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EXTRA-TERRESTRIAL ^{53}Mn IN ANTARCTIC ICE

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The reasons why ^{53}Mn (a cosmogenic radionuclide with a half-life of 3.7×10^6 y) appears as one of the best indicators of the presence of interplanetary dust are summarized. This paper reports the detection of ^{53}Mn in pre-1952 snow samples collected on the Eastern Antarctic Plateau in the vicinity of Plateau Station. The measurements were carried out by neutron activation and X-ray spectrometry on three samples weighing a few hundred kg and covering each the time interval 1935–1950. The specific activity of ^{53}Mn was found to be (0.82 ± 0.17) disint.min^{-1}/10^3 tons of snow, corresponding to a deposition rate at Plateau Station of $(2.2 \pm 0.5) \times 10^{-5}$ disint. min^{-1} m^{-2} y^{-1}. The mean global deposition rate would be three times higher if ^{53}Mn were assumed to behave in the same way as stratospheric ^{90}Sr. By comparing this figure with existing data on the meteorite flux reaching the earth and with the galactic and solar production rates of ^{53}Mn, it is concluded that the bulk of the ^{53}Mn found at Plateau Station is associated with interplanetary dust in which it had been produced by the action of solar protons on iron. The deposition rate of extra-terrestrial dust-borne iron must be between 1.3×10^{-5} and 1.3×10^{-4} g m^{-2} y^{-1} at Plateau Station. These results support jointly with other studies the concept of an interplanetary zodiacal cloud of dust with a chemical composition and density not essentially different from chondritic meteorites, with a relatively 'flat' grain size distribution and a mass influx to the earth of the order of 10^5 tons/y.

1. Introduction

We report here the results of ^{53}Mn measurements in Antarctic snow. This investigation forms part of a research programme under way at the *Centre des Faibles Radioactivités*, Gif-sur-Yvette (France) and the *Université Libre de Bruxelles* (Belgium) on the geochemistry of the polar ice sheets and in particular on the identification of extra-terrestrial material in this ice [1–3].

It is generally agreed that most of the extra-terrestrial material captured by the earth consists of interplanetary dust, and that the contribution of meteorites is low although poorly defined. Attempts so far made to study this material and to evaluate its deposition on the earth's surface have led to no definite results. The main reason is the difficulty of identifying the extra-terrestrial component and of separating it from terrestrial materials in which it is highly diluted, even in media such as pelagic sediments and polar snows where the sedimentation rate of terrestrial material is reduced to the minimum.

Wasson [4] and Lal and Venkatavaradan [5] have shown that the presence of extra-terrestrial dust would be demonstrated unequivocally if radioactive nuclides produced by the action of high energy (5–100 MeV) solar protons on dust present in interplanetary space were detected in the materials being investigated. Most of these radionuclides can also be formed by spallation reactions due to galactic cosmic radiation, but this mode of production, predominant in meteorites is entirely negligible in objects smaller than 1 cm as a result of the difference in energy and range between galactic protons (several tens of cm) and solar protons (a few mm).

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All the radionuclides formed are not equally suitable for identifying extra-terrestrial dust. We first discard those which are produced also on earth by a different natural nuclear process. Of the remaining ones, only two nuclides display suitable characteristics as regards rate of production and half-life: ^{53}Mn (electron capture, $T = 3.7 \times 10^6$ y) and ^{59}Ni (electron capture, $T = 8 \times 10^4$ y). ^{26}Al (β^+, $T = 0.7 \times 10^6$ y) has attracted much attention because of its high rate of production and its β^+ radiation which is easier to detect than the X-rays emitted by the two other nuclides. Unfortunately, it is also produced by spallation of atmospheric argon at a rate which seems similar to that expected for the rate of deposition of ^{26}Al linked to extra-terrestrial dust [2, 6, 7].

The detection and measurement of ^{53}Mn or ^{59}Ni in terrestrial sediments would not only provide irrefutable proof of the presence of extra-terrestrial dust but would also give valuable information regarding the astrophysical parameters discussed later which govern the concentration and deposition of these nuclides. Research carried out in this field has so far led to the identification of ^{26}Al and the evaluation of its activity in pelagic sediments [7, 8] and in polar snows [9]. The corresponding rates of fallout vary from 9×10^{-4} to 1×10^{-4} disint. min^{-1} m^{-2} y^{-1}. Despite their scatter, these results – including the negative one by Tanaka et al [10]– could not be considered as contradictory when allowance is made for natural variations and experimental uncertainties. However, the authors disagree regarding the interpretation of their results: Mc Corkell et al. [9] consider that virtually all the ^{26}Al found is of atmospheric origin, while Amin et al. [7] and Wasson et al. [8] believe it to be of extra-terrestrial origin. Later on, Yokoyama [11] showed on the basis of a new ^{26}Al/^{10}Be atmospheric production ratio that no definite conclusion could be drawn from these results regarding the extra-terrestrial origin of the ^{26}Al found. This problem does not arise for ^{59}Ni or ^{53}Mn for which any significant natural production on earth seems very unlikely. It cannot be excluded that significant amounts of these nuclides have been generated by the large-scale thermonuclear bomb tests which have taken place in the atmosphere since 1952; however, there is no published information which confirms or denies this possibility. In any event, it is possible to completely avoid interference of this kind by processing pre-1952 atmospheric precipitations, which can be found at the present time in the polar ice-sheets.

These considerations led us in 1967, with the support of the U.S. National Science Foundation, to consider measuring ^{53}Mn in snow taken from the central area of the Eastern Antarctic Plateau, in the neighbourhood of the U.S. Plateau Station. This station, built during the southern summer of 1965–1966, at 79°15'S, 40°30'E and an altitude of 3,700 m [12] has extremely interesting features. Terrestrial contributions are minimized on account of its remoteness from marine, continental or industrial sources. The average annual temperature is –60°C and the maximum temperature never exceeds 0°C. Consequently the precipitated materials are perfectly preserved and do not diffuse. The extremely low rate of snow accumulation, 2.7 g cm^{-2} y^{-1} [13], makes it possible without too much difficulty to reach firn layers deposited before 1952. Moreover, the concentration of extra-terrestrial iron in these snows was already estimated to be of the order of 0.4 mg t^{-1} by Vosters et al. [3] on the basis of chemical arguments.

Taking account of this figure and of calculated production rates [5, 14], a specific activity of the order of one disint. min^{-1}/10^3 tons of snow may be expected both for ^{53}Mn and ^{59}Ni. The available logistic facilities, in spite of their importance, would not allow measuring these nuclides by direct counting. But, it did seem possible to measure the ^{53}Mn on a 10^2 kg sample by neutron activation [15, 16]. In this case the detection limit is set by the production of ^{54}Mn (T = 312 days) from the side reactions ^{55}Mn (n,2n) and ^{54}Fe (n,p) induced by fast neutrons. It is therefore essential to minimize the contribution of fast neutrons in the thermal neutron flux as well as the amount of stable iron and Mn in the sample being irradiated. As regards this latter condition, it should be recalled that the natural concentration of the elements of interest in this snow is of the order of one part in 10^9, that of iron in particular being 3 mg t^{-1} and that of Mn 0.05 mg t^{-1} [3].

Therefore, the strictest precautions are required in handling hundreds of litres of ice and meltwater without introducing more than a few μg of manganese. Minimizing and estimating losses give rise to equally serious problems and, what is worse, can be overcome only at the price of increasing the risk of contamination. In particular, the complete dissolution of a Mn

fraction possibly bound to particulate matter can be ascertained only after vigorous chemical attack, which releases unacceptable amounts of iron and Mn from the containers. Faced with these conflicting requirements, we deliberately stressed the control of contamination, perhaps at the expense of the certainty of complete dissolution of the ^{53}Mn. If, for this reason, we cannot discard any possibility of error by default we believe this error to be small and that it can in no way affect the order of magnitude of the result given here.

2. Experimental procedure

The samples of firn were taken in January 1968 from a site about 1200 m SSE upwind of Plateau Station. A 3 m deep pit was dug by hand using an aluminium alloy spade. Three tons of snow were collected in the depth interval 145–275 cm, corresponding to the period 1950–1935 (to within one or two years). The snow placed in polyethylene bags was kept frozen until analysis. Three samples, in the 100 kg range and covering each the above mentioned depth interval, were processed in turn.

All handling operations prior to the neutron irradiation were carried out in a dust-free laboratory. Tests were made to ascertain that the reagents and the containers did not introduce significant amounts of Mn. At each stage of the chemical separations, the total amount of Mn present was measured in aliquots by neutron activation and atomic absorption, in order to verify the chemical yields and the absence of accidental contamination.

The samples were melted in a polyethylene tank in the presence of HCl (final molarity = 2×10^{-3}). Experiments carried out elsewhere (Apers, private communication, 1969) with ^{54}Mn have shown that under these conditions of acidity and Mn concentration no adsorption of Mn takes place on polyethylene and that isotopic exchange is insured between ^{55}Mn and ^{53}Mn. The major elements were measured in the melt water. The values found (namely, in mg t^{-1}, iron = 3, Mn = 0.05, Ni = 0.03) are within 10% of those measured by Vosters et al. [3] on their 'ultra clean' samples. This agreement is taken as a confirmation that there has been neither significant loss nor serious contamination of these elements during sampling and melting, and also that the dissolution of any iron or Mn in particulate form was almost total. The melting tank was carefully washed with a few litres of 0.5 M HCl. The meltwater was passed over successive ionic exchange columns containing each about 200 g of Dowex 50. The Mn was eluted together with other cations, including iron, by 3 M HCl. This solution, together with the tank rinse solution, was evaporated down in a quartz flask to a few ml which were brought to 50 ml of 8 M HCl and passed twice over an anionic exchange column (Dowex 1). The effluent, which contained practically all the Mn and less than 2 μg of iron was transferred into a 1 ml quartz vial where it was evaporated to dryness. The iron retained in the column was eluted and it was ascertained that no ^{55}Fe, and hence ^{54}Mn, from thermonuclear bomb tests was detectable. At the end of this stage, 80% of the Mn initially present in the meltwater and the washing solution was recovered (actually the chemical yield was close to 100%, about 20% had to be used for chemical controls, mainly in order to measure the iron content in the final concentrate by atomic absorption). Given the conditions above described, we consider that this chemical yield applies as well to ^{53}Mn.

The quartz vial was sealed and irradiated for several weeks in an experimental channel of the EL 3 reactor at Saclay where the flux of about 2×10^{12} n cm^{-2} sec^{-1} was found to have a particularly low ratio of fast to thermal neutrons. The integrated neutron flux was determined each time using a cobalt monitor. In the case of sample 3, a zinc monitor (T = 245 days) was added at each end of the vial in order to check the homogeneity of the flux. A few mg of Mn and iron were also irradiated with each sample in view of measuring the ^{54}Mn production from these elements. After the completion of the snow samples analyses, a standard solution of ^{53}Mn (kindly provided by Dr. M. Honda) was irradiated together with a cobalt and a zinc monitor. From the results an activation factor of $(320 \pm 50)/10^{18}$ n cm^{-2} was calculated (with σ Co = 37 barns and σ Zn = 0.77 barns) in agreement with the figure (360 ± 30) published by Honda and Imamura [16].

After a one-month cooling period, the contents of the vial were dissolved. A wide range of carriers including a few mg of Mn free of detectable ^{54}Mn were added and the ^{54}Mn was radiochemically separated using a method based upon the specific extraction

TABLE I
^{53}Mn in snow samples from Plateau Station (79° 15'S-40° 30'E)*

Sample	Weight of sample	Overal chemical yield	Irrad. time	Integrated flux	^{54}Mn activity measured	^{54}Mn activity at end of irradiation	^{54}Mn activity due to side reactions**			^{54}Mn activity from ^{53}Mn	^{53}Mn activity in snow ***
							^{55}Mn (n,2n) ^{54}Mn	^{54}Fe (n,p) ^{54}Mn	Total		
no.	kg	%	days	10^{18} n cm^{-2}	c min^{-1}	disint. min^{-1}	disint. min^{-1}			disint. min^{-1}	10^{-3} disint. min^{-1} t^{-1}
1	126 ±1.4	17.7 ±2.7	21	3.72 ±0.27	0.0030 ±0.0020	0.096 ±0.064	0.003 ±0.001	0.004 ±0.003	0.007 ±0.003	0.09 ±0.06	3.1 ± 2.3
A	193 ±1.7	43.0 ±4.4	61	10.80 ±0.78	0.0109 ±0.0032	0.305 ±0.089	0.019 ±0.004	0.062 ±0.051	0.081 ±0.051	0.22 ±0.10	0.77 ± 0.37
2 B		31.2 ±3.7			0.0069 ±0.0017	0.275 ±0.068	0.014 ±0.003	0.045 ±0.037	0.059 ±0.037	0.22 ±0.08	1.02 ± 0.40
Mean value 2A – 2B											0.89 ± 0.27
3	377 ±2.4	26.0 ±4.7	67.5	11.66 ±1.16	0.0158 ±0.0025	0.327 ±0.052	0.027 ±0.006	0.002 ±0.001	0.028 ±0.006	0.30 ±0.05	0.78 ± 0.22
Mean value 2 – 3											0.82 ± 0.17

* The error represents one standard deviation. It takes into account counting statistics and an estimated uncertainty arising from the other parameters (chemical yield, neutron flux, . . .).

** Average production rates of ^{54}Mn from ^{55}Mn and ^{54}Fe are, for an integrated flux of 10^{18} n cm^{-2}: 0.28 ± 0.02 disint. min^{-1} per mg Mn and 0.22 ± 0.02 disint. min^{-1} per mg Fe.

*** An activation factor of (360 ± 30) per 10^{18} n cm^{-2} was used as proposed by Honda and Imamura [16].

of the tetraphenyl-permanganate complex in nitrobenzene [17]. Three complete purification cycles were carried out, yielding a decontamination factor greater than 10^8, a residual β activity less than 0.5 disint. min^{-1}, and a Mn recovery yield of about 80%.

The $MnO_2 \cdot nH_2O$ finally obtained was made into a suspension in alcohol and deposited by sedimentation onto a gold leaf. The 5.4 keV X-ray emitted by ^{54}Mn was measured in a small proportional counter operated in anti-coincidence, derived from a counter previously described by Leger et al. [18]. The counting system was stabilized by a CSGZ Intertechnique device using a ^{55}Fe source as reference. The counter background was 0.020 c min^{-1} keV^{-1} for an overall detection efficiency of about 8%. The measurements were carried out over periods between 4 and 6 weeks with integration of individual measurements lasting 10^3 min.

In view of the very low activities involved, the resolution of the counter was not sufficient to detect contamination by the two adjacent X-ray emitters: ^{51}Cr and ^{55}Fe. However, ^{51}Cr can be eliminated because of its half-life (27.8 days). To ascertain the absence of ^{55}Fe, sample 2 was given a further purification cycle after counting (A) and then counted again (B). It can be seen in table 1 that the specific activity remained constant within the limits of error.

3. Results

The three samples which were processed in turn gave increasingly accurate results (table 1) due to the combined effect of a number of factors: increase in the amounts of snow used, in chemical yield and in irradiation time, as well as to an improved flux determination and to a lower content in iron. The relatively low yield for sample 3 was caused by an accidental loss of irradiated material.

The ^{53}Mn activity in the three samples is seen to be in good agreement within the limits of error, the average value being (0.82 ± 0.17) disint. $min^{-1}/10^3$ tons of snow.

These results bring the first experimental evidence for the presence of ^{53}Mn in terrestrial sediments. We will now discuss briefly the following points: global deposition rate of ^{53}Mn on the earth; reasons which led us to believe that the major part of the detected ^{53}Mn is associated with the deposition of interplanetary dust; implications on the astrophysical parameters involved, mainly the size distribution of the particles in space.

4. Discussion of results

4.1. Global deposition of ^{53}Mn

The average snow accumulation at Plateau Station is 2.7 g cm^{-2} y^{-1} [13]. Therefore, during the period 1935–1950, the deposition rate of ^{53}Mn was equal to $(2.2 \pm 0.5) \times 10^{-5}$ disint. min^{-1} m^{-2} y^{-1}. However, it would not be appropriate to extrapolate this value to the entire earth. Even if it is assumed that the distribution of ^{53}Mn is isotropic at the top of the atmosphere, all the evidence suggests that a significant, although not well known, proportion of the extra-terrestrial material is volatilized as soon as it enters the atmosphere and becomes fixed on the stratospheric aerosols which fate it shares. Its deposition will then follow the well-known pattern of maximum concentration in temperature latitudes and minimum concentration in the equatorial and polar regions. It follows that the deposition measured at Plateau Station should represent a lower limit of the average deposition over the earth's surface. In the present state of knowledge, it is difficult to determine the extrapolation factor with any accuracy. Brocas and Picciotto [1], taking as a model the behavior of ^{90}Sr injected into the stratosphere by the thermonuclear bomb explosions, evaluated the ratio of the average global deposition rate to the local deposition at the South Pole as 2.5. In the light of more recent results [19] concerning ^{90}Sr deposition on the Eastern Antarctic Plateau, it seems that this figure should be raised to 3, and that, moreover, this ratio remains practically constant over the whole region between the South Pole and Plateau Station in spite of variations in the snow accumulation rate. The average global deposition of ^{53}Mn would then be $(6.6 \pm 1.5) \times 10^{-5}$ disint. min^{-1} m^{-2} y^{-1}, insofar as the comparison with stratospheric ^{90}Sr is valid.

4.2. Origin of the ^{53}Mn

As shown above, the ^{53}Mn found at Plateau Sta-

tion cannot be a product of the thermonuclear bomb tests. It could then arise from two distinct extra-terrestrial sources: the interplanetary dust on the one hand and the material volatilized or dispersed from meteorites during their flight through the atmosphere on the other. In our view, the bulk of the ^{53}Mn should be associated with interplanetary dust. Indeed, in the meteoritic material, supposed chondritic in the average, the mean specific activity of ^{53}Mn could not exceed 150 disint. min^{-1} kg^{-1}, whether this material originated from the surface layer where the action of solar protons will be predominant or from deeper layers where the sole interactions are due to galactic radiation [5, 20, 21]. If the ^{53}Mn deposition rate we inferred were due exclusively to the meteoritic contribution, this would require that, between 1935 and 1950, over 2×10^5 tons of chondritic material were volatilized every year in the atmosphere. There is still great uncertainty as to the flux of meteorites reaching the earth, as well as to the fraction volatilized in the atmosphere, but it does appear very unlikely that the total meteoritic influx exceeds 4×10^3 tons/y [22] i.e., less than 2% of the required value. The objection might be made that this evaluation takes no account of objects of which the preatmospheric mass would be less than say 10^3 g. Very little is known of the mass distribution below this value. However, under the assumption that the distribution worked out by Hawkins [23] in the range 10^3 to 10^9 g can be extrapolated down to 10^{-3} g, the global meteoritic influx would be only twice as high.

4.3. Undersaturation factor and size distribution of the dust

The formation of ^{53}Mn results essentially from four reactions involving iron isotopes [4, 5]: ^{54}Fe (p,2p); ^{54}Fe (p,pn); ^{54}Fe (p,2n); ^{56}Fe (p,α). The production rate depends on a number of parameters which have been discussed in detail by Wasson [4], Lal and Venkatavaradan [5] and Venkatavaradan [14]. Besides the excitation functions of the above reactions, the parameters involved are: the flux and energy spectrum of the solar protons, the flux variation with distance from the sun, the point of origin of the dust in the solar system and the size distribution of the dust which, in its turn, determines its residence time in interplanetary space. When Lal and Venkatavaradan published their calculations, most of these parameters were not well known. Since then, the measurements of Finkel et al. [20] on the concentration of cosmogenic nuclides in lunar rocks have shown that of the production rates calculated by Lal and Venkatavaradan, the one best fitting the experimental results was that corresponding to a proton flux of 100 p cm^{-2} sec^{-1} and a value of $R_0 = 100$ MV for the characteristic rigidity, that is to say a ^{53}Mn production rate of 3.5 atoms min^{-1} g^{-1} Fe, the value we shall use in this discussion.

The specific activity should be less than the production rate since, by all accounts, the residence time of the dust in interplanetary space is too short for secular equilibrium to be reached. The undersaturation factor is a complex function of the irradiation time, which is itself a function of the size distribution of the dust, of its place of origin in the solar system, and of the variation of the solar proton flux with the distance from the sun. The calculations of Venkatavaradan [14] show that the undersaturation factor is practically independent of the point of origin of the dust. On the other hand, the effect of the other parameters in the range of plausible values introduces an uncertainty of about an order of magnitude. Venkatavaradan considered two functions for the variation of proton flux with distance (r) from the sun, one involving $1/r$ and the other $1/r^2$. He also envisaged two size distributions, derived from observations on the zodiacal light. They both involve functions of the form: $dN = C S^{-p} dS$ where N is the number of particles and S their radius; however, the first distribution [24] gives a value of 2.6 to the population index p and the second [25] takes $p = 4.0$.

In the first case, the calculation leads to an undersaturation factor of 33% or 52% depending on whether a flux variation proportional to $1/r$ or to $1/r^2$ is adopted. Similarly in the second case, the undersaturation factor is 6% or 10%. These calculations are based upon a value of 2×10^6 y for the ^{53}Mn half-life. Using the new value of 3.7×10^6 y [16] would lead to lower undersaturation factors. By all accounts, the undersaturation factor must be somewhere between 5% and 50%. Consequently, the ^{53}Mn activity measured in the snow at Plateau Station implies a concentration of extra-terrestrial dust-borne iron of between 0.5 and 5 mg t^{-1}.

It appears particularly fruitful to compare these

results with those of Vosters et al. [3]. These authors measured the concentration of a number of chemical elements including iron and nickel, in twelve samples of about 2 kg, collected with the utmost precautions in the immediate neighbourhood (a few m) and over the same depth range as our own samples. They found a total iron concentration of 3 mg t^{-1}, which is perfectly compatible with our findings.

Thus it is possible to assign limits which are relatively safe, although still covering a fairly wide interval, to the deposition rate of extra-terrestrial iron at Plateau Station. This must be between 1.3×10^{-5} and 8.1×10^{-5} g m^{-2} y^{-1}. If we take as valid the factor 3 proposed above to take account of geographical variations in deposition, the global deposition of extra-terrestrial iron associated with interplanetary dust must therefore be between 2×10^4 and 1.2×10^5 t y^{-1}.

If this dust is taken to have a chondritic composition – according to the usual practice which however is not based upon any experimental evidence – the total flux of interplanetary dust falling on the earth must lie between 1×10^5 and 6×10^5 t y^{-1}.

It should be noted that on the basis of the assumption of a chondritic composition for the extra-terrestrial material contained in the snow, Vosters et al. [3] conclude that the concentration of extra-terrestrial iron in this snow is 0.4 mg t^{-1}. In this case the undersaturation factor would be fixed at (58 ± 12)% and the deposition of extra-terrestrial dust at Plateau Station at 5.6×10^{-5} g m^{-2} y^{-1}, i.e. a global flux of 8.4×10^4 t y^{-1}.

This undersaturation factor would also support the idea that the particles containing most of the ^{53}Mn follow a relatively 'flat' distribution law. However, it should be noted, as pointed out by Singer [26], that the particles responsible for the zodiacal light fall into a smaller range of sizes than those containing most of the ^{53}Mn and that there is no a priori reason to suppose that a single distribution law covers these two size ranges.

On the basis of measured values of ^{53}Mn deposition, it is possible to evaluate roughly the amount of ^{26}Al that should be associated with it. Using the production rates proposed by Lal and Venkatavaradan and a geographical extrapolation factor of 3 and also assuming that the dust has a chondritic composition, we find that the global deposition of extra-terrestrial ^{26}Al would be of the order of 10^{-4} disint. min^{-1} m^{-2} y^{-1}, a figure which is close to the ^{26}Al production rate in the atmosphere.

5. Conclusions

To conclude, the work described here provides indisputable evidence of the presence on the surface of the Earth of ^{53}Mn, a radionuclide linked specifically to interplanetary dust. The interpretation of our results introduces a large number of parameters, for the most part closely interdependent and of which some are still very poorly understood. Although, for these reasons, this work provides no final answer to any of the many questions arising over interplanetary dust, it does determine certain basic conditions of which any theory on this subject will have to account. As concerns the many models suggested (see Singer [26] for example), it gives firm support to the concept of a zodiacal cloud of dust with sizes distributed according to a 'flat' law, with chemical composition and density not basically different from those of chondritic meteorites and with a deposition rate on earth of the order of 10^5 t y^{-1}.

The simultaneous determination of ^{26}Al, ^{53}Mn and ^{59}Ni would be a decisive step forward. Such an operation on the Antarctic ice cap involves handling several thousands of tons of ice but, by providing unequivocal values for the average residence time and the silicon-iron ratio in interplanetary dust, it would throw light on most of the questions still unresolved in this field.

Finally, the demonstration made here of the possibility of measuring ^{53}Mn using ice samples of the order of 100 kg opens the way to the study of variations in deposition of this nuclide over the last one hundred thousand years or more, by analysing deep ice cores retrieved in the polar ice sheets.

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