content amounts to ~ $4.2 \times 10^{-5} cm^3$ per gram of dust. If we assume an average K concentration of 1000 parts per million, a K-Ar age of 3.5×10^9 years results.

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Pattern of Bombardment-Produced Radionuclides in Rock 10017 and in Lunar Soil

Abstract. *A large number of radionuclides have been measured as a function of depth in lunar rock 10017 and in bulk fines. Data are reported on ^{10}Be, ^{22}Na, ^{26}Al, ^{36}Cl, ^{49}V, ^{53}Mn, ^{54}Mn, ^{55}Fe, ^{56}Co, ^{57}Co, and ^{59}Ni and on upper limits for ^{46}Sc, ^{48}V, ^{51}Cr, and ^{60}Co. The results for several nuclides show striking evidence of excess surface production attributable to solar flare particles. Data for short-lived species, ^{56}Co, ^{57}Co, ^{54}Mn, ^{55}Fe, and ^{22}Na, appear consistent with fluxes from known recent events. Long-lived species demonstrate the existence of solar flare protons and alphas at least for the last 10^5 to 10^6 years, at fluxes comparable to those now observed.*

Measurements of the concentration of short- and long-lived radionuclides in meteorites have been useful in unraveling the history of the high-energy radiation which produced them and of the target objects (*1*, *2*). Similar studies on lunar surface materials have the same potential usefulness, with some special advantages. A lunar sample has suffered no ablation, and in principle its recent orientation and depth are known. The planet which they sample is a single, intensely studied one at a known orbital position.

We attempted to study processes of gardening by impact or other processes on the lunar surface, through times of exposure and depth variations, and to decipher the fossil record of solar and galactic high-energy particles in the earth-moon region. Since the longest half-lives available are less than 3×10^6 years, and since measured bombardment ages are generally much longer than this (*3*, *4*), it appears that the samples so far received are, for our present purposes, relatively fixed targets. Thus we have generally concentrated on the objective of using the rock and soil samples to study the high-energy particle emission of the sun on a time scale up to millions of years. Nevertheless, a combination of radionuclide, rare gas, and track data can still give much information on turnover rates.

We received two lunar samples. First, 100 g of bulk fines (10084,16), <1 mm size, was received on 12 September (52 days after lunar landing). The second sample, two chunks of rock 10017 (a coarse-grained rock of mass 980 g) was received on 3 October. Figure 1a shows a drawing of this rock, with an outline of the approximate position of the specimens. The rock had two plausible surface orientations; most photographs made in the preliminary examination were in what we now consider a position inverted relative to its orientation on the moon (*5*).

Samples were subjected to grinding (rock), magnetic separation (rock and dust), and leaching (dust). They were dissolved in $HF-HNO_3$, with carrier addition where appropriate, in a Teflon still under flowing N_2. Chlorine and Si were recovered in the distillate. The residue from the distillation was heated with H_2SO_4 in a Pt dish to remove HF; HCl was added, and Fe was extracted. The "main chemistry" was completed by anion- and cation-exchange column steps. This main chemistry was carried out, with variations, for two 50-g portions of soil and for six subdivisions of the rock (Fig. 1b). The resulting fractions, containing usually one element of interest, sometimes two or three, were purified chemically and radiochemically.

Chemical analyses for calculation of chemical yields and for following the progress of separations and recoveries were carried out by atomic absorption spectrometry. Agreement was good between these analyses and those available to us from others, with a few exceptions. In some cases the analytical data of others were used for final yield calculations. Overall yields were usually in

Table 1. Summary of counting results (dpm/kg). Limits (2σ) in bulk fines sample: ^7Be, 250; ^{46}Sc, 9; ^{48}V, 38; ^{51}Cr, 45; and ^{60}Co, 5.

Nuclide	Half-life	Rock 10017					Bulk fines	
		T4DF (0 to 4 mm)	T4U (4 to 12 mm)	T4I (12 to 30 mm)	T3SI (60 mm)	Houston RCL*	This work	Houston RCL
^{10}Be	$2.5 \times 10^6 y$	16 ± 2		16 ± 2			15.7 ± 1.6	
^{22}Na	2.6 y	76 ± 10	43 ± 6	37 ± 6			55 ± 7	51 ± 9
^{26}Al	$7.4 \times 10^5 y$	133 ± 15	70 ± 10	57 ± 9	31 ± 5	39 ± 7	108 ± 17	$120 (\pm 19)$
^{36}Cl	$3 \times 10^5 y$	12 ± 5	16 ± 2	16 ± 2	63 ± 12	$73 (\pm 13)$	17.0 ± 1.6	
^{49}V	330 d						7.4 ± 2.0	
^{53}Mn	$\sim 2 \times 10^6 y$	107 ± 21	45 ± 13	52 ± 10				
^{54}Mn	303 d	33 ± 17	34 ± 22	10 ± 11		$34 (\pm 13)$	18 ± 10	28 ± 9
^{53}Mn and ^{54}Mn			98 ± 15	88 ± 10			82 ± 11	
^{55}Fe	2.6 y	420 ± 44	94 ± 27	93 ± 30	97 ± 24		195 ± 22	
^{56}Co	77 d	125 ± 20	< 16	< 8		$26 (\pm 6)$	44 ± 6	40 ± 10
^{57}Co	270 d	5.6 ± 2.6	< 1.7	< 2.4			< 1.8	
^{59}Ni	$8 \times 10^4 y$	< 4					2.6 ± 0.8	

* Houston 10017 data were measured using the whole rock (*14*).

the range of 80 percent, varying with the element, though in some cases of low specific activity (^{26}Al and ^{55}Fe) only a fraction of the available material was purified and counted. In some cases small amounts of radioactive impurities were seen in the counting, and where necessary these samples were recycled.

Several detector systems were used, depending on the decay scheme of each species and the demands of schedule. The most important were: (i) the Evans counter (E), a β-γ system developed from a design by N. Bhandari, consisting of a small Geiger counter (external sample) in the well of a 7.5 by 7.5 cm NaI(Tl) crystal; (ii) the Loosli counter (H), also modified from a design by Bhandari (*6*), a small internal pressurized proportional counter in a 10 by 10-cm NaI(Tl) well crystal, for x-ray, x-γ, and β-γ measurements; (iii) the Tanaka counter (T) (*7*), a Geiger-plastic scintillator package for β-counting and spectrometry; (iv) the Lal counter (L), from a design by Rajagopalan (*8*), a pair of Geiger counters with fixed absorber between, also for β-counting; (v) the Delany counters (D), internal x-ray counters of large area, 60 and 160 cm^2; and (vi) a 40-cm^3 solid-state Ge(Li) crystal (SS) in an annulus of NaI(Tl) for gamma counting.

Samples were counted on Cu or plastic holders, with the use of a Millipore deposition technique or electroplating, both of which gave extremely smooth and uniform deposits. Counting standards were prepared from standardized solutions. Where possible, the form and thickness of samples and standards were approximately matched. Beyond this, self-absorption corrections were calculated by standard methods. Backgrounds were taken where possible with the same or similar chemical species, carried through the same chemistry as the samples. In several cases, samples were counted on more than one system, generally with concordant results. All these procedures were carried out more or less completely several times on terrestrial materials before arrival of the sample.

The counting results to date are summarized in Table 1. The errors quoted include all known sources: counting statistics, chemical yield, calibration, and others. Ratios, especially of the same nuclide in different samples, may be more accurate. Table 1 also shows the results obtained at Houston for comparable materials, where available. In general the agreement is good.

Table 2. Comparison of observed and calculated activities in Bruderheim and Harleton meteorites and in rock 10017 (units dpm/kg).

Nuclide	Bruderheim			Harleton		Rock 10017 (12 to 30 mm)	
	Obs.	Calc.		Obs.	Calc. $\alpha = 600$	Obs.	Calc. $\alpha = 600$
		$\alpha = 400$	S(10)				
^{55}Fe	340	140	280	$\leqslant$ 180	92	93	37
^{54}Mn	100	56	92	38	41	10	16
^{53}Mn	85	56	68	45	42	52	17
^{49}V	34	25	25	20	22	7*	9
^{36}Cl	7.5	6.5	7.2	7.0	6	16	9
^{26}Al	60	54	65	45	41	57	33
^{22}Na	90	88	94	64	67	37	29
^{10}Be	19	24	24	21	23	16	15

* Estimated from bulk fines.

The gentle acid leaching of the dust sample, before the main chemistry, dissolved a large fraction of the Ni and Co in the sample, with a little Fe and other elements. A small amount of Cl was isolated and showed no ^{36}Cl activity. The tabulated activities of ^{56}Co and ^{59}Ni are based on chemical yield for the undissolved portion; we assume that Fe was the only significant target.

The ratios of activities in surface samples to those in deep samples (Table 1) are of special interest. For all accurately measured nuclides the ratio of sample T4DF to T4I is substantially greater than 1. It is particularly large for ^{55}Fe, and in the extreme case of ^{56}Co no activity is observed below the top sample. A comparison of fines and deep samples shows a similar but smaller effect. We infer that the mean depth of the dust below the undisturbed surface is of the order of 2 cm. All these data seem to show the importance of solar proton irradiation near the surface.

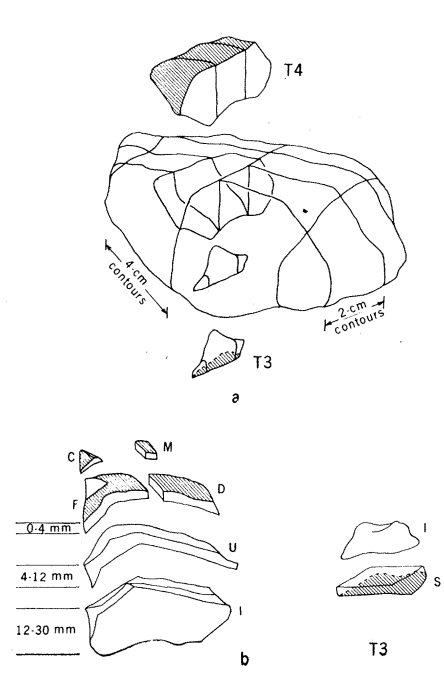

Fig. 1. (a) Rock 10017, as oriented by counting data, showing the location of our two samples. (b) Exploded drawing of samples T4 and T3, showing location and orientation of subsamples divided by us.

The ^{59}Ni is significant as showing the presence of solar α-particles. It is produced uniquely by ^{56}Fe (a,n) ^{59}Ni.

For the production of radionuclides in these materials, then, we must consider two sources: the galactic cosmic radiation and solar particles. The galactic production can be calculated on several models (*1*, *2*). It is reasonable to assume for this component the same constancy with time observed in meteorites (*1*).

We have used a model developed from that of Arnold *et al.* (*9*). Primary and secondary spectral shapes were chosen, usually of the form $\kappa/(\alpha + E)^{2.5}$. Smaller values of α correspond to a softer spectrum (more secondaries, greater depth). The absolute intensity is related to that at the surface by assuming that particles >1 Bev move linearly with a mean attenuation length of 155 g/cm^2, and normalizing the spectra above that point. The value of α must decrease smoothly with depth. Excitation functions are taken from experiment, analogy, or reference (*9*, *10*) for each product nuclide and important target nuclides. Integration gives the production in disintegrations per minute per kilogram of lunar material.

The model was tested with data for the Bruderheim and Harleton chondrites (*1*). A value of α=400 Mev was found to give the best fit for Bruderheim

Table 3. Short-lived nuclides in 0 to 4 mm layer from solar proton data: Experiment versus calculation.

Nuclide	Half-life	Expt. (dpm/cm^2)	Calculated (dpm/cm^2)
^{56}Co	77 d	0.18	0.13*, 0.36†
^{57}Co	270 d	0.01	0.004*, 0.01†
^{54}Mn	303 d	~0.02	0.01*, 0.02†
^{55}Fe	2.6 y	0.5	0.5
^{22}Na	2.6 y	0.06	0.12

* See (*15*). † See (*16*).

(Table 2); for Harleton the best relative and absolute fit was for α=600. A low energy tail, of the AHL S(10) (*9*) form, gave improved agreement for ^{55}Fe, but made little change otherwise. Since for primaries α=1 Bev, and since even at the top surface secondaries must be important, we chose a value of α=700 at zero depth, falling to α=500 for the deepest samples of rock 17.

With this spectrum normalized for 2π bombardment as above, we then computed the expected galactic production for the deep sample T4I (Table 2). It must be emphasized that this procedure, while consistent with available physical information, is in effect a method of interpolation, in which the soundness of the model is determined empirically in meteorites, and the results are translated to the different composition and geometry of the lunar samples. We believe the results are not more than 30 percent in error on the average.

It can be seen that some species are well accounted for by galactic production. Others show a marked excess.

Solar particles have lower energies than galactic cosmic rays. They occur during flares in short bursts, with spectral shapes generally well represented for our purposes in exponential rigidity form (*10*). Since secondary particles are not important at these energies, we adopt a different model for production calculations. Particles are assumed to lose energy almost entirely by ionization, and production is integrated along the path, giving results in dpm/cm^2 for a given total flux above 10 Mev and mean rigidity R_0. For ^{56}Co, a product of the low energy ^{56}Fe(p,n) ^{56}Co reaction, and the observed low mean rigidity (~50 MV), all the production occurs in the outer 4 mm. Thus we need not calculate angular dependence or interior spectral shape. For higher energy products and higher R_0 a more elaborate program is used to calculate dpm/cm^2 for given depth ranges.

For short-lived products we have compared our data with predictions from known flares (Table 3). It can be seen in particular that the results for ^{56}Co are well accounted for by flares—mostly by the event of 12 April 1969. The other short-lived species are also present in roughly the calculated amounts; somewhat less for ^{22}Na.

From the activities of the long-lived species ^{36}Cl, ^{26}Al, ^{53}Mn, and ^{10}Be, we can see that solar protons have been present for the past million years in amounts comparable to those observed recently. Only the high-energy product ^{10}Be fails to show a large excess over galactic production (Table 2). But before we can make definite conclusions about the flux and rigidity of past solar events we must examine the excess production observed in the deep samples. At present, without detailed rare gas and nuclear track information and precise ^{53}Mn measurements by activation analysis, we can explain our data in two ways. Model (A) assumes that there has been no gardening or erosion for rock 10017 over 10^7 years or so. This would lead us to believe in a mean flux of solar protons similar to the presently observed one (of order 30/cm^{2}sec), but higher mean energy or rigidity, $R_0 \geqslant 150$ MV, over millions of years, and perhaps in different fluxes in different periods. Model (B) assumes that the higher interior values are those characteristic of production at deeper levels in the moon and that the exposure of the present surface is relatively recent ($\leqslant 5 \times 10^5$ years age). This leads to a higher flux and lower rigidity in the period of exposure to produce the observed surface excess. Thus in either case the mean solar proton flux appears to have been as high as (or even higher than) that observed at present.

Several other topics remain to be discussed. The comparison with rare gas data shows a complex radiation history, discussed by Marti *et al.* (*4*). The Gd isotope data (*4, 11*) will give an independent integral of the neutron flux. Nuclear track data (*12, 13*) confirm the location of the upper rock surface. The lunar sample results have implications for meteorite studies, and for cosmic dust influx rates, both as deduced from limits on erosion rates, and for comparison with ^{26}Al data in sediments.

We conclude that: (i) Our data on short-lived nuclides in the lunar surface can be explained by known bombarding fluxes: the cosmic radiation and solar flare particles. (ii) The long-lived nuclides show a similar pattern. Hence we conclude that solar flares, and presumably sunspots, have been occurring for at least the last 10^5 to 10^6 years. (iii) While details of flux and mean rigidity depend on data not yet available, the mean solar particle flux appears to have been comparable to or perhaps greater than that observed at present.

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