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Cosmogenic Radioactivities in the
Peace River and Harleton Chondrites

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Abstract

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The results of measurements of cosmogenic radionuclides in the Peace River and Harleton chondrites are reported. Activity ratios of short- and long-lived nuclides are compared with ratios observed in other freshly-fallen meteorites. Long-term temporal constancy of the cosmic-ray flux is indicated and the spatial constancy of cosmic radiation throughout meteoritic orbits is supported. The eleven-year solar cycle does not appear to have had any significant effect on activities with half-lives on the order of a year or longer, such as Mn^{54} and Na^{22} . The activities are compared with thick-target bombardments which indicate that the pre-atmospheric size of Harleton was smaller than that of the Peace River, Bruderheim, and Ehole meteorites.



Introduction

Long-lived radionuclides such as Be^{10} , C^{14} , Al^{26} , Cl^{36} , Mn^{53} , and Ni^{59} have been extensively studied in iron and stone meteorites during the last few years (Honda et al., 1961; Suess and Wanke, 1962; Goel and Kohman, 1963; Kohman and Goel, 1963; Rowe et al., 1963; Kaye, 1963; Honda and Arnold, 1964; Cressy, 1964). These investigations have increased knowledge of the relative productions of these nuclides under different shielding conditions, suggested long-term temporal constancy of cosmic radiation, and provided a means of calculating long terrestrial ages, and occasionally short cosmic-ray exposure ages.

Freshly-fallen meteorites, because of their short-lived radionuclide contents, provide unique opportunities for direct comparisons of production rates by cosmic rays and by artificial bombardments. These comparisons can lead to better determinations of the spatial and temporal constancy of cosmic radiation, meteorite shielding, terrestrial ages, and exposure ages.

Previous measurements of long- and short-lived radionuclides in freshly-fallen meteorites have been summarized by Honda and Arnold, (1964); Fireman et al., (1963); and Davis et al., (1963). This paper reports an extensive investigation of cosmogenic radionuclides in the Peace River chondrite along with results from the Harleton chondrite which were reported in preliminary form at the 43rd annual American Geophysical Union Meeting, 1962.

The Peace River chondrite fell March 31, 1963, in Alberta, Canada. It is a gray chondrite which apparently belongs to the Urey-Craig low-iron group. Seven fragments totaling 49 kg were recovered. A description of its fall and recovery is given by Folinsbee and Bayrock, (1964). We received

a number of pieces through the courtesy of Dr. Folinsbee in May 1963.

Experimental Procedures

The sample fragments were ground to pass an 80 mesh sieve, and the powder, 412 grams, was leached with hot dilute HNO_3 . AgCl was collected from the solution, and the chloride purified by ion-exchange and sublimation. The insoluble residue was brought into solution, after combining with the HNO_3 filtrate above, by successive treatments with HF , H_2SO_4 , HNO_3 , and HCl . After an aliquot was taken for chemical analysis, carriers for Be, Sc, and V were added. Fe was extracted into ether. Co was separated by anion exchange from 9M HCl . V and Ti were collected by extraction of their cupferrates into chloroform, followed by precipitation of $\text{Ti}(\text{OH})_4$ with NaOH . After purification of the Ti, Sc^{44} was allowed to grow to equilibrium, and scandium was separated from the titanium and purified. Be, Al, Sc, Cr, and Mn hydroxides were precipitated from the meteorite solution with $\text{NH}_4\text{OH} + \text{H}_2\text{O}_2$. These hydroxides were dissolved in HNO_3 and MnO_2 was precipitated with NaBrO_3 . Al, Be, and Sc hydroxides were collected from the chromate solution, and then BaCrO_4 was precipitated. Al, Be, and Sc were separated by cation exchange. NiS was precipitated from the first NH_4OH filtrate, along with some MnS . Mn was separated from Ni with NaBrO_3 in HNO_3 and combined with the previous MnO_2 fraction. For details of the chemical separations and specific element purifications see Cressy, (1964). Ni, Mn, Al, and Ti chemical yields were

determined from analysis of the aliquot, while the other yields were based on a chemical analysis of the Peace River chondrite (Baadsgaard et al., 1964).

The counting apparatus and techniques have been described elsewhere (Cressy, 1964; Shedlovsky and Kaye, 1963). Al^{26} was determined by coincidence counting of the positron annihilation radiation. Gamma spectrometry was used for measuring Mn^{54} and $\text{Co}^{56,57,58}$. A gas-filled x-ray proportional counter was used to measure V^{49} , Cr^{51} , $\text{Mn}^{53,54}$, $\text{Co}^{56,57,58}$, and Ni^{59} . Low-level Geiger counters were used to measure Be^{10} , Cl^{36} , Sc^{44} milked from Ti^{44} , Sc^{46} , V^{48} , and Co^{60} . Be^{10} and Cl^{36} were recycled to constant specific activity while decay curves were determined for Sc^{44} , Sc^{46} , and V^{48} .

Results and Discussion

Table 1 lists the results obtained for the Peace River and Harleton chondrites along with data from other laboratories. In addition, data primarily from Honda and Arnold (1964) on two other chondrites of recent fall are given for comparison.

Our Peace River and Harleton results are generally very similar except for Cr^{51} , Mn^{53} , and Mn^{54} ; the Peace River Sc^{46} result and our Harleton Co^{60} value, although well determined, seem anomalously high. The lower values in Harleton indicate that it was a small body in space. Thick-target bombardments of mock stone meteorites show similar broad build-up factors

for Cr^{51} and Mn^{54} production. These are maximized at ~ 35 and $\sim 75 \text{ g/cm}^2$ depths for 1 and 3 Gev protons respectively (Shedlovsky and Rayudu, 1963). Although the errors are large, the Cr^{51} , Mn^{53} , and Mn^{54} values of Peace River, Bruderheim, and Ehole relative to Harleton are all close to 2, the Mn^{53} ratios being somewhat lower. This strongly suggests that of these four meteorites, Harleton had the smallest pre-atmospheric size, and that the small recovered Ehole fragment came from a larger body. It is to be noted that the iron content of Ehole is $\sim 30\%$, whereas in Peace River, Bruderheim, and Harleton it is $\sim 22.5\%$ in each case. Since Mn^{53} and Mn^{54} in a meteorite are produced mainly from the iron, the normalized ratio Ehole/Harleton is ~ 1.6 for these nuclides, with a fairly large standard deviation. This normalized value (neglecting the large uncertainty) agrees well with the Peace River/Harleton and Bruderheim/Harleton Mn^{53} ratios, but is appreciably lower than these meteorite ratios in the case of Mn^{54} . This suggests that the Mn^{53} production maximum may be broader than that of Mn^{54} , and that the Ehole fragment was exposed in its pre-atmospheric body to a less well developed nuclear cascade than were the Peace River and Bruderheim samples.

The agreement between the three sets of Harleton results is gratifying except for Co^{60} . Our Cl^{36} results on Peace River and Harleton are lower limits as they represent only the leachable chloride before decomposition of the silicate matrix.

A long term factor-of-two temporal constancy of cosmic-ray intensity has been proposed by several investigators primarily as a result of comparisons

of specific activities in the Aroos iron meteorite with predicted activity levels (Arnold et al., 1961; Geiss et al., 1962; Honda and Arnold, 1964). Comparing a given meteorite activity with a predicted production rate presumes that the effect of meteorite shielding is known. The problem of shielding can be eliminated by studying an activity ratio in which the shielding effects are similar for both nuclides. Thick-target bombardments have shown that production rates of most products from iron decrease exponentially with depth with the same slope (Honda, 1962; Shedlovsky and Rayudu, 1964). Low-energy products, such as Mn^{54} , Mn^{52} , and Cr^{51} , show broad build-up factors at the front of the target followed by the same exponential decrease. These experiments predict that the production ratio of Mn^{53}/Mn^{54} should be nearly constant with depth in a meteorite.

In Table 2, activity ratios for a number of radionuclides in recently fallen meteorites and predicted ratios from thick-target bombardments are given. The Peace River and Harleton ratios are derived from our data except where noted. The ratios for Bruderheim and Ehole are taken from Honda and Arnold, (1964) except as noted. Specific activity ratios were calculated only in cases where the two activities were measured in the same meteorite sample.

The weighted average of the Mn^{53}/Mn^{54} meteorite values in Table 2 is $0.94 \pm .08$, in good agreement with the predicted ratio. To the extent that the 1:1 predicted production ratio is correct, the recent cosmic-ray flux is the same as the flux averaged over the last few million years, the

half lives of Mn^{53} and Mn^{54} being 2×10^6 y and 314 d respectively. It should be noted that the Mn^{54} activity could be affected by the 11-year solar cycle; yet six measurements of the Mn^{53}/Mn^{54} ratio, in five meteorites (including the Aroos iron with a ratio of 1.09 ± 0.15 , reported by Honda and Arnold, 1964) having individual activity levels that vary by a factor of ten, is remarkably constant. It appears that the solar cycle effect, at least from 1958 to 1963, within which period the five meteorites fell, was virtually negligible.

Another possible nuclide pair for the study of temporal constancy of cosmic radiation is Al^{26} ($t_{1/2} = 7.4 \times 10^5$ y): Na^{22} ($t_{1/2} = 2.58$ y). The activities of these nuclides have been measured in a number of meteorites and the Al^{26}/Na^{22} ratio in six of the eight measurements is constant at 0.7. The situation is complicated by the fact that these two nuclides depend to different extents on several target atoms for their production, Al^{26} being produced primarily from silicon ($Si^{28}(p,2pn)$) while Na^{22} is produced from both magnesium ($Mg^{24}(p,2pn)$) and silicon ($Si^{28}(p,4p3n)$). Thick-target bombardments of simulated stone meteorites show similar Na^{22} depth dependences from magnesium and silicon. This plus the essentially constant silicon and magnesium contents of stone meteorites indicates that the Na^{22} production rate should be virtually independent of chemical composition. Because of the similarity in nuclear reactions involved, the Al^{26} depth dependence is expected to be similar to that of Na^{22} . Thus it is reasonable to expect the Al^{26}/Na^{22} activity ratio to be constant in stone meteorites.

If the results of previous investigations plus the agreement between the predicted and observed Mn^{53}/Mn^{54} and Al^{26}/Na^{22} activity ratios can be taken as indicative of temporal constancy of cosmic radiation, then the question of the spatial constancy of cosmic radiation can be studied. Convenient nuclide pairs are V^{49} ($t_{1/2} = 330d$): V^{48} ($t_{1/2} = 16.1d$), V^{49} : Cr^{51} ($t_{1/2} = 27.8d$), Mn^{54} : Cr^{51} , Ar^{37} ($t_{1/2} = 35d$): Ar^{39} ($t_{1/2} = 325y$), Co^{57} ($t_{1/2} = 270d$): $Co^{56,58}$ ($t_{1/2} = 77, 72d$), V^{49} : Sc^{46} ($t_{1/2} = 85d$). If regular discrepancies can be established between the ratios in a number of meteorites and the predicted ratios, from bombardment data for example, then spatial fluctuations of the cosmic-ray intensity may be indicated. Unusually high ratios of short-lived to long-lived nuclides in a given meteorite may be caused by solar flare protons.

The Ar^{37}/Ar^{39} activity ratio has received the greatest attention to date (Davis et al., 1963; Fireman et al., 1963). Eight measurements of this ratio in five stone meteorites have given values between 1.0 and 2.3 to be compared with measured production ratios which vary from 2.1 at the surface to 1.2 at $50g/cm^2$ depth. Davis et al. (1963) conclude that the cosmic-ray intensity at several astronomical units from the sun is within 15% of the intensity at the earth's orbit. This conclusion is supported by Ar^{37} - Ar^{39} measurements on the separated metal phases of the Harkerston (Stoennner and Davis, 1962; Tilles et al., 1962) and Peace River meteorites (Fireman and DeFelice, 1964). The Ar^{37}/Ar^{39} activity ratio is expected to be more constant in the iron phase, since calcium, which varies by a

factor of two in abundance in the stone phase, contributes significantly to the production of Ar^{37} . The observed activity ratios of 0.8 and 0.9 respectively in the Harleton and Peace River metal phases are in good agreement with the experimental value of 0.8 from proton bombardments of iron targets.

If the above conclusion of the general spatial constancy of cosmic-ray flux over meteoritic orbits is valid, then the other nuclide ratios previously listed should bear this out. The $\text{V}^{49}/\text{V}^{48}$, $\text{V}^{49}/\text{Cr}^{51}$, and $\text{Mn}^{54}/\text{Cr}^{51}$ ratios for Peace River, Harleton, and Bruderheim are all reasonably constant, having weighted averages of 0.97 ± 0.11 , 0.18 ± 0.03 , and 0.76 ± 0.12 respectively. Comparison with the expected ratios are good for the $\text{Mn}^{54}/\text{Cr}^{51}$ and, to a lesser extent, $\text{Co}^{57}/\text{Co}^{56,58}$ cases, and within a factor of two for the most of the $\text{V}^{49}/\text{V}^{48}$ and $\text{V}^{49}/\text{Cr}^{51}$ measurements. Further measurements of all of these ratios in freshly-fallen meteorites are needed before one can answer the question of spatial constancy of cosmic radiation. The trend however is in agreement with the more precise $\text{Ar}^{37}/\text{Ar}^{39}$ measurements and supports the spatial constancy of cosmic radiation over meteorite orbits.

Recovered samples of artificial earth satellites have yielded both radioactive and stable nuclides. H^3 , He^3 , Ar^{37} , Co^{57} , Ag^{106} , Xe^{127} , and Bi^{205} have been observed in Discoverer satellites orbited for a few days (Fireman et al., 1961; 1963; Stoener and Davis, 1961; Wasson, 1961; 1962; Keith and Turkevich, 1962; Schaeffer and Zähringer, 1962). H^3 , P^{32} , Ar^{37} ,

Sc^{46} , $\text{V}^{48,49}$, Cr^{51} , Mn^{54} , Fe^{55} , $\text{Co}^{56,57,58}$, have been measured in a fragment of Sputnik 4 orbited for 843 days (Shedlovsky and Kaye, 1963; DeFelice et al., 1963; Kammerer et al., 1962; Rowe et al, 1964; Wasson, 1964). The production of such nuclides in satellites is attributed to a combination of solar flare protons, galactic cosmic-rays and Van Allen protons. The effects caused by these various protons is not completely resolved.

The irradiation of meteorites by the above sources at present is best understood for galactic cosmic-rays. The effect of solar flares on meteorites is still ambiguous. $\text{Ar}^{37}/\text{Ar}^{39}$ ratios in Hamlet and Ehole, both of which fell following a solar flare (Davis et al., 1963), were high. However, the errors of the measurements for this ratio in the meteorites and the bombardments overlap, leaving the question in doubt. The ratios in Table 2 for Hamlet and Ehole unfortunately are not precise enough to show a flare effect; it would be surprising to see an effect with Na^{22} in any event, because of its relatively long half-life. No measurements of sufficiently short-lived radionuclides in these two meteorites have been reported; there are indications however that the $\text{Co}^{56,58}$ activities were enhanced in the Hamlet meteorite (A. Turkevich, private communication). During the next several years the solar activity should be increasing, affording a better opportunity to observe solar flare effects on freshly-fallen meteorites.

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Table 1. Cosmogenic Activities in Recently Fallen Stone Meteorites

Date of Fall Recovered Mass	Peace River 3/31/63 49 kg	Harleton 5/30/61 8.4 kg	Bruderheim 3/4/60 300 kg	Ehole 8/31/61 2.4 kg
Be^{10}	27 ± 1	26 ± 1 21 ± 2 (1)	19 ± 2 (1)	19 ± 2 (1)
Al^{26}	57 ± 1 39 ± 4 (3)	51 ± 4 45 ± 5 (1) 43 ± 3 (2)	60 ± 6 (1) 57 ± 2 (2)	70 ± 7 (1) 33 ± 2 (2)
Cl^{36}	$\geq 3.0 \pm 0.2$	$\geq 2.1 \pm 0.2$ 7.0 ± 0.7 (1)	7.5 ± 0.8 (1)	7.8 ± 1.0 (1)
Ti^{44}	≤ 1.0	1.4 ± 0.2 (1)	2.0 ± 0.2 (1)	
Sc^{46}	35 ± 1	16 ± 4 5.4 ± 0.7 (1) 11 ± 4 (2)*	6.2 ± 0.6 (1)	
V^{48}	18 ± 2	14 ± 2 17 ± 2 (1)	34 ± 7 (1)	
V^{49}	14 ± 2	25 ± 8 20 ± 6 (1)	34 ± 2 (1)	
Cr^{51}	140 ± 35	60 ± 20 (1)	110 ± 27 (1)	
Mn^{53}	91 ± 9	55 ± 5 44 ± 8 (1)	85 ± 17 (1)	110 ± 20 (1)
Mn^{54}	103 ± 5	45 ± 9 38 ± 5 (1) 47 ± 3 (2)	100 ± 13 (1) 82 ± 7 (2)	90 ± 20 (1)

Table 1. Cosmogenic Activities in Recently Fallen Stone Meteorites (Continued)

Date of Fall Recovered Mass	Peace River 3/31/63 49 kg	Harleton 5/30/61 8.4 kg	Bruderheim 3/4/60 300 kg	Ehole 8/31/61 3.4 kg
^{60}Co	19 ± 4 30 ± 6 (3)	15 ± 12	1'	14 ± 6 (1)
^{60}Co			1.1 ± 1 (1)	
^{59}Ni	21 ± 3	6.5 ± 0.7 (1) 1.3 ± 4		
^{59}Co	2.1 ± 0.4	1.5 ± 0.5 (1)	9 ± 1 (1)	4.8 ± 1.2 (1)

* Includes $\text{Co}^{58,59}$

(1) Honda and Arnold [1964].

(2) Rowe et al., [1963].

(3) Fireman and DeFelice [1964].

