
Extraterrestrial He in Sediments: From Recorder of Asteroid Collisions to Timekeeper of Global Environmental Changes

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Abstract

Most ^3He in deep-sea sediments is derived from fine-grained extraterrestrial matter known as interplanetary dust particles (IDPs). These particles, typically $<50\ \mu\text{m}$ in diameter, are sufficiently small to retain solar wind-implanted He with high $^3\text{He}/^4\text{He}$ ratios during atmospheric entry heating. This extraterrestrial ^3He ($^3\text{He}_{\text{ET}}$) is retained in sediments for geologically long durations, having been detected in sedimentary rocks as old as 480 Ma. As a tracer of fine-grained extraterrestrial material, $^3\text{He}_{\text{ET}}$ offers unique insights into solar system events associated with increased IDP fluxes, including asteroid break-up events and comet showers. Studies have used $^3\text{He}_{\text{ET}}$ to identify IDP flux changes associated with a Miocene asteroid break-up event and a likely comet shower in the Eocene. During much of the Cenozoic, $^3\text{He}_{\text{ET}}$ fluxes have remained relatively constant over million-year timescales, enabling $^3\text{He}_{\text{ET}}$ to be used as a constant flux proxy for calculating sedimentary mass accumulation rates and constraining sedimentary age models. We review studies employing $^3\text{He}_{\text{ET}}$ -based accumulation rates to estimate the duration of carbonate dissolution events associated with the K/Pg boundary and Paleocene-Eocene Thermal Maximum. Additionally, $^3\text{He}_{\text{ET}}$ has been used to quantify sub-orbital variability in fluxes of paleoproductivity proxies and windblown dust. In order to better interpret existing records and guide the application of $^3\text{He}_{\text{ET}}$ in novel settings, future work requires constraining the carrier phase(s) of $^3\text{He}_{\text{ET}}$ responsible for long-term retention in sediments, better characterizing the He isotopic composition of the terrigenous

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end-member, and understanding why observed extraterrestrial ^3He fluxes do not match the predicted variability of IDP accretion rate over orbital timescales.

1 Introduction

The presence of extraterrestrial material in marine sediments was first proposed by Murray (1876) based upon his observations of magnetic spherules in open-ocean surface sediments collected during the Challenger expedition. Later investigations found similar spherules at high altitudes in the atmosphere and in ice sheets, terrestrial deposits and geologically old marine sediments (Laevastu and Mellis 1955; Fireman and Kistner 1961; Thiel and Schmidt 1961; Fredriksson and Gowdy 1963). Geochemical and mineralogical investigations of cut and polished samples provided evidence consistent with an extraterrestrial origin for at least some of these spherules, including metallic iron cores and high nickel contents (Fredriksson 1956; Fredriksson and Martin 1963). In parallel with this work, studies of meteorites established that extraterrestrial materials contained noble gas signatures clearly distinguishable from terrestrial samples, marked most notably by high ^3He concentrations and $^3\text{He}/^4\text{He}$ ratios (e.g., Tilles 1962; Zähringer 1962). Drawing upon these findings, Merrihue (1964) demonstrated that the He isotopic composition of a red clay sample from the south tropical Pacific was inconsistent with any known terrestrial source and was similar to that found in extraterrestrial samples.

Subsequent work has established that most ^3He in open-ocean sediments is extraterrestrial, and that this extraterrestrial ^3He ($^3\text{He}_{\text{ET}}$) is dominantly brought to Earth's surface by fine (generally $<50\ \mu\text{m}$) interplanetary dust particles (IDPs). IDPs consist of a combination of asteroid and comet debris and carry implanted solar ions with characteristically high $^3\text{He}/^4\text{He}$ ratios. Though all extraterrestrial matter initially contains implanted He, coarser particles attain higher temperatures during atmospheric entry, causing them to lose most of their $^3\text{He}_{\text{ET}}$ before

reaching the Earth's surface. The atmosphere thus acts as a filter, allowing only fine particles to retain $^3\text{He}_{\text{ET}}$.

Over the past 30 years $^3\text{He}_{\text{ET}}$ has been identified in Quaternary marine sediments (e.g., Takayanagi and Ozima 1987; Farley and Patterson 1995; Marcantonio et al. 1995) and ice cores (Brook et al. 2000); in marine sediments spanning the Cenozoic (Farley 1995); and in sedimentary rocks as old as 480 Ma (Patterson et al. 1998). When combined with sedimentary accumulation rate estimates, $^3\text{He}_{\text{ET}}$ data provide insight into past changes in the flux of extraterrestrial material associated with asteroid break-up events and comet showers (Farley 1995; Farley et al. 1998, 2005, 2006; Mukhopadhyay et al. 2001a, b). Because $^3\text{He}_{\text{ET}}$ traces the accretion of IDPs dominantly smaller than $50\ \mu\text{m}$, $^3\text{He}_{\text{ET}}$ fluxes offer a view of accretion rates of extraterrestrial matter that is distinct from that provided by platinum group elements (PGEs) such as iridium, which trace the total extraterrestrial mass flux; large impactors leave PGE anomalies in the sedimentary record but contribute negligible amounts of $^3\text{He}_{\text{ET}}$ (e.g., Mukhopadhyay et al. 2001a, b). Likewise, collisions in the asteroid belt substantially enhance IDP accretion rates—and thus, $^3\text{He}_{\text{ET}}$ fluxes—without delivering large impactors (Farley et al. 2006).

As $^3\text{He}_{\text{ET}}$ accumulation rates have been found to be roughly constant on million-year timescales through most of the Cenozoic, $^3\text{He}_{\text{ET}}$ can also be used as a constant-flux proxy for calculating sedimentary accumulation rates (Marcantonio et al. 1995, 1996; Mukhopadhyay et al. 2001b, Farley and Eltgroth 2003). This approach has proved particularly valuable in portions of the sedimentary record marked by dramatic changes in sedimentation (e.g., Mukhopadhyay et al. 2001b; Farley and Eltgroth 2003; Murphy et al. 2010). Even in periods of more typical pelagic sedimentation, $^3\text{He}_{\text{ET}}$ -based accumulation rates

allow calculation of fluxes for paleoclimate studies with sub-orbital resolution that are largely independent of age model errors, changes in carbonate preservation and lateral advection of sediments (Winckler et al. 2005; Marcantonio et al. 2009; McGee et al. 2010; Torfstein et al. 2010).

In this chapter we review the current understanding of the sources of IDPs and the carrier phases of extraterrestrial He responsible for long-term retention of He in marine and terrestrial deposits. We then discuss applications of ${}^3\text{He}_{\text{ET}}$ to questions of past changes in IDP flux and marine sediment accumulation. We close the chapter with a set of open questions that provide directions for future work.

2 Origin of Extraterrestrial He in IDPs

Interplanetary dust particles originate as asteroid and comet debris and range in size from $\sim 1 \mu\text{m}$ to 1 mm, with a peak in mass flux near Earth at a diameter of $\sim 200 \mu\text{m}$ (Love and Brownlee 1993). IDPs are typically porous aggregates of silicate minerals and carbonaceous matter (Nier and Schlutter 1990), with densities averaging $\sim 2 \text{ g/cm}^3$ and ranging from 1 to 6 g/cm^3 (Love et al. 1994). Poynting-Robertson and solar wind drag cause IDP orbits to decay toward the Sun on timescales of 10^4 – 10^5 years (Burns et al. 1979). This short lifetime requires continuous supply of new IDPs to the inner solar system. IDP sources include asteroid collisions and comets, though the relative importance of these sources is as yet uncertain. Kortenkamp and Dermott (1998b) argued that much of the zodiacal cloud, the diffuse cloud of dust in the plane of the solar system, originates from collisions of a limited number of asteroid families. More recently, modeling by Nesvorný et al. (2010) suggested that Jupiter-family comets might be the most important source of IDPs at distances closer than 5 AU.

As IDPs spiral into the sun they are bombarded by solar wind (SW), which imparts high noble gas concentrations and solar isotopic ratios. SW ions are primarily composed of H and He with particle energies on the order of 1 keV/n (Futagami et al.

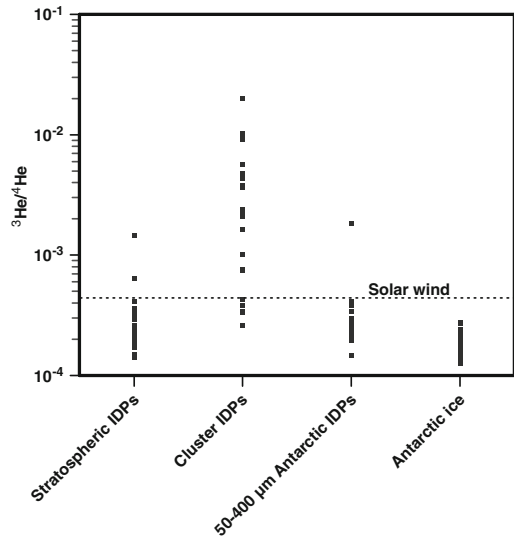


Fig. 1 ${}^3\text{He}/{}^4\text{He}$ ratios of IDPs collected from the stratosphere and from Antarctic ice. Dashed line indicates the ${}^3\text{He}/{}^4\text{He}$ ratio of solar wind (4.48×10^{-4}). Note that most IDPs have ${}^3\text{He}/{}^4\text{He}$ ratios similar to or lower than solar wind, while cluster IDPs (fragments of large IDPs) commonly have much higher ${}^3\text{He}/{}^4\text{He}$ ratios, perhaps due to spallogenic He production by galactic cosmic rays. Stratospheric IDPs: Nier et al. (1990), Nier and Schlutter (1992). Cluster IDPs: Nier and Schlutter (1993). 50–400 μm Antarctic IDPs: Stuart et al. (1999). Antarctic ice: Brook et al. (2000)

1990) and ${}^3\text{He}/{}^4\text{He}$ ratios of $4.48 (\pm 0.15) \times 10^{-4}$ (Benkert et al. 1993). ${}^3\text{He}/{}^4\text{He}$ ratios in IDPs sampled in the stratosphere reach the solar wind value of 4.4×10^{-4} , although the majority of the particles have ratios ~ 2.0 – 2.7×10^{-4} (Fig. 1) (Nier and Schlutter 1990, 1992; Pepin et al. 2000). The lower-than-SW ${}^3\text{He}/{}^4\text{He}$ ratios in most IDPs have led to suggestions that SW He is lost during atmospheric entry and that IDP He instead reflects a distinct population of higher energy solar ions with lower ${}^3\text{He}/{}^4\text{He}$ ratios, called solar energetic particles (SEP; Amari and Ozima 1988; Nier et al. 1990; Pepin et al. 2000). However, study of solar ions implanted during the Genesis mission has recently demonstrated that the heavier isotope is implanted deeper, resulting in depth-dependent isotopic fractionation such that lighter compositions are observed near the surface (Grimberg et al. 2006; Wieler et al. 2007). If so, the lower ${}^3\text{He}/{}^4\text{He}$ ratios observed in IDPs might simply indicate preferential loss of the

more shallowly implanted component of solar wind ions during atmospheric entry heating rather than a separate SEP component.

Though most data support a solar source of He in IDPs, some fragments of cluster IDPs collected in the stratosphere—large ($\sim 40 \mu\text{m}$) IDPs that break up when impacting the collector surface—have ${}^3\text{He}/{}^4\text{He}$ ratios of up to $\sim 1 \times 10^{-2}$ (Fig. 1) (Nier and Schlutter 1993; Pepin et al. 2000, 2001). Spallation reactions from galactic cosmic rays (GCR) or solar cosmic rays (SCR) appear to be the only plausible source of these ${}^3\text{He}/{}^4\text{He}$ ratios. In some cases, ${}^3\text{He}/{}^4\text{He}$ ratios in deep-sea ferromanganese crusts also indicate the presence of GCR-produced ${}^3\text{He}$ (Basu et al. 2006). However, the ${}^3\text{He}$ flux implied from ferromanganese crusts is a few orders of magnitude lower than that inferred from marine sediments. Furthermore, in magnetic separates from deep sea sediments the measured Ne isotopic compositions appear to be primarily a two component mixture of SW and atmospheric Ne (Fukumoto et al. 1986; Matsuda et al. 1990). Thus, GCR- or SCR-produced ${}^3\text{He}$ appears not to be a major component of the extraterrestrial ${}^3\text{He}$ inventory.

3 IDPs in Terrestrial Deposits

3.1 Determination of Extraterrestrial Noble Gases

In marine sediments and ice cores He is often a two component mixture of extraterrestrial He from IDPs and terrigenous He present in detrital minerals (Fig. 2a). The concentration of extraterrestrial ${}^3\text{He}$ can be estimated using the following equation (Marcantonio et al. 1995):

$$[{}^3\text{He}_{ET}] = \left(\frac{1 - \frac{{}^3\text{He}/{}^4\text{He}_{TERR}}{{}^3\text{He}/{}^4\text{He}_{meas}}}{{}^3\text{He}/{}^4\text{He}_{IDP}} \right) \cdot [{}^3\text{He}_{meas}] \quad (1)$$

where brackets indicate concentrations, ‘meas’ indicates the measured values, and ${}^3\text{He}/{}^4\text{He}_{TERR}$ and ${}^3\text{He}/{}^4\text{He}_{IDP}$ denote the ratios for the terrigenous and extraterrestrial endmembers, respectively.

The fraction of ${}^3\text{He}_{ET}$ in the sediments is usually insensitive to the choice of whether the IDP ${}^3\text{He}/{}^4\text{He}$ ratio is 4.48×10^{-4} (SW value; Benkert et al. 1993) or the average value of 2.4×10^{-4} observed in stratospheric IDPs (Nier and Schlutter 1990; 1992). The ${}^3\text{He}/{}^4\text{He}_{TERR}$ ratio typically varies between $2\text{--}4 \times 10^{-8}$ but occasionally may reach 10^{-7} . When the measured ${}^3\text{He}/{}^4\text{He}$ ratios in the sediments are in the 10^{-5} range, the deconvolution is insensitive to the choice of the terrigenous endmember (Fig. 2b; Marcantonio et al. 1995; Farley and Patterson 1995; Mukhopadhyay et al. 2001a; Higgins et al. 2002; Winckler et al. 2005). However, when measured ratios are in the low 10^{-6} to 10^{-7} range, such as in continental margin sediments and other settings with high siliciclastic inputs, studies must consider the sensitivity of ${}^3\text{He}_{ET}$ concentrations to uncertainties in the ${}^3\text{He}/{}^4\text{He}_{TERR}$ ratio (e.g., McGee et al. 2010).

The ${}^3\text{He}/{}^4\text{He}_{TERR}$ ratio is usually governed by the coupled production of ${}^4\text{He}$ from U and Th decay and ${}^3\text{He}$ from the reaction ${}^6\text{Li}(n,\alpha){}^3\text{H} \rightarrow {}^3\text{He}$, where the source of the neutrons is the decay of U and Th (Andrews 1985; Mamyurin and Tolstikhin 1984). The ${}^3\text{He}/{}^4\text{He}$ production ratio in crustal materials is $\leq 4 \times 10^{-8}$ (Andrews 1985; Mamyurin and Tolstikhin 1984), four orders of magnitude lower than ${}^3\text{He}/{}^4\text{He}_{ET}$. This value is similar to values measured for bulk Chinese loess (Marcantonio et al. 1998; Farley 2000; Du et al. 2007; McGee 2010). Some terrigenous samples, however, have ${}^3\text{He}/{}^4\text{He}$ ratios of $\sim 10^{-7}$, a factor of 10 higher than the average production rate for the upper continental crust (Marcantonio et al. 1998). Recent work has demonstrated that the fine fraction of Chinese loess ($< 4 \mu\text{m}$) has a factor of 10 higher ${}^3\text{He}/{}^4\text{He}$ ratios ($\sim 10^{-7}$) than bulk loess, suggesting that the grain size of terrigenous inputs should be taken into account in choosing ${}^3\text{He}/{}^4\text{He}_{TERR}$ (McGee 2010).

A systematic understanding of what controls terrigenous ${}^3\text{He}/{}^4\text{He}$ ratios would help in determining ${}^3\text{He}/{}^4\text{He}_{TERR}$ for various settings, but at present, the source of high ($\geq 10^{-7}$) ${}^3\text{He}/{}^4\text{He}$ ratios in terrigenous sediments is debated. Air-derived ${}^3\text{He}$ appears to be ruled out in sediments even when the measured ${}^3\text{He}/{}^4\text{He}$ ratios are

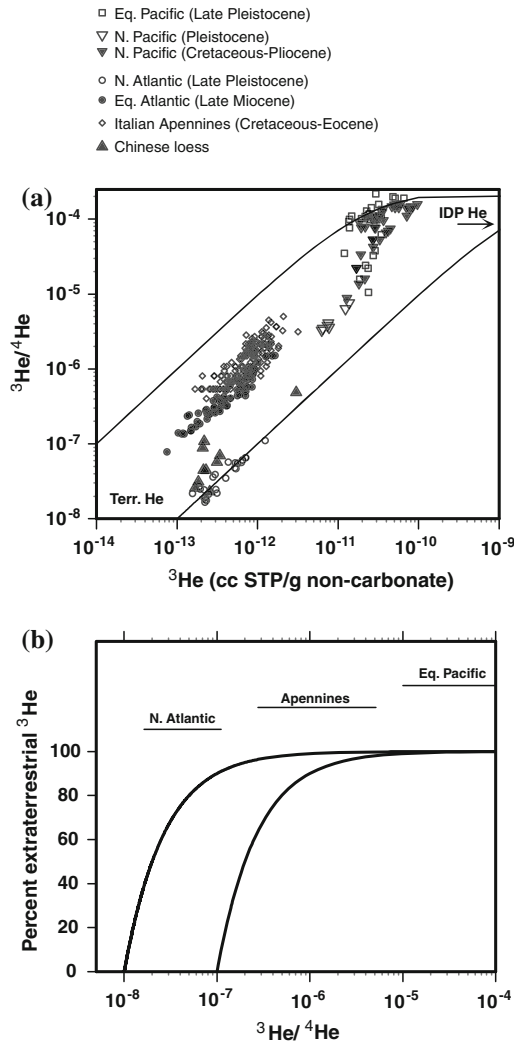


Fig. 2 Mixing between extraterrestrial and terrigenous He in terrestrial sediments. **a** He isotope data from deposits spanning from the late Cretaceous to the Holocene (after Marcantonio et al. 1998). Lines depict mixing between IDP He (${}^3\text{He}/{}^4\text{He} = 2.4 \times 10^{-8}$) and two hypothetical terrigenous endmembers. Chinese loess data are shown as a potential terrigenous endmember for North Pacific sediments. ${}^3\text{He}$ concentrations are calculated for the non-carbonate fraction of sediments. Even in IDP-rich samples, sedimentary ${}^3\text{He}/{}^4\text{He}$ ratios typically do not exceed 2.4×10^{-4} . **b** Plot showing the fraction of ${}^3\text{He}$ that is

extraterrestrial for different measured ${}^3\text{He}/{}^4\text{He}$ ratios (after Farley 2000). Lines indicate terrigenous endmembers with ${}^3\text{He}/{}^4\text{He}$ ratios of 10^{-7} and 10^{-8} ; most studies assume ${}^3\text{He}/{}^4\text{He}$ ratios of $2\text{--}4 \times 10^{-8}$ for the terrigenous endmember. Ranges of measured ${}^3\text{He}/{}^4\text{He}$ ratios from selected studies are shown for reference. Equatorial Pacific: Marcantonio et al. (1995); North Pacific: Farley (1995). North Atlantic: McGee et al. (2010). Equatorial Atlantic: Farley et al. (2006). Italian Apennines: Mukhopadhyay et al. (2001a). Chinese loess: Marcantonio et al. (1998), Farley (2000)

close to atmospheric (1.4×10^{-6}). Measurements of Ne concentrations in sediments multiplied by the He/Ne ratio of air indicate that in sediments at DSDP Site 607 (Farley and Patterson 1995), LL44-GPC3 (Farley 2000), and

Gubbio in the Italian Apennines (Mukhopadhyay et al. 2001a) the percentage of air-derived ${}^3\text{He}$ is $\leq 1\%$. On the other hand, mantle He has been suggested as an important contributor (Marcantonio et al. 1998; Tolstikhin and Drubetskoy

1975) with some volcanic rocks having $^3\text{He}/^4\text{He}$ ratios on the order of 10^{-5} . Cosmogenic He produced within minerals such as olivines and pyroxene can also have high $^3\text{He}/^4\text{He}$ ratios. Based on in-vacuo crushing of mantle minerals (e.g., Kurz et al. 1996), Farley (2000) argued that most mantle He ($\sim 90\%$) would be lost by the time the volcanic material is ground down to grain sizes of a few to few 10 s of microns for transport to the ocean. Additionally, while the globally averaged cosmogenic ^3He production is similar to the flux of $^3\text{He}_{\text{ET}}$, Farley (2000) argued that cosmogenic He inventory in sediments must be low, since most terrigenous materials are actually derived from a very small percentage of the Earth's surface, and the most common terrigenous minerals (quartz, feldspars, clays) have very low He retentivity. An additional possibility for relatively high $^3\text{He}/^4\text{He}$ ratios in crustal material is that terrigenous ^3He and ^4He may be held in separate phases that respond differently to weathering, leading some deposits to have elevated $^3\text{He}/^4\text{He}$ ratios due to preferential loss of radiogenic ^4He (Tolstikhin et al. 1996).

One key uncertainty in determining the correct $^3\text{He}/^4\text{He}_{\text{TERR}}$ for the deconvolution (Eq. 1) is the possibility that samples taken to represent the terrigenous endmember contain IDPs. To the extent that IDPs retain $^3\text{He}_{\text{ET}}$ during weathering and transport, $^3\text{He}/^4\text{He}$ ratios in fan sediments and loess deposits—sediments often taken to reflect the terrigenous endmember—may be higher than the true $^3\text{He}/^4\text{He}_{\text{TERR}}$ due to the presence of IDPs. As an example, Marcantonio et al. (1998) found relatively high $^3\text{He}/^4\text{He}$ ratios in Amazon fan sediments ($\sim 1-2 \times 10^{-7}$). These rapidly accumulating sediments should have low concentrations of directly in-falling IDPs. It is not known whether the high ratios reflect terrigenous sources (possibly enriched in mantle He) (Marcantonio et al. 1998) or additions of IDPs deposited in the Amazon watershed and transported to the fan (Farley 2000). Likewise, the observation that the fine fraction of Chinese loess has higher $^3\text{He}/^4\text{He}$ ratios than bulk loess (McGee 2010) could result from the fact that IDPs would be concentrated in the fine fraction.

In cases where $^3\text{He}_{\text{ET}}$ values are sensitive to $^3\text{He}/^4\text{He}_{\text{TERR}}$ and in which $^3\text{He}/^4\text{He}_{\text{TERR}}$ is not known, three approaches have been taken to estimate $^3\text{He}/^4\text{He}_{\text{TERR}}$. The first and most basic approach is to take the lowest $^3\text{He}/^4\text{He}$ value in the dataset as an upper bound on $^3\text{He}/^4\text{He}_{\text{TERR}}$ (McGee et al. 2010). Second, as IDPs are concentrated in the magnetic fraction of sediments, the $^3\text{He}/^4\text{He}$ ratio of the nonmagnetic fraction has also been used as an upper bound on $^3\text{He}/^4\text{He}_{\text{TERR}}$ (Fourre 2004). Finally, it has been suggested that step heating of samples may be able to separate terrigenous and extraterrestrial ^3He , based upon the observation in early studies that ^4He is predominantly released at low temperatures ($\sim 400\text{ }^\circ\text{C}$), while ^3He is mostly released at $>600\text{ }^\circ\text{C}$ (Hiyagon et al. 1994; Farley 2000). In this approach, He released below a given temperature (e.g., $500\text{ }^\circ\text{C}$) is taken to be terrigenous, while the remainder of He is taken to be extraterrestrial (Farley 2000); however, recent work employing additional heating steps between 300 and $500\text{ }^\circ\text{C}$ has found evidence for extraterrestrial He loss at temperatures of $<500\text{ }^\circ\text{C}$ (see Sect. 3.2) (Mukhopadhyay and Farley 2006). These recent results suggest that the step heating approach may underestimate $^3\text{He}_{\text{ET}}$ concentrations as $15-30\%$ of $^3\text{He}_{\text{ET}}$ may be released at temperatures $<500\text{ }^\circ\text{C}$.

For studies in which the choice of $^3\text{He}/^4\text{He}_{\text{TERR}}$ is critical but $^3\text{He}/^4\text{He}_{\text{TERR}}$ is not known, both lower bound estimates ($^3\text{He}/^4\text{He}_{\text{TERR}} = 2 \times 10^{-8}$) and upper bound estimates using the approaches listed above should be presented. Such an approach would make it clear to the reader whether the assumptions with regards to the $^3\text{He}/^4\text{He}_{\text{TERR}}$ affect the overall conclusions of the work.

3.2 Carrier Phase of $^3\text{He}_{\text{ET}}$ in Terrestrial Deposits

$^3\text{He}_{\text{ET}}$ in the geological record is retained for at least 480 Ma (Patterson et al. 1998). However, the identity of the phase(s) responsible for long-term ^3He retention remains unclear, which presents a challenge in understanding how changing redox conditions or sedimentary diagenesis

affect the retention of ^3He . Based on magnetic separations and chemical dissolution experiments performed on Quaternary sediments, magnetite and a second non-magnetic phase, probably a silicate, have been suggested as carriers of $^3\text{He}_{\text{ET}}$ (Fukumoto et al. 1986; Amari and Ozima 1988; Matsuda et al. 1990). The suggestion of magnetite as one of the carriers is, however, problematic. Magnetite in IDPs is not primary but formed during atmospheric entry heating through oxidation of Fe–Ni sulfides, olivine, pyroxene, and poorly ordered silicates (Fraundorf et al. 1982; Brownlee 1985; Bradley et al. 1988). Because magnetite formation during entry heating will involve breaking chemical bonds and diffusion of oxygen, pervasive loss of ^3He from the material undergoing the chemical transformation might be expected. ^3He retention in silicates is also problematic as the dominant silicate phases in IDPs (olivines and pyroxenes) would be susceptible to diagenetic alteration on the seafloor, while the layer lattice silicates would have low retentivity for ^3He . Finally, sequential chemical leaching on recent sediments suggests that refractory phases such as diamond, graphite, SiC, and Al_2O_3 do not account for more than 10 % of the $^3\text{He}_{\text{ET}}$ (Fukumoto et al. 1986).

More recently, Mukhopadhyay and Farley (2006) carried out chemical leaching experiments of the magnetic and non-magnetic fraction of geologically old sediments (up to 90 Ma). These experiments indicate that similar amounts of ^3He are lost from the magnetic and non-magnetic fractions and rule out organic matter and refractory phases such as diamond, graphite, SiC, and Al_2O_3 as the carriers responsible for long-term retention of $^3\text{He}_{\text{ET}}$. In addition, based on IRM acquisition, Mukhopadhyay and Farley (2006) showed that the magnetic moment of the magnetic and non-magnetic fraction is similar. The result suggests that the magnetite separation technique they and all previous workers used results in approximately half of the magnetite still residing in the non-magnetic fraction. The chemical leaching and the magnetic properties of the magnetic and non-magnetic fraction of the sediments combined suggest a single carrier that

might be Fe–Ni metal, Fe–Ni sulfides, or possibly magnetite but more likely a phase that is associated with magnetite. In IDPs collected from the stratosphere olivine and pyroxene grains are frequently rimmed by magnetite a few to a few tens of nm thick that forms during atmospheric entry heating (Bradley et al. 1988). Because magnetite is stable on the ocean floor for tens of millions of years, the magnetite rims may armor the olivines, pyroxenes or poorly ordered silicates against chemical alteration and may also explain the association of the ^3He carrier(s) with magnetite.

The presence of a single carrier phase is also strongly supported by step heating experiments (Fig. 3) on the magnetic and non-magnetic fraction of the sediments that demonstrate that the ^3He release patterns in the two sediment fractions are identical (Mukhopadhyay and Farley 2006). The step heating experiments indicate two release peaks, one at low temperature (350–400 °C) and one at higher temperature (600–750 °C). Previous work (Amari and Ozima 1985, 1988; Hiyagon 1994) had not noticed the low temperature peak as the step heating experiments started at temperatures in excess of 500 °C. While Farley (2000) had observed a low temperature release peak from step heating experiments in sediments from Site 607B, the $^3\text{He}/^4\text{He}$ ratio for the low temperature release peak was 0.1 R_A and hence, the release was associated with crustal He. Additionally, the new step heating experiments (Mukhopadhyay and Farley 2006) indicate a higher retentivity for ^3He over geologic time. For example, extrapolation diffusivities obtained from high-temperature step heating experiments (e.g., Amari and Ozima 1985) to seafloor indicates that greater than 99 % of the extraterrestrial ^3He is expected to be lost in 50 Ma, which is at odds with the pelagic clay ^3He record from the Central Pacific GPC3 core (Farley 1995). The new step heating experiments indicate that about 20 % of the ^3He will be lost through diffusion at seafloor temperatures after 50 Ma, while sedimentary rocks exposed on the Earth's surface for the same amount of time would lose up to 60 % (Mukhopadhyay and Farley 2006). Additionally, if temperatures exceed 70 °C during sediment diagenesis

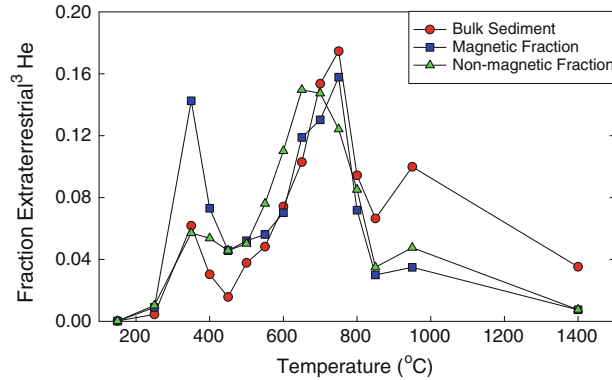


Fig. 3 Extraterrestrial ^3He release patterns for bulk, magnetic and non-magnetic fractions of sediments from step heating experiments (Adapted from Mukhopadhyay and Farley 2006). Bulk sediments are from LL44-GPC3 and DSDP Site 596B, while the magnetic and non-magnetic fractions are from LL44-GPC3 sediments.

Since multiple experiments for bulk, magnetic and non-magnetic fractions were carried out by Mukhopadhyay and Farley (2006), only the average release pattern for the three sediment types has been shown. Note the very strong similarity in the release patterns for all three sediment types

extensive ^3He loss is expected to occur (>90 %) after only a few hundred ka. Thus, care must be taken to compare the extraterrestrial ^3He record from different sites and tectonic environments.

3.3 Atmospheric Entry Heating and Grain Size Distribution of $^3\text{He}_{\text{ET}}$ -Bearing IDPs in Terrestrial Deposits

Atmospheric entry heating modifies the extraterrestrial noble gas content of incoming IDPs. Heating during entry causes thermal decomposition of phyllosilicates within IDPs as well as diffusive loss and bubble rupture, all of which contribute to He losses (Stuart et al. 1999). Maximum entry temperatures are proportional to mass and entry velocity; as a result, larger IDPs and IDPs derived from comets (which tend to have higher geocentric velocities than asteroid-derived IDPs) are more likely to be degassed before reaching Earth's surface (Flynn 1989; Love and Brownlee 1991). Hence, ^3He in sediments will not reflect the relative abundance of cometary versus asteroid IDPs that enter the Earth's atmosphere but will rather be biased towards the accretion of asteroidal IDPs.

Farley et al. (1997) quantitatively modeled the grain size distribution of IDPs capable of retaining

$^3\text{He}_{\text{ET}}$ after atmospheric entry. Based upon the assumption that IDPs have implanted $^3\text{He}_{\text{ET}}$ to depths of few hundred nanometers—i.e., ^3He is surface area-correlated rather than volume-correlated—and that $^3\text{He}_{\text{ET}}$ is lost in IDPs heated to more than ~ 600 °C, Farley et al. (1997) predicted that $^3\text{He}_{\text{ET}}$ in terrestrial sediments should primarily (>70 %) be contained within IDPs between 3 and 35 μm in diameter. If $^3\text{He}_{\text{ET}}$ is instead assumed to be volume-correlated, a much broader grain size distribution results, with >70 % of $^3\text{He}_{\text{ET}}$ contained with grains ranging from ~ 5 to 150 μm (Farley et al. 1997). Overall, Farley et al. (1997) find that as a result of atmospheric entry heating, $^3\text{He}_{\text{ET}}$ -bearing IDPs represent only ~ 0.5 % of the total IDP mass flux to Earth of $40 \pm 20 \times 10^6$ g/a (Love and Brownlee 1993) and ~ 4 % of the total IDP surface area flux.

Since the entry-heating model predicts the size distribution of ^3He -bearing particles in sediment (particles not heated to >600 °C), for a given IDP flux and sediment accumulation rate, one can calculate the minimum volume of sediment required to statistically sample the He-bearing IDPs so as not to underestimate the ^3He flux. Additionally, predictions can be made of the reproducibility of ^3He measurements in sediment aliquots of different sizes (i.e., different area-time products) depending upon whether ^3He is surface area-correlated

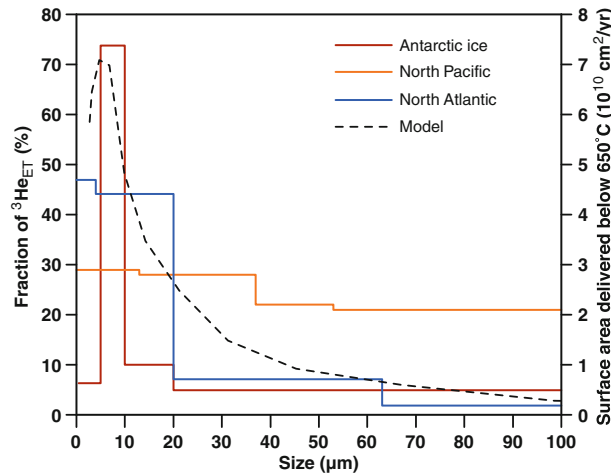


Fig. 4 Grain size distribution of ${}^3\text{He}_{\text{ET}}$ in terrestrial deposits and a model of surface area delivery of He-retentive IDPs. Data (*solid lines*) reflects the percent of the total ${}^3\text{He}_{\text{ET}}$ in a given grain size fraction. Data from Holocene Antarctic ice (Brook et al. 2009) and 33 Ma North Pacific sediments (Mukhopadhyay and Farley

2006) represent one sample each, while data from the North Atlantic represent the average of six samples from the last glacial period and Holocene (McGee et al. 2010). Model results (*dashed line*) reflect the total IDP surface area delivered by different particle sizes heated to less than 650 °C during atmospheric entry (Farley et al. 1997)

or volume-correlated (Farley et al. 1997). The reproducibility of replicate samples matches predictions from model results if ${}^3\text{He}_{\text{ET}}$ is a surface-area correlated component rather than a volume-correlated component (Farley et al. 1997; Patterson and Farley 1998; Mukhopadhyay et al. 2001a). This suggests that ${}^3\text{He}_{\text{ET}}$ is primarily contained in a large number of fine grains rather than a small number of large grains. Studies of grain size fractions of deep-sea sediments and ice core particulates also support a correlation of ${}^3\text{He}_{\text{ET}}$ content with IDP surface area, finding ~60–90 % of the total ${}^3\text{He}_{\text{ET}}$ within size fractions <35 μm (Fig. 4) (Mukhopadhyay and Farley 2006; Brook et al. 2009; McGee et al. 2010).

Though all three studies find that ${}^3\text{He}_{\text{ET}}$ is primarily contained in grains <35 μm , each provides a slightly different estimate of the grain size distribution of ${}^3\text{He}_{\text{ET}}$ in terrestrial deposits. Data from a sample of Holocene Antarctic ice indicate a sharp peak in ${}^3\text{He}_{\text{ET}}$ -bearing grains between 5 and 10 μm (Brook et al. 2009); analyses of six samples of Late Quaternary North Atlantic drift sediments indicate a broader peak between 0 and 20 μm (McGee et al. 2010); and data from a sample of 33 Ma North Pacific red clay suggest a broader distribution, with roughly equal ${}^3\text{He}_{\text{ET}}$

inventories in the 0–13 and 13–37 μm fractions and only slightly lower ${}^3\text{He}_{\text{ET}}$ inventories in the 37–53 μm and >53 μm fractions (Mukhopadhyay and Farley 2006). The differences in these studies may have to do with differences in the time-area product of the samples used (samples with higher time-area products, such as red clays, are more likely to sample rare large IDPs) or with the different methods used for grain size separation. Brook et al. (2009) passed ice melt through filters to separate grain size fractions, while McGee et al. (2010) used settling after adding a chemical dispersant to reduce flocculation of fine grains, and Mukhopadhyay and Farley (2006) used settling without dispersant addition. Settling, and particularly settling without dispersants, is unlikely to quantitatively remove fine grains from coarser size fractions, potentially leading to an overestimation of ${}^3\text{He}_{\text{ET}}$ inventories in coarser fractions.

Some large (50–400 μm) IDPs from Antarctic ice appear to have retained ${}^3\text{He}_{\text{ET}}$ of both solar and spallogenic origin (Fig. 1) (Stuart et al. 1999), but the grain size and reproducibility measurements cited above suggest that these rare large He-retentive particles do not contribute substantially to the total ${}^3\text{He}_{\text{ET}}$ flux. Finally,

we note that Lal and Jull (2005) proposed that instead of ${}^3\text{He}$ in IDPs, spallogenic He within fragments released by meteorites during atmospheric entry is a dominant source of ${}^3\text{He}_{\text{ET}}$ in sediments. The hypothesis of Lal and Jull (2005) makes two predictions: (1) approximately 50 % of the ${}^3\text{He}$ is in sediments coarser than 50 μm and (2) high ${}^3\text{He}/{}^4\text{He}$ ratios in sediments should be associated with spallogenic Ne. As noted above, the grain size distribution data and reproducibility of ${}^3\text{He}_{\text{ET}}$ measurements do not support 50 % of ${}^3\text{He}_{\text{ET}}$ being in grain sizes larger than 50 μm . Further, high ${}^3\text{He}/{}^4\text{He}$ ratios of $2\text{--}3 \times 10^{-4}$ in sediments have been found to be associated with solar wind Ne and not spallogenic Ne (Fukumoto et al. 1986; Matsuda et al. 1990). Hence, data do not support the hypothesis that fragmentation of meteorites in the atmosphere is the dominant contributor to ${}^3\text{He}$; they are instead best explained by ${}^3\text{He}_{\text{ET}}$ in marine sediments and terrestrial ice being a surface-area correlated component in IDPs.

4 Extraterrestrial He in the Geologic Record

4.1 Extraterrestrial He as a Tracer of Past Variations in IDP Flux

As a tracer of the IDP flux, ${}^3\text{He}_{\text{ET}}$ offers a view into past accretion rates of extraterrestrial matter that is quite different from that provided by iridium and other PGEs, which trace the total extraterrestrial mass flux. As demonstrated below, large impacts that leave PGE anomalies may or may not be accompanied by elevated IDP fluxes (Farley et al. 1998; Mukhopadhyay et al. 2001a, b), and elevated IDP fluxes are not always accompanied by large impacts (Farley et al. 2006).

${}^3\text{He}_{\text{ET}}$ fluxes ($f_{\text{He}_{\text{ET}}}$) are calculated by multiplying the measured ${}^3\text{He}_{\text{ET}}$ concentration by the sedimentary mass accumulation rate (MAR):

$$f_{\text{He}_{\text{ET}}} = ([{}^3\text{He}_{\text{ET}}] \cdot \text{MAR})/R \quad (2)$$

where $[{}^3\text{He}_{\text{ET}}]$ is the ${}^3\text{He}_{\text{ET}}$ concentration in the sediment (see Eq. 1), MAR is the sedimentary

mass accumulation rate and R is the fractional retentivity of ${}^3\text{He}$ (Farley 1995). Though assumptions of constant inputs of hydrogenous cobalt have occasionally been used for MAR calculation (Farley 1995), MARs are usually derived from age models:

$$\text{MAR} = \frac{\rho \cdot \Delta z}{\Delta t} \quad (3)$$

where ρ is the dry bulk density, Δz is a depth interval, and Δt is the time associated with that depth interval. Age models are typically determined from geologic epochs, magnetic chrons or astronomical tuning (e.g., Farley 1995; Farley et al. 1998, 2006; Mukhopadhyay et al. 2001a) and are thus subject to errors in the ages of epoch or chron boundaries or in the identification of astronomical cycles. Age model-based MARs also do not account for sedimentary inputs by lateral advection or slumping. As a result of potential age model errors and spurious ${}^3\text{He}_{\text{ET}}$ flux changes caused by changes in lateral advection of sediments, some studies have sought to replicate ${}^3\text{He}_{\text{ET}}$ flux excursions in multiple cores or sections (e.g., Farley et al. 1998, 2006). Finally, ${}^3\text{He}_{\text{ET}}$ retentivity in the sedimentary record over geological time is not well quantified, and thus the value of R is not known. As a result, the absolute ${}^3\text{He}_{\text{ET}}$ flux cannot be determined. Rather, one always calculates the product $f_{\text{He}_{\text{ET}}} \cdot R$ and the product is termed the implied ${}^3\text{He}_{\text{ET}}$ flux (e.g., Farley et al. 1998; Mukhopadhyay et al. 2001a). We note that ${}^3\text{He}_{\text{ET}}$ loss by diffusion is expected to increase with age; i.e., R should decrease monotonically with age. However, because ${}^3\text{He}_{\text{ET}}$ is retained in the geological record for at least 480 Ma (Patterson et al. 1998), assuming an invariant R in a given sedimentary setting is probably valid over the geologically short (\sim a few to a few tens of Ma) timescales over which IDP flux variations would be expected to occur (Farley et al. 1998, 2006). Thus, relative variations in ${}^3\text{He}_{\text{ET}}$ fluxes in the sedimentary record over million-year durations are robust even though the absolute values of the fluxes may not be well defined.

The discovery of ${}^3\text{He}_{\text{ET}}$ in 480 Ma Ordovician limestones (Patterson et al. 1998) suggests that

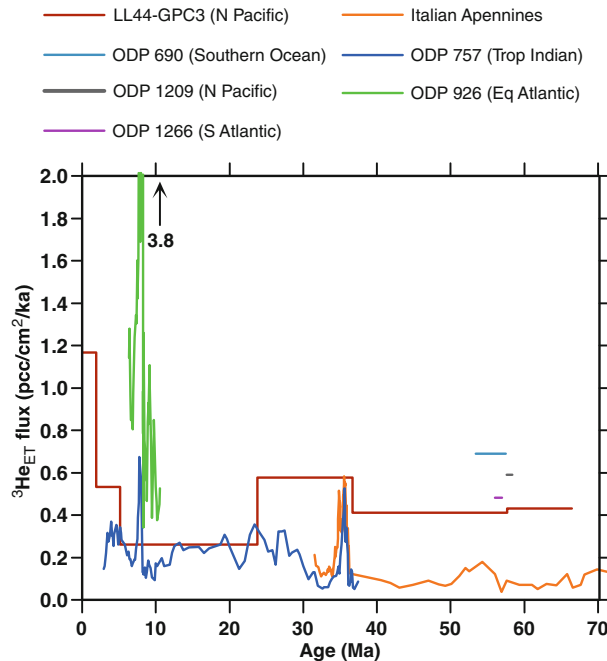


Fig. 5 Records of $^3\text{He}_{\text{ET}}$ flux from the late Cretaceous to the Pleistocene. Records from LL44-GPC3, ODP 690, ODP 1209 and ODP 1266 represent the average $^3\text{He}_{\text{ET}}$ fluxes across dated intervals. Records from ODP 757 and ODP 926 reflect the 3-point moving average of “instantaneous” $^3\text{He}_{\text{ET}}$ fluxes (i.e., the product of a sample’s $^3\text{He}_{\text{ET}}$ concentration and MAR derived from the age model). Data from the Italian Apennines reflect the authors’ best estimate based on chron-averaged $^3\text{He}_{\text{ET}}$ fluxes, $^3\text{He}_{\text{ET}}$

concentrations normalized to the non-carbonate fraction, and $^3\text{He}/^4\text{He}$ ratios. The short-lived flux increases at 35 and 8 Ma are shown in greater detail in Fig. 6. Differences in absolute fluxes between records are likely to reflect differences in $^3\text{He}_{\text{ET}}$ preservation or sediment focusing. LL44-GPC3: Farley (1995). ODP 690: Farley and Eltgroth (2003). ODP 1209: Marcantonio et al. (2009). ODP 1266: Murphy et al. (2010). Italian Apennines: Mukhopadhyay et al. (2001a). ODP 757 and ODP 926: Farley et al. (2006)

IDP accretion rates could be reconstructed over most of the Phanerozoic. The implied accretion rate of extraterrestrial ^3He in the Late Ordovician is $\sim 0.5 \pm 0.2$ pcc STP/cm²/ka, which is similar to the average flux over the Cenozoic. The Ordovician sediments further demonstrate that $^3\text{He}_{\text{ET}}$ in sediments is largely carried by IDPs, as co-existing meteorites in the sedimentary section have He concentrations that are similar to or only slightly higher than the ^3He concentration of the host limestones.

At the Permo-Triassic boundary (P/Tr) sections at Meishan, China and Sasayama, Japan, $^3\text{He}_{\text{ET}}$ has been reported, although not in IDPs but rather in fullerenes delivered through a bolide impact (Becker et al. 2001). However, Farley and Mukhopadhyay’s (2001) measurements of ^3He from the Meishan section did not detect $^3\text{He}_{\text{ET}}$ at or near the vicinity of the P/Tr boundary and thus, they found no evidence for fullerene-hosted

$^3\text{He}_{\text{ET}}$. Additional work by Farley et al. (2005) from the Opal Creek P/Tr section in Canada also found no evidence of extraterrestrial ^3He , and hence, fullerene-hosted $^3\text{He}_{\text{ET}}$. As a result, whether or not fullerene-hosted $^3\text{He}_{\text{ET}}$ is present at the P/Tr boundary remains an open question.

In spite of $^3\text{He}_{\text{ET}}$ being retained for the past 480 Ma, ^3He fluxes are relatively well characterized only for the Cenozoic. The Cenozoic history of $^3\text{He}_{\text{ET}}$ fluxes was first investigated by Farley (1995) in core LL44-GPC3 from the central North Pacific. The study found roughly constant $^3\text{He}_{\text{ET}}$ fluxes averaged over epochs from the Late Cretaceous to the Pliocene ($\sim 0.5 \pm 0.2$ pcc STP/cm²/ka), with highest fluxes in the Oligocene and lowest fluxes in the Miocene (Fig. 5). $^3\text{He}_{\text{ET}}$ fluxes were also calculated by normalizing to Co concentrations in the sediment, based on the assumption that hydrogenous Co accumulates at a constant rate (Kyte et al. 1993).

Co-based fluxes of ${}^3\text{He}_{\text{ET}}$ can be calculated at much higher resolution than epoch-averaged fluxes and show high short-term variability, including peaks just before the K/Pg boundary and during the early Eocene and latest Eocene/early Oligocene. Of these, the first is related to Co-based accumulation rates and is not reflected in ${}^3\text{He}_{\text{ET}}$ concentrations, while the latter two are marked by increased ${}^3\text{He}_{\text{ET}}$ concentrations in the sediments. One of the most dramatic features of the GPC3 record is a factor of 2 increase in the ${}^3\text{He}_{\text{ET}}$ flux from the Pliocene to the Quaternary. This change could reflect (1) a change in IDP flux, (2) diffusional loss of ${}^3\text{He}_{\text{ET}}$, (3) an age model error that places the Quaternary/Pliocene boundary too high in the core, or (4) increased lateral advection of sediments to the site during the Quaternary. The Quaternary/Pliocene change in ${}^3\text{He}_{\text{ET}}$ flux has not been studied at other sites and offers an important avenue for future work.

Subsequent work investigated the Cretaceous-Eocene portion of the record in greater detail in pelagic limestones exposed in the Umbria-Marche basin of the Italian Apennines (Farley et al. 1998; Mukhopadhyay et al. 2001a). The mean ${}^3\text{He}_{\text{ET}}$ flux in the Apennine sections is a factor of 3–5 lower than the mean flux over the same period in the North Pacific core studied by Farley (1995), suggesting either ${}^3\text{He}_{\text{ET}}$ loss during the burial and uplift of this section (Mukhopadhyay and Farley 2006) or systematically higher lateral advection of sediments at the North Pacific site. Mukhopadhyay et al. (2001a) found approximately constant ${}^3\text{He}_{\text{ET}}$ fluxes from the late Cretaceous through the early Eocene in pelagic limestones in the Italian Apennines. Possible departures from a constant flux occur near the Paleocene/Eocene boundary, with a 2–4-fold increase in ${}^3\text{He}_{\text{ET}}$ flux, and during the early Eocene, when a gradual decrease in flux is indicated; however, there are concerns about tectonic disturbances and slumping in this section (Mukhopadhyay et al. 2001a). Significantly, there is no evidence for an increase in ${}^3\text{He}_{\text{ET}}$ flux associated with the K/Pg boundary in either the Apennines or an expanded K/Pg section in Morocco. A single asteroid or comet impact is not accompanied by increased

accretion rate of IDPs, while a shower of comets generated by perturbation of the Oort cloud leads to an increased terrestrial IDP accretion rate associated with multiple impacts (Farley et al. 1998). Hence, the K/Pg ${}^3\text{He}$ results suggest that the impactor was not part of a comet shower but rather an asteroid or a lone comet (Mukhopadhyay et al. 2001a, b).

In the latest Eocene, ${}^3\text{He}_{\text{ET}}$ fluxes reconstructed from limestones in the Italian Apennines and from an Indian Ocean sediment core document a factor of ~ 5.5 increase in the IDP accretion rate, with peaks at ~ 36 and 35 Ma (Farley et al. 1998, 2006). The peaks are accompanied by spikes in Ir concentration in the Apennine section and roughly correspond to the ages of the Chesapeake Bay and Popigai impact structures and tektite layers found in sediments around the world (Fig. 6). The implied increase in IDP flux begins 0.7 Ma before the first Ir spike and gradually decays for almost 1 Ma after the second spike. Two potential mechanisms can account for the simultaneous increase in IDP accretion rates and the delivery of large impactors: a comet shower associated with a perturbation of the Oort cloud or a large collision in the asteroid belt. Farley et al. (1998) preferred the comet shower hypothesis because (1) the observed variations in the dust flux matched Hut et al.'s (1987) predictions of dust enhancement in the inner solar system associated with a comet shower generated by perturbation of the Oort cloud and (2) comet showers would necessarily lead to an increase in both dust and large impactors in the inner solar system. While large collisions in the asteroid belt can enhance dust accretion rates to the Earth, simultaneous delivery of large impactors and dust is not predicted a priori, unless the collision occurs in the close vicinity of one of the several secular and mean-motion resonances in the asteroid belt from which large objects can be ejected on Earth-crossing orbits on timescales of <1 Ma (e.g., Gladman et al. 1997). Furthermore, it is not clear whether the enhancement in dust accretion rate from asteroid belt collisions would match the observed pattern seen in the late Eocene (Farley et al. 1998). Recent work, however, has suggested an

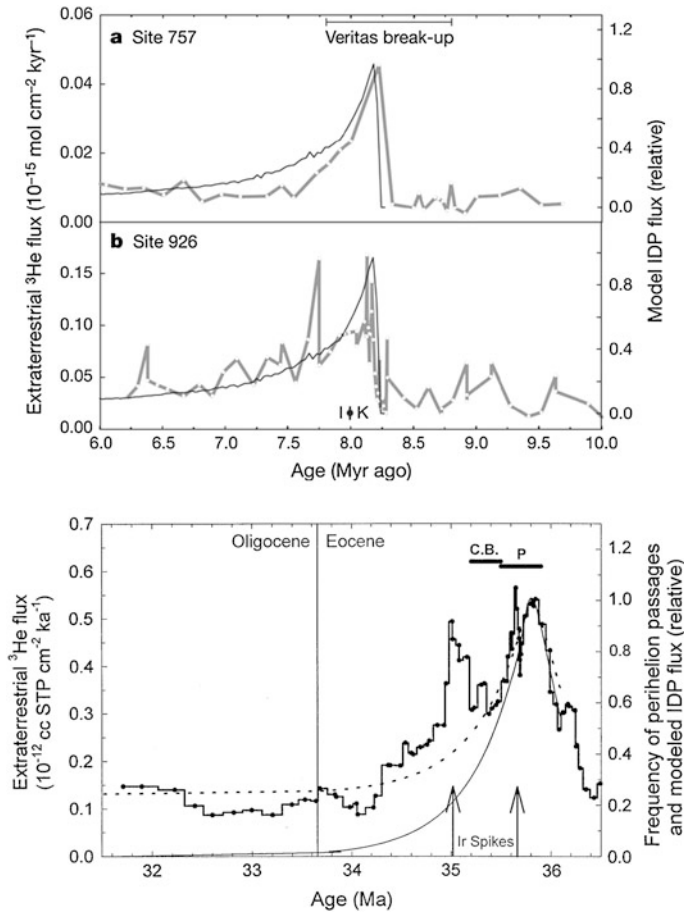


Fig. 6 Insights into past solar system events from variations in the $^3\text{He}_{\text{ET}}$ flux. *Top*—Comparison of late Miocene $^3\text{He}_{\text{ET}}$ fluxes at two sites with flux changes predicted from a model of IDP creation and transport after the Veritas asteroid break-up event (Farley et al. 2006). An independent estimate for the age of this event is shown at the top of the figure. I/K denotes the transition from kaolinite to illite as the dominant clay mineral at Site 926. *Bottom*—A Late Eocene increase in

$^3\text{He}_{\text{ET}}$ flux measured in samples from the Italian Apennines (Farley et al. 1998). Independent ages of increases in Ir concentration in marine sediments (a marker of extraterrestrial impacts) and of the Chesapeake Bay and Popagai impact structures are shown. The solid line indicates the modeled increase in IDP flux associated with a comet shower initiated by a perturbation of the Oort cloud; the dashed line is the same result shifted up to reflect pre- and post-event baseline IDP fluxes

L-chondrite composition for the Popigai impactor (Tagle and Claeys 2004) suggesting that the increase in IDP flux may have an asteroidal origin.

$^3\text{He}_{\text{ET}}$ flux again increased transiently in the late Miocene (8.2 Ma) (Fig. 6). The $^3\text{He}_{\text{ET}}$ flux pattern recorded in sediment cores from both the Indian and Atlantic oceans is broadly similar to that of the late Eocene (Farley et al. 2006); in detail, however, the increase in $^3\text{He}_{\text{ET}}$ during the Miocene occurs more abruptly than in the

Eocene event, rising a factor of ~ 4 in <100 ka, followed by a ~ 1.5 Ma decay to pre-event levels. Unlike the Eocene event, there are no known large impacts of this age. Farley et al. (2006) provided evidence that the event reflects the breakup of a parent asteroid with a diameter greater than 100 km to form the Veritas family of asteroid fragments, an event that has been independently dated to 8.3 ± 0.5 Ma (Nesvorný et al. 2003). The Veritas family currently orbits at a distance of 3.17 AU and continues to be a

major source of IDPs to the zodiacal cloud (Nesvorný et al. 2006). The breakup event is likely to have formed abundant IDPs, but it is unlikely to have sent any large fragments into Earth-impacting orbits (Farley et al. 2006). Though the link between the Veritas breakup and the $^3\text{He}_{\text{ET}}$ flux increase is compelling, remaining questions include (1) why models indicate a more prolonged decay of IDP flux following the event than is observed and (2) whether a similar peak in $^3\text{He}_{\text{ET}}$ flux is associated with the Karin asteroid breakup event dated to ~ 5.8 Ma (Nesvorný et al. 2006).

Several studies have investigated the ^3He -based IDP accretion rate during the Quaternary, as cyclic variations in IDP accretion rate were suggested as a driver of the 100 ka glacial cycle (Muller and Macdonald 1995). Modeling of the terrestrial accretion rate of asteroid dust predicts 100 ka variations in IDP accretion rate of up to a factor of 2, associated with changes in Earth's orbital inclination (Kortenkamp and Dermott 1998a). $^3\text{He}_{\text{ET}}$ fluxes from the Atlantic and Pacific oceans over the last 2 Ma based on MARs derived from benthic $\delta^{18}\text{O}$ data do suggest a 100 ka periodicity in $^3\text{He}_{\text{ET}}$ fluxes over the past 700 ka (Farley and Patterson 1995; Patterson and Farley 1998). Peak fluxes occur during interglacial periods with variations in amplitude ranging from a factor of ~ 1.5 to ~ 3.5 between cycles and between cores. Surprisingly, however, the peaks in ^3He fluxes are $\sim 180^\circ$ (50 ka) out of phase with the predicted accretion rate of IDPs based on orbital variations (Kortenkamp and Dermott 1998a). Hence, either the observed peaks in $^3\text{He}_{\text{ET}}$ fluxes are not a consequence of varying IDP accretion rate from space, or the models that predict IDP accretion rates based on variations in Earth's orbital parameters (Kortenkamp and Dermott 1998a) may not be fully capturing all of the accretion processes. For example, Dermott et al. (2001) suggested that accretion of asteroidal dust from the resonant ring of dust around the Earth may be a possible mechanism for inducing a 50 ka lag in the IDP accretion rate. This hypothesis has yet to be verified.

The apparent variations in $^3\text{He}_{\text{ET}}$ flux during the late Quaternary have also been called into question based on observations that the ratio of $^3\text{He}_{\text{ET}}$ to scavenged ^{230}Th —a tracer of sedimentary flux largely independent of age model errors and lateral advection of sediments—remains constant within $\pm 40\%$ (one standard deviation of the mean) through glacial-interglacial cycles in equatorial Pacific sediments, suggesting relatively constant $^3\text{He}_{\text{ET}}$ flux (Marcantonio et al. 1995, 1996, 1999, 2001b; Higgins et al. 2002) (see Sect. 4.2 for additional discussion). Further, Winckler et al. (2004) found that age model-based $^3\text{He}_{\text{ET}}$ fluxes vary with a 41 ka periodicity in the early Quaternary; this period matches that of contemporaneous glacial-interglacial variability but does not match that of changes in orbital inclination. Winckler and Fischer (2006) also found no changes in $^3\text{He}_{\text{ET}}$ fluxes in Antarctic ice from the last glacial period to the Holocene. Together, these results suggest that the observed 100 ka cycles in late Quaternary $^3\text{He}_{\text{ET}}$ accumulation in marine sediments are a consequence rather than a cause of climate variability, perhaps related to glacial-interglacial changes in sediment focusing or systematic age model errors caused by glacial-interglacial changes in carbonate dissolution (Marcantonio et al. 1996, 2001b; Higgins et al. 2002; Winckler et al. 2004). Variations in extraterrestrial ^3He flux of smaller magnitude ($\pm 40\%$), however, cannot yet be ruled out.

The variations in the ^3He -based IDP accretion history discussed above have provided important observational evidence for hypothesized solar system events, such as the possible comet shower in the Late Eocene (Farley et al. 1998), the collision disruption of an asteroid to create the Veritas family 8.3 at Ma (Farley et al. 2006), and the nature of the impactor at the K/Pg boundary (single impactor vs. comet shower). Furthermore, the ^3He -based accretion history over the past 70 Ma strongly refutes the speculative hypothesis that mass extinctions in the geological record are driven by quasi-periodic comet showers (Hut et al. 1987). While a comet shower may have occurred in the Late Eocene,

not a single extinction event over the past 70 Ma appears to be associated with comet showers, and one of the largest mass extinction events of the Phanerozoic—the K/Pg boundary event—is clearly not associated with a comet shower (Mukhopadhyay et al. 2001a, b).

4.2 Use of Extraterrestrial ^3He to Calculate Sedimentary Accumulation Rates

When the $^3\text{He}_{\text{ET}}$ concentration in a sedimentary section varies, it implies either a variation in the flux from space or changing MAR. If the flux from space is taken to be constant, then Eq. 2 can be inverted to solve for the sediment MAR. A constant $^3\text{He}_{\text{ET}}$ flux from space can often be inferred when $^3\text{He}_{\text{ET}}$ concentrations co-vary with concentrations of a terrigenous sedimentary component (e.g., terrestrial ^4He , Fe, or the non-carbonate fraction) or with concentrations of ^{230}Th scavenged from the water column. $^3\text{He}_{\text{ET}}$ -based MARs differ in several important respects from MARs derived from age models. First, $^3\text{He}_{\text{ET}}$ -based MARs can be calculated at each sample depth for which $^3\text{He}_{\text{ET}}$ measurements are available, while MARs based on age models are calculated as average values between age model tie points. Second, over the last 400 ka (when $^3\text{He}_{\text{ET}}$ fluxes have been determined by ^{230}Th -normalization, as described below), absolute $^3\text{He}_{\text{ET}}$ -based MARs are independent of sedimentary age models. Finally, if $^3\text{He}_{\text{ET}}$ -bearing IDPs are transported with laterally advected sediments, sediment focusing or winnowing will not substantially change the measured $^3\text{He}_{\text{ET}}$ concentration at a site; $^3\text{He}_{\text{ET}}$ -based MARs will then “see through” sediment focusing and provide estimates of the vertical rain rate of sediment at a site (e.g., Marcantonio et al. 2001b; McGee et al. 2010). In periods prior to 400 ka, when $^3\text{He}_{\text{ET}}$ fluxes must be calculated from sedimentary age models, the $^3\text{He}_{\text{ET}}$ flux estimates may be biased by systematic focusing or winnowing in the sediment, affecting the absolute value of $^3\text{He}_{\text{ET}}$ -based MARs but not the relative changes in MARs.

A critical first step in constructing ^3He -based MARs is testing whether $^3\text{He}_{\text{ET}}$ fluxes are in fact constant over a given period. In late Quaternary marine sediments, $^3\text{He}_{\text{ET}}$ fluxes have been calculated using ^{230}Th -normalization (Marcantonio et al. 1995, 1996; Higgins et al. 2002). Briefly, ^{230}Th -normalization relies on the approximation that ^{230}Th produced in the water column from the decay of dissolved ^{234}U ($^{230}\text{Th}_{\text{xs}}$) is scavenged by sinking particles more quickly than it can be laterally mixed as a dissolved species. The flux of $^{230}\text{Th}_{\text{xs}}$ to the seafloor is then taken to be equal to the (known) production rate of ^{230}Th in the water column, and MARs can be calculated as the ratio of the ^{230}Th production rate to the decay-corrected $^{230}\text{Th}_{\text{xs}}$ concentration in the sediment (for a review of ^{230}Th normalization, see Francois et al. 2004). ^{230}Th -based MARs have the same properties as $^3\text{He}_{\text{ET}}$ -based MARs mentioned above: namely, they can be computed at high resolution; they are largely independent of age model errors; and they should be only minimally affected by lateral advection of sediments. ^{230}Th -normalization can only be used in sediments from the last ~ 500 ka due to the radioactive decay of ^{230}Th (half-life 75.7 ka).

Marcantonio et al. (1995, 1996, 2001b) and Higgins et al. (2002) found $^3\text{He}_{\text{ET}}/^{230}\text{Th}_{\text{xs}}$ ratios in sediments to be constant to within $\pm 40\%$ throughout the equatorial Pacific Ocean over the last 200 ka with no systematic glacial-interglacial variability, supporting a near-constant flux of $^3\text{He}_{\text{ET}}$ during this time period. When $^{230}\text{Th}_{\text{xs}}$ measurements are used to calculate MARs, these results indicate an average $^3\text{He}_{\text{ET}}$ accumulation rate of 0.8 ± 0.3 pcc STP/cm²/ka in the late Quaternary (Fig. 7; all flux estimates are given as mean \pm one standard deviation of the mean). Marcantonio et al. (1999) determined a similar $^{230}\text{Th}_{\text{xs}}$ -based $^3\text{He}_{\text{ET}}$ flux in an eastern equatorial Indian Ocean core for the past 200 ka (1.1 ± 0.4 pcc STP/cm²/ka). In a core from the Arabian Sea, $^{230}\text{Th}_{\text{xs}}$ -based $^3\text{He}_{\text{ET}}$ fluxes are 0.4 ± 0.3 pcc STP/cm²/ka over the past 23 ka, lower than observed at other sites, for reasons that are not clear (see discussion below) (Marcantonio et al. 2001a).

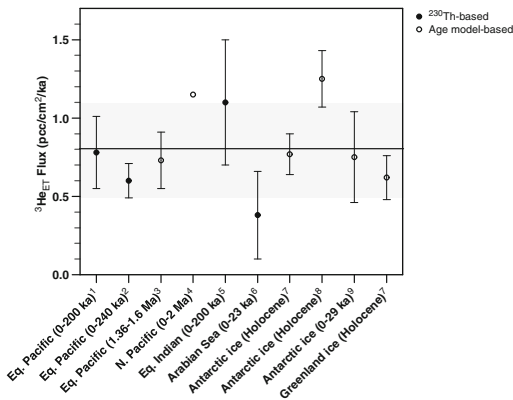


Fig. 7 Estimates of the Quaternary $^3\text{He}_{\text{ET}}$ flux. Fluxes are estimated using both sedimentary and ice core age models (*open circles*) and using ^{230}Th -normalization (*closed circles*). 1-sigma confidence intervals are shown. Most estimates are consistent with a flux of 0.8 ± 0.3 pcc STP $^3\text{He}_{\text{ET}}/\text{cm}^2/\text{ka}$ (indicated by the *line and shaded box*). The results do not suggest a latitudinal dependence of $^3\text{He}_{\text{ET}}$ flux. Data sources: Marcantonio et al. (1995), Higgins et al. (2002), Winckler et al. (2004), ⁴Farley (1995), Marcantonio et al. (1999), Marcantonio et al. (2001a), Brook et al. (2000), Brook et al. (2009, Winckler and Fischer (2006)

Independent corroboration of $^3\text{He}_{\text{ET}}$ flux estimates from marine sediments comes from measurements of $^3\text{He}_{\text{ET}}$ in ice cores, which find $^3\text{He}_{\text{ET}}$ accumulation rates ranging from 0.62 ± 0.14 to 1.25 ± 0.18 pcc STP/cm²/ka in four sets of samples of ice from Greenland and Antarctica over the last 30 ka (Brook et al. 2000, 2009; Winckler and Fischer 2006). Similar fluxes have also been found using accumulation rates inferred from an age model in Quaternary samples from the North Pacific core studied by Farley (1995) (~ 1.2 pcc STP/cm²/ka) and in equatorial Pacific sediments from the early Quaternary between 1.36 and 1.6 Ma (0.73 ± 0.18 pcc STP/cm²/ka) (Winckler et al. 2004). Taken together, these results suggest approximately constant ($\pm 40\%$) $^3\text{He}_{\text{ET}}$ fluxes over the past 1.6 Ma and minimal latitudinal variations in $^3\text{He}_{\text{ET}}$ flux, providing a basis for using $^3\text{He}_{\text{ET}}$ for calculating MARs over this time period. Importantly, these studies establish $^3\text{He}_{\text{ET}}$ as the only constant flux proxy available for paleoflux studies in sediments prior to 500 ka.

$^3\text{He}_{\text{ET}}$ can also be used as a constant flux proxy for MAR calculations in studies ranging back to at least the K/Pg boundary, if $^3\text{He}_{\text{ET}}$ fluxes from space and the fraction of original $^3\text{He}_{\text{ET}}$ lost to diffusion are roughly constant over the timescale of interest. The first assumption does not hold during the late Eocene and late Miocene (see Sect. 4.1), and the second may not be appropriate in transitions between oxic and anoxic sediments, but in much of the late Cretaceous and Cenozoic they appear reasonable. In the carbonate-rich sections that have been studied, $^3\text{He}_{\text{ET}}$ concentrations are relatively constant with respect to the non-carbonate fraction, a rough indicator of relative sedimentation rates, supporting an approximately constant $^3\text{He}_{\text{ET}}$ flux (e.g., Mukhopadhyay et al. 2001a, b; Farley and Eltgroth 2003; Marcantonio et al. 2009).

$^3\text{He}_{\text{ET}}$ fluxes for these earlier periods are typically determined in portions of a given sedimentary section in which the age model is well constrained (usually by orbital tuning or magnetostratigraphy) and that either are adjacent to or span the period of interest. These $^3\text{He}_{\text{ET}}$ fluxes are then applied to the interval of interest, which is often a time of substantial changes in sedimentation when orbital signals are in doubt. Absolute $^3\text{He}_{\text{ET}}$ fluxes for a given time period vary from site to site due to differences in $^3\text{He}_{\text{ET}}$ preservation or sediment focusing (Fig. 5); for example, $^3\text{He}_{\text{ET}}$ fluxes for the late Eocene and early Paleocene are a factor of 3 or more higher in sediment cores from the central and western North Pacific, South Atlantic and Southern Ocean (Farley 1995; Farley and Eltgroth 2003; Marcantonio et al. 2009; Murphy et al. 2010) than in a sediment core from Blake Nose in the North Atlantic (Farley and Eltgroth 2003). To the extent that these differences reflect differences in sediment focusing (which increases $^3\text{He}_{\text{ET}}$ fluxes during a dated interval by laterally advecting IDPs to a core site), $^3\text{He}_{\text{ET}}$ -based MARs will be systematically biased low in sites with high focusing but relative changes in accumulation rates will still be robust. As noted above, absolute $^3\text{He}_{\text{ET}}$ -based MARs are not independent of the

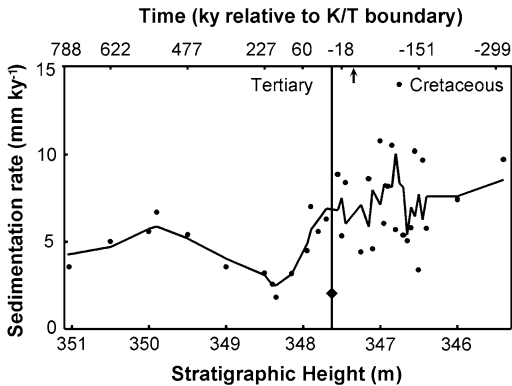


Fig. 8 $^3\text{He}_{\text{ET}}$ -based sedimentation rates for the Cretaceous-Paleogene boundary section at Gubbio, Italy (Mukhopadhyay et al. 2001b). Points indicate instantaneous sedimentation rates, while the line is the three-point moving average. The *diamond* indicates the sedimentation rate in the K/Pg (K/T in the figure) boundary clay. Note that the sedimentation rate in the boundary clay is only a factor of ~ 3 lower than in surrounding carbonate-rich layers. Based upon results at this and two other sections, the authors determined a duration for the K/Pg boundary clay of only 8–11 ka. By combining sedimentation rate estimates with measurements of stratigraphic height, time relative to the K/Pg boundary can be estimated in the rest of the section

chronology used to determine the mean $^3\text{He}_{\text{ET}}$ flux, but relative MAR changes are independent of the chronology within a given core.

$^3\text{He}_{\text{ET}}$ has been used as a constant flux proxy to estimate the duration of two prominent intervals of carbonate dissolution in the late Cretaceous and early Cenozoic: the K/Pg boundary clay and clays associated with the Paleocene-Eocene Thermal Maximum (PETM). Mukhopadhyay et al. (2001b) calculated the mean $^3\text{He}_{\text{ET}}$ flux immediately prior to the K/Pg boundary from Gubbio in the Italian Apennines and then used this flux combined with $^3\text{He}_{\text{ET}}$ concentration data within the boundary clay to determine sedimentation rates and the duration of the K/Pg boundary event (Fig. 8). This was the original purpose of measuring Ir in the boundary clay (Alvarez et al. 1980), but while the asteroid impact created an Ir spike at the boundary, $^3\text{He}_{\text{ET}}$ shows little change. This is to be expected as the large impactor should have been degassed and therefore would not have left a ^3He signature in

the sedimentary record. Dividing the density and thickness of the clay by the $^3\text{He}_{\text{ET}}$ -based MAR of the clay, Mukhopadhyay et al. (2001b) found durations for the boundary event of only 7.9 ± 1.0 ka and 10.9 ± 1.6 ka in two Apennine sections and 11.3 ± 2.3 ka in an expanded K/Pg section in Tunisia.

Farley and Eltgroth (2003) and Murphy et al. (2010) used a similar approach to estimate the duration of the PETM carbonate dissolution event in sediment cores from the Southern Ocean (ODP Site 690), South Atlantic (ODP Site 1266) and North Atlantic (ODP Site 1051). All three cores had age models established by astronomical calibration (in which variations in sediment composition are tuned to orbital changes) that had previously been used to estimate the durations of the onset, peak and recovery phases of the carbon isotope excursions and carbonate dissolution associated with the event. Farley and Eltgroth (2003) determined a mean $^3\text{He}_{\text{ET}}$ flux during two magnetic chrons spanning the PETM, while Murphy et al. (2010) determined the $^3\text{He}_{\text{ET}}$ flux over six eccentricity cycles preceding the event. These mean fluxes were then combined with $^3\text{He}_{\text{ET}}$ concentration data from PETM sediments to estimate the durations of each phase of the event. In each core, the $^3\text{He}_{\text{ET}}$ -based age model suggests that the peak duration of the carbon isotope excursion is longer, and the recovery period shorter, compared to results from cyclostratigraphy (Fig. 9) (Röhl et al. 2000, 2007; Farley and Eltgroth 2003; Murphy et al. 2010). If the $^3\text{He}_{\text{ET}}$ age models are correct, the longer duration of the CIE suggests sustained release of light carbon following the initial rapid burst of light carbon into the ocean-atmosphere system (Murphy et al. 2010). Determination of whether the $^3\text{He}_{\text{ET}}$ or cyclostratigraphic age-model is correct is also essential for evaluation of hypothesized mechanisms for removing excess carbon from the ocean-atmosphere system during and after the PETM. Additionally, the $^3\text{He}_{\text{ET}}$ age-model from Site 690 (Farley and Eltgroth 2003) suggests a faster pace of the PETM event compared to Site 1266 (Murphy

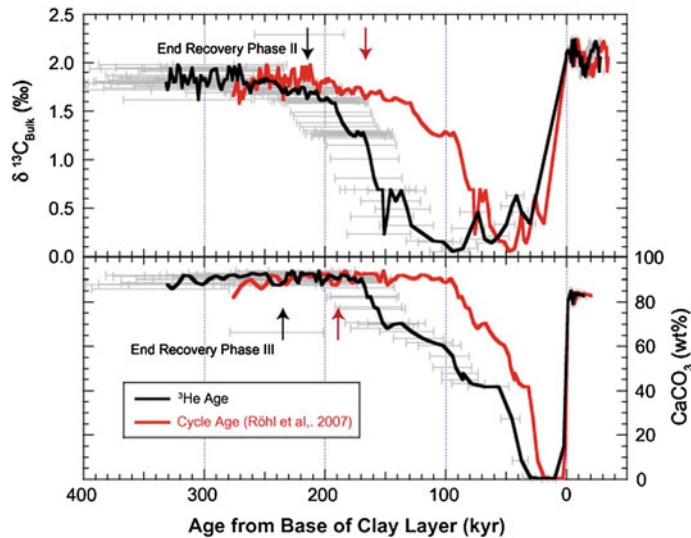


Fig. 9 Age models for the Paleocene-Eocene Thermal Maximum (PETM) based on $^3\text{He}_{\text{ET}}$ (black) and cyclostratigraphy (red) (Murphy et al. 2010). The panels show the $\delta^{13}\text{C}$ and calcium carbonate preservation changes associated with the PETM at ODP Site 1266. Grey error bars reflect the 2σ uncertainty in the $^3\text{He}_{\text{ET}}$ -based age model based on the uncertainty of the $^3\text{He}_{\text{ET}}$ flux calibration for the interval. Note that in the $^3\text{He}_{\text{ET}}$ -based

age model, the duration of the carbon isotope excursion is significantly longer than in the cyclostratigraphic age model, the recovery from the $\delta^{13}\text{C}$ excursion is more rapid, and the return to high carbonate preservation is slower. Arrows indicate the relative ages of the ends of two phases of the PETM recovery estimated by the two methods, with the $^3\text{He}_{\text{ET}}$ -based model indicating a slightly longer (~ 45 ka) total duration for the PETM event

et al. 2010). The reason for this discrepancy is not completely clear, but may be related to the $^3\text{He}_{\text{ET}}$ flux calibration from Site 690.

$^3\text{He}_{\text{ET}}$ -based MARs have also provided the basis for a number of paleoflux studies. As $^3\text{He}_{\text{ET}}$ -MARs can be calculated at much higher resolution than age model-based MARs, they allow novel insights into variations in past fluxes—for example, distinguishing whether an increase in the terrigenous fraction of sediments is due to increased terrigenous flux or decreased dilution by other sedimentary constituents. $^3\text{He}_{\text{ET}}$ data from Farley and Eltgroth's (2003) study have been used to determine accumulation rates of excess barium, a productivity proxy, in order to demonstrate that marine productivity did not play a major role in lowering atmospheric CO_2 levels after the PETM (Torfstein et al. 2010). Marcantonio et al. (2009) used $^3\text{He}_{\text{ET}}$ data to determine relative changes in bulk MARs and fluxes of dust and calcium carbonate during the late Eocene prior to the PETM. In addition to identifying orbitally-paced variations in

carbonate preservation, this study found that orbital variations in aeolian dust deposition in this greenhouse climate were similar in magnitude to those observed in the icehouse climate of the late Pleistocene. Similarly, Winckler et al. (2005) used $^3\text{He}_{\text{ET}}$ -based MARs to determine fluxes of productivity-related trace elements (barium, aluminum, phosphorous) and dust in the equatorial Pacific during a 1 Ma period spanning the mid-Pleistocene transition (MPT). While previous studies normalized productivity proxies to titanium and concluded that productivity increased during the MPT, $^3\text{He}_{\text{ET}}$ -normalized fluxes of productivity proxies indicate no change in productivity across the MPT and instead indicate lower fluxes of titanium and ^4He from aeolian dust (Winckler et al. 2005).

Given the fact that sedimentary $^3\text{He}_{\text{ET}}$ appears to be predominantly contained in particles $< 20 \mu\text{m}$ in diameter (see Sect. 3.3), one concern in using $^3\text{He}_{\text{ET}}$ as a constant flux proxy is that grain size fractionation during lateral advection of sediments by ocean currents

(sediment focusing) will enrich focused sediments in $^3\text{He}_{\text{ET}}$ -bearing IDPs. This fractionation would increase $^3\text{He}_{\text{ET}}$ concentrations in sites of sediment focusing while decreasing concentrations in winnowed sediments; $^3\text{He}_{\text{ET}}$ -based MARs would then be biased low by sediment focusing and biased high by winnowing. Such an effect was suggested by Marcantonio et al. (2001a), who observed $^3\text{He}_{\text{ET}}/^{230}\text{Th}_{\text{xs}}$ ratios in an Indian Ocean core a factor of 1.8 lower than at other sites. To test this suggestion, McGee et al. (2010) measured He and U-Th isotopes in sediments from two cores on the Blake Ridge in the western North Atlantic. The cores are <10 km apart and should thus receive similar vertical fluxes of sediment, but one core received much greater inputs of focused sediments over the past 20 ka. Despite substantial differences in focusing, $^3\text{He}_{\text{ET}}$ -based MARs agree between the two sites and are largely consistent with $^{230}\text{Th}_{\text{xs}}$ -based MARs. Though the uncertainties in $^3\text{He}_{\text{ET}}$ -based MARs in this study are large due to uncertainties in the correction for terrigenous ^3He , the results suggest that $^3\text{He}_{\text{ET}}$ -based MARs are not substantially affected by lateral advection.

The work summarized above builds on the observation that implied $^3\text{He}_{\text{ET}}$ fluxes at a given site are largely constant over Ma timescales, allowing $^3\text{He}_{\text{ET}}$ to be used as a constant flux proxy for determining sedimentary mass accumulation rates. In studies seeking to determine variations in fluxes of sedimentary constituents, such as dust and paleoproductivity proxies, ^3He -based MARs currently provide the only means of obtaining data that are independent of age models and capable of resolving sub-orbital flux variations. (Many studies multiply point-by-point concentration data by longer-term average fluxes determined from age models and give the appearance of sub-orbital resolution, but in reality these changes in concentration could reflect either short-term changes in flux or short-term changes in dilution by other sedimentary constituents with no change in flux.) Applications of ^3He -normalization have provided novel insights into the K/Pg and PETM events and into

changes in productivity and dust flux during the late Eocene and the mid-Pleistocene transition. Even so, the use of $^3\text{He}_{\text{ET}}$ as a constant flux proxy has been relatively limited compared to its potential applications.

5 Summary and Future Work

Measurement of $^3\text{He}_{\text{ET}}$ in marine sediments has provided important insights into past changes in the accretion rates of IDPs. Additionally, $^3\text{He}_{\text{ET}}$ has shown promise as a constant flux proxy capable of quantifying sub-orbital variability in past sedimentary fluxes and in constraining sedimentary age models, particularly during periods of carbonate dissolution. In looking ahead to future work, several questions offer promising avenues of research. These include:

- Can we improve our ability to distinguish $^3\text{He}_{\text{ET}}$ from terrigenous ^3He in detrital-rich, rapidly accumulating sediments such as continental margin deposits or lake sediments?
- What phases are responsible for the long-term retention of $^3\text{He}_{\text{ET}}$ in marine sediments, and is the stability of these phases affected by changing redox conditions? Is $^3\text{He}_{\text{ET}}$ retained in anoxic sediments, allowing $^3\text{He}_{\text{ET}}$ -based studies of ocean anoxic events?
- What is the relative importance of asteroidal vs. cometary sources of IDPs? Why do the measured $^3\text{He}_{\text{ET}}$ fluxes not match the predicted variability in IDP accretion rate over orbital timescales?
- What is the pace of environmental change during major climatic transitions? Several major transitions—including those associated with the Eocene–Oligocene, Oligocene–Miocene, and Pliocene–Pleistocene boundaries—have not yet been studied at high resolution, and $^3\text{He}_{\text{ET}}$ is perhaps the best timekeeper available for such studies.

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