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Interstellar ⁶⁰Fe on the Surface of the Moon

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A dying massive star ends in a supernova explosion ejecting a large fraction of its mass into the interstellar medium. If this happens nearby, part of the ejecta might end on Solar System bodies and, in fact, radioactive ⁶⁰Fe has been detected on the Pacific ocean floor in about 2 Ma old layers. Here, we report on the detection of this isotope also in lunar samples, originating presumably from the same event. The concentration of the cosmic ray produced isotope ⁵³Mn, measured in the same samples, proves the supernova origin of the ⁶⁰Fe. From the ⁶⁰Fe concentrations found we deduce a reliable value for the local interstellar fluence in the range of 1×10^8 at/cm². Thus, we obtain constraints on the recent and nearby supernova(e).

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Introduction.—The debris of a supernova (SN) explosion can travel great distances, depending on the density of the interstellar medium. If such an explosion occurs close enough to our Solar System, SN dust particles may overcome the opposing solar wind pressure and reach the inner part of the heliosphere [1,2].

The surfaces of Solar System bodies (e.g., Earth or Moon) may collect interstellar material. The possibility to detect long-lived SN-produced radionuclides on Earth has been discussed by Korschinek et al. [3] and Ellis et al. [4]. Knie et al. [1,5], and Fitoussi et al. [6] succeeded to identify an ⁶⁰Fe $[T_{1/2} = (2.61 \pm 0.04)$ Ma] (weighted average of Refs. [7,8]) signature in a 2 Ma old hydrogenetic ferromanganese crust from the central Pacific Ocean, which they attributed to the deposition of freshly synthesized SN material onto Earth. Recently, Kachelrieß et al. [9] investigated cosmic ray spectra and found indications that they carry the signature of a nearby SN explosion about 2 Ma ago as well. In terrestrial samples, the exogenous atoms are typically widely dispersed and diluted both in transit through the atmosphere and later by the action of wind and water. On the other hand, lunar soil samples may retain a more concentrated signal as these atmospheric processes do not operate on the Moon. Well documented soil cores from the Apollo collection are available, but lunar soils have several potential weaknesses as collectors of ⁶⁰Fe of SN origin. First, ⁶⁰Fe unrelated to SNe is formed on the lunar surface by spallation reactions induced by solar and by Galactic cosmic rays (SCRs and GCRs) when they react, mainly, with the heavy isotopes of Ni. Although the concentration of Ni in lunar material is in general low, a superposition of cosmogenic and interstellar ⁶⁰Fe must be considered. Second, the lunar regolith is dynamic and may not preserve an undisturbed record of any dust deposited there. Impacts by micrometeorites (objects with masses less than 1 mg) constantly stir and mix the topmost few millimeters of the lunar surface [10,11]. Over time, this gardening would smear a SN layer over a range of depths. Sporadic impacts by full-size meteorites contribute to the gardening on a larger scale by pulverizing and melting material [12], by excavating or burying clumps of matter, and by inducing down-slope movements. In any one column of lunar soil, the probability of significant effects is largest close to the surface, but decreases rapidly with depth and for shorter (less than a few megayears) periods of exposure. Third, during the sampling and/or handling of some lunar cores the upper portions of the material can be lost or disturbed.

Gardening and matters related to core recovery and processing can be addressed by selecting samples whose recent gardening histories are known independently. In particular, the vertical stability of a core on a time scale of 1–4 Ma can be assessed through measurements of depth profiles for the cosmogenic radionuclides ²⁶Al and ⁵³Mn, which have half lives of $T_{1/2} = 0.7$ Ma [13] and $T_{1/2} = 3.7$ Ma [14], respectively. One compares the depth profiles expected in an undisturbed column with measured activities to establish whether loss of material or excessive mixing might have taken place during the last 10 Ma. These depth profiles tell us how deeply we must sample a core in order to capture a sizeable fraction of any reworked surface material.

Samples.—From the Apollo 12 mission $(3^{\circ}00'44.60'' \text{ S}, 23^{\circ}25'17.65'' \text{ W})$, we received two near-surface samples (top 1.2 cm) taken with the double drive tube 12025/8. From the Apollo 15 mission $(26^{\circ}07'55.99'' \text{ N}, 3^{\circ}38'01.90'' \text{ E})$, we obtained four samples from the core 15008, with depths ranging from the top half centimeter down to 10 cm. Both

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profiles show an undersaturated activity of ⁵³Mn in the top centimeters, followed by a smooth profile at greater depths [15,16]. From the Apollo 16 mission (Descartes plateau 8°58′22.84″ S, 15°30′00.68″ E) we received three samples, one sample each from skimmed soil 69921, the underlying scooped soil 69941, and the underboulder soil 69961. The uppermost sample of these three, 69921, shows rather high ²⁶Al and ⁵³Mn activities, which presumably have been produced by solar cosmic rays on the surface of a boulder from which material eroded and fell onto the surface of the adjacent regolith [17]. For more details on the samples, please refer to Ref. [18].

⁶⁰Fe *concentrations.*—At present, only accelerator mass spectrometry has the capability to determine ⁶⁰Fe concentrations in subgram lunar samples. The measurements were performed at the GAMS setup at the Maier-Leibnitz Laboratorium in Garching, Germany, which has a sensitivity for measuring ⁶⁰Fe/Fe atom ratios of 10^{-16} and below [18–20]. The measured ⁶⁰Fe depth profiles are shown in Fig. 1 (see also Table I in Ref. [18]). Figure 1(a) shows the ratio of ⁶⁰Fe to total iron compared with the background level, indicated with a dashed line. This level was determined by the measurement of a processing blank sample (⁶⁰Fe free). Twelve samples show concentrations



FIG. 1. This graph shows, as a function of depth, on a logarithmic scale, (a) the concentration of 60 Fe/Fe compared with the blank level (dashed line) during these measurements and (b) disintegrations per minute per kilogram of Ni [dpm/(kg Ni)], compared with the average activity measured in meteorites [thin shaded region at 0.5 dpm/(kg Ni)], published up to this date (Refs. [21–24]; see Ref. [18] for a summary of these). The labeling of the data points is specified in Tables I and II in Ref. [18]. The empty triangles and squares show the calculation of the expected contribution for the SCR spallation production of 60 Fe in the positions of the Apollo 12 and 16 landings, respectively. The error bars indicate the 1 σ confidence level [25].

samples have no detectable ⁶⁰Fe. The large errors are due to the low counting statistics [25] associated with the low 60 Fe/Fe atom ratios observed. Since the production of 60 Fe by cosmic radiation also has to be taken into account, we show in Fig. 1(b) the 60 Fe activities per kilogram of nickel, the main target element, calculated using the nickel concentration (see Table I in Ref. [18]). In both figures, the depths in g/cm² were calculated using measured depths in centimeters and the bulk density for each set of samples: 1.92 g/cm² for core 12025, 1.65 g/cm³ for 15008, and 1.50 g/cm³ for 60007/6 [11]; for samples 69921/41/61 a typical value of 1.50 g/cm³ was used because of a lack of information on the density of these samples.

well above the blank level (${}^{60}\text{Fe}/\text{Fe} = 2 \times 10^{-16}$). Two

Contribution from cosmic ray production.-It is important to assess the possible contributions of SCRs and GCRs to the concentrations of ⁶⁰Fe present in the lunar samples. Here, it is not the isotopic ratio that is relevant but the ratio with respect to the dominant target elements. Traditionally, the number of radionuclei is expressed as their activity, in disintegrations per minute (dpm). The expected saturation activity due to irradiation with SCRs was calculated with the TALYS code [26] in a similar way as described in Ref. [27]. Included in these calculations are the cross sections for the nuclear reactions $^{nat}Fe(\alpha, X)^{60}Fe$, ^{nat}Ni $(p, X)^{60}$ Fe, and ^{nat}Ni $(\alpha, X)^{60}$ Fe. The solar-proton flux was assumed to decrease with $\exp(-R/R_0)$, where R =pc/(Ze) is the magnetic rigidity and $R_0 = 90$ MV is the rigidity parameter, and to have an omnidirectional flux \geq 10 MeV of $73p/(cm^2 s)$. For the solar alpha particles we assumed $R_0 = 75$ MV and an omnidirectional flux for ≥ 10 MeV of $7.5\alpha/(\text{cm}^2 \text{ s})$. Higher values of R_0 imply larger fractions of high-energy particles. The results for the production of ⁶⁰Fe by particles for $1 \le E_p \le 100$ MeV, at the surface and just below, are shown in Fig. 1(b). The empty triangles and squares show the calculations of the expected SCR production of ⁶⁰Fe in the positions of the Apollo 12 and 16 samples, respectively. SCR production is largest at the surface and then decreases rapidly with increasing depth. At the depths estimated for our shallowest samples, 0.4 g/cm², the measured 60 Fe activities of 1-20 dpm/(kg Ni) exceed the calculations by a factor of 30 or more; the factor is even larger for the samples taken from greater depths. We conclude that the SCR production of ⁶⁰Fe is at most 10% in near-surface samples and much less than 10% in samples at greater depth.

GCR contributions to 60 Fe may arise in two ways, either through the presence in the lunar soil of a meteoritic component that contains 60 Fe, or as the result of *in situ* production. We can constrain the former by comparing the 60 Fe in the lunar soils with the 60 Fe activities measured in meteorites (see Refs. [21–24]; a summary of these can also be found in Table III in Ref. [18]). The meteorite activities will differ from lunar ones because of differences in the average irradiation geometries. The weighted mean for meteorites is $(0.35 \pm 0.08) \text{ dpm}/(\text{kg Ni})$, which may be compared with the weighted mean of $(20\pm5) \text{ dpm}/(\text{kgNi})$ for the three most active lunar soil samples [Fig. 1(b)]. We conclude that less than 10% of the ⁶⁰Fe measured in these lunar samples results from meteoritic contamination. The ⁵³Mn activities of the lunar soils can further constrain the direct production of ⁶⁰Fe by GCRs [18]. Figure 2 shows the 60 Fe activities for 11 of the lunar soil samples (1–11) in comparison to their ⁵³Mn activities, both relative to their dominant target element. The 53Mn in lunar samples is formed mostly by GCRs through the (p, α) reaction with ⁵⁶Fe (91.8% abundance). The possible contribution from any SN material is negligible. Published calculations [28,29] for the ⁶⁰Fe/⁵³Mn ratios (atomic) expected in supernovae are roughly in the range of 0.1 to 1. In comparison, we observe a mean activity in the lunar soils of around 10 dpm/(kg Ni) [Fig. 1(b)] for 60 Fe, and of around 350 dpm/(kgFe) for ⁵³Mn (Fig. 2), which corresponds to an ${}^{60}\text{Fe}/{}^{53}\text{Mn}$ atom ratio of around 10^{-4} . Thus, any contribution from ⁵³Mn formed in a SN is overwhelmed by recent GCR production of ⁵³Mn on the Moon.

Figure 2 shows also the empirical relationship between 60 Fe and 53 Mn activities measured in meteorites, which, despite having different elemental compositions from those of our lunar samples, are relevant because production derives from Fe and Ni in both types of material. All of these meteorites, except possibly Dermbach [$A(^{60}$ Fe) = $0.38^{+0.08}_{-0.07}$ dpm/(kg Ni)] [18], have cosmic-ray exposure



FIG. 2. Measured activities of 60 Fe versus 53 Mn in meteoritic and lunar samples. Units are disintegrations per minute per kilogram of Fe and Ni, for 53 Mn and 60 Fe, respectively. Samples 1 through 11 (filled points) are lunar samples; the other values (error bars only) are for iron meteorites. The labeling of the data points is specified in Table I in Ref. [18]. The shaded bar indicates the error band for cosmogenically produced 53 Mn and 60 Fe activities in meteorites. The error bars indicate the 1 σ confidence level [25].

ages greater than 10 Ma, which implies that the ⁵³Mn and ⁶⁰Fe activities had attained their saturation values at the time of fall. As terrestrial ages for iron meteorites are typically less than 0.1 Ma, the measured activities are likely to have decayed only by a few percent since the meteorites fell. Most of the variation of a factor of 6 in ⁵³Mn and a factor of about 4 in ⁶⁰Fe reflects differences in the preatmospheric sizes of the precursor meteoroids and the depths of the samples within them. The samples with the lowest activities, Dermbach and Tlacotepec [18], presumably came from the largest meteoroids. Since the scatter of the activities is mainly due to differences in meteoroid geometry, we would expect a rather constant ratio if both activities are normalized to their respective target elements. Figure 2 also shows the activity of 60 Fe/(kg Ni) versus that of ${}^{53}Mn/(kg Fe)$ of the lunar samples along with the results for iron meteorites. The meteoritic data (Fig. 2 and Table III in Ref. [18]) yield a constant activity ratio of 60 Fe/(kg Ni) to 53 Mn/(kg Fe) of 0.00268 \pm 0.00035, with a γ^2 /d.o.f = 8.5/6 (used to inflate the quoted error). To prove the constancy of this ratio is an achievement by itself. This ratio is expected to apply to the Moon as well.

We may now compare the data for the lunar soils with the expectations based on the ⁶⁰Fe/⁵³Mn activity ratio measured for the meteoritic data, whose uncertainty band is shown by the shaded line in Fig. 2. The data points for most of the lunar samples lie well above the expected relationship. Only three of the lunar data points (3, 8, and 11) have 60 Fe/(kg Ni) to 53 Mn/(kg Fe) activity ratios compatible with cosmogenic production. These samples have also a complicated history, as described in Ref. [18], and are therefore excluded from further comparison. Because of the cosmogenic origin of ⁵³Mn in the lunar samples, we clearly can state a surplus of ⁶⁰Fe in the lunar soils. The three data points with the highest ⁶⁰Fe activities show an average activity ratio of 0.047(12), a factor of 17 and about 4σ higher than for the meteorites. The same significance holds true for five and eight data points with average ratios 0.032(6) and 0.0138(25), respectively.

With the simultaneous measurement of ⁵³Mn concentrations we can safely conclude that these lunar samples carry ⁶⁰Fe that has not been formed by cosmic radiation but in one or more SN explosions. Our finding contradicts the arguments of Fry et al. [30], who suggested that practically all SN-produced ⁶⁰Fe would escape into space after impact on the Moon. They relied on an experiment [31] that irradiated steel with dust particles formed of iron and observed the number of evaporated atoms, but could not distinguish whether the gas was produced from the projectile or the target. The lunar regolith as a target has certainly a much higher porosity than steel and the projectile material stays at the surface much more likely. A proof of this is the Interstellar Preliminary Examination (ISPE) experiment on the Stardust spacecraft, which has collected at least seven dust particles of presumably interstellar origin [32]. Even those collected in aluminum (four craters with about a 0.4 μ m diameter) could be analyzed concerning their elemental composition, proving that a large part of the projectile material stayed in the crater.

Interstellar ⁶⁰Fe *fluence.*—The deposition of ⁶⁰Fe on the lunar surface must have happened on a time scale of megayears, since already considerable gardening has happened and, on the other hand, cannot have happened more than some three half-lives, i.e., 8 Ma, ago, to still be detectable. Thus, it is likely that it coincides with the ⁶⁰Fe surplus in ferromanganese crusts [5,6], which was collected between 1.7 and 2.6 Ma ago, based on the dating [6] rescaled with the new value for the ¹⁰Be half-life [1.387(12) Ma [33,34]].

Any ⁶⁰Fe signal is expected to be distributed downward due to gardening of the lunar surface [10]. In a time period of around 2.2 Ma, gardening down to a few g/cm^2 (assuming a typical density of the lunar regolith of 1.50 g/cm^3) is expected. It is reasonable, therefore, to integrate the measured ⁶⁰Fe concentration over this range, in order to estimate the local fluence of ⁶⁰Fe. Nevertheless we found also elevated concentrations of ⁶⁰Fe down to a depth of 18.75 g/cm² [12] (see Fig. 3), indicating possible excavations by meteorites and/or down-slope movements [18]. Thus, an inclusion of these deep samples yields a conservative upper limit for the integration to obtain a local interstellar fluence of 60 Fe, Φ_{LIF} . As the lower limit (smallest depth) we adopted that of sample 4. Consequently, from the data (Fig. 3) we can estimate a range for the 60 Fe abundance on the Moon between 1×10^7 and 6×10^7 at/cm²; these values include a correction of less than 10% of the original signal for cosmogenic 60 Fe based on a fixed ratio of 60 Fe dpm/(kg Ni) to 53 Mn dpm/(kg Fe) of 0.00268 \pm 0.00035.



FIG. 3. Estimation of the local fluence of 60 Fe on the Moon's surface. The dashed curves represent two different integration scenarios. They symbolize a lower and an upper limit. The labeling of the data points is detailed in Table I in Ref. [18]. The error bars indicate the 1σ confidence level [25].

To calculate the original $\Phi_{\rm LIF},$ a correction for radioactive decay since the time of deposition, 1.7-2.6 Ma, is required. We obtain a decay corrected Φ_{LIF} of ⁶⁰Fe at 1 AU from the Sun between 0.8×10^8 and 4×10^8 at/cm². Our estimate assumes that the ⁶⁰Fe is uniformly spread over the lunar surface; the 60 Fe Φ_{LIF} is 4 times the fluence on the Moon (radius of a spherical surface to its two-dimensional projection). We have neglected here losses through evaporation. If there are losses then the above fluence value has to be viewed as a lower limit. Knie et al. [5] previously estimated a larger value for Φ_{LIF} of 20×10^8 at/cm². This earlier estimate was partially based on the ⁵³Mn fluence measured in ice by Bibron et al. [35], which was used to deduce an uptake factor of ⁶⁰Fe into the ferromanganese crust. Since the estimate of Knie et al., revisions to the halflives of ⁶⁰Fe [7,8] and ¹⁰Be [33,34,36] have been made, and the age of the ferromanganese crust was revised from 2.8 to 2.2 Ma. In addition, a recent measurement of the ⁵³Mn fluence by Auer et al. [37] results in a value that is about 40 times smaller than the earlier value of Bibron *et al.* [35]. Applying these newly revised parameters to the data of Knie *et al.* [5] reduces their estimate to 0.5×10^8 at/cm², which coincides roughly with the lower limit of our lunar value. Our estimate also includes the revised values for the two half-lives and the ferromanganese crustal age, and it does not depend on the ⁵³Mn fluence in ice. The observed ⁶⁰Fe signature on the Moon is a clear indication of the deposition of SN material. Our results can be used to improve simulations of the close stellar and interstellar medium several megayears ago [38] as well as estimations of the deposition of other long lived radionuclides such as ²³⁷Np $(T_{1/2} = 2.144 \times 10^6 \text{ a})$ and the heavier long lived transuranium isotopes ²⁴⁴Pu and ²⁴⁷Cm $(T_{1/2} = 8 \times 10^7 \text{ a})$ and $T_{1/2} = 15.6 \times 10^7$ a, respectively).

Conclusions.—In conclusion, we detected ⁶⁰Fe concentrations on the lunar surface, which exceed the calculated values for SCR and GCR production by roughly 1 order of magnitude. Cosmic ray production of ⁶⁰Fe, either *in situ* or through deposition of irradiated meteorites, can be predicted by the determination of ⁵³Mn concentrations in the same lunar samples and the determination of both concentrations in a number of meteorites, which show a constant ratio irrespective of the size and position of the meteoroids in the parent body. We argue that enhanced ⁶⁰Fe concentrations observed in lunar surface samples have the same origin as the ⁶⁰Fe signature observed in deep-sea ferromanganese crusts, namely, a deposition of freshly synthesized material by one or more SN explosions. Assuming that deposition occurred synchronously in both solar system archives, between 1.7-2.6 Ma ago, we can arrive at a more reliable, local ⁶⁰Fe Φ_{LIF} at that time than was previously possible because no estimate of uptake efficiency is involved as is the case for the ferromanganese crusts.

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