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Assessment of ⁵³Mn deposition on Earth via accelerator mass spectrometry

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HIGHLIGHTS

- A novel approach to assess the ⁵³Mn deposition on Earth is proposed.
- 1400 l of snow from Antarctica were chemically processed to extract Mn and Be.
- ⁵³Mn/⁵⁵Mn and ¹⁰Be/⁹Be isotope ratios were measured by accelerator mass spectrometry.
- An upper limit for the ⁵³Mn global flux is established.

ABSTRACT

The ⁵³Mn flux onto Earth is a quantity relevant for different extraterrestrial and astrophysical questions. It is a proxy for related fluxes, such as supernova-produced material or interplanetary dust particles. In this work, we performed a first attempt to assess the ⁵³Mn flux by measuring the ⁵³Mn/¹⁰Be isotopic ratio in a 1400 L sample of molten Antarctic snow by AMS (Accelerator Mass Spectrometry). Using the ¹⁰Be production rate in the atmosphere, an upper limit of 5.5×10^3 atoms cm⁻² yr⁻¹ was estimated for the deposition of extraterrestrial ⁵³Mn. This result is compatible with one of the two discrepant values existing in the literature.

1. Introduction

Earth continuously receives solid extraterrestrial material originating predominantly from asteroids, debris, and comets sublimation. These objects are usually classified by their size and most of them are in the form of interplanetary dust particles (IDPs) with radii between 5 and 250 μ m (Cremonese et al., 2012). The radionuclides produced in these unshielded particles –through nuclear reactions induced by cosmic rays- are excellent tracers for the deposition of IDPs on Earth. In particular, ⁵³Mn is a long-lived radionuclide (T_{1/2} = 3.7 Myr; Honda and Imamura, 1971) mainly produced through ^{nat}Fe(p, x) and ^{nat}Ni(p, x) reactions (Merchel et al., 2000). ⁵³Mn is poorly produced on Earth (see Section 4.2), and that is the reason why it is probably the most useful tracer to investigate several astrophysical issues. Two of them are the assessment of interstellar material flux produced by supernovae and the determination of the total IDPs flux entering into Earth.

The deposition of supernova-produced material has been studied with 60 Fe (T_{1/2} = 2.6 Myr; Wallner et al., 2015) depth profiles in ocean ferromanganese crust by Knie et al., (1999, 2004). In these studies, the major uncertainty to estimate the flux of 60 Fe comes from the uptake of iron from the ocean water into the ferromanganese crust. The iron uptake factor can be calculated relative to the manganese uptake factor, which can be assessed by the 53 Mn flux (Knie et al., 2004; Fitoussi et al., 2008).

The 53 Mn flux is also useful for the assessment of IDPs influx, which can be estimated by means of the ratio between the 53 Mn global flux and its concentration in IDPs, providing a new and independent approach. The amount of accreted IDPs is today still poorly constrained,

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with an estimated range from 2 to 100 kt per year, obtained by different techniques (Plane, 2012 and references therein). IDPs fallen to Earth are accessible samples of extraterrestrial dust within the solar system. Moreover, the study of IDPs in geological environments may provide a retrospective insight into the solar system evolution (Genge et al., 2017 and references therein).

Decay counting techniques are unable to reach the necessary sensitivity to quantify the 53 Mn content. To overcome this difficulty, thermal neutron activation analysis has been used (Bibron et al., 1974; Imamura et al., 1979; Herpers, Sarafin, 1987). However, Accelerator Mass Spectrometry (AMS) –the most sensitive technique for the measurement of 53 Mn (Knie et al., 2004; Poutivtsev et al., 2010)– offers an alternative approach.

In this work, we propose to determine the 53 Mn global flux assuming that the 53 Mn/ 10 Be production ratio is roughly constant worldwide. The normalization with the 10 Be content circumvents uncertainties on atmospheric transport, deposition patterns, snow accumulation or erosion rates. As a proof of principle, we applied the proposed approach to 1400 L of molten snow collected at the Argentinian Carlini base in Antarctica. This location is close to the latitudinal range (50–60°S) where the flux of both 53 Mn and 10 Be is strongly enhanced (Dhomse et al., 2013).

2. Transport and deposition model

While entering the atmosphere, IDPs undergo a series of processes which include meteoric ablation and vaporization of the 90% on average of their mineral constituents (Taylor et al., 1998; Vondrak et al., 2008). The resulting metallic vapors form oxides or hydroxides and are polymerized to nanometer-sized particles known as meteoric smoke particles (MSPs).

From a general circulation model applied to the transport of MSPs to the lower stratosphere, Saunders et al. (2012) concluded that the MSPs are rapidly carried down to the stratosphere through the polar vortex, where they are assimilated by sulphate aerosols. Then, they are redistributed to middle-latitudes in the stratosphere. Finally, they enter the troposphere and are preferentially deposited on the Earth surface at latitudes between 50°S and 60°S preferentially. For a discussion of recent MSP transport see Dhomse et al. (2013).

In order to trace the atmospheric transport, deposition, snow accumulation and erosion processes, it is advantageous to use another cosmogenic long-lived radionuclide. ¹⁰Be is mainly produced by spallation of N and O nuclei present in the atmosphere (stratosphere and troposphere) with a global production rate of 6.6×10^5 atoms cm⁻² yr⁻¹ (Masarik and Beer, 2009). ¹⁰Be from extraterrestrial origin can be dismissed in comparison with the total ¹⁰Be inventory (McHargue and Damon, 1991).

¹⁰Be produced in the stratosphere has a residence time around 12–16 months in this atmospheric layer before entering troposphere, while ¹⁰Be in the troposphere is only three weeks (McHargue and Damon, 1991) (Fig. 1).

The MSPs meridional mixing, the ¹⁰Be attachment to ambient aerosols, and the long residence time of these aerosols, all processes taking place in the stratosphere, allow us to assume a uniform stratospheric ${}^{53}\text{Mn}/{}^{10}\text{Be}$ ratio worldwide. On the other hand, the residence time of ${}^{10}\text{Be}$ in the troposphere is too short to assume complete latitudinal mixing. So, when aerosols enter this atmospheric layer, the ${}^{53}\text{Mn}/{}^{10}\text{Be}$ ratio is altered and is no longer independent of the latitude.

The model proposed in this work assesses 53 Mn flux considering two different processes: a) the 53 Mn and 10 Be mixing in the stratosphere that allows to relate 53 Mn flux with the 10 Be global atmospheric production rate and then, b) the atmospheric transport and deposition that allows to relate the stratospheric 10 Be contribution with the local 10 Be deposition.

Following the arguments given above, we can assume that 53 Mn follows the same transport mechanisms as stratospheric 10 Be, therefore

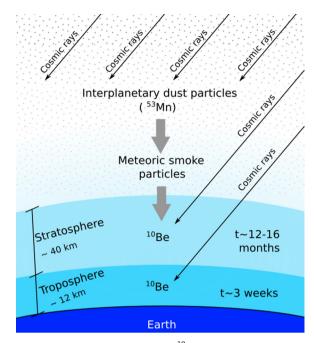


Fig. 1. Atmosphere layers' scheme where ¹⁰Be is produced. The entry of extraterrestrial ⁵³Mn produced in IDPs is also represented.

the ¹⁰Be tropospheric contribution must be subtracted from the global ¹⁰Be production rate. Thus, considering that 65% of the ¹⁰Be production takes place in the stratosphere, and the remaining 35% is produced in the troposphere (Heikkilä et al., 2009), the ¹⁰Be stratospheric production rate $Pr_s(^{10}Be)$ yields 4.3 × 10⁵ atoms cm⁻² yr⁻¹.

On the other hand, to subtract the contribution of tropospheric ¹⁰Be deposited at the sampling site, it has to be considered that according to Heikkilä et al. (2009) only 70% of the ¹⁰Be deposited in that site comes from the stratosphere. Hence, the measured ¹⁰Be concentration must be multiplied by a factor f = 0.7 to get rid of the ¹⁰Be originating in the troposphere that was deposited at the sampling site. Then, the ⁵³Mn flux onto Earth can be determined as

$$\Phi(^{53}Mn) = [(^{53}Mn \times Pr_s(^{10}Be)/(^{10}Be \times f)]/A$$
(1)

where A = 0.9 represents the fraction of ablated and vaporized IDPs entering the atmosphere. Thus, the global ⁵³Mn deposition rate on Earth can be assessed from the ⁵³Mn/¹⁰Be measured ratio in the snow sample.

3. Experimental procedure

3.1. Sampling

We studied a snow sample collected from Isla 25 de Mayo (King George Island) near the Antarctic Peninsula, in the ice field close to the Argentinian Carlini Base ($62^{\circ}14'18''S 58^{\circ}40'00''W$). This area presents a net average accumulation rate (water equivalent, w.e.) of 590 mm yr⁻¹ (Rückamp et al., 2011). For the sampling year (2011), the weather station at the base has reported an annual precipitation rate of 330 mm w.e (SMN, 2016). The snow pit was about 1 m deep with a surface area of 4.5 m^2 . The latitude of the sampling site ($62^{\circ} 14' \text{ S}$) is close to the latitudinal range where the deposition of MSPs is most effective (50–60°S) (Dhomse et al., 2013).

The high precipitation rate at the sampling site in comparison to higher latitudes in Antarctica implies a dilution of the 53 Mn content. Thus, a large amount of snow (1400 L) was collected to achieve sufficient sensitivity for the detection of 53 Mn. The snow was gathered with a plastic shovel, transported to the base and stored in polyethylene bags. After melting the snow in plastic trays, it was stored in seven

Table 1

Mean average concentration values (given in ppb) for some stable isotopes present in the sample measured by ICP-MS.

Ве	Mn	Fe	Ni
0.016(2)	3.24(2)	18.27(45)	0.080(4)

200 L polyethylene tanks.

3.2. Chemical processing

The 1400 L of molten snow were chemically processed at the TANDAR Laboratory in Buenos Aires. To allow the AMS measurement of the ¹⁰Be and ⁵³Mn content in this sample, the chemical treatment had to extract beryllium and manganese from the sample. On the other hand, boron and chromium contained in the sample had to be suppressed, as ¹⁰B and ⁵³Cr are, respectively, interfering stable isobars of ¹⁰Be and ⁵³Mn. Be, Mn, Fe and Ni were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). These results are shown in Table 1.

Thus, the entire sample contained approximately 23 µg of beryllium and 4.5 mg of manganese. Manganese amount is close to the minimum necessary for a carrier-free chemical procedure and AMS measurement. A larger amount of stable manganese would imply a ⁵³Mn/⁵⁵Mn ratio below the current AMS detection limit (⁵³Mn/⁵⁵Mn > 10⁻¹⁴; Poutivtsev et al., 2010).

For the ¹⁰Be/⁹Be AMS measurements an extra amount of 5.96(1) mg of beryllium was added to the entire sample. The isotopic ratio of the added Be material was ¹⁰Be/⁹Be = 9.16(76) $\times 10^{-15}$, measured at VERA (Steier et al., 2004).

Since we are interested in the main source of 53 Mn –the ablated component of IDPs– the entire sample was passed through a 1 µm poresize polypropylene filter to ensure that only this fraction of IDPs remained in the solution. The filtration excludes coarse particles such as the non-ablated IDPs fraction which represent a small contribution of 10% to the 53 Mn inventory and erosion debris of continental origin which contains 53 Mn-poor manganese. A contribution of the latter would have strongly decreased the sample 53 Mn/ 55 Mn ratio below the AMS detection limits. Thus, even if the measured 53 Mn/ 55 Mn ratio may not strictly reflects the actual ratio of the extraterrestrial material, the 53 Mn/ 10 Be ratio of the ablated material –from which the 53 Mn deposition is derived– should not be affected by the filtration procedure.

The sample volume was acidified adding seven liters of concentrated high-purity hydrochloric acid, reaching a concentration of 0.05 M HCl and it was then reduced using ten parallel ion exchange columns (5 cm in diameter), each of these containing 5 cm of Dowex 50WX8, 100-200 mesh cations resin. Mn, Be and the other cations were eluted with 4 L of 6 M HCl, with a recovery of \sim 80% (determined through the concentrations measured by ICP-MS before and after the elution). In this way, the cations present in the 1400 L sample were concentrated in a 4L acidic solution. For a further extraction, the remaining sample was split by precipitation with NH₃ at pH~9 into two different fractions: a precipitate containing Be (and Al, Fe, etc.) and the aqueous fraction containing Mn (and Ca, Ni, etc.). The separation of Be and Mn from the other elements in each fraction was performed by ion exchange columns according to the procedure described by Merchel and Herpers (1999) adapted to the present sample size. Iron was extracted before the separation step using diisopropyl ether to prevent saturation of the ion exchange resins. Manganese was precipitated as MnO(OH)₂ from solution with KClO₃, washed three times with distilled water (18.2 MΩ cm), dried in an oven at 80 °C for 2 h, and then heated in a furnace at 500 °C for 2 h. The level of the interfering stable isobars of ¹⁰Be and ⁵³Mn (¹⁰B and ⁵³Cr) was reduced by means of this chemical procedure down to ppm levels. To assure thermal and electrical conductivity in the ion source, the sample material was mixed with Ag powder, in about 1:1 proportion by volume and pressed into two Ag sample holders.

3.3. Accelerator mass spectrometry measurements

The measurement of the 53 Mn/ 55 Mn ratio was performed using the Munich 14-MV tandem accelerator, which is able to provide the necessary energy for the suppression of the stable isobar ⁵³Cr by means of the gas-filled magnet GAMS (Knie et al., 1997). In this gas, Cr and Mn isobars have different mean charge states due to electron-exchange collisions and therefore follow different trajectories inside the magnet. In addition, an ionization chamber with segmented anode provides position-angle signal and discrimination in atomic number by the measurement of partial energy losses (Poutivtsev et al., 2010). For assessing the contribution of ⁵³Cr to the background a MnO₂ sample spiked with 1000 ppm of chromium was used, resulting in a suppression factor of 10⁻⁶. The sample (divided in two holders) yielded mean ⁵⁵MnO⁻ currents of about 100 nA after the first mass selection, running for two hours until exhaustion. A terminal voltage of 12.3 MV was employed to accelerate ⁵³Mn¹¹⁺ ions to an energy of 145 MeV. After applying all software gates, two events of ⁵³Mn could be identified. The ⁵³Mn concentration was finally determined relative to a standard sample (GRANT GLS), which has a ⁵³Mn/⁵⁵Mn ratio of 2.83(14) $\times 10^{-10}$

The ${}^{10}\text{Be}/{}^{9}\text{Be}$ ratio was measured at the Spanish Accelerator for Radionuclides Analysis (SARA), the 1 MV AMS facility at the Centro Nacional de Aceleradores in Seville, Spain (Ruiz-Gómez et al., 2010).

The ¹⁰B isobaric interference was suppressed by a factor of $\sim 10^5$ by using a 100 nm thick silicon nitride passive absorber placed at the entrance of a 120° electrostatic analyzer. Background contribution from residual ¹⁰B events was discriminated from ¹⁰Be by means of a two-anode ionization chamber. It is worth to note that the isotopic ratio ¹⁰Be/⁹Be of the blank material used is far below the expected ratio in the sample (¹⁰Be/⁹Be $\sim 10^{-9}$), which is well above the AMS detection limit (¹⁰Be/⁹Be $\sim 10^{-14}$) at SARA.

4. Results

4.1. AMS results

Due to the low concentration of ⁵³Mn, and despite the large volume of the sample, only two counts compatible with this radionuclide were observed. Since the expected background contribution from ⁵³Cr was determined to be 1.35 events, an upper limit for the $^{53}\mathrm{Mn}/^{55}\mathrm{Mn}$ isotopic ratio was established at 7.6 \times 10⁻¹³. This value was calculated according to the Feldman, Cousins (1998) prescription. Considering the Mn concentration (Table 1), this result means that the ⁵³Mn concentration in the sample was less than 2.7×10^4 atoms per liter, with a 1- σ confidence interval. On the contrary, the ${}^{10}\text{Be}/{}^{9}\text{Be}$ ratio in the sample was well above the detection limit, and therefore an accurate value could be achieved. After carrier correction, the isotopic ratio was determined to be ${}^{10}\text{Be}/{}^{9}\text{Be} = 3.04(12) \times 10^{-9}$, which corresponds to $3.36(14) \times 10^6$ atoms per liter of sample. Then, the upper limit for the ratio of interest results to be 53 Mn/ 10 Be < 8 × 10⁻³. It is worth to note that, since it is an upper limit, this value would not be incremented by a background correction.

4.2. ⁵³Mn global flux

Extraterrestrial ⁵³Mn is mainly produced by nuclear reactions of high-energy cosmic rays in IDPs, while terrestrial ⁵³Mn can be produced in iron-rich rocks. The latter could also contribute to ⁵³Mn content in the snow after erosion and transport processes. Schaefer et al. (2006) measured the terrestrial production rate as 103(11) atoms yr⁻¹(g Fe)⁻¹ and then Fujioka et al. (2010) fully validated this result. Thus, the iron content in our sample (about 28 mg according to Table 1) allow us to

estimate an upper limit to the amount of 53 Mn produced in an iron matrix, eroded by wind or rain and transported as dust into the snow sample. In the case the iron stems from rocks with very low erosion rates (< 10 cm/Myr), the rocks could have been exposed to cosmic rays for much longer periods than the 53 Mn lifetime. In that case, the iron present in the sample could have reached an equilibrium content of 53 Mn in the order of 10⁷ atoms for our 1400 L sample, which would be comparable to the amount expected by IDP deposition. However, any erosion process with rates larger than 10 cm/Myr would make the contribution of the *in situ* production much lower than the extraterrestrial 53 Mn component.

Since each of the three parameters involved in Eq. (1) (A, *f*, and $Pr_s(^{10}\text{Be})$) has a relative uncertainty of 15%, the uncertainty in the ^{53}Mn global flux is dominated by the upper limit obtained for the $^{53}\text{Mn}/^{10}\text{Be}$ ratio. Thus, the upper limit for the ^{53}Mn global flux is $\Phi(^{53}\text{Mn}) < 5.5 \times 10^3$ atoms cm⁻² yr⁻¹. This result is compatible with the one reported by Imamura et al. (1979). In that work, the author studied deep-sea sediment cores using neutron activation technique with isotope enrichment and they published a ^{53}Mn global flux of 1.96(84) $\times 10^3$ atoms cm⁻² yr⁻¹. On the other hand, the upper limit for the global flux obtained in our work is three standard deviations away from the value reported by Bibron et al. (1974), which is 18(4) $\times 10^3$ atoms cm⁻² yr⁻¹ (determined by neutron activation and X-ray spectrometry using snow samples from the Antarctic Plateau).

It has to be pointed out that the ${}^{53}\text{Mn}/{}^{10}\text{Be}$ ratio reported by Imamura et al. (1979), namely, 1.6(0.7) × 10⁻³, taken from a deep-sea sediment core at 10°57′S, 169°59′W, it agrees with our limit from Antarctica at 62°14′S, 58°40′W. This supports the hypothesis that the ${}^{53}\text{Mn}/{}^{10}\text{Be}$ ratio can be assumed to be roughly uniform worldwide.

4.3. Astrophysical implications

It was mentioned above (Section 1), that the total amount of IDPs accreted on Earth could also be deduced from the 53 Mn global flux as the ratio between this flux and the concentration of this radionuclide in IDPs. The latter value is very difficult to be assessed, as it depends on the trajectory of the IDPs in space, among other variables. An estimation of the 53 Mn content can be appraised considering that most of IDPs are originated in the Jupiter Family Comets orbiting between less than one and several astronomic units (AU) (Di Sisto et al., 2009; Nesvorný et al., 2010; Carrillo-Sánchez et al., 2016). Therefore, a reliable estimate of the 53 Mn content in IDPs requires the integration of solar and galactic cosmic ray contributions using the Jupiter Family Comets trajectories, which is far from the scope of this work.

Just as a rough estimation, we can consider that the IDPs origin is 3 AU away from Earth and that their trajectory is governed by the Poynting-Robertson effect (Kortenkamp and Dermott, 1998). Due to their dimensions, IDPs slowly spiral towards the Sun due to the Poynting-Robertson drag and the solar wind. Then, knowing the chemical composition of the IDPs, their exposure-time in space and the production-rate of a given radionuclide (⁵³Mn in this case) one can estimate the concentration of such radionuclide as done in Gómez-Guzmán et al. (2015). Each contribution (solar and galactic) to the production rate data and the elemental abundances in IDPs, assuming that IDPs have a similar composition to CI chondrite meteorites. Both galactic and solar productions rates are considered independent of the size of the IDPs in the calculations because homogeneous production within a given grain was assumed (Trappitsch and Leya, 2013).

The contribution from solar cosmic rays is obtained integrating the equation of motion over the spiral trajectory. According with the arguments given above, the solar production rate of ⁵³Mn at 1 AU corresponds to 963 dpm/kg. This value was obtained using the production rate on Fe (5.2×10^3 dpm/kg(Fe)) from Table 1 from Trappitsch and Leya (2013) and assuming the CI composition for IDPs. Considering that the flux of solar cosmic rays decreases with the square of the

distance from the Sun, its contribution to the 53 Mn concentration was calculated to be about 49 dpm/kg for IDPs travelling during times around 1.5 Myr from 3 AU (the distance of the asteroid belt) to 1 AU.

The production rate of ⁵³Mn in IDPs due to galactic cosmic rays has been estimated to be around 558 dpm/kg(Fe) in secular equilibrium at 1 AU (Reedy, 1989). Considering again CI composition it results in 102.7 dpm/kg. From this value, the contribution from galactic cosmic rays for IDPs coming from 3 AU down to 1 AU was estimated to be about 19 dpm/kg. Therefore, the total effective concentration of ⁵³Mn in IDPs, integrating over the mass distribution extracted from Cremonese (2012), corresponds to a value of 68 dpm/kg (1.9×10^{14} ⁵³Mn at/ kg). Using this value and the upper limit for ⁵³Mn global flux, it is possible to compute 150 kt/yr as an upper limit for the global flux of IDP onto Earth. This result is a factor 1.5 greater than the IPDs flux range upper limit reported in the literature (Plane, 2012).

Other potential application of the global ⁵³Mn deposition on Earth might be the determination of the interstellar material flux associated with supernovae. Knie et al., (1999, 2004) identified a peak in a ⁶⁰Fe depth profile in the ocean crusts in layers corresponding to an age of about 1.7–2.6 Myr. The 1- σ upper limit for the ⁵³Mn flux obtained in this work, allows us to calculate a lower limit for the Mn uptake factor (U_{Mn}) . This factor can be calculated as the ratio between the ⁵³Mn flux incorporated into the crust, $\Phi_{crust,53Mn}$ = 1.7 \times 10^2 atoms cm $^{-2}$ yr $^{-1}$ (measured by Knie et al., 2004) and the total flux constrained in our work, $\Phi_{tot} < 5.5 \times 10^3$ atom $cm^{-2}~yr^{-1},$ yielding $U_{Mn} >$ 0.03. With this constraint and using known concentration of elemental Fe and Mn in ocean water and in the crust, we calculated a lower limit for the iron uptake factor for that crust of $U_{Fe} > 0.005$. This agrees with the value reported by Knie et al. (2004), namely, $U_{Fe} \sim 0.006$. However, more recent works (Wallner et al., 2016; Breitschwerdt et al., 2016; Fry et al., 2016) estimate significantly higher values for the Fe uptake ($U_{Fe} \sim 0.1$) and consider an efficiency between 0.4% and 9% for ⁶⁰Fe atoms to get trapped in dust particles of the interstellar medium and to be able to enter the Solar System without being deflected by solar wind or interplanetary magnetic field.

5. Conclusions

This contribution presents a novel approach to assess the ⁵³Mn deposition on Earth. The two values found in the literature for the ⁵³Mn flux were both obtained by neutron activation and differ in one order of magnitude between each other. Our upper limit for the ⁵³Mn global flux, $\Phi(^{53}Mn) < 5.5 \times 10^3$ atoms cm⁻² yr⁻¹, agrees with the result reported by Imamura et al. (1979) and is incompatible with the one given by Bibron et al. (1974). A lower limit for the iron uptake factor in marine crust and an upper limit for the flux of incoming IDPs were derived from the upper limit for the ⁵³Mn global flux.

The transport and deposition model proposed in this work depends on not well determined quantities, like the fraction of stratospheric produced ¹⁰Be atoms from the total ¹⁰Be deposited at the sampling site and the fraction of ablated and vaporized IDPs entering the atmosphere. Although these and other values are still not well known, the methodology presented is a promising approach for addressing extraterrestrial and astrophysical issues related to extraterrestrial ⁵³Mn deposition on Earth. In order to reduce the uncertainties, it would be desirable to study samples with a higher concentration of ⁵³Mn such as snow from Antarctic latitudes with lower precipitation rates.

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