



## Galactic-Cosmic-Ray-Produced $^3\text{He}$ in a Ferromanganese Crust: Any Supernova $^{60}\text{Fe}$ Excess on Earth?

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An excess of  $^{60}\text{Fe}$  in  $2.4\text{--}3.2 \times 10^6$  year old ferromanganese crust (237KD) from the deep Pacific Ocean has been considered as evidence for the delivery of debris from a nearby supernova explosion to Earth. Extremely high  $^3\text{He}/^4\text{He}$  (up to  $6.12 \times 10^{-3}$ ) and  $^3\text{He}$  concentrations (up to  $8 \times 10^9$  atoms/g) measured in 237KD cannot be supernova-derived. The helium is produced by galactic cosmic rays (GCR) and delivered in micrometeorites that have survived atmospheric entry to be trapped by the crust.  $^{60}\text{Fe}$  is produced by GCR reactions on Ni in extraterrestrial material. The maximum  $^3\text{He}/^{60}\text{Fe}$  of 237KD (80–850) is comparable to the GCR  $^3\text{He}/^{60}\text{Fe}$  production ratio (400–500) predicted for Ni-bearing minerals in iron meteorites. The excess  $^{60}\text{Fe}$  can be plausibly explained by the presence of micrometeorites trapped by the crust, rather than injection from a supernova source.

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Nearby supernova (SN) explosions may have profound implications for the Earth's biosphere. The enhanced flux of cosmic rays produced by a SN explosion, depending on duration and intensity, may deplete atmospheric ozone layer and produce a "cosmic-ray winter" due to increased cloud cover [1–3]. SN explosions that are close enough to Earth to have influence on climate, i.e., within approximately 30 pc from the sun, occur only a few times every  $10^8$  years [4]. The ability to detect nearby SN events in the geological record provides a test of SN frequency and may allow the effect they have on the Earth's climate to be investigated.

Ancient SN explosion debris can be detectable from anomalously high concentrations of radioactive isotopes like  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{53}\text{Mn}$ ,  $^{59}\text{Ni}$ , and  $^{60}\text{Fe}$  produced during the explosion or in dust swept up by the ejecta as it traverses the interstellar medium and then deposited as debris on Earth [5]. Excesses of  $^{60}\text{Fe}$  have been measured in slowly accumulating ferromanganese (FeMn) crusts from the deep Pacific Ocean [3,6]. The largest  $^{60}\text{Fe}$  excesses have been recorded in a layer in crust 237KD deposited between 2.4 and 3.2 Ma [3]. This has been attributed to the deposition of ejecta from a type-II SN ( $M \approx 30$  solar masses) at approximately 40 pc [3]. Confirming this observation with other radioisotopes has proved challenging as the intrinsically low concentrations make detection difficult [3,7]. For instance, a small excess of  $^{244}\text{Pu}$  has been measured in 237KD [7], but the uncertainty is large, leaving the  $^{60}\text{Fe}$  excess in 237KD as the strongest evidence for deposition of material of ancient SN debris.

In addition to SN,  $^{60}\text{Fe}$  is produced by reactions of galactic cosmic rays (GCR) with Ni in extraterrestrial material during exposure in space [8]. Production rates are high enough to generate measurable  $^{60}\text{Fe}$  excesses in

Ni-bearing minerals in iron meteorites [9,10]. It has been known for decades that ocean floor FeMn nodules incorporate extraterrestrial particles [11]. It is possible to estimate the contribution of GCR-produced  $^{60}\text{Fe}$  by measuring the concentration of nuclides that are exclusively produced by GCR. The helium isotope record of 237KD results from incorporation of micrometeorites that have been exposed in space for 100's of millions of years [12]. Here we compile the 16 published analyses (in the original study, the crust was named VA13/2) with 52 new He measurements in order to quantify the GCR-He contribution [13]. By comparing production rates in possible extraterrestrial materials, we estimate the contribution of GCR- $^{60}\text{Fe}$  from micrometeorites and test the supernova hypothesis.

Helium isotopes in oceanic sediments trace the contribution of solid extraterrestrial material to Earth [14–16]. This is evident from the  $^3\text{He}/^4\text{He}$  of ferromanganese crusts. It is unlikely that seafloor sediments incorporate SN He. If  $36 \times 10^{52}$   $^3\text{He}$  atoms ( $\approx 9 \times 10^{-4}$  solar masses) are produced during SN explosion [17], the flux distributed over a sphere with radius 40 pc for 1 kyr is  $2 \times 10^9$  atoms/cm<sup>2</sup> per year. A trivial fraction of this will penetrate the heliosphere and make it to the Earth as is evident from the dominance of the solar cosmic ray-derived  $^3\text{He}$  flux to Earth despite it being lower than the GCR  $^3\text{He}$  flux in the interstellar medium by 20 times [18]. Further, He solubility in seawater is extremely low (e.g., Pacific deep water =  $1.2 \times 10^{12}$  atoms/g [19]), and seawater-dissolved noble gases do not partition significantly into growing minerals [15]; therefore, high-energy  $^3\text{He}$  ions that make it to Earth will not be incorporated into growing FeMn crusts. The predicted SN  $^3\text{He}/^4\text{He}$  ( $\sim 1.0 \times 10^{-4}$ , or  $73R_A$ , where  $R_A$  is the atmosphere value of  $1.39 \times 10^{-6}$ ) [17] is significantly lower than in 237KD where values exceed  $4400R_A$  [Fig. 1(a)].

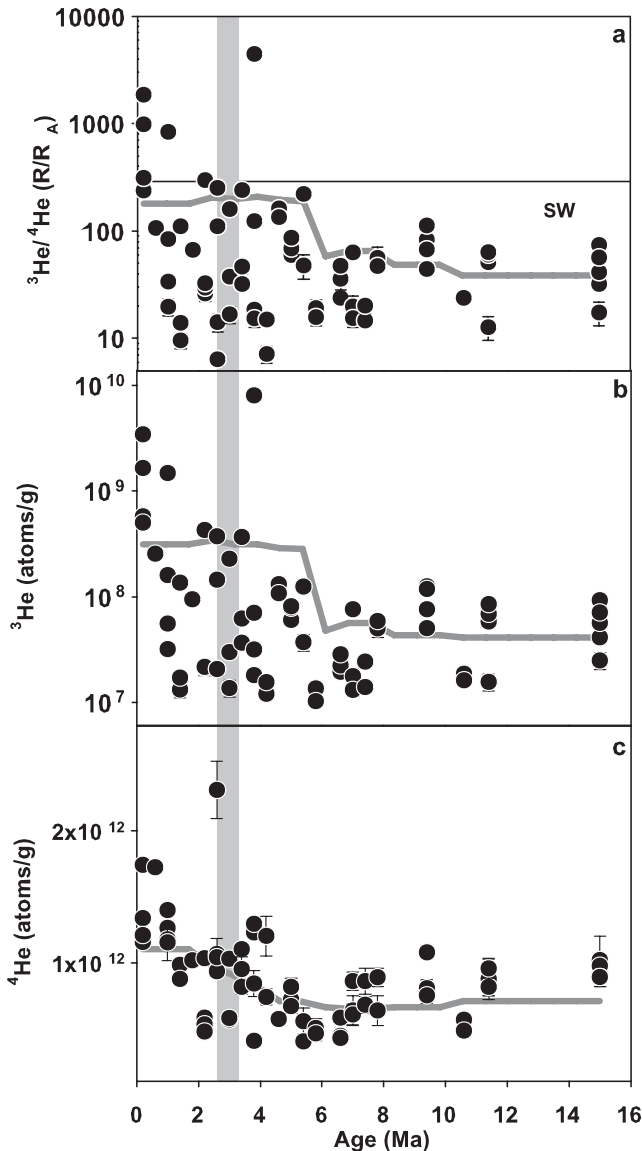


FIG. 1. Plot of sample age versus (a)  ${}^3\text{He}/{}^4\text{He}$ , (b)  ${}^3\text{He}$ , and (c)  ${}^4\text{He}$ . Solid lines depict smooth curves for sample proportion of 0.4 over 20 intervals.  ${}^3\text{He}/{}^4\text{He}$ ,  ${}^3\text{He}$ , and  ${}^4\text{He}$  increase at 4–5 Ma. The increase in  ${}^3\text{He}$  and  ${}^4\text{He}$  concentrations is due to incorporation of more extraterrestrial and detrital grains, probably due to decrease in current strength. The presence of GCR He in the extraterrestrial dust is evident from the very high  ${}^3\text{He}/{}^4\text{He}$  recorded for many post-4 Ma samples. The  ${}^3\text{He}$  increase is not coupled with the  ${}^{60}\text{Fe}$ -rich supernova layers (gray shaded bar) predicted by [3].

Crust 237KD was dredged from a depth of 4.8 km in the Central Pacific Ocean ( $9^\circ 18'N$ ,  $146^\circ 03'W$ ) [20]. The average growth rate is estimated to be 2.5 mm/Myr over the last 16 Myr [21]. We have measured He isotopes in 5–10 mg samples from 23, 1 mm-thick layers back to 15 Ma. Each layer integrates approximately 0.4 Myr of crust growth.  ${}^3\text{He}/{}^4\text{He}$  range from  $5.5 \pm 0.8$  to  $4440 \pm 89R_A$ .  ${}^4\text{He}$  concentrations show limited variation ( $0.4$  to  $2.3 \times$

$10^{12}$  atoms/g) compared to  ${}^3\text{He}$  ( $0.8$  to  $800 \times 10^7$  atoms/g) (Fig. 1). This is consistent with mixing between an extraterrestrial component (essentially pure  ${}^3\text{He}$ ) delivered in extraterrestrial dust and a radiogenic He component (essentially pure  ${}^4\text{He}$ ) delivered in wind-borne terrestrial dust [12].  ${}^3\text{He}/{}^4\text{He}$  variation is governed largely by variation in  ${}^3\text{He}$  concentrations. The extraterrestrial  ${}^3\text{He}$  flux to Earth recorded by the crust is approximately 100 times lower than in bulk oceanic sediments [12]. This reflects the low trapping efficiency of the crust which probably results from the high water current speed and steep slope at the site of accumulation.

The He isotope record of 237KD shows a significant change at 4–5 Ma (Fig. 1). Prior to this time,  ${}^3\text{He}$  concentrations varied by little over an order of magnitude, and no  ${}^3\text{He}/{}^4\text{He}$  was higher than the solar wind value ( $290R_A$ ; [22]). In the last 4 Myr,  ${}^3\text{He}$  concentrations increased significantly and displayed more than 100-fold variation [Fig. 1(b)]. The average  ${}^3\text{He}/{}^4\text{He}$  ( $330R_A$ ;  $n = 32$ ) was considerably higher than the average of  $>4$  Myr samples ( $54R_A$ ;  $n = 36$ ). Six samples have  ${}^3\text{He}/{}^4\text{He}$  equal to, or higher than, solar wind-He, indicating the dominance of GCR-He ( $1.2 \times 10^5 R_A$  [23]). The highest  ${}^3\text{He}/{}^4\text{He}$  ( $4440R_A$ ) is greater than the maximum measured in stratospheric interplanetary dust particles (IDPs) ( $2220R_A$ ) [24]. It is the highest ratio measured in a terrestrial sample and can only be due to the incorporation of GCR-He. The  ${}^3\text{He}$  enhancement is not due to an increase in the cosmic ray intensity or the flux of extraterrestrial dust to Earth as it not recorded by extensive studies of the oceanic sediment record [e.g. [25]]. The enhanced  ${}^3\text{He}$  (and  ${}^4\text{He}$  [Fig. 1(c)]) is probably due to increased efficiency of trapping dense micrometeorites that settle through the oceans. The  ${}^3\text{He}$  and  ${}^4\text{He}$  increase may be due to a decrease of water current velocity. This is consistent with the decreased deep water mass exchange between the Pacific and Atlantic Oceans during closure of the Panama gateway between 8 and 5 Ma [26].

The trapping of more extraterrestrial particles after 4–5 Ma is associated with a greater variability of  ${}^3\text{He}/{}^4\text{He}$  in replicate measurement of the individual crust layers [Fig. 1(a)]. The statistical variability of He concentrations between replicate analyses of the same sample is estimated by the fractional difference (FD) following:

$$\text{FD} = \frac{|(a - b)|}{\left[\frac{(a+b)}{2}\right]}$$

where  $a$  and  $b$  are the He concentration in subsamples of comparable masses (5–10 mg) from the same layer. FD values depend on the ratio  $a/b$  [12]. Samples of 237KD older than 4 Myr have a broadly Gaussian distribution of FD peaking at 0.5. Only 9% of values are less than 0.2, and none exceeds 1.5 [Fig. 2(a)]. This is consistent with the pattern predicted for oceanic sediments [27] and implies that subsamples tend to have a similar number of particles

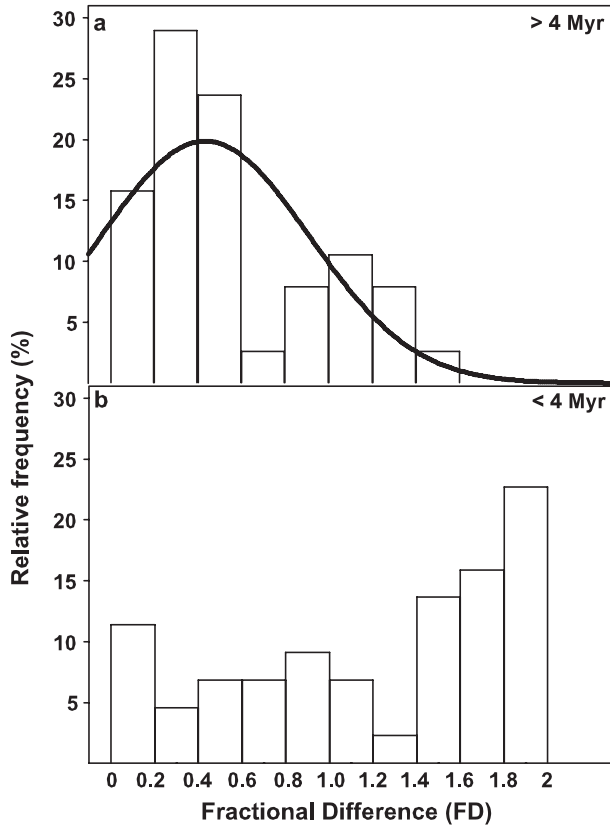


FIG. 2. Fractional differences (FD) of  $^3\text{He}$  concentration between replicate analysis of crust layers (a)  $>4$  Ma and (b)  $<4$  Ma. Samples  $>4$  Ma have a broadly Gaussian distribution approximated by the solid curve. The peak between 1.8–2 for FD distribution of post-4 Ma samples indicate that the He budget is dominated by occasional, extremely GCR-He-rich particles.

with similar  $^3\text{He}$  concentrations. The skew to high values is due to under-sampling of He-rich particles. Samples that are less than 4 Ma have a large component of extreme FD values [Fig. 2(b)]; 16% have  $\text{FD} < 0.2$ , and 20% have  $\text{FD} > 1.6$ . This is highly unlikely to be sampled by normal distribution. The likeliest explanation is that high FD values are generated by the incorporation of a small number of extremely  $^3\text{He}$ -rich particles. The association of high  $^3\text{He}$  concentrations and GCR- $^3\text{He}/^4\text{He}$  implies that the occasional particles have long exposure durations in space.

The occasional incorporation of GCR-He-rich particles has important implications for the origin of excess  $^{60}\text{Fe}$  in 237KD [3]. The production rate of  $^{60}\text{Fe}$  ( $P_{60}$ ) has been determined in pure Ni targets in a 10 cm-radius iron sphere isotropically irradiated with 1.6 GeV protons [28].  $P_{60}$  (normalized to 1 primary proton/cm $^2$ /s) varies little with depth and integrated over the radius of the sphere corresponds to  $0.6 \times 10^8$  atoms/g per Myr [28]. Although this differs from GCR production rates in space, it can be compared to the production rate of  $^3\text{He}$  ( $P_3$ ) determined in the same experiment [28].  $P_3$  ( $2.8\text{--}3.5 \times 10^{10}$  atoms/g

per Myr) shows little discernible change with depth and, importantly, there is no difference in the  $^3\text{He}$  production rate from Ni and Fe [29]. Consequently, the experiment predicts that  $(P_3/P_{60})_{\text{GCR}}$  in a pure Ni phase in iron meteorites is 450–550. This will scale linearly with the Ni concentration. The  $(P_3/P_{60})_{\text{GCR}}$  in chondritic meteorites will be significantly higher as  $P_3$  is approximately twice that of iron meteorites [29], and Ni contents are commonly less than 1% [30].

$^3\text{He}$  and  $^{60}\text{Fe}$  have been determined in the iron meteorites Odessa and Tlacotepec [10,31].  $^3\text{He}$  ranges from  $4.2 \times 10^{13}$  atoms/g (Odessa) to  $1.5 \times 10^{14}$  atoms/g (Tlacotepec). For cosmic ray exposure ages of 875 Ma and 945 Ma for Odessa and Tlacotepec, respectively [31],  $^3\text{He}$  production rates are  $4.8 \times 10^{10}$  atoms/g per Myr (Odessa) and  $1.5 \times 10^{11}$  atoms/g per Myr (Tlacotepec). For Ni contents of 7 and 18% and  $^{60}\text{Fe}$  of 2.0 dpm/kg Ni and 1.2 dpm/kg Ni for Odessa and Tlacotepec, respectively [10],  $P_{60}$  ranges from  $7.6 \times 10^7$  atoms/g per Myr (Odessa) to  $1.1 \times 10^8$  atoms/g per Myr (Tlacotepec). The resultant  $(P_3/P_{60})_{\text{GCR}}$  (630–1360) are broadly consistent with the experimentally-determined values and the lower Ni concentration.

The  $^3\text{He}$  concentration in the four crust samples where GCR-He dominates (i.e.,  $^3\text{He}/^4\text{He} > 290R_A$ ) is  $1.5\text{--}8.0 \times 10^9$  atoms/g. Excess  $^{60}\text{Fe}$  was measured in three of the 14 layers spanning the last 5 Myr of 237KD [3]. The average  $^{60}\text{Fe}$  concentration in these three layers (after background correction of  $2.4 \times 10^{-16}$  [3] and assuming 20% Fe in the crust) is  $3.7 \times 10^6$  atoms/g. Radioactive decay of  $^{60}\text{Fe}$  ( $t_{1/2} \approx 1.49$  Myr [9]) has reduced this by 67–77% since  $2.8 \pm 0.4$  Ma [3], leading to an initial  $^{60}\text{Fe}$  concentration of  $0.9\text{--}1.9 \times 10^7$  atoms/g. If the  $^3\text{He}$  and  $^{60}\text{Fe}$  are delivered in the same particle, the  $^3\text{He}/^{60}\text{Fe}$  is 80–850. This overlaps the predicted  $(^3\text{He}/^{60}\text{Fe})_{\text{GCR}}$  of FeNi alloy minerals such as taenite (40–50% Ni) that are common in the ataxite class of iron meteorites. Further, it is similar to the production rate ratio in iron meteorites and provides strong evidence for a significant, if not dominant, contribution of GCR-produced  $^{60}\text{Fe}$  in crust 237KD. The  $^3\text{He}/^{60}\text{Fe}$  of the 237KD is a low estimate of the true value as He is substantially degassed from incoming micrometeorites during atmospheric entry [27]. For a cutoff temperature of 600 °C for He loss, only 0.5% of the mass of the incoming particles still retain He by arrival at Earth surface [27].

Using measured  $^{60}\text{Fe}$  in Dermbach [10], the  $^{60}\text{Fe}$  excess in 237KD can be accounted by one 500  $\mu\text{m}$ -diameter or greater micrometeorite, or 2–4, 320–360  $\mu\text{m}$  particles. Although 500  $\mu\text{m}$  micrometeorites are not common, magnetic cosmic spherules up to 700  $\mu\text{m}$  diameter have been extracted from seafloor sediments as well as from Antarctic and Greenland ice (e.g., [32,33]). Importantly, FeNi alloys are the densest common minerals in meteorites (taenite  $> 8$  g/cc) and a 500  $\mu\text{m}$ -diameter micrometeorite will settle approximately 5 times faster than equivalent-

sized chondritic fragments, and 7500 times faster than the  $<8 \mu\text{m}$ -diameter zircons that contribute the  $^4\text{He}$  [34]. The FD distribution of  $^4\text{He}$  in both the post- and pre-4 Ma crust layers are largely between 0 and 0.4. This implies that  $^4\text{He}$  was transported by a large number of particles with rather constant  $^4\text{He}$  concentrations (probably micron-sized zircon [34]) and contrasts strongly with  $^3\text{He}$  record. This supports the notion that the  $^3\text{He}$  FD variation is more sensitive to subtle changes in ocean circulation.

Support for the micrometeorite origin of the GCR- $^{60}\text{Fe}$  comes from the presence of small excesses of  $^{53}\text{Mn}$  in 237KD that can be accounted for by spallation of Fe and Ni in meteorites [6]. The experimentally-determined GCR- $^{53}\text{Mn}$  production rate ( $P_{53}$ ) ranges from  $3.1\text{--}4.8 \times 10^{10}$  atoms/g per Myr [34]. The equivalent  $P_3/P_{53}$  ranges from 1 to 5 [28,29]. The  $^{53}\text{Mn}$  flux in 237KD (5–10 mm) ( $6.4 \times 10^8/\text{cm}^2/\text{Myr}$  [6]) corresponds to  $1 \times 10^9$  atoms  $^{53}\text{Mn}/\text{g}$  crust. For  $t_{1/2} = 3.7$  Myr [6] and a terrestrial residence time of 2–4 Myr,  $^3\text{He}/^{53}\text{Mn}$  ranges from 0.6 to 4.3. Again, this overlaps the theoretical value of GCR production and implies that the crust  $^{53}\text{Mn}$  is contributed by fragments of iron meteorite. Additionally, the outer layers of 237KD have Os isotopic compositions that are significantly lower than seawater values, strongly indicative of a meteoritic component [35].

In summary, we find that over the last 4–5 Myr, FeMn crust 237KD has incorporated micrometeorites with extremely high GCR-He concentrations. Consequently, the  $^{60}\text{Fe}$  excesses in the crust are probably due to the incorporation of Ni-rich extraterrestrial particles that were exposed in space for a few 100's Myr. The delivery of Ni-rich micrometeorites may be more sporadic than the delivery of He-rich particles if they originate as ablation debris from Ni-rich iron meteorites. These results strongly question whether  $^{60}\text{Fe}$  excesses provide evidence for nearby supernova explosion, but imply that it may find use as a tracer of the origin of meteoritic debris on Earth.

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