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COSMOGENIC RADIONUCLIDES IN THE SOŁTMANY (L6) METEORITE

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Abstract: Cosmogenic radionuclides were measured in two specimens of the Sołtmany chondrite (L6) that fell on April 30, 2011. The first fragment (154.9 g) was measured 12 days after the fall and the second piece (120 g), 53 days after the fall. Both fragments were measured by means of non-destructive gamma ray spectroscopy. The first specimen was examined with an ultra-low background high purity germanium (HPGe) detector in a deep underground laboratory. A standard low-background HPGe detector was used to examine the second fragment in a ground level laboratory. Twelve cosmogenic nuclides were detected in the activity range of 0.030 m·Bq g^{-1} until 1.5 m·Bq g^{-1}. Their activities place constraints on the exposure history of the meteorite and reflect the effect of solar modulation of galactic cosmic rays during the solar maximum. On the activities of expected radionuclides ^{60}Co (< 0.0075 m·Bq g^{-1}) and ^{44}Ti (< 0.023 m·Bq g^{-1}) only upper limits could be given. Sołtmany is part of a group of only 14 meteorites where ^{52}Mn (5.591 d half life) could be determined.

Keywords: meteorite, ordinary chondrite, short-lived radionuclides, cosmogenic radionuclides, gamma-ray spectrometry, ^{52}Mn

INTRODUCTION

Cosmogenic radioactive and stable nuclides in chondrites have preserved important records of their exposure history during the last ten million years (e.g., Michel, 1999; Vogt et al., 1990; Caffee et al., 1988). The activity of the typical neutron-capture product, ^{60}Co has been used as an indicator of meteorites' preatmospheric size (e.g., Eberhardt et al., 1963; Spergel et al., 1986), and the activities of spallation products, including ^{22}Na, ^{54}Mn, ^{26}Al, etc., reflect irradiation conditions such as cosmic-ray shielding (e.g., Bhandari et al., 1993; Michel et al., 1995), exposure age (e.g., Heimann et al., 1974; Herpers and Englert, 1983), and the total flux of cosmic-rays (e.g., Evans et al., 1982). Production rates of cosmogenic radionuclides are influenced by many parameters. They are dependent on the composition of the meteorite. Each cosmogenically produced radionuclide has only a few target nuclides from which they are produced. Each radionuclide's production rate is determined by the abundances of its respective target nuclides (e.g. for ^{22}Na the target nuclides are Mg, Al, and Si, whereas for ^{54}Mn it is mainly iron, and for ^{60}Co it is mainly cobalt). Another important factor affecting radionuclide production is the size of the meteoroid. If the object is relatively small, the production rate increases with depth, to the body's center. Once the meteoroid's diameter exceeds approximately 80 cm, self-shielding limits radioisotope production in the inner part of the meteoroid and the production rates decrease towards the center. It is thus important to estimate, if possible, the so-called shielding depth of the sample, as it can significantly influence the production rate at the sample's depth within the meteoroid. Radionuclides and stable isotopes are produced by the interactions of both Solar (SCR) and Galactic (GCR) Cosmic Ray particles with extraterrestrial materials. These cosmogenic

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nuclide archives can be analyzed to give information about the history of both the object and the cosmic rays themselves. Production of cosmogenic nuclides by solar protons is limited to the top few centimeters of the lunar or meteorite's surface, while GCR particles penetrate deeply into an object. The contribution to the overall cosmogenic nuclide production due to secondary neutrons produced in GCR reactions is significant, especially at greater depths. Short-lived radionuclides such as ^{48}V (15.9735 d), ^{56}Co (77.236 d), ^{46}Sc (83.788 d), ^{54}Mn (312.13 d), and ^{22}Na (2.6027 a), have been used to estimate the solar proton fluxes in a single solar cycle, to determine the flux of an individual solar proton event, and to study solar modulation effects on the cosmic ray flux. Finally, there is variation of the flux of galactic cosmic rays due to time and space (orbit of the meteoroid). Long-term variations are usually averaged out in very long lived radionuclides. Short-term variations primarily affect short-lived radionuclides. Meteoroids exposed locally to the influence of the Sun can exhibit enhanced concentrations of short-lived radionuclides due to the 11-year solar activity cycle. Thus, one would expect meteorites that fall during solar activity minima to have higher radionuclide concentrations than those that fall during solar activity maxima (see Evans et al. (1982) for an example).

In Poland, only one meteorite fall in addition to Sołtmany has been recovered in the last 20 years (Baszkówka, an L5 chondrite, fell August 25, 1994), and to our knowledge no measurements of γ-emitting radionuclides have been carried out on it.

In this paper, we present the results obtained by non-destructive γ-ray measurements of two specimens of the Sołtmany meteorite (chondrite (L6), fell April 30, 2011 in Sołtmany, a small village near Giżycko in northeastern Poland) and discuss its exposure history including the period of time immediately prior to its collision with Earth.

EXPERIMENTAL METHODS

The concentrations of short-lived cosmogenic radionuclides, as well as ^{26}Al (half-life = 7.17·10^5 a) and natural radioactivity were measured in a 154.9 g fragment of the Sołtmany meteorite at the STELLA (SubTerranean Low Level Assay) underground facility of the Laboratori Nazionali del Gran Sasso (LNGS) (Arpesella, 1996) in Italy. Measurements were started on an ultra low background high-purity germanium (ULB HPGe) detector (coaxial p-type, 120% relative efficiency, thin Cu window, shielded with 25 cm of lead and 5 cm of copper, flushed with radon-free nitrogen) within 12 days of the meteorite fall, so that short-lived radionuclides, such as ^{52}Mn (half-life = 5.591 days) and ^{48}V (half-life = 15.9735 days) could be detected. Unfortunately, too much time had passed since the fall to detect very short-lived nuclides such as ^{24}Na, ^{28}Mg, ^{43}K and ^{57}Ni (with half-lives of 15-38 hours), which have thus far been detected in only a few meteorite falls (Bhandari et al., 1989, Komura et al., 2002). The counting efficiencies of each radionuclide are calculated using a Monte Carlo code that has been validated through measurements and analyses of samples of well-known radionuclide activities and geometry. This method is described in more detail in Welten et al. (2012) and references therein. The uncertainties in the radionuclide activities are dominated by the uncertainties in the counting efficiency, which are conservatively estimated at 10%. This estimate is based on Monte Carlo simulations taking into account the uncertainty in geometry, density and chemical composition of meteorite specimens. The measurement period was about 21 days, from May 12th to 31st, 2011. The measured activities of the cosmogenic radionuclides in the Sołtmany meteorite, normalized to the time of fall, are given in Table 1. U and Th were observed to have 6 (295.2 keV and 351 keV for ^{214}Pb; 609.3 keV, 1120.3 keV, 1764.5 keV and 2204.0 keV for ^{214}Bi) and 7 single peaks (338.6 keV, 911.2 keV, 964/968 keV for ^{228}Ac; 238.6 keV for ^{212}Pb, 727.2 keV for ^{212}Bi, 583.2 keV and 2614.5 keV for ^{208}Tl), respectively, and then the weighted average was calculated. Assuming secular equilibrium within the uranium chain between ^{226}Ra (parent nuclide for ^{214}Pb and

Fig. 1. The larger fragment of the Sołtmany meteorite (154.9 g) examined at the LNGS.

^{214}Bi) and ^{238}U (U), and within the thorium chain between ^{228}Ra (parent nuclide for ^{228}Ac), ^{228}Th (parent nuclide for ^{212}Pb, ^{212}Bi and ^{208}Tl) and ^{232}Th (Th) concentrations for U and Th have been calculated. A second, prism-like piece of Sołtmany meteorite (120 g) was measured in a gamma spectrometric measurement at the Institute of Nuclear Physics of the Polish Academy of Sciences (IFJ PAN), in Kraków (Poland). The measurements were completed using a 30% relative efficiency HPGe Ortec Pop-top detector equipped with a beryllium window, shielded by a 17 cm thick XIX century steel shield. The spectrometer was calibrated using a specially prepared source, which was designed to have very similar shape to the examined piece of meteorite, placed in the same position with respect to the detector. The source named ERL-1 contained washed, fine quartz sand carefully mixed with powdered uranium ore (from USAEC, NBL, Standard 3-B), monazite sand (from USAEC, NBL, Standard 7-A), and KCl (from POCH, analytical grade). A correction for the activity concentration in sand was applied. The same mixture (ERL-1) was placed in a Marinelli beaker (800 g) in order to verify the calculated activity concentration of natural radionuclides in the prepared prism-like standard (the one approximating the shape and dimensions of analyzed piece of meteorite). The activity concentration in the ERL-1 standard was additionally verified by preparing a vessel in the form of a disc 50 mm in diameter and 5 mm tall, filled with the same ERL-1 mixture (17.48 g). It was measured against a multi-gamma standard used for air filter calibration (SZN-3, provided by Polatom, Świerk) of the same size, which contained a certified amount of ^{241}Am, ^{109}Cd, ^{57}Co, ^{113}Sn, ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co. This test gave results that were consistent within 25% deviation at maximum (for the radionuclides of the Th and U series it is below 10%), probably caused by sample heterogeneities. The measurement for the second, smaller piece lasted for about 6 days, from 22 to 28 June 2011, so approximately 53 days after fall of the meteorite (30 April 2011).

RESULTS AND DISCUSSION

The activities of cosmogenic nuclides at the time of fall are summarized in Table 1 in comparison to the data of other recent ordinary chondrites (Bhandari et al. 1989, Bischoff et al., 2011) as well as to a range of typical values taken from Evans et al., 1982, Cressy, Jr., 1970 and Shedlovsky et al., 1967. All values are decay corrected to the time of fall. The average composition for L chondrites as given in Wasson, J. T. and Kallemeyn, G. W. (1988) was used for the following discussion.

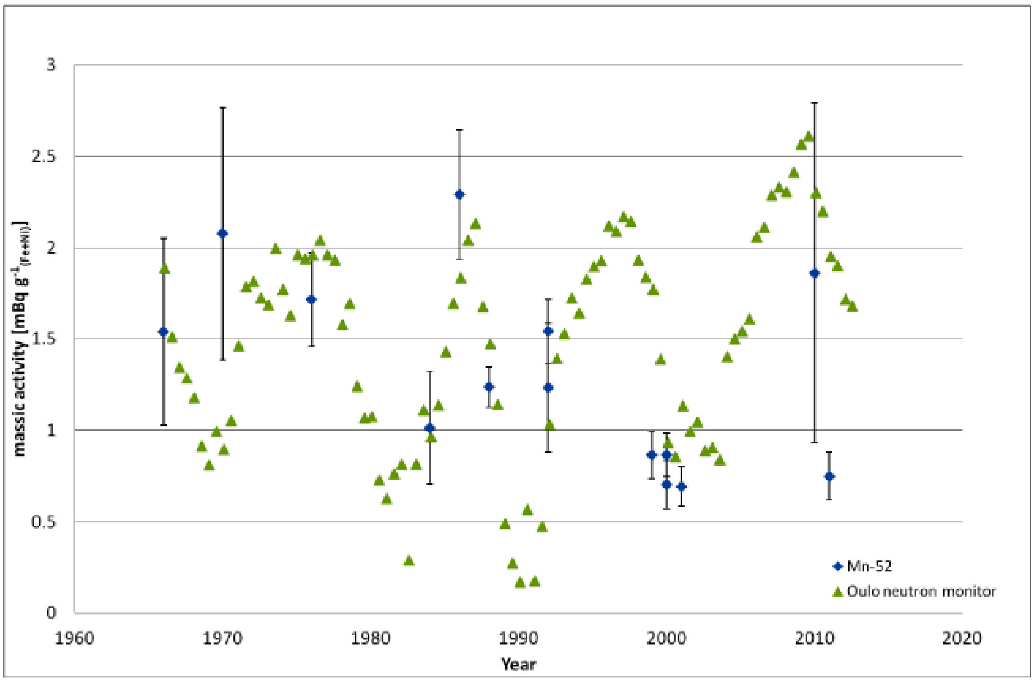

Fig. 2. The massic activities of ^{52}Mn as a function of the time of fall, taken from Table 2 and normalized to the average concentration of the target isotopes iron and nickel for each meteorite type (sourced from the publications listed in the caption of the table, and, if not available therein, from Wasson and Kallemeyn, 1988). Overlying is the neutron monitor data of Oulo in arbitrary units with monthly averages (Cosmic Ray Station of the University of Oulu, 2013).

COSMIC-RAY EXPOSURE HISTORY

The gamma-ray measurements of Sołtmany show the presence of short-lived cosmogenic radionuclides with half-lives ranging from about 5.591 days (^{52}Mn) to 2.6027 years (^{22}Na). The presence of short-lived ^{52}Mn has been detected in only a handful of chondrites, including for example Itawa Bhopji (L3-5), Kobe (CK4), Mihonoseki (L6), and Torino (H6) (Bhandari et al. 2002; Komura et al. 2002; Shima et al. 1993; Bhandari et al. 1989). All available data for ^{52}Mn in meteorites together with the results from Sołtmany are summarized in Table 2. The concentrations of short-lived radionuclides at the time of fall are a function of shielding conditions of the meteorite as well as of the solar modulation of the galactic cosmic-ray flux, which follows an 11-year cycle (Evans et al., 1982). As can be seen in Fig. 2 the data points of all meteorites (concentration of ^{52}Mn normalized to the Fe/Ni content of the meteorite) are rather well in agreement with the variations of the Solar Cycle. The value of Sołtmany is lower than expected. Indeed, most of its short-lived radionuclides are somewhat lower than the range for medium-sized chondrites, such as Torino (R=20 cm), Bruderheim (R=30 cm), Mbale and Villalbeto de la Peña (R=35-40 cm), suggesting a pre-atmospheric radius of less than 20 cm for the Sołtmany chondrite. As there are no calculations available for the production rate of ^{52}Mn in meteorites, we tried to establish an alternative way for checking the consistency of the available data for this radionuclide. From the most recent measurements of cross sections for the production of ^{52}Mn and ^{54}Mn through reactions (p,x) and (n,x) on the main targets, iron and nickel (Sisterson and Vincent, 2006), one would expect a ratio of the activities of ^{52}Mn/^{54}Mn, somewhat smaller than about 0.4 in the meteorites, where both radioisotopes have been measured. This comes from the fact that 0.4 is the ratio of cross sections for the reaction path Fe(p,x)Mn, whereas for the reactions Fe(n,x)Mn the ratio is about 0.15. Most of the ^{52}Mn and ^{54}Mn are produced on the surface by the SCR protons and only a partly in the bulk of the meteorite by secondary neutrons, their amount depending also on the size of the meteorite. Thus, the overall ratio will be less than 0.4, but bigger than 0.15. If we do this exercise with the meteorites listed in Table 2, taking the data from the publications listed in the Table caption, we get as mean value 0.29 ± 0.15, where the uncertainty is simply the standard deviation. This average agrees well with the expected results.

Since ^{22}Na and ^{26}Al in chondrites are produced by very similar reaction mechanisms from major elements (Mg, Al and Si), the activity ratio of ^{22}Na to ^{26}Al, P(^{22}Na)/P(^{26}Al), is rather independent of shielding conditions. However, due to the short half-life of ^{22}Na, its concentration in recent meteorite falls depends on the recent GCR flux, whereas the concentration of ^{26}Al reflects the average GCR flux over the past ~1 Ma. The ^{22}Na/^{26}Al activity ratio in observed chon-

Table 1. Massic activities of cosmogenic radionuclides (decay corrected to the time of fall) in the Sołtmany meteorite in comparison to the Jesenice L6 chondrite (Bischoff et al., 2011), the Torino H6 chondrite (Bandhari et al., 1989), and to a typical range for ordinary chondrites (taken from Evans et al., 1982). The first two columns give the results for the specimens measured at LNGS (12 d after fall) and IFJPAN (53 d after fall), respectively. Uncertainties given are combined standard uncertainties with the expansion factor of k=1. (n.a. is standing for not available).

Radionuclide	Half-life	Activity concentration in [mBq g^{-1}]				
		Sołtmany (LNGS)	Sołtmany (IFJ PAN)	Jesenice (sample F9)	Torino (sample A)	Typical range for chondrites
^{52}Mn	5.591 d	0.17 (3)	< 320	n.a.	0.338 (30)	0.15-0.43
^{48}V	15.9735 d	0.300 (24)	< 3.2	n.a.	0.346 (25)	0.083-0.57
^{51}Cr	27.703 d	0.83 (9)	< 11	n.a.	1.27 (12)	0.47-1.83
^{59}Fe	44.495 d	0.03 (1)	< 1.1	n.a.	< 0.058	n.a.
^{7}Be	53.22 d	1.28 (11)	< 5.4	6 (1)	0.98 (10)	0.50-2.05
^{58}Co	70.38 d	0.094 (8)	< 0.50	0.24 (7)	0.183 (12)	0.017-0.28
^{56}Co	77.236 d	0.081 (7)	< 0.55	0.20 (6)	0.128 (13)	0.050-0.15
^{46}Sc	83.788 d	0.138 (11)	< 0.44	0.23 (7)	0.173 (33)	0.033-0.17
^{57}Co	271.8 d	0.156 (12)	< 0.29	0.18 (4)	0.272 (17)	0.043-0.29
^{54}Mn	312.13 d	1.26 (9)	1.53 (14)	1.4 (2)	2.02 (3)	0.47-2.17
^{22}Na	2.6027 a	1.51 (11)	1.28 (12)	1.4 (2)	1.33 (2)	0.67-4.33
^{60}Co	5.2710 a	< 0.008	< 0.25	< 0.037	0.047 (5)	0-1.67
^{44}Ti	60.0 a	< 0.023	< 0.19	0.03 (1)	0.037 (7)	n.a.
^{26}Al	717000 a	0.82 (6)	0.87 (10)	0.8 (1)	0.900 (18)	0.63-1.33

Table 2. Massic activities of ^{52}Mn (decay corrected to the time of fall) in meteorites, including the Sołtmany meteorite. The data sets are ordered in ascending order for the date of fall. The data for the meteorites other than Sołtmany are taken from (Cressy, 1970; Cressy, 1971; Bhandari et al, 1978; Heusser et al., 1985; Yabuki et al., 1985; Bandhari et al., 1989; Shima et al., 1993; Jenniskens et al., 1994; Neder et al. 2001; Bhandari et al., 2002; Komura et al., 2002; Murty et al., 2004; Kita et al., 2013)

Meteorite	Fall (Date and Time at Coordinated Universal Time (UTC))	Type	Activity concentration of ^{52}Mn in [$mBq \cdot g^{-1}$]	Solar activity
Saint- Séverin	June 27, 1966, 14:40 UTC	LL6	0.25 (10)	Solar maximum
Lost City	January 4, 1970, 02:14 UTC	H5	0.6 (2)	Solar maximum
Dhajala	January 28, 1976, 15:10 UTC	H3	0.47 (7)	Solar minimum
Aomori	June 30, 1984, 04:50 UTC	L6	0.23 (7)	Solar minimum
Kokubunji	July 29, 1986, 10:00 UTC	L6	0.52 (8)	Solar minimum
Torino	May 18, 1988, 12:40 UTC	H6	0.338 (30)	Solar minimum
Mbale	August 14, 1992, 12:40 UTC	L5-6	0.28 (8)	Solar maximum
Mihonoseki	December 10, 1992, 12:00 UTC	L6	0.35 (4)	Solar maximum
Kobe	September 26, 1999, 11:21 UTC	CK4	0.20 (3)	Solar minimum
Morávka	May 6, 2000, 11:52 UTC	H5	0.22 (3)	Solar maximum
Itawa Bhopji	May 30, 2000, 08:15 UTC	L3-5	0.16 (3)	Solar maximum
Devgaon	February 12, 2001, 10:30 UTC	H3-4	0.19 (3)	Solar maximum
Mifflin	April 14, 2010, 03:07 UTC.	L5	0.4 (2)	Solar minimum
Sołtmany	April 30, 2011, 04:03 UTC	L6	0.17 (3)	Solar minimum

drite falls are generally in the range from 1-2 (Evans et al. 1982; Bhandari et al. 2002), with most of the variations being due to variations in the GCR flux as a function of the time of fall within the 11-year solar cycle. Higher $^{22}Na/^{26}Al$ ratios may be due to a complex CRE history, as shown by the high ratios in Jilin (4.4 ± 0.4) (Heusser et al., 1985), Dhajala (2.2 ± 0.2) (Bhandari et al., 1978) and Kobe (2.0 ± 0.2) (Komura et al., 2002), chondrites with well documented complex CRE histories, or due to a large contribution of SCR produced ^{22}Na, as is the case for the very small Salem LL chondrite (Evans et al., 1987). The average value of (1.8 ± 0.2) for the $^{22}Na/^{26}Al$ ratio in the Sołtmany meteorite is consistent with what is expected as the fall occurred during the actual period of the end of minimum solar activity and the beginning of the new maximum solar activity (see e.g. Bhandari et al., 2002).

Cosmogenic ^{60}Co in chondrites is predominantly produced by capture of thermal neutrons on ^{59}Co, a reaction pathway that is very sensitive to size and depth (Spergel et al., 1986). The Sołtmany meteorite shows no ^{60}Co activity above the sensitivity of the detector (< 0.008 $mBq \cdot g^{-1}$), which according to Eberhardt et al., 1963 (normalizing the activity concentration of ^{60}Co to the Co content of the meteorite) suggests that the preatmospheric size of this meteorite was small (radius < 15 cm) or that the specimen was located near the surface region of the meteoroid.

Summarizing the results, when we compare the radionuclide concentrations with cosmic ray production estimations for ^{26}Al (Leya and Masarik, 2009), ^{60}Co (Eberhardt et al., 1963), ^{54}Mn (Kohman and Bender, 1967), and ^{22}Na (Bhandari et al., 1993, Murty et al., 1998), the best agreement is obtained (in the sequence of the given isotopes) for pre-atmospheric radii of less than 15 cm, less than 15 cm, less than 13 cm and less than 15 cm. These numbers are obtained by interpreting the measured massic activities as production rates (PR). Those values are then compared to the calculated production rates for different radii of meteorites at different depths given in literature, and interpolating the measured values yields corresponding radii of the measured meteorite. The strongest constraint comes from ^{60}Co, because the production rates observed for the other radionuclides would also be consistent with exposure in deep shielding positions at radii beyond 100 cm. Our estimations are based upon the average flux of primary cosmic ray intensity; no corrections have been applied for potential deviations. In any case, this effect would result in even smaller radii due to enhanced PRs at least for the relevant periods for ^{22}Na and ^{54}Mn production before the fall of Sołtmany.

For the primordial radionuclides, the measured concentrations of U = $(10 \pm 1) \cdot 10^{-9}$ $g \cdot g^{-1}$ and Th = $(42 \pm 2) \cdot 10^{-9}$ $g \cdot g^{-1}$, are consistent with the average L-chondrite concentrations given in Wasson and Kallemeyn (1988), Lodders and Fegley (1998), and McSween and Huss (2010). The measured ^{40}K activity yields a K content of $(840 \pm 60) \cdot 10^{-6}$ $g \cdot g^{-1}$, also consistent with average L-chondrite values given in the same literature as above.

SUMMARY

Twelve cosmogenic nuclides (^{52}Mn, ^{48}V, ^{51}Cr, ^{59}Fe, ^{7}Be, ^{58}Co, ^{56}Co, ^{46}Sc, ^{57}Co, ^{54}Mn, ^{22}Na, and ^{26}Al) were measured via "extremely" low background γ-ray measurement of the Sołtmany meteorite. Three additional nuclides (^{54}Mn, ^{22}Na, and ^{26}Al) were counted at ground conditions. For the activities of ^{60}Co, and ^{44}Ti, only upper limits were determined. The low activities of neutron-induced ^{60}Co (< 0.008 mBq·g^{-1}) and spallation reaction products suggest two possibilities: 1) the preatmospheric size of this meteorite was rather small (r < 15 cm) or 2) this fragment was from near the surface region of a bigger meteoroid. Sołtmany is one of only 14 meteorites in which the concentration of ^{52}Mn has been quantitatively measured and published. The rather low content of this very short-lived radionuclide, together with the fact that the other short-lived radionuclides are somewhat lower than the average of similar meteorites, suggests that the meteorite was rather small before entering the Earth's atmosphere.

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