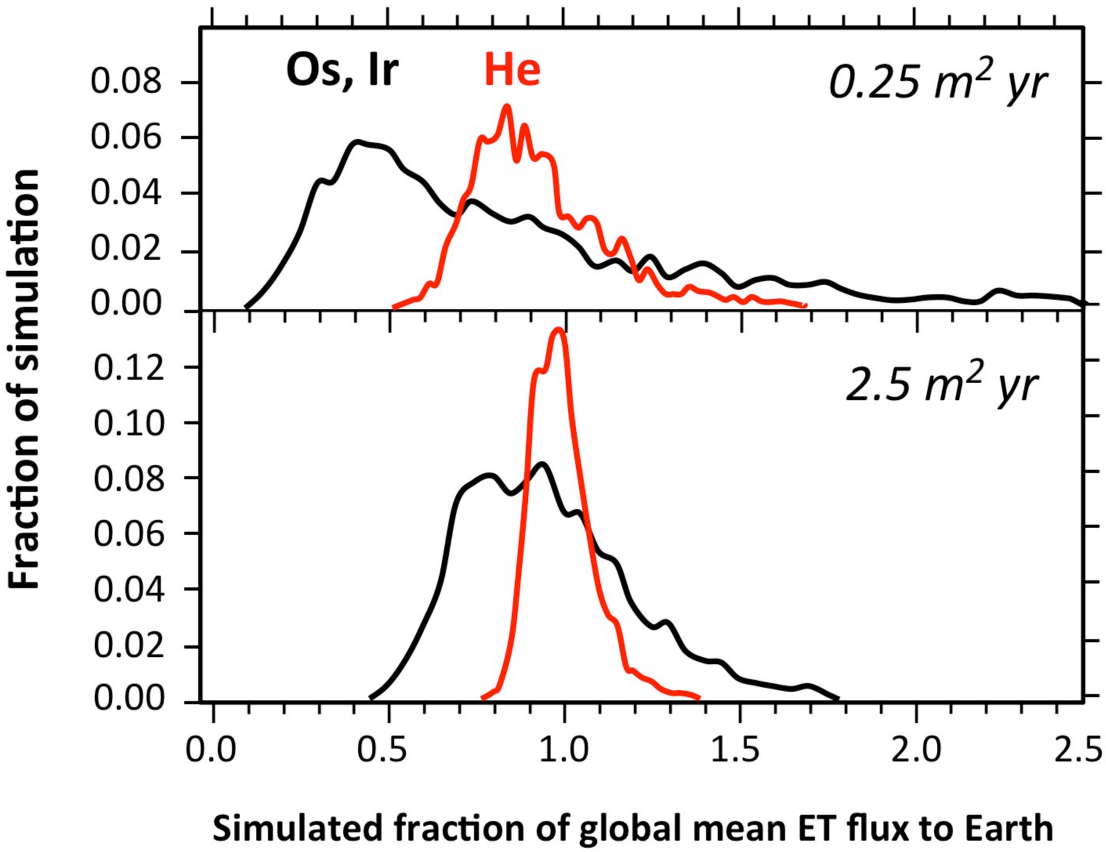

FIGURE 3 Monte Carlo simulation of bias introduced by sampling the population of extraterrestrial (ET) particles for a specific area–time (m^2 y) product. Black line represents refractory tracers that are a function of particle mass (Os, Ir). Red line represents solar wind tracers (He) that are a function of surface area. Whereas a sample representing 2.5 m^2 y (bottom panel) captures the extraterrestrial fluxes of He, Os and Ir without bias (simulated fractions ~1.0), a sample representing only 0.25 m^2 y (top panel) likely samples less than half (simulated fraction ~0.5) of the true Os and Ir flux, without significantly biasing the results for He (simulated fraction ~0.9). MODIFIED AFTER FARLEY ET AL. (1997) AND PEUCKER-EHRENBRINK (2001).

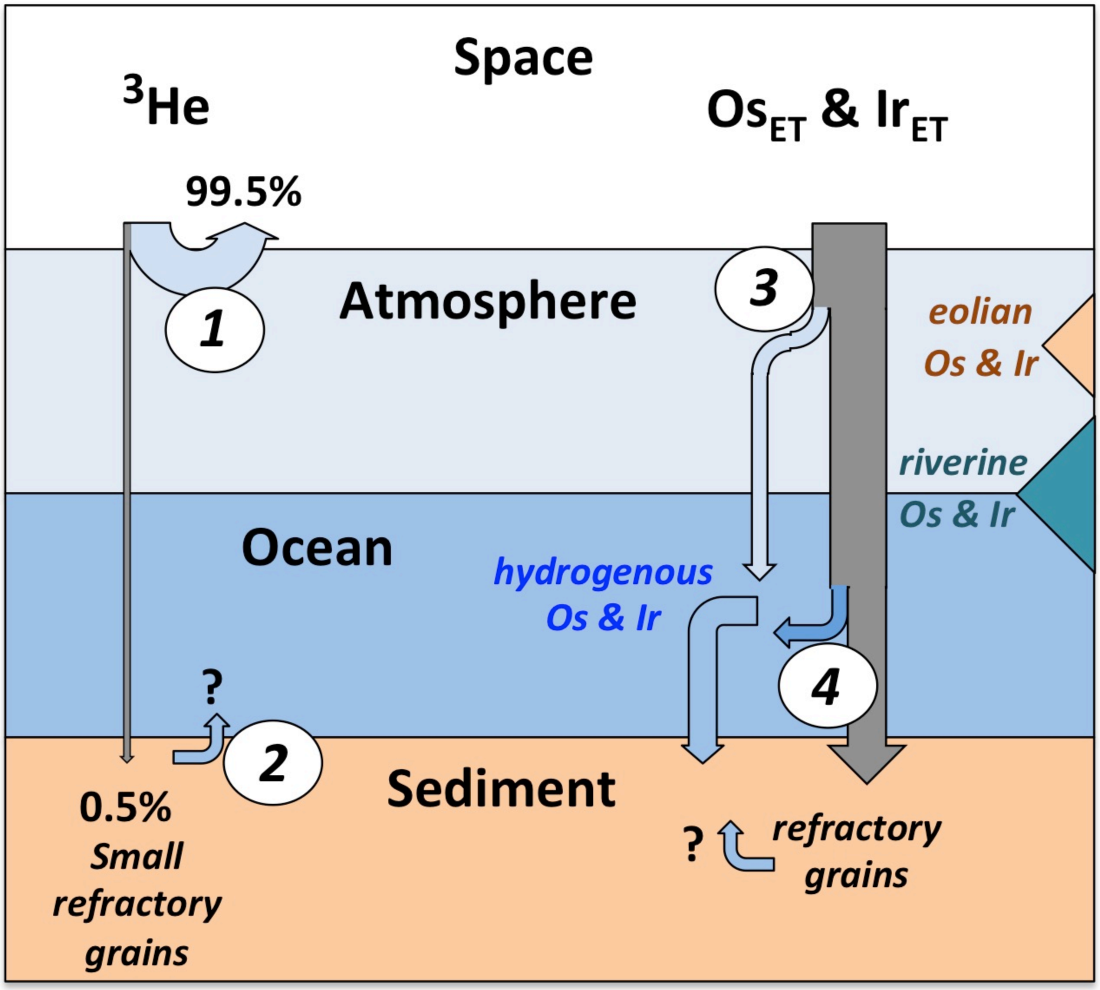

FIGURE 4 A schematic representation of the fate of extraterrestrial (ET) ^{3}He, Ir and Os. (**1**) The vast majority of ^{3}He that impinges upon Earth's atmosphere is lost during ablation of larger particles (>35 μm) and subsequent gravitational escape. (**2**) Whereas there is potential loss of ^{3}He from small cosmic dust particles deposited on the seafloor, empirical evidence demonstrates that elevated ^{3}He levels persist in seafloor sediments for well over 100 My. (**3**) Large fractions of Ir and (presumably) Os are vaporized from incoming cosmic dust particles. (**4**) Dissolution of Ir and Os from cosmic dust particles in the water column may further reduce the fraction of extraterrestrial Ir and Os that reaches the seafloor as refractory material. It is noteworthy that the soluble components of extraterrestrial Ir and Os are comingled with terrestrial backgrounds of Ir and Os dissolved in seawater, making the soluble extraterrestrial component difficult to trace. For ^{3}He, terrestrial background can be largely neglected in many marine sedimentary environments.

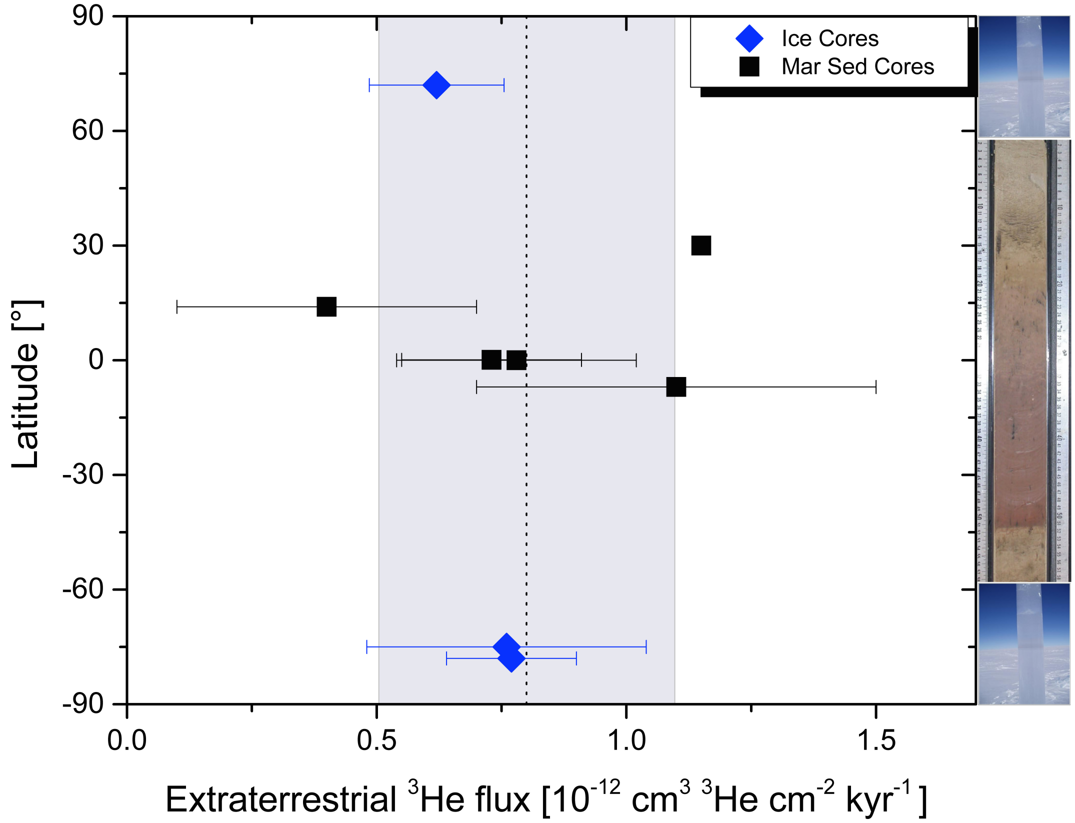

FIGURE 5 Latitudinal distribution of Holocene extraterrestrial helium flux on Earth, reconstructed from marine sediment cores as represented by black squares [DATA FROM FARLEY (1995), MARCANTONIO ET AL. (1995, 1999, 2001), AND WINCKLER ET AL. (2004)] and from ice cores as represented by blue diamonds [DATA FROM BROOK ET AL. (2000) AND WINCKLER AND FISCHER (2006)]. The grey box indicates the average extraterrestrial ^{3}He accumulation rate during the late Quaternary (0.8 ± 0.3 10^{-12} cm^{3} STP cm^{-2} ky^{-1}; McGee and Mukhopadhyay 2013). There is no systematic pattern of spatial variability evident in the extraterrestrial ^{3}He fluxes. Pictures at right are symbolic of collection environment. ICE PHOTO COURTESY OF IDDO, UNIVERSITY OF WISCONSIN-MADISON; SEDIMENT PHOTO COURTESY OF DANIELA SCHMIDT, UNIVERSITY OF BRISTOL.

TABLE 1 Assumed concentrations of geochemical tracers of extraterrestrial Ir, Os, and ^{3}He in cosmic dust tabulated to illustrate strong tracer enrichment in the extraterrestrial component compared to common terrestrial material. Normalization to Al further amplifies this contrast. The unit for ^{3}He is "cubic centimeters at standard temperature and pressure per gram."

	Al (wt%)	**Ir (ng/g)**	**Os (ng/g)**	**^{3}He (cc STP/g)**
Cosmic Dust	0.9	455	490	$\approx 10^{-5}$
Upper Crustal Material	7.3	0.02	0.03	$\approx 10^{-13}$