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## Supernova-produced $^{53}\text{Mn}$ on Earth

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### Abstract:

For the time period from 1.5 to 4 Myr before present we found in deep ocean ferromanganese crusts a  $^{53}\text{Mn}$  excess concentration in terms of  $^{53}\text{Mn}/\text{Mn}$  of about  $4 \times 10^{-14}$  over that expected for cosmogenic production. We conclude that this  $^{53}\text{Mn}$  is of supernova origin because it is detected in the same time window, about 2.5 Myr ago, where  $^{60}\text{Fe}$  has been found earlier. This overabundance confirms the supernova origin of that  $^{60}\text{Fe}$ . For the first time supernova-formed  $^{53}\text{Mn}$  has been detected and it is the second positively identified radioisotope from the same supernova. The ratio  $^{53}\text{Mn}/^{60}\text{Fe}$  of about 14 is consistent with that expected for a SN with a 11 - 25  $M_{\odot}$  progenitor mass and solar metallicity.

Stars heavier than about 10 solar masses ( $M_{\odot}$ ) end their life in a supernova explosion (SN). At the same time they eject large amounts of stable- and radio-nuclides into the interstellar medium. This happened close to the solar system several times during the last few millions of years (Myr) [1]. One of the expelled nuclides is the long-lived nuclide  $^{60}\text{Fe}$  ( $T_{1/2} = 2.61 \pm 0.04$  Myr) [2,3]. If the explosion is close enough to the solar system, some material can enter the solar system [4, 5]. Traces of  $^{60}\text{Fe}$  have already been found in Earth's reservoirs like ferromanganese crusts and ocean sediments, and even on the Moon [6 - 11]. Several indications support the SN origin of the  $^{60}\text{Fe}$  [1, 12]. However, there is also the possibility of  $^{60}\text{Fe}$  being formed in asymptotic giant branch (AGB) stars [13]. Despite having lower nucleosynthesis yields, they could be the origin of the observed  $^{60}\text{Fe}$ , as well. An unambiguous, only SN formed radionuclide, such as  $^{53}\text{Mn}$ , detected in the same samples as the  $^{60}\text{Fe}$ , can solve this open question. Therefore the concomitant finding of  $^{53}\text{Mn}$  ( $T_{1/2} = 3.7 \pm 0.4$  Myr) [14] would be a compelling support for the SN origin of this  $^{60}\text{Fe}$ . The proton-rich nuclide  $^{53}\text{Mn}$  is generated primarily during so called silicon burning before the supernova explosion. During this process also  $^{53}\text{Fe}$  is formed [15] that decays in a few minutes to  $^{53}\text{Mn}$  and both end up eventually as stable  $^{53}\text{Cr}$ . Until now,  $^{53}\text{Mn}$ , formed by nucleosynthesis, has not been detected in the interstellar space because  $^{53}\text{Mn}$  decays by electron capture directly to the ground state of  $^{53}\text{Cr}$ , hence only low-energy x-rays are emitted, not detectable by space borne detectors. This is different to isotopes like  $^{60}\text{Fe}$  [16] or  $^{26}\text{Al}$  [17] whose 1173keV and 1333keV ( $^{60}\text{Fe}$ ), or 1809 keV ( $^{26}\text{Al}$ ) gamma lines following  $\beta$ -decay have been observed in the Galaxy.

A feasible way to detect  $^{53}\text{Mn}$  in Earth's reservoirs is, as in the case of the finding of  $^{60}\text{Fe}$ , direct atom counting by accelerator mass spectrometry (AMS). Unfortunately, and in contrast to  $^{60}\text{Fe}$ , the dominate fraction of the  $^{53}\text{Mn}$  in the solar system is produced in dust that originates from asteroid collisions or comets. Individual contributions [18], and the absolute amounts deposited on Earth are still under debate. In these dust particles,  $^{53}\text{Mn}$  is mainly formed by cosmic ray reactions mostly on iron via the nuclear reactions  $^{\text{nat}}\text{Fe}(p,x)^{53}\text{Mn}$  or  $^{\text{nat}}\text{Fe}(n,x)^{53}\text{Mn}$  (with neutrons from the secondary component of the cosmic rays) and on nickel. However, due to its lower abundance, nickel contributes only less than 5% [19, 20]. Compared to interplanetary  $^{53}\text{Mn}$  influx on Earth the  $^{53}\text{Mn}$  influx via interstellar dust [21] represents only a very small contribution, and makes its detection challenging considering that the overwhelming bulk of  $^{53}\text{Mn}$  is cosmogenically produced within the Solar System.

Reservoirs collecting the  $^{53}\text{Mn}$  from the dust, and preserving time information of the flux over millions of years, are ferromanganese crusts [22] and seafloor sediments [23]. As the terrestrial  $^{55}\text{Mn}$  abundance remains constant over time in these reservoirs, knowledge of  $^{53}\text{Mn}/^{55}\text{Mn}$  ratios in dated layers of such archives reveals the dust flux over very long periods. Here, we have focused on

ferromanganese crusts. Applying AMS we measured time profiles of  $^{53}\text{Mn}/\text{Mn}$  isotope ratios over time periods of more than 10Myr. The dating of the different layers in crusts was performed by means of  $^{53}\text{Mn}$  and  $^{10}\text{Be}$  [24].  $^{10}\text{Be}$  is formed by cosmic-ray induced nuclear reactions on nitrogen and oxygen in Earth's atmosphere at approximately constant rates over Myr time-periods and becomes eventually incorporated into deep-ocean crusts. It has a half-life of 1.387 My [26, 27]. In contrast to  $^{53}\text{Mn}$ ,  $^{10}\text{Be}$  is routinely measured with AMS with high precision. There have been three previous attempts to measure extraterrestrial  $^{53}\text{Mn}$  [23, 28, 29], but none measured a time dependence.

At present, only AMS systems with high-energy accelerators in conjunction with a gas-filled magnet have the ability to suppress the interfering isobar  $^{53}\text{Cr}$  sufficiently to determine  $^{53}\text{Mn}/\text{Mn}$  ratios at levels down to about  $10^{-14}$  in milligram amounts of sample material.  $\text{MnO}^-$  beams are extracted from an ion source. The ions are mass separated and accelerated to energies of ~150 – 180 MeV and  $^{53}\text{Mn}$  finally detected as single events in an energy-sensitive particle detector. Details of the measurements can be found elsewhere [30].

We measured four different crusts for their  $^{53}\text{Mn}$  content, all of them of hydrogenetic origin, and from different locations in the Pacific Ocean. Two, 29DR-32 and 29DR-45, were from the Midway Atoll (28°13'N, 177°22'W) from depths of 2938 and 1589 m [31], respectively. One, 4DR [32], from the Donizetti Ridge (32°14,99'N, 159°56,99'W from a depth of 5120 m) and the fourth, (237KD), from the Central Pacific (9°18'N, 146°03'W) from a depth of 4830 m [33].

Typical mass concentrations in the crusts were about 20% for Mn and in the range of 10 - 15 ppm for Cr. The Mn fraction was extracted using chemical treatments as described in [30]. Thus, around 100 mg of crust material yielded ~15 - 20 mg  $\text{MnO}_2$  sample material available for the subsequent AMS measurements. Around 15 samples (each about 100 mg crust material) representing 2 mm depth intervals (and representing ~1Myr time integral) were taken from each crust, which resulted in more than 60 samples totally. The measured isotope ratios  $^{53}\text{Mn}/\text{Mn}$  are plotted in Fig. 1 and listed in the supplement [34]. In addition, also iron had been separated for two crusts for the determination of  $^{60}\text{Fe}/\text{Fe}$  ratios as reported in ref. [7] and [11].

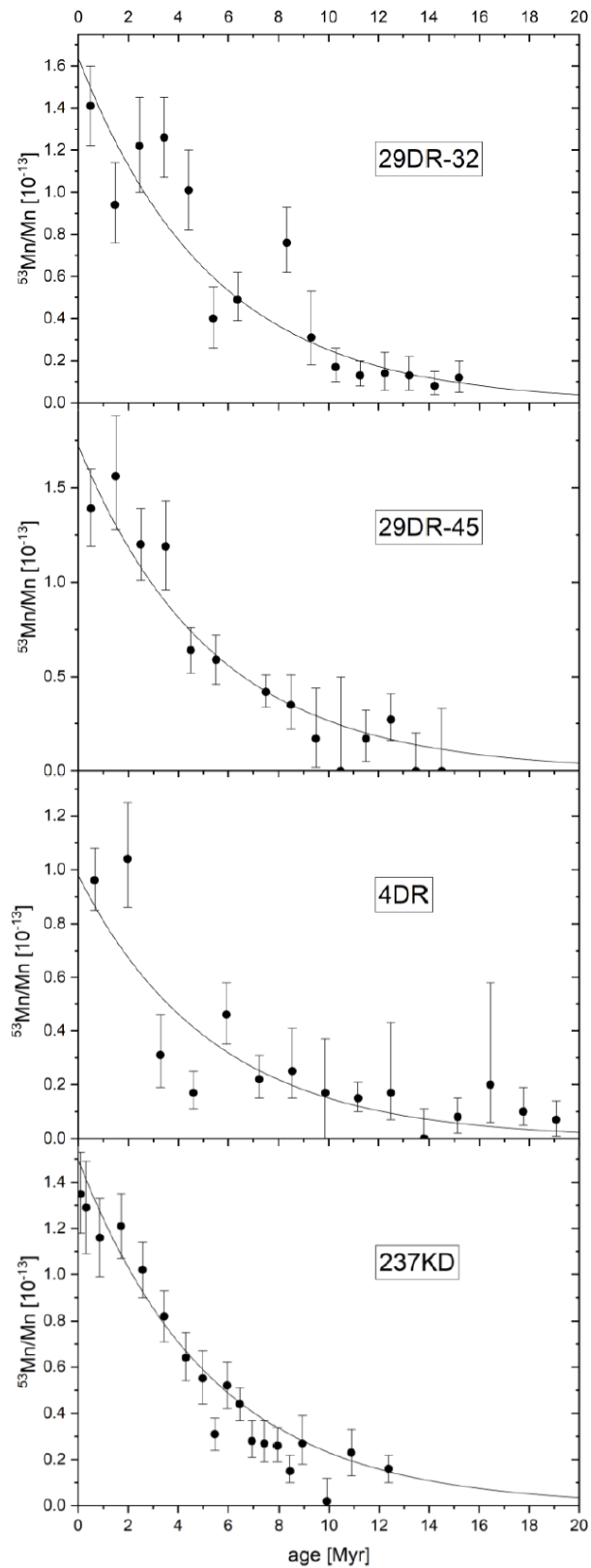


Fig. 1 shows the measured isotope ratios  $^{53}\text{Mn}/\text{Mn}$  at different depths/ages in the crusts. Top to bottom are for the four crusts from the Pacific: the two from Midway Atoll, (29DR-32 and 29DR-45), one from the Donizetti Ridge (4DR) and one from Central Pacific (237KD). Error bars for small event numbers were calculated following the prescription of Feldman and Cousins [35]. The  $1\sigma$ -limits are drawn.

The crust 237KD, where the enhanced  $^{60}\text{Fe}/\text{Fe}$  ratio about 2.5 Myr ago was reported [7], was dated with  $^{10}\text{Be}$  [25]. We used these data but the improved value of the  $^{10}\text{Be}$  half-life of 1.387 Myr [26,27] for the time scale given in Fig. 1. The  $^{53}\text{Mn}/\text{Mn}$  ratios of all crusts were fitted to the function

$$C = C_0 \cdot \exp(-d \cdot \ln 2 / (gr \cdot T_{1/2}))$$

Here,  $C$  is the ratio at depth  $d$ ,  $C_0$  the result of the fit at the surface,  $gr$  is the growth rate and  $T_{1/2} = 3.74$  Myr [14] the half-life of  $^{53}\text{Mn}$ . Then, the age of a sample equals  $d/gr$  and the uncertainty of the exponent is subsumed into the uncertainty of  $gr$ . The results of the least-squares fits for the two adjusted parameters are listed in table 1.

crust	29DR-32	29DR-45	4DR	237KD
growth rate [mm/Myr]	2.04(25)	2.00(18)	1.52(27)	2.55(22)
$C_0$ [ $10^{-13}$ ]	1.64(19)	1.72(14)	0.98(16)	1.50(11)
$\chi^2/\text{dof}$	1.51	0.52	1.78	1.14

Table 1: Results for growth rate and initial concentration of the four crusts. The values of the normalized  $\chi^2$  are also given.

Although there are considerable fluctuations, especially at ages older than 7 Myr, all crusts show an exponential behavior of the  $^{53}\text{Mn}/\text{Mn}$  ratio as a function of depth due to the half-life of  $^{53}\text{Mn}$ . In one case the value of  $\chi^2/\text{dof}$  is considerably larger than unity.

To check the validity of dating by  $^{53}\text{Mn}$ , we compare the dating by  $^{10}\text{Be}$  with  $^{53}\text{Mn}$  for the case of the crust 237 KD. The growth rate for  $^{10}\text{Be}$  dating, was 2.32 mm/Myr in [25], but if we fit these  $^{10}\text{Be}$  data between 2mm and 12mm, where the growth rate seems constant and use the most precise half-life of  $^{10}\text{Be}$ , namely 1.387 Myr [26,27], we obtain a growth rate of 2.56(13) mm/Myr. Within the uncertainties the  $^{53}\text{Mn}$  data yield the same values for the growth rate, 2.55(22) mm/Myr, meaning that the adopted half-life value for  $^{53}\text{Mn}$  is supported. Therefore, the dating of the other three crusts was based on  $^{53}\text{Mn}$  data only.

In order to deduce the contribution of interstellar, SN-produced  $^{53}\text{Mn}$ , we select the time range between 1.5 Myr and 4 Myr where the highest  $^{60}\text{Fe}/\text{Fe}$  ratios have been found [7,9,10]. These data were compared with the concentrations obtained for younger and older time periods. The rather wide time window was chosen to allow for uncertainties in the dating of the individual samples which is mainly caused by the inhomogeneous structure of the different crusts. In a first step we calculated all  $^{53}\text{Mn}/\text{Mn}$  ratios at the time of incorporation by means of their age (i.e. corrected for radioactive decay). In a second step we merged the data from the four crusts as shown in Fig. 2.

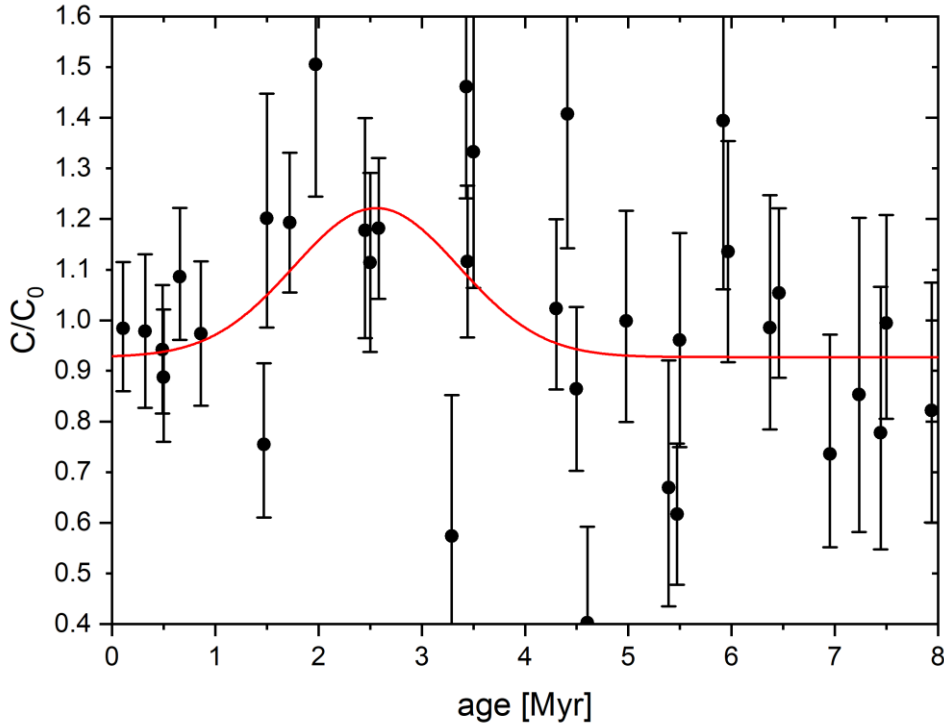


Fig.2. Merged  $^{53}\text{Mn}/\text{Mn}$  ratios ( $C/C_0$ ) from the different crusts at the time of incorporation. The red curve is the result of a fit of a Gaussian with fixed width  $\sigma=0.8$  Myr.

In the next step, we formed the weighted average of the data in the relevant time interval  $T_2$ , where the influx of  $^{60}\text{Fe}$  has been detected [6,7,9-11], from 1.5 until 4 Myr, and the adjacent intervals,  $T_1$  from 0 until 1.5 Myr and  $T_3$  from 4 Myr until 5.5 Myr. The fact that the decay-corrected concentrations at  $T_1$  and  $T_3$  are smaller than one is the result of fitting to a pure exponential and underlines the excess at  $T_2$ . The ratio of the concentration at  $T_2$  and the average at  $T_1$  and  $T_3$  yields  $\frac{2C(T_2)}{C(T_1)+C(T_3)} = 1.34 \pm 0.09$ . This is more than  $3\sigma$  larger than unity.

As an alternative, we fitted the data in Fig. 2 (from 0 to 13 Myr) to a constant  $b$  and a Gaussian

$$\frac{C}{C_0} = b + a \cdot \exp(-(t - t_p)^2 / (2\sigma^2))$$

with fixed width  $\sigma=0.8$  Myr on the basis of the  $^{60}\text{Fe}$  data [9,10], and varied the position  $t_p$  and height  $a$  as well as the base value  $b$ . The result is plotted in Fig. 2 and the parameters are:

$$b = 0.928 (39),$$

$$a = 0.29 (10),$$

$$t_p = 2.56 (33) \text{ Myr.}$$

The resulting  $\chi^2=58.9$  is larger than the 50 degrees of freedom. However, the Gaussian is probably not the correct shape of the distribution. Again, the peak has a significance of about  $3\sigma$ .

Now, we can compare our measured values with nucleosynthesis models, in order to get a hint on the SN progenitor. We assume that the  $^{53}\text{Mn}$  ejected from a SN follows the same path as the  $^{60}\text{Fe}$  until it becomes incorporated into the ferromanganese crusts. We have measured a ratio  $^{60}\text{Fe}/\text{Fe} \sim 3.4 \times 10^{-15}$  in the crusts [7] for the time range  $(2.58 \pm 0.43)$  Myr, calculated back to the time of deposition. The additional  $^{53}\text{Mn}/\text{Mn}$  ratio above the cosmogenically formed ratio is  $0.95 \cdot a \cdot b \cdot C_0 = (3.8 \pm 1.5) \times 10^{-14}$ , the factor 0.95 represents the average value of the fitted Gaussian in the same range  $\pm 0.43$  Myr as evaluated for the  $^{60}\text{Fe}$  data. As weighted average of the

four  $C_0$  values from Table 1 we obtain  $C_0 = (1.47 \pm 0.15) \times 10^{-13}$ , where the error is inflated by the square root of the normalized  $\chi^2$ . If we consider a Fe/Mn ratio in the crusts of  $\sim 0.8$  (it varies between 0.6 and 0.9 [22]), we get for the  $^{53}\text{Mn}/^{60}\text{Fe}$  ratio a value of about 14. However, the uptake factors into the crust for Mn and Fe can be quite different. In [7] the uptake factor for Mn was estimated 7 times greater than for Fe. If this were the case, the relative mass yields of  $^{53}\text{Mn}$  to  $^{60}\text{Fe}$  in the interstellar dust would be 2:1. As well, the ratio of the mass yields  $^{53}\text{Mn}/^{60}\text{Fe}$ , calculated by Woosley and Weaver [15], varies between about 1 and 20 for a SN with a mass of the progenitor star between 11 and 25  $M_\odot$  and solar metallicity. For the time being, we only can state that the observed ratio of  $^{53}\text{Mn}/^{60}\text{Fe}$  in the crusts is in the expected range.

Since the excess of  $^{53}\text{Mn}$  is detected in the same samples and time range in which  $^{60}\text{Fe}$  has been identified, it confirms the SN origin of that  $^{60}\text{Fe}$ . Thus,  $^{53}\text{Mn}$  is the second radioisotope from the same SN where  $^{60}\text{Fe}$  has been detected, and it is for the first time that  $^{53}\text{Mn}$ , formed by nucleosynthesis during a SN, has been found. This finding might initiate further searches in deep sea sediments or manganese crusts for long lived radionuclides, like the proton-rich nuclei  $^{92}\text{Nb}$  ( $T_{1/2} = 35$  Myr) or  $^{146}\text{Sm}$  ( $T_{1/2} = 68$  Myr), to corroborate nucleosynthesis calculations. Up to now, the search for SN-produced  $^{26}\text{Al}$  only resulted in an upper limit [36].

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