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The Dyadic Radionuclide System ^{60}Fe / ^{53}Mn to Distinguish Interstellar from Interplanetary ^{60}Fe

Dominik Koll[1,2,3,*], *Thomas* Faestermann[3], *Gunther* Korschinek[3], *Ingo* Leya[4], *Silke* Merchel[2,5], and *Anton* Wallner[1,2]

[1]NPAA, Research School of Physics, The Australian National University, Canberra, ACT 2601, Australia.
[2]Accelerator Mass Spectrometry & Isotope Research, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany.
[3]Department of Physics, Technical University of Munich, 85748 Garching, Germany.
[4]Physical Institute - Space Research and Planetology, University of Bern, 3012 Bern, Switzerland.
[5]Isotope Physics, Faculty of Physics, University of Vienna, 1090 Vienna, Austria.

Abstract. The discovery of live ^{60}Fe in a deep-sea crust with proposed interstellar origin followed by evidence for elevated interplanetary ^{3}He in the same crust raised the question on how to unambiguously identify the true production site of the identified ^{60}Fe. Here, we show the implementation of the dyadic radionuclide system ^{60}Fe / ^{53}Mn to serve as a tool for the identification of surplus interstellar ^{60}Fe over interplanetary production. The recent updates in experimental ^{60}Fe and ^{53}Mn data from iron meteorites as well as in production rate models confirm the validity and robustness of this dyadic system for future applications.

1 Introduction

Direct detection of freshly-synthesized supernova nuclides is one of the gold standards to study stars and their impact on the solar system. The radionuclide ^{60}Fe proved to be an excellent candidate because of its relatively long half-life of 2.6 Myr [1, 2], allowing accumulation and detection over several million years.

The first indication of live ^{60}Fe in a deep-sea ferromanganese crust [3] followed by the clear discovery in a 2 Myr old layer [4] raised the question on its astrophysical origin. The proposed supernova (interstellar) origin [4] was subsequently challenged. Elevated levels of ^{3}He in the same crust were used as an argument to propose that the discovered ^{60}Fe was produced by cosmic-ray spallation in (micro-) meteorites and not by stars [5]. Further evidence for a global ^{60}Fe signal [6] was used as an indirect argument for the supernova hypothesis. ^{60}Fe was subsequently identified on timescales of years, thousands of years and millions of years in several archives, see [7] for a recent summary.

The clear identification of the astrophysical production site of any discovered ^{60}Fe requires the exclusion of anthropogenic ^{60}Fe, which was conclusively presented in [8]. Cosmogenic production is governed by cosmic-ray fluxes, target sizes and compositions, and production rates for radionuclides by primary and secondary cosmic rays outside Earth's atmosphere. The most viable candidates to establish cosmic-ray production rates of ^{60}Fe are meteorites, which could be used to distinguish between a rather constant cosmic dust influx plus single large object events such as meteoroids, comets or asteroids and solitary supernova ^{60}Fe influxes.

It is critical to deduce the absolute amount of interplanetary dust influx into a sample. This makes it particularly challenging, because a constant background flux of ^{60}Fe bearing cosmic dust modulated by single large events can mimic supernova influxes. Note, for deep-sea crusts we integrate over several hundred thousand to millions of years, which makes rare events like meteorite falls even over global scales probable. Therefore, a tool to unambiguously identify interstellar over cosmogenic ^{60}Fe is needed. Here, we show that by additionally measuring the long-lived radionuclide ^{53}Mn ($t_{1/2} = 3.7$ Myr), the dyadic radionuclide system ^{60}Fe / ^{53}Mn could be used to overcome these problems and to disentangle interstellar from interplanetary ^{60}Fe influxes. It is worth mentioning that one can estimate the maximum cosmogenic ^{60}Fe concentration in a sample by measuring the stable target element Ni and assuming all of the Ni present is extraterrestrial Ni. This estimation is crude but practical for large ^{60}Fe influxes and low stable Ni abundances and fails for samples with low extraterrestrial but high terrestrial influxes. The hereby proposed system is applicable in any case.

2 Cosmogenic ^{60}Fe and ^{53}Mn from meteorites

In the last few years, we were able to establish a substantial meteorite database for both radionuclides ^{60}Fe and ^{53}Mn using accelerator mass spectrometry (AMS) [9]. AMS is the most sensitive experimental approach to directly determine concentrations of both radionuclides in terrestrial and extraterrestrial samples. Radiochemical neutron activation analysis

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[*] e-mail: d.koll@hzdr.de

and low-level counting have been used in the past to measure both radionuclides in meteorites but are not sensitive enough to search for supernova traces. For ^{60}Fe in particular, one needs to consider waiting times of decades after chemical purification for the in-growth of the daughter nucleus ^{60}Co to achieve secular equilibrium [10]. Note, large databases for the isotope ^{53}Mn exist without ^{60}Fe data [11]. AMS does not require any waiting time since live radionuclides are detected and not their decay. Currently, the only AMS facility in the world capable of measuring both radionuclides at natural levels is the Heavy Ion Accelerator Facility of ANU [2]. The Munich accelerator lab [12], which was pioneering the measurements of both radionuclides using a gas-filled magnet, has been closed in 2020. Clearly, there is the need for existing or new labs to establish new capabilities for ^{60}Fe and ^{53}Mn AMS.

The database with more than 30 single iron meteorites is complemented by an updated production rate model based on the INCL (Liège Intra Nuclear Cascade) code [9]. The predicted production rate ratio is $^{60}\text{Fe}/^{53}\text{Mn} = (2.5 \pm 0.1) \times 10^{-3}$ in units of dpm kg^{-1}[Ni] / dpm kg^{-1}[Fe]. The common units of production rates are specific activities in disintegrations per minute per kg of target element or target mass, in this case stable Ni for ^{60}Fe and stable Fe for ^{53}Mn. The stable element content in terrestrial samples is dominated by terrestrial elements. The specific activity as a measure for the interplanetary influx is therefore not suitable, because the extraterrestrial target element concentration is not quantifiable. Hence, it is necessary to estimate reasonable values of extraterrestrial Ni / Fe in terrestrial samples. Importantly, only the ratio of stable Ni to Fe is needed and not the concentration of both elements. For terrestrial samples, the amount of cosmogenic radionuclides present is much smaller compared to extraterrestrial samples, where intrinsic production is dominant. This relaxes the requirement on the exact knowledge of Ni / Fe.

Using the half-lives of 2.6 Myr and 3.7 Myr, we back-calculate a half-life and target composition independent and directly measurable production rate ratio of $^{60}\text{Fe}/^{53}\text{Mn} = (1.77 \pm 0.07) \times 10^{-3} \times \text{Ni}/\text{Fe}$ in units of at / at and wt.%. By factoring out Ni / Fe, the data becomes comparable between samples with different target element concentrations. This allows us to compare the model prediction with extraterrestrial samples (known extraterrestrial Ni / Fe) and with terrestrial archives (unknown extraterrestrial Ni / Fe). The possible values for extraterrestrial Ni / Fe in terrestrial samples are conservatively estimated to be less than 1. Solar system, chondritic and meteorite abundances show the clear trend of $\text{Ni} \ll \text{Fe}$, most commonly $\text{Ni}/\text{Fe} = 0.055$ [13]. Iron meteorites, which are metal-rich and elevated in Ni, generally show a higher Ni / Fe of around 0.1. For comparison, Earth's crust has a low Ni to Fe ratio of 0.002, whereas the ratio in sea water is 0.28, but with low concentration of both metals [14]. Using the most common abundances of stable elements in the solar system, the cosmogenic ^{60}Fe to ^{53}Mn atomic ratio obtained in a measurement would be around 10^{-4}.

In contrast to that, the stellar atomic nucleosynthesis ratio is estimated to be 10^{0} - 10^{-2} [15]. The hypothetical interstellar ^{60}Fe would dominate over cosmogenic ^{60}Fe as an excess, whereas any supernova ^{53}Mn is only a fraction of the bulk cosmogenic ^{53}Mn. This makes any accompanied supernova ^{53}Mn challenging to identify over cosmogenic background with only one attempt so far by AMS [16]. Therefore, interstellar ^{60}Fe would appear as an enhanced ^{60}Fe to ^{53}Mn ratio over the cosmogenic baseline, which is dominated by cosmogenic ^{53}Mn.

3 Interstellar ^{60}Fe identification

The very first utilization of this principle happened alongside the first indication of supernova ^{60}Fe in a deep-sea crust [3]. There, it was already estimated, that the atomic ratio of cosmogenic ^{60}Fe to ^{53}Mn should be in the order of 10^{-4}. This strong experimental argument was not adopted in the following studies, possibly due to limited data on extraterrestrial production rates and meteorite samples. Seventeen years later in 2016, this dyadic radionuclide system yielded evidence for supernova ^{60}Fe on the surface of the Moon [17]. Lunar regolith was analyzed for ^{60}Fe and ^{53}Mn and compared to four different meteorites before the database was significantly extended. The specific activity ratio was calculated, since the lunar surface itself is the target for the cosmic ray interactions. The meteorite activity ratio was determined to be $^{60}\text{Fe}/^{53}\text{Mn} = (2.68 \pm 0.35) \times 10^{-3}$ in units of dpm kg^{-1}[Ni] / dpm kg^{-1}[Fe]. This agrees perfectly with the current model value of $(2.5 \pm 0.1) \times 10^{-3}$ and the meteorite database value of 2.6×10^{-3}. The lunar surface ratio was determined to be $(10 - 50) \times 10^{-3}$, much higher than for meteorites. The measured atomic $^{60}\text{Fe}/^{53}\text{Mn}$ of lunar material would be misleading because of the low lunar Ni / Fe of 0.001 - 0.01, which is similar to the ratio on Earth and significantly lower than for meteorites. The specific activity reveals the clear enhancement, which shows that for lunar material the knowledge of Ni / Fe is crucial.

More recently, ^{60}Fe was discovered in Antarctic surface snow [8]. Besides ruling out anthropogenic production, the identification of its astrophysical origin was important since the Antarctic snow covered only the most recent 20 years of terrestrial history and could therefore reveal an influx right at this moment. Here, the specific activity ratio approach was not possible, because the stable elements in the sample are of terrestrial origin. The same meteorite data from [17] was converted to atomic ratios with a value of $^{60}\text{Fe}/^{53}\text{Mn} = (1.9 \pm 0.2) \times 10^{-3} \times \text{Ni}/\text{Fe}$ in units of at / at and wt.%. Similarly, this again agrees perfectly with the outlined model value of $(1.77 \pm 0.07) \times 10^{-3} \times \text{Ni}/\text{Fe}$ and the meteorite database value of $1.8 \times 10^{-3} \times \text{Ni}/\text{Fe}$. The measured ^{60}Fe to ^{53}Mn ratio in Antarctic snow was around 17×10^{-3}. The extraterrestrial Ni / Fe was conservatively estimated to be less than 1. This yields a more than two orders of magnitude enhancement over cosmogenic background for chondritic composition $\text{Ni}/\text{Fe} = 0.055$. There is still a clear enhancement even for the extreme case of $\text{Ni}/\text{Fe} = 1$. The interstellar origin of the discovered ^{60}Fe becomes now even clearer with the updated model and the significantly extended meteorite database (Figure 1).

Table 1: Summary of ^{60}Fe / ^{53}Mn ratios from the theoretical model, the meteorite database and the samples lunar regolith as well as Antarctic snow. Depending on the origin of the analyzed sample, one has to compare any measured data with the first or the second ^{60}Fe / ^{53}Mn ratio for extraterrestrial or terrestrial samples, respectively. The unit dpm / kg X refers to the specific activity ratio with X being the respective target element for the production of ^{60}Fe or ^{53}Mn. In particular for lunar regolith, the measured atomic ratio is misleading because of the low intrinsic Ni / Fe. ^{60}Fe / ^{53}Mn ratios around 10^{-3} indicate cosmogenic production, whereas higher ratios indicate a surplus of interstellar ^{60}Fe.

	Sample	**Ni / Fe**	^{60}**Fe /** ^{53}**Mn** [dpm / kg X]	^{60}**Fe /** ^{53}**Mn** [at/at] × Ni / Fe
Leya *et al.* [9]	Model	. . .	$(2.5 \pm 0.1) \times 10^{-3}$	. . .
This work	based on [9]	. . .	. . .	$(1.77 \pm 0.07) \times 10^{-3}$
Knie *et al.* [18]	Dermbach Met.	0.91	2.3×10^{-3}	1.6×10^{-3}
Knie *et al.* [18]	Emery Met.	0.09	2.6×10^{-3}	1.8×10^{-3}
Ott *et al.* [19]	Gebel Kamil Met.	0.24	2.7×10^{-3}	1.9×10^{-3}
Fimiani *et al.* [17]	NWA 6369 Met.	0.07	2.2×10^{-3}	1.5×10^{-3}
Leya *et al.* [9]	Database (27 Met.)	0.1	2.6×10^{-3}	1.8×10^{-3}
				[at/at]
Fimiani *et al.* [17]	Lunar regolith	0.001 - 0.01	$(10 - 50) \times 10^{-3}$	$10^{-5} - 10^{-4}$
Koll *et al.* [8]	Antarctic snow	. . .	. . .	17×10^{-3}

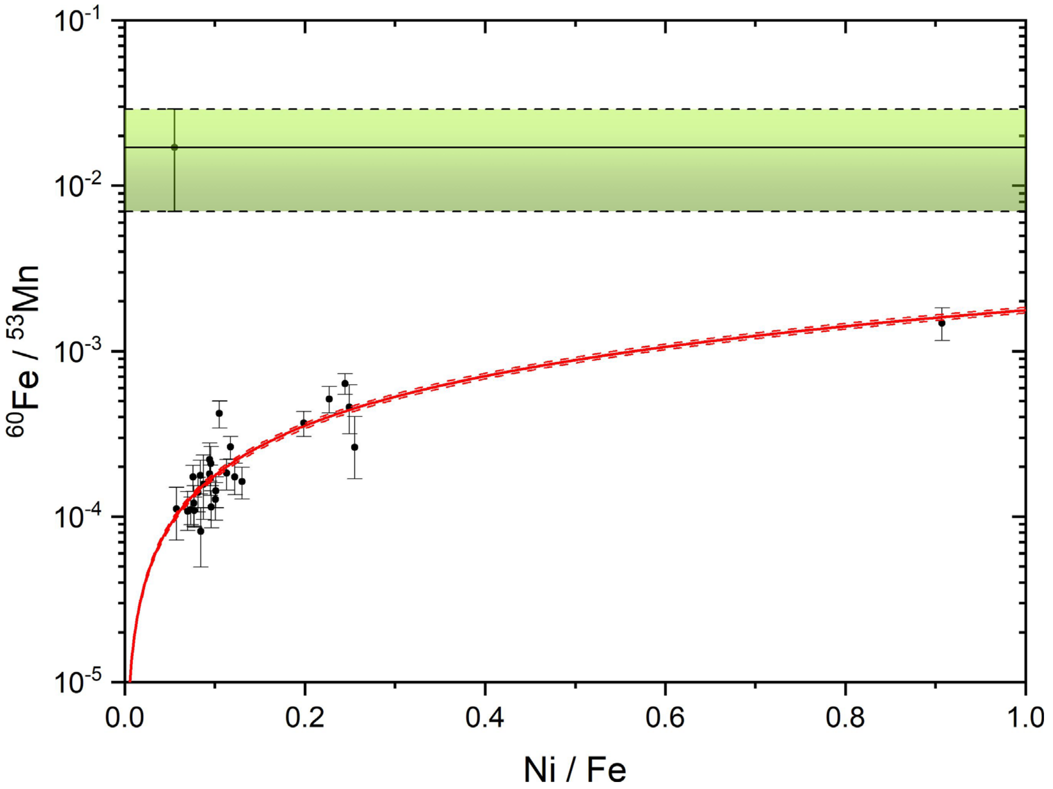

Figure 1: Antarctic snow data [8] in green with the updated, independent meteorite model in red and available meteorite data in black [9]. The enhancement of ^{60}Fe over the expected cosmogenic ratio is clearly visible. The datapoint is exemplarily displayed for the most likely stable element abundance Ni / Fe = 0.055.

By establishing a significant meteorite database for both radionuclides and having an independent model, the dyadic radionuclide system ^{60}Fe/^{53}Mn is now robust enough to be widely applied for the search of interstellar ^{60}Fe in any terrestrial or extraterrestrial archive. The model values for the production rate ratios are in very good agreement with now 31 single meteorite measurements, see Table 1. This system allows to unambiguously identify supernova produced ^{60}Fe over cosmogenic background.

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