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^{14}C depth profiles in Apollo 15 and 17 cores and lunar rock 68815

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Abstract—Accelerator mass spectrometry (AMS) was used to measure the activity vs. depth profiles of ^{14}C produced by both solar cosmic rays (SCR) and galactic cosmic rays (GCR) in Apollo 15 lunar cores 15001-6 and 15008, Apollo 17 core 76001, and lunar rock 68815. Calculated GCR production rates are in good agreement with ^{14}C measurements at depths below ~10 cm. Carbon-14 produced by solar protons was observed in the top few cm of the Apollo 15 cores and lunar rock 68815, with near-surface values as high as 66 dpm/kg in 68815. Only low levels of SCR-produced ^{14}C were observed in the Apollo 17 core 76001. New cross sections for production of ^{14}C by proton spallation on O, Si, Al, Mg, Fe, and Ni were measured using AMS. These cross sections are essential for the analysis of the measured ^{14}C depth profiles. The best fit to the activity-depth profiles for solar-proton-produced ^{14}C measured in the tops of both the Apollo 15 cores and 68815 was obtained for an exponential rigidity spectral shape R_0 of 110–115 MV and a 4π flux (J_{10}, Ep > 10 MeV) of 103–108 protons/cm^2/s. These values of R_0 are higher, indicating a harder rigidity, and the solar-proton fluxes are higher than those determined from ^{10}Be, ^{26}Al, and ^{53}Mn measurements. *Copyright © 1998 Elsevier Science Ltd*

1. INTRODUCTION

Many radionuclides are produced by spallation reactions in meteorites and other materials such as those in the lunar surface, which are exposed to cosmic-ray particles. Cosmic-ray production of various nuclides and studies of its effects in meteorites and lunar samples has been summarized by Reedy et al. (1983), Vogt et al. (1990), and Herzog (1994). Cosmogenic nuclides are produced by the high-energy (~0.1–10 GeV) particles in the galactic cosmic rays (GCR), by secondary particles made by GCR, and by the particles in the solar cosmic rays (SCR), which are energetic (~10–100 MeV) particles emitted irregularly by the Sun (Reedy and Arnold, 1972; Reedy and Marti, 1991; Herzog, 1994). The production of spallation-produced isotopes depends on the particle flux at the sample location during irradiation in space. In addition to the energy distribution of nuclear particles, important parameters are the cross sections for a nuclide's production from various elements as a function of energy and the chemical composition of the sample.

Due to their long exposure times, lunar samples provide a continuous record of GCR and SCR intensities and spectral distributions. Lunar cores are also very useful in studies of the production of nuclides by GCR particles in very large objects. Lunar samples contain excellent records of SCR effects and variations of SCR fluxes in the past (Reedy, 1980; Reedy and Marti, 1991). Radionuclides in lunar samples that have been measured and that can be used to study SCR fluxes in the past include ^{14}C with a half-life ($t_{1/2}$) of 5.73 ka (Begemann et al., 1972; Boeckl, 1972; Jull et al., 1995), ^{59}Ni ($t_{1/2}$ 76 ka; Lanzerotti et al., 1973), ^{41}Ca ($t_{1/2}$ 100 ka; Fink et al., 1998), ^{81}Kr ($t_{1/2}$ 229 ka; Reedy and Marti, 1991), ^{36}Cl ($t_{1/2}$ 300 ka; Nishiizumi et al., 1989, 1995), ^{26}Al ($t_{1/2}$ 700 ka; Kohl et al., 1978; Nishiizumi et al., 1990, 1995; Fink et al., 1998), ^{10}Be ($t_{1/2}$ 1.5 Ma; Nishiizumi et al., 1988, 1990, 1995, 1997; Fink et al., 1998), and ^{53}Mn ($t_{1/2}$ 3.7 Ma; Kohl et al., 1978; Nishiizumi et al., 1990). The stable noble-gas isotopes ^{21}Ne, ^{22}Ne, and ^{38}Ar have also been used by Rao et al. (1994) to study past SCR particle fluxes. However, the interpretation of the SCR-produced nuclides in lunar samples has often been made more difficult by the lack of cross sections for the relevant nuclear reactions (Reedy and Marti, 1991; Sisterson et al., 1991a, 1992, 1994, 1996). Some new values for relevant ^{14}C production cross sections, not previously published, are reported in this paper.

About 30,000 years of continuous exposure is required to reach a saturation (maximum) level of ^{14}C. This length of time is much less than the typical exposure ages of lunar samples (millions of years) and relocation of material by erosion or gardening (see Gault et al., 1974), which is a few mm per million years (Langevin et al., 1982). Early ^{14}C work, done with gas counters, on samples from the top few centimeters of lunar rocks 12002 (Boeckl, 1972) and 12053 (Begemann et al., 1972) indicated high levels at the very surface. These data had relatively large uncertainties due to the sample size limitations of the counters used. Boeckl (1972) proposed that higher surface ^{14}C values, compared to values at a few cm depth, could be explained by an enhanced solar-proton 4π flux of 200 protons/cm^2/s ($E_p > 10$ MeV; $R_0 = 100$ MV) over the last ~10 ka. Begemann et al. (1972) had suggested that the very surface layer of lunar samples could be implanted with solar ^{14}C, and this could account for the enhanced surface activity in rock 12002.

Measurements on scooped soil 10084 (Begemann et al., 1970; Born, 1973; Fireman et al., 1976, 1977; Fireman, 1978) and the trench soil samples 73221, 73241, and 73261 from the

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Table 1. Published values of ^{14}C in lunar rocks and soil

Depth (g/cm^2)	^{14}C (dpm/kg)	
Rock 12002[1]		
0–0.28	72 ± 11	Boeckl (1972)
0.28–0.52	50 ± 7	
0.52–1.04	44 ± 6	
1.04–2.08	61 ± 9	
2.08–5.6	29 ± 4	
5.6–11.2	27 ± 3	
Rock 12053		Begemann et al. (1972);
0–1.5	72 ± 7	Born (1973)
1.5–6	33 ± 3	
6–19.5	30 ± 3	
Soil 10084		
0–7.5	52 ± 4	Born (1973)
	50.9 ± 3.9	Fireman et al. (1977)
	33.8 ± 5.2	
	53.0 ± 0.6	Jull et al. (1995)
Soil 73221		
0–2	57.5 ± 4.7	Fireman et al. (1977)
	50.8 ± 0.8	Jull et al. (1995)
Soil 73241		
2–8	26.9 ± 3.3	Fireman et al. (1977)
	28.0 ± 0.3	Jull et al. (1995)
Soil 73261		
8–16	21.7 ± 3.8	Fireman et al. (1977)
	26.8 ± 0.4	Jull et al. (1995)

[1] The results have been rounded off from the values published.

Apollo 17 site (Fireman et al., 1976, 1977) have been reported, using gas counting methods and are listed in Table 1. Using AMS, Jull et al. (1995) reported on a follow-up study of the work of Fireman et al. (1977), who found evidence for an implanted ^{14}C component in lunar surface soils. In the study of Jull et al. (1995), acid etching of Apollo 17 soil samples (73221, 73241, and 73261) and a surface patina sample of 68815 were studied. The newer results confirmed evidence for an implanted ^{14}C component in the surface soil and rock.

In summary, the previous work with ^{14}C in lunar samples has identified three extraterrestrial sources of the ^{14}C observed in lunar samples: production by nuclear reactions induced by GCR or SCR particles, or implantation from an external source. These three sources of ^{14}C in lunar samples will be discussed further. Preliminary reports of some of the work discussed in this paper have been presented at Lunar and Planetary Science Conferences (Jull et al., 1991, 1992; Jull and Cloudt, 1996).

1.1. GCR-Produced ^{14}C in Lunar Samples

Reedy and Arnold (1972) developed a model for calculation of the production of ^{14}C by cosmic-ray effects in the lunar surface. Even with revised cross sections (see Jull et al., 1989b; Imamura et al., 1990; Sisterson et al., 1991a,b, 1994, 1996) for the GCR production of ^{14}C, using the model of Reedy and Arnold (1972) did not result in a good fit to lunar samples. Born (1973) and Rao et al. (1994) found it necessary to increase the production rates they calculated with the Reedy and Arnold (1972) model by up to 25% to fit their experimental data. In this paper, we will discuss results from the model of Masarik and Reedy (1994) for lunar GCR particle fluxes and revised cross sections (Jull et al., 1989b; Imamura et al., 1990; Sisterson et al., 1996). This model of Masarik and Reedy (1994) was previously used to calculate production rates of ^{14}C by GCR particles in meteorites as a function of depth and agreed with measured values to within 10% (Jull et al., 1994, Wieler et al., 1996).

1.2. Solar Cosmic Rays

Solar-cosmic-ray particles, ~98% protons, with energies of tens to hundreds of MeV, have a range of ~1 cm in rocks. The SCR flux can be approximated (Reedy and Arnold, 1972) as a distribution in rigidity units of the form

$$dJ/dR = k \exp(-R/R_0) \quad (1)$$

where J is the flux, R is the rigidity (pc/Ze) of the particles, R_0 is a spectral shape parameter, expressed in units of megavolts (MV), and k is a constant. It is conventional to quote fluxes for SCR proton energies >10 MeV and 4π solid angle, defined as J_{10} in this paper, with units of protons/cm^2/s. Values of R_0 in the range 70–125 MV have often been fitted to nuclide data from lunar samples (see Reedy and Marti, 1991; Rao et al., 1994; Fink et al., 1998).

1.3. Implanted Energetic Particles

The solar wind is a stream of particles, mostly protons, emitted by the sun with an average flux at 1AU of $\sim 2 \times 10^8$ protons/cm^2/s (Keays et al., 1970). These particles are ~1 keV/amu in energy and have a range of a few tens of nm in rock. As already discussed, Begemann et al. (1972) had suggested that the very surface layer of lunar samples could be implanted with solar ^{14}C. Fireman et al. (1976) found higher than expected levels of ^{14}C in the 600–1000°C temperature fractions of the Apollo 11 soil 10084 and the Apollo 17 trench soils 73221–73261 (see Table 1), which they interpreted as implanted solar wind ^{14}C. Fireman et al. (1976) concluded that the surface-correlated ^{14}C in lunar samples, if implanted from the solar wind, would imply a $^{14}C/^{1}H$ ratio in the solar wind of $\sim 5 \times 10^{-11}$. They also noted that this ratio is much more than expected by most models for nuclear reactions in the solar surface regions. In a recent paper, Jull et al. (1995) reported on an experiment that confirmed the existence of the implanted ^{14}C component first recognized by Fireman et al. (1976). Jull et al. (1995) concluded that there was an implanted ^{14}C flux on the very surfaces of lunar rocks and soil of 4 to 7×10^{-6} $^{14}C/cm^2/s$, which is equivalent to a $2–3.5 \times 10^{-14}$ $^{14}C/H$ ratio. In this paper, we report measurements of cosmogenic ^{14}C in two Apollo 15 cores (the deep drill core 15001–15006 and drive tube 15008, which was the top half of a double drive tube), in Apollo 17 core (76001) and in lunar rock 68815. Depth vs. activity profiles of other radionuclides have previously been measured in these samples. In rock 68815, Kohl et al. (1978) measured ^{53}Mn and ^{26}Al, Nishiizumi et al. (1988) determined ^{10}Be, and Reedy and Marti (1991) reported results for ^{81}Kr. In these Apollo 15 cores, Nishiizumi et al. (1984a,b, 1989, 1990, 1997) have determined ^{26}Al, ^{10}Be, ^{36}Cl, ^{41}Ca, and ^{53}Mn; Imamura et al. (1973) determined ^{53}Mn and Fruchter et al. (1976, 1982) measured ^{26}Al. Nishiizumi et al. (1990) have measured ^{10}Be, ^{26}Al, ^{36}Cl, and ^{53}Mn in core 76001. Rao et al.

Table 2. Cross sections for ^{14}C production

Proton Energy (MeV)	O(p,x)^{14}C (mb)	Si(p,x)^{14}C (mb)	^{27}Al(p,x)^{14}C (mb)	Mg(p,x)^{14}C (mb)	Fe(p,x)^{14}C (mb)	Ni(p,x)^{14}C (mb)
500	2.190 ± 0.119	1.030 ± 0.056	1.480 ± 0.081	1.030 ± 0.056	0.166 ± 0.010	0.126 ± 0.007
400	2.120 ± 0.130	0.651 ± 0.035	0.853 ± 0.047	1.070 ± 0.058	0.100 ± 0.006	0.073 ± 0.004
300	1.900 ± 0.130	0.256 ± 0.015	0.753 ± 0.042	1.100 ± 0.062	0.033 ± 0.002	0.025 ± 0.004
200	1.510 ± 0.130	0.160 ± 0.015	0.586 ± 0.032	0.691 ± 0.038	0.024 ± 0.001	0.016 ± 0.001
159.4 ± 1.0			0.541 ± 0.029	0.437 ± 0.026		
159.0 ± 1.0				0.697 ± 0.038	0.016 ± 0.002	
158.4 ± 1.0	1.940 ± 0.138					
156.6 ± 1.0		0.207 ± 0.019				
149.4 ± 1.0	2.070 ± 0.146					
147.5 ± 1.0		0.177 ± 0.017				
139.7 ± 2.0					0.008 ± 0.0004	0.005 ± 0.0003
130.5 ± 2.0			0.571 ± 0.031	0.601 ± 0.034		
129.3 ± 2.0	1.980 ± 0.141					
127.5 ± 2.0		0.174 ± 0.010				
101.2 ± 3.0				0.385 ± 0.022		
99.1 ± 3.0	1.980 ± 0.141					
96.6 ± 3.0		0.090 ± 0.007				
89.1 ± 3.0	1.900 ± 0.135					
86.3 ± 3.0		0.065 ± 0.005				
81.0 ± 3.5			0.351 ± 0.019	0.320 ± 0.018		
79.0 ± 3.5	2.150 ± 0.152					
76.0 ± 3.5		0.031 ± 0.002				
68.9 ± 3.5	2.230 ± 0.158					
67.5			0.283 ± 0.016			
67.0				0.264 ± 0.015		
66.0		~0.013				
65.5 ± 4.0		0.020 ± 0.002				
63.8 ± 4.0	2.110 ± 0.149					
60.2 ± 4.0		0.010 ± 0.001				
60.0			0.112 ± 0.007			
58.7 ± 4.0	2.510 ± 0.178					
54.9 ± 4.0		0.006 ± 0.001				
52.5			0.041 ± 0.003			
50.0		~0.001				
45.7			0.137 ± 0.008			
44.4	1.76 ± 0.095					
40.2			0.088 ± 0.005			
38.2	1.38 ± 0.074					
33	0.908 ± 0.049					
28	0.085 ± 0.004					

(1994) measured the stable noble-gas isotopes ^{21}Ne, ^{22}Ne, and ^{38}Ar to study past SCR particle fluxes in 68815. We can use these other measurements to compare estimated solar-cosmic-ray fluxes deduced from ^{14}C with the fluxes derived from the longer-lived species for these and other lunar samples.

2. METHODS

Cosmogenic ^{14}C is extracted from rocks by fusion of the rock powder with Fe (which is used as a combustion accelerator) in an oxidizing environment. The sample is crushed, and several grams of iron chips are added. The sample is preheated to 500°C in air to remove contaminants due to organics and adsorbed CO_2. The sample is then heated to melting in a flow of oxygen in an radio-frequency furnace. Any evolved gases are passed over MnO_2 to remove sulfur compounds and CuO/Pt at 450°C to oxidize all carbonaceous gases to CO_2. The volume of the gas is measured, and the gas is diluted with about 0.5–2.5 cm^3 of ^{14}C-free CO_2. The CO_2 produced is converted to graphite by catalytic reduction over Fe (Donahue et al., 1990b). The graphite powder (usually about 0.5 mg) is pressed into an accelerator target. Samples of graphite as small as 100 micrograms are then analyzed by AMS. The basic studies on Bruderheim, other meteorites, and blank rock samples were published in Jull et al. (1989a, 1993). A complete description of the AMS analyses and calculations is given by Donahue et al. (1990a,b).

3. RESULTS AND DISCUSSION

3.1. New Cross Section Measurements

In order to estimate solar-cosmic-ray fluxes from measured production rates, we need good cross-section data for ^{14}C production from the elements found in meteorites and lunar rocks and cores. Until recently, few of the needed cross sections had been measured (Tamers and Delibrias, 1961; Sisterson et al., 1994, 1996). Measurements of cross sections are reported in Table 2 for ^{27}Al(p,x)^{14}C, Mg(p,x)^{14}C, Fe(p,x)^{14}C, and Ni(p,x)^{14}C. Thin-target foil-irradiation techniques were used which limited energy losses in each target to $<$2 MeV per foil and $<$10 MeV for an entire stack of targets. Experimental details of the irradiations have been summarized by Sisterson et al. (1994, 1996). Revised values, corrected for naturally-occurring ^{14}C in the target material, for the previously-reported important cross sections O(p,x)^{14}C and Si(p,x)^{14}C are included

Table 3. Results of ^{14}C measurements on Apollo 15 soil cores

Sample	Depth	(g/cm^2)	Mass (g)	^{14}C (dpm/kg)
15008,227	0–0.5 cm	0.413 ± 0.413	0.0983	44.0 ± 0.6
15008,228	0.5–1 cm	1.238 ± 0.413	0.0923	42.3 ± 0.6
15008,229	1.0–1.5 cm	2.063 ± 0.413	0.1109	40.3 ± 0.5
15008,230	1.5–2 cm	2.888 ± 0.413	0.1157	37.6 ± 0.5
15008,231	2.5–3.0 cm	4.538 ± 0.413	0.0916	33.5 ± 0.6
15008,232	3.5–4 cm	6.188 ± 0.413	0.1167	32.2 ± 0.5
15008,233	4.5–5 cm	7.838 ± 0.413	0.1034	32.8 ± 0.5
15008,234	9.5–10 cm	16.088 ± 0.413	0.0930	36.6 ± 0.5
15008,235	19.5–20 cm	32.588 ± 0.413	0.1068	38.5 ± 0.5
15006,284	0.7–1.2 cm	1.568 ± 0.413	0.0191	39.6 ± 1.7
15006,283	4.7–5.2 cm	8.168 ± 0.413	0.0686	31.9 ± 0.5
			0.0292	26.1 ± 4.0
			Weighted mean:	31.8 ± 0.8
15006,282	9.7–10.2 cm	16.42 ± 0.41	0.0704	32.8 ± 0.8
15006,281	19.7–20.2 cm	32.92 ± 0.41	0.0708	36.1 ± 0.6
15005,566	50–50.5 cm	82.91 ± 0.41	0.1034	34.8 ± 0.5
			0.0947	27.5 ± 1.3
			Weighted mean:	33.9 ± 2.4
15004,220	99.9–100.4 cm	165.2 ± 0.4	0.1446	21.6 ± 0.3
			0.1031	22.4 ± 1.2
			Weighted mean:	21.6 ± 0.3
15001,370	200.3–200.7 cm	330.8 ± 0.3	0.14812	10.7 ± 0.3

in Table 2 (Sisterson et al., 1994). Now that these cross sections are known, calculations show that >95% of the ^{14}C produced by solar protons in rock 68815 is via the reaction O(p,x)^{14}C (Sisterson et al., 1996). The cross sections reported here for the O(p,x)^{14}C reaction show that the early measurements of Tamers and Delibrias (1961) were essentially correct, when normalized by Audouze et al. (1967). As described below, using the new cross section values given in Table 2 as input to the theoretical calculations leads to better estimates for the solar-proton flux deduced from ^{14}C production profiles measured in lunar rocks and cores.

3.2. ^{14}C in the Apollo 15 Cores

We have made a series of measurements of ^{14}C concentration on samples from different depths from the Apollo 15 deep drill core 15001/6 and the drive tube 15008. The data are summarized in Table 3. The bulk density of the core when extruded from the coring devise was taken to be 1.65 g/cm^3, as used by Nishiizumi et al. (1990). A consistent set of results were obtained, although there may be a small offset of the longer core data from the 15008 core measurements. Some different cores densities have been reported elsewhere (Carrier et al., 1991), but these are not relevant to the discussion of irradiation conditions on the Moon.

3.2.1. GCR component

The measurements of ^{14}C in the longer core (15001-6) in Fig. 1 indicate the expected shape as a function of depth (Armstrong and Alsmiller, 1971; Reedy and Arnold, 1972). We initially tried to use the Reedy and Arnold (1972) model for lunar GCR particle fluxes and revised cross sections (see Jull et al., 1989b; Imamura et al., 1990; Sisterson et al., 1991a,b, 1994, 1996) to calculate the GCR production of ^{14}C. Using the new cross sections resulted in a GCR production rate about 10% higher than that originally calculated in Reedy and Arnold (1972). However, to fit the experimental data presented here, these calculated GCR production rates would still have to be increased by additional factors of ~1.25. Rao et al. (1994) also concluded that their estimate of GCR production rates had to be increased relative to Reedy-Arnold. In his work, Born (1973) raised the GCR production rates of Reedy and Arnold (1972) by 19%, and similar estimates were also used by Begemann et al. (1972).

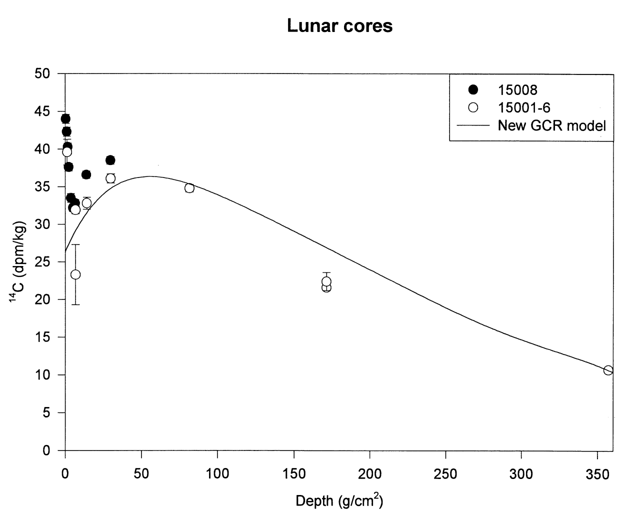

Fig. 1. ^{14}C concentration (dpm/kg) as a function of depth (g/cm^2) for the Apollo 15 cores. The open circles represent the deep drill core 15001/6; the filled circles represent drive tube 15008. The calculated GCR profile using the LCS model of Masarik and Reedy (1994) is shown as the solid line, using a bulk density of 1.65 g/cm^3 (Nishiizumi et al., 1990). As discussed in the text, the GCR profile has been increased by 5% to give the best agreement with the observed profile. Samples from core 15006 do not show evidence for the displacement of the top few cm of the core reported by Fruchter et al (1976) to explain their ^{26}Al data.

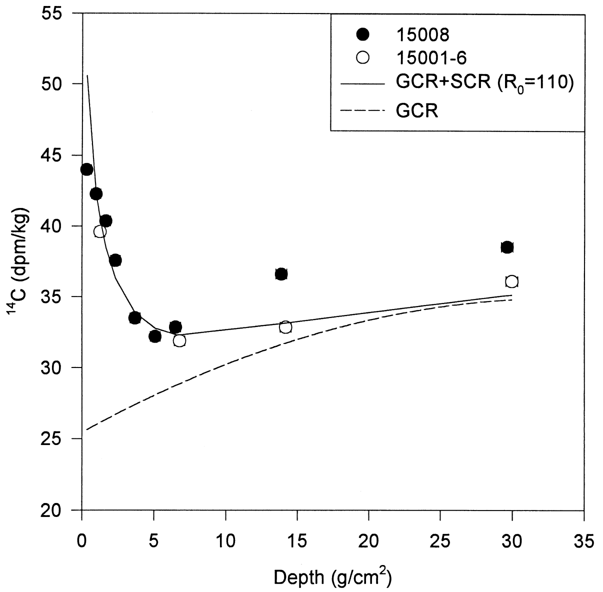

Fig. 2. ^{14}C concentration (dpm/kg) in the top 20 cm of cores 15008 (●) and 15006 (○). Best-fit calculations for the combined effect of SCR and GCR, using the SCR production for R_0 of 110 MV with J_{10} of 108p/cm^2/s. The calculated GCR curve is also plotted as the dashed line.

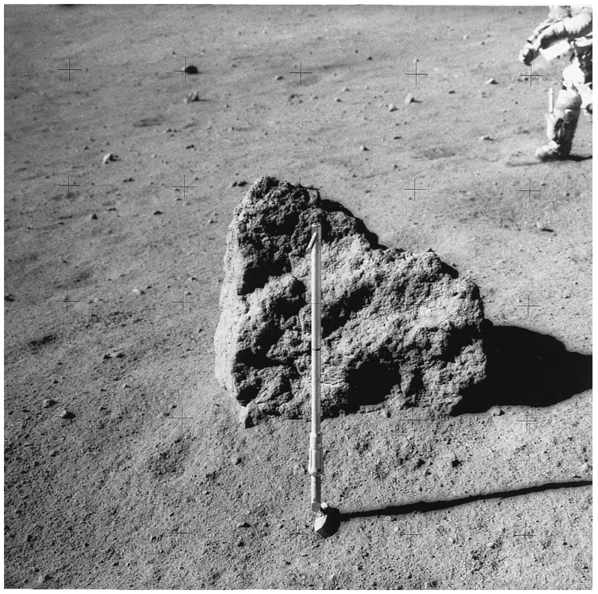

Fig. 3. Rock 68815 was removed from the top of this boulder at the Apollo 16 landing site. NASA photo no. AS16-108-17699

In order to resolve these model discrepancies, we embarked on a new calculation of GCR production rates using LCS, the LAHET Code System (e.g., Masarik and Reedy, 1994). LAHET (the Los Alamos High Energy Transport) is a improved version of the High Energy Transport Code that is often used for cosmic-ray studies (e.g., Armstrong and Alsmiller, 1971). These new calculations gave good agreement with experimental values, only requiring a small adjustment of 1.05 to raise the model GCR production rates to observed values (see Fig. 1). These new calculations mean that lunar LCS calculations for GCR ^{14}C production and similar calculations for meteorites, such as Knyahinya (Jull et al., 1994), are now in reasonable agreement. Nevertheless, to define the GCR production-rate curve unambiguously, considerably more data over the depth of the core will be required. For example, sampling at more depths would help define the curve in the region of the point at 165g/cm^2 shown in Fig. 1.

3.2.2. SCR component

Only the GCR component within 50 g/cm^2 of the surface is important to understanding the SCR component. Once we have normalized this component, we can analyze the excess values close to the surface and determine if they are due to SCR or other effects. The ^{14}C depth dependence for 0–35 g/cm^2 is shown in Fig. 2 . Using our GCR normalization, we could fit the observed ^{14}C depth profile with several solar-proton flux and rigidity parameter (R_0) combinations. We have calculated fluxes and rigidities using the latest cross sections for ^{14}C production from protons (Sisterson et al., 1991a,b, 1994, 1996 and this work) and the SCR calculational procedures of Reedy and Arnold (1972). We will discuss these calculations in detail in conjunction with the rock data in the following section. For the core samples, the best fits were obtained for a value of R_0 of 110 MV and with J_{10} flux of 108 protons/cm^2/s using the constant density model (1.65 g/cm^3). We obtained ninety-eight protons/cm^2/s using a variable density model (Carrier et al, 1991). Even with a varying density model, it is not possible to reconcile the points from core 15008 at 15 and 30 g/cm^2 with those of core 15006. Fruchter et al. (1976) reported on an apparent disturbance in ^{26}Al and ^{22}Na measurements at the top of core 15006. The fit to 15006 is satisfactory, and the data are not consistent with the proposal of Fruchter et al. (1976) that the top 3 g/cm^2 of this core have been removed. However, only one point (15006, 284) would be displaced off the trend in Fig. 2 if the assertion of Fruchter et al. (1976) were correct.

3.3. ^{14}C in Lunar Rock 68815

Our measurements on the Apollo 15 cores encouraged us to look at ^{14}C in a series of samples from lunar rock 68815. With a rock, we should obtain a complete SCR profile independent of any gardening or loss of material from the core top. Erosion by micrometeorites, of the order of mm/million years, will not have any effect on the ^{14}C. Rock 68815 was removed from the top of a large boulder at the Apollo 16 site (see Fig. 3). Results of our measurements are listed in Table 4. The density of the rock used was 2.8 g/cm^3 (Kohl et al., 1978). The results show that our measurements give a similar trend of ^{14}C with depth in the rock as observed previously in lunar rock 12053 by Begemann et al. (1972) and in rock 12002 by Boeckl (1972) but with much greater detail. Our values on 68815 using AMS are generally more precise in depth (due to the much smaller samples) and in ^{14}C content (due to the higher AMS sensitivity) than those from previous work. Also studied were two surface patina samples that covered a larger area of rock than the column cut. The geometry of the samples is shown in Figs. 4 and 5. The surface patina samples give ^{14}C values that bracket the 0–1 mm surface column sample. The ^{14}C values from the

Table 4. ^{14}C concentrations in rock 68815

Sample	Depth	(g/cm^2)[1]	Mass (g)	^{14}C (dpm/kg)
68815,298	Surface patina	0	0.0219	66.0 ± 1.6
68815,296	Surface patina	0	0.0324	51.5 ± 1.1
68815,301	0–1 mn	0.14 ± 0.14	0.0110	64.0 ± 2.0
68815,316	2.06–3.56 mm	0.79 ± 0.27	0.0257	44.4 ± 1.0
68815,319	5.62–6.62 mm	1.71 ± 0.18	0.0649	40.0 ± 0.5
68815,307	7–8 mm	2.1 ± 0.2	0.0338	47.2 ± 1.2
			0.0154	40.0 ± 3.1
			Weighted mean:	46.3 ± 2.4
68815,324	1.10–1.3 cm	3.4 ± 0.3	0.0711	40.5 ± 1.0
68815,327	2.25–2.5 cm	6.65 ± 0.35	0.0474	30.8 ± 1.0
68815,330	4.1–4.3 cm	11.8 ± 0.3	0.0683	33.8 ± 0.5

[1] The density of the rock is taken as 2.8 g/cm^3 (Kohl et al., 1978).

0–1 mm sample appear to agree well with the earlier work on 12002 and 12053. Our results do not show the very high surface value observed by Jull et al. (1995), which was based on an etch of a few tens of nm of the surface using a dilute HF solution. This component must be present, but does not contribute significantly to the observations here. It may have been removed on pretreatment of the sample to 500°C; however, further studies which compare these two extraction methods will resolve this apparent discrepancy.

The measurements for ^{14}C production in 68815 are illustrated in Fig. 6. There are several important points to consider from these data. The best fit was obtained using all samples down to 2 g/cm^2 except the surface patina sample 68815, 296, which yielded 51.5 dpm/kg. We believe this point may be low as the surface patina of a lunar rock can include glass splashes and may be covered with dust, so that this particular sample may not necessarily indicative of SCR production of ^{14}C. The best fit for these five samples had a standard deviation for their observed/calculated ratios of 20% with no change in this ratio as a function of depth. As a result, we find the best fits to the data for R_0 of 115 MV and J_{10} of 103 protons/ cm^2/s. However, we can also make other reasonable fits for different values of R_0 from 100 to 130 MV, with equivalent values of J_{10} of 130 to 88p/cm^2/s. All fits gave a flux of 19 p/cm^2/s for J ($E > 57$MeV). We also tried several other types of fits, such as power-law spectra in both energy and rigidity, but the best fits were found for spectra with the model energy distribution shown in Eqn. 1 used by Reedy and Arnold (1972). The best fit R_0 was that with the smallest standard deviation of the observed/calculated ratios of SCR activities using calculated SCR production rates for a wide range of Ro values. The average ratio for the best-fit Ro was then used to adjust the arbitrary solar-proton flux used in the calculation to get the best-fit flux. For the Apollo 15 cores, the best fit used the data between 0.8

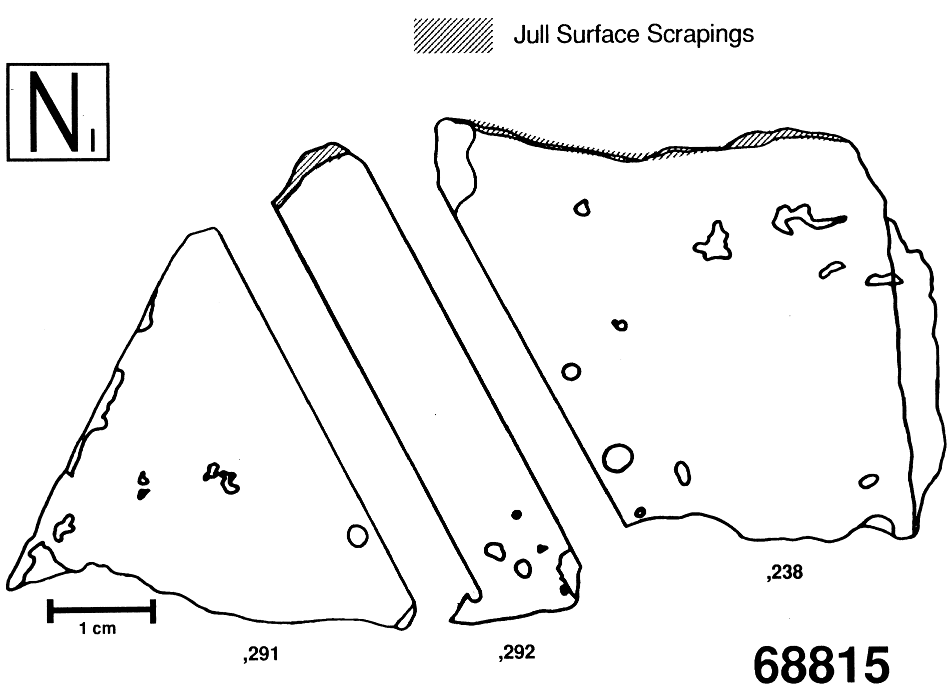

Fig. 4. Diagram of cutting of sample column from rock 68815. Courtesy NASA Johnson Space Center.

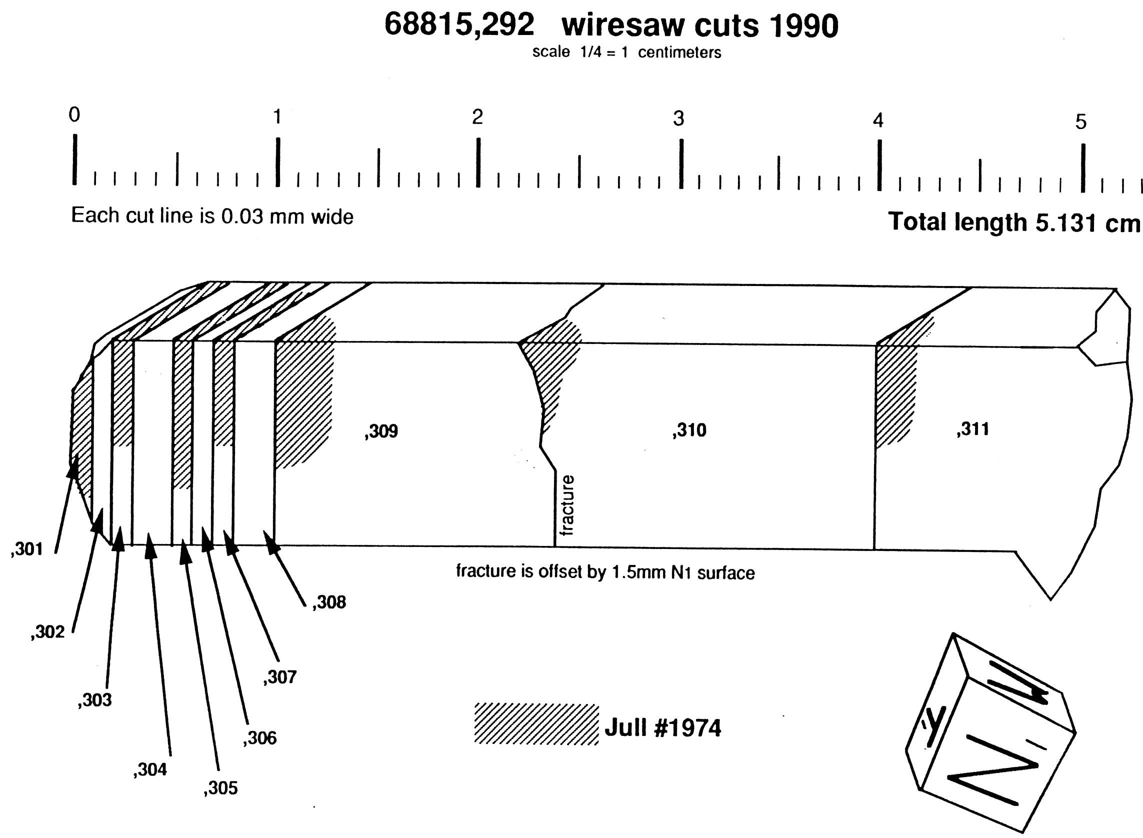

Fig. 5. Detail of depth slices cut from column 68815,292. Note that the sample numbers indicated here in some cases refer to the NASA parent numbers of the sub-samples used in our study. Samples 68815, 301 and 307 are the same. Other sample numbers used in the text correspond to the following parent numbers in brackets: 316 (303), 319 (305), 324 (309), 327 (310), 330 (311). Courtesy NASA Johnson Space Center.

and 5 g/cm^2 (the ^{14}C activity of the surface sample being significantly lower than calculated). The standard deviation of the observed/calculated ratio for these five samples was 11%. The ratio of observed to calculated SCR activities for the best fit did not show any systematic trend with depth. Deeper samples are consistent with this best fit.

Further, we can easily fit most data points from rock 68815 to the SCR profile deduced from the Apollo 15 core data. We expect the implanted component discussed by Jull et al. (1995) may affect the observations on the very surface, but our samples may sample too coarse depth increments to observe this very-surface effect observed in that HF-etching experiment.

3.4. ^{14}C in Apollo Core 76001

We have also obtained ^{14}C information as a function of depth in Apollo 17 drive tube core, 76001. This core was described as undisturbed by NASA documentation and was collected at a break in the slope of North Massif, on an 11° slope and close to a complex of boulders (Muehlberger et al., 1973, Nagle et al., 1979). Results are listed in Table 5 and Fig. 7. As can be seen in Fig. 7, the results show a disturbance of the core data in the top few g/cm^2 of this core and a lack of ^{14}C production by SCR. Samples below about 6 g/cm^2 show more normal behavior, although ^{14}C activities are still below those of

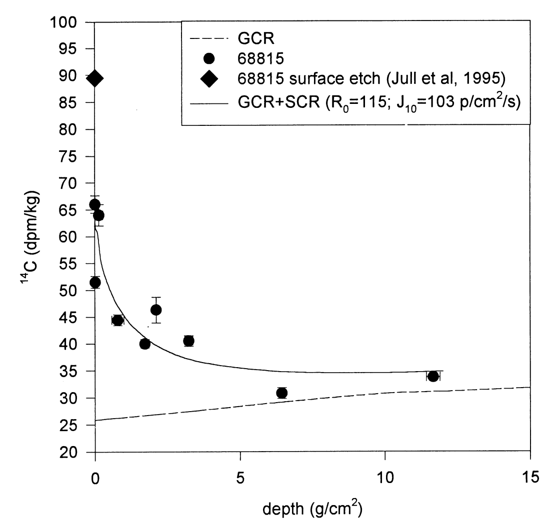

Fig. 6. ^{14}C (dpm/kg) as a function of depth in Apollo 16 rock 68815,292. The plot shows the experimental measurements (●) compared to the best fit of SCR of R_0 = 115 MV and J_0 = 103 p/cm^2/s, plus GCR (solid line), as well as the calculated GCR production, plotted as the dashed line. Bulk density of the rock is 2.8g/cm^3 (Kohl et al, 1978). Also shown is the higher surface value measured by Jull et al. (1995) from acid etching of the rock surface. This sample shows the effects of surface implantation by solar-wind ^{14}C in the top few nm.

Table 5. ^{14}C Results on Apollo 17 Station 6 Core, 76001

Sample	Depth (cm)	$(g/cm^2)^1$	Wt (mg)	^{14}C (dpm/kg)
76001,397	0–0.1	0.089 ± 0.089	88.5	32.1 ± 1.3
76001,398	0.1–0.2	0.27 ± 0.08	109.4	28.1 ± 1.1
76001,399	0.2–0.3	0.45 ± 0.08	114.1	31.2 ± 0.9
76001,400	0.3–0.4	0.62 ± 0.08	103.1	26.3 ± 1.1
76001,394	0.4–1.0	1.25 ± 0.5	82.3	38.3 ± 1.4
76001,401	1.0–1.5	2.23 ± 0.4	100.9	27.3 ± 1.5
76001,395	2.0–2.5	4.01 ± 0.4	87.9	31.3 ± 1.3
76001,396	2.5–3.0	4.90 ± 0.4	75.3	32.0 ± 1.5
76001,402	3.0–3.5	5.79 ± 0.4	114.1	27.7 ± 0.4
76001,403	5.0–5.5	9.35 ± 0.4	117.6	28.9 ± 0.6
76001,404	10.0–10.5	18.3 ± 0.4	96.9	27.9 ± 1.6
76001,405	20.0–20.5	36.0 ± 0.4	100.9	31.4 ± 0.5

[1] Assumed density: 1.78 g/cm^3

the Apollo 15 cores. This result is very similar to the observations of disturbances in ^{36}Cl in core 76001 (Nishiizumi et al., 1990). These radionuclides show a lack of SCR production. Evans et al. (1980) observed an excess of ^{26}Al and ^{53}Mn in the core for depths between about 2 and 20 g/cm^2 but a definite deficit below 2 g/cm^2. These profiles can be attributed to downslope movement affecting the concentration of the longer-lived nuclides (Evans et al., 1980; Nishiizumi et al., 1990). An explanation of these results would be disturbance of the core, perhaps by deposition of material irradiated at greater depth (to account for the longer-lived nuclides). This episode must have occurred recently enough on the Moon to allow for a minimum of SCR-produced ^{14}C and also ^{36}Cl (Nishiizumi et al., 1990). The results from core 76001 cannot be used to estimate SCR fluxes, due to this disturbance, but indicate the importance of an undisturbed core for such measurements.

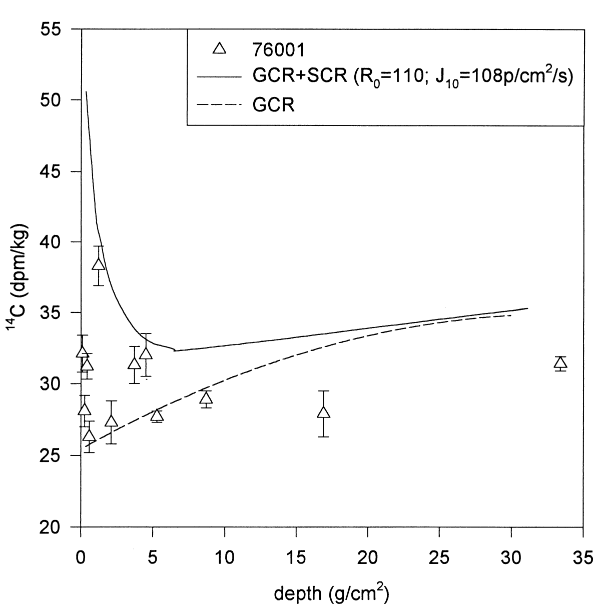

Fig. 7. ^{14}C (dpm/kg) as a function of depth in Apollo 16 station 6 core, 76001. Experimental measurements are shown (Δ), along with best-fit calculations, from Apollo 15 cores, for the combined effect of SCR using the value of R_0 of 110 MV and J_{10} of 108p/cm^2/s and GCR are plotted as a solid line. The calculated GCR curve is also plotted as the dashed line.

3.5. Comparisons to Other Radionuclides

In Table 6, we list for comparison the R_0 and flux estimates obtained from measurements of several long-lived and stable isotopes in lunar rocks. The values of R_0 and flux obtained in this work from ^{14}C measurements are, in general, larger than the equivalent values from studies using longer-lived nuclides. In addition, for some nuclides, cross sections are not sufficiently well determined to allow us to determine uniquely both R_0 and J_{10}. Nevertheless, the fact that the rigidity and fluxes for the relatively short-lived ^{14}C are higher, in general, than other nuclides, indicates that the SCR flux over the last ~30,000 years must have been greater than for longer intervals. The scatter in the integral fluxes above 30 and 60 MeV for entries in Table 6 are less than those for >10 MeV for the last few million years, which suggests that the fluxes for E > 30 and >60 MeV are better determined. As noted above, the integral flux >57 MeV was the one best determined from all fits to our ^{14}C data. Our best fits for SCR ^{14}C resulted in higher R_0 of 110–115 MV and a J_{10} flux of 108–98 protons/cm^2/s, respectively. In the following section, we discuss the other nuclide data and their relevance to this conclusion.

3.5.1. Comparison to results for nuclides of half-life > 10^6 yr

Kohl et al. (1978) estimated the value of J_{10} to produce the ^{53}Mn and ^{26}Al observed in 68815 to be about 70 protons/cm^2/s with a spectral shape parameter R_0 of 100 MV, but they noted that other combinations of flux (>10 MeV), R_0, and erosion rates would also work. The ^{10}Be data (Nishiizumi et al., 1988) cannot be fitted with R_0 = 100 MV, but all three nuclides, ^{10}Be, ^{26}Al, and ^{53}Mn could be fitted with lower values of R_0. However, at that time there were no cross sections below 135 MeV for the production of ^{10}Be, and Nishiizumi et al. (1988) used the estimated cross sections of Tuniz et al. (1984), thus their deduced solar-proton fluxes based on ^{10}Be were fairly uncertain. Recent measurements of cross sections for the production of ^{10}Be by protons (for example, Sisterson et al., 1997a,b,c; Bodemann et al., 1993; Schiekel et al., 1996) give a production rate for ^{10}Be in the surface layer of 68815 to be ~25% higher than those calculated using the Tuniz et al. (1984) data set. This result yields a solar proton spectral parameter of R_0 = 70 MV and J_{10} of 100 protons/cm^2/s. This confirms that both the cross sections used and the fluxes deduced from the ^{10}Be data for

Table 6. Solar-proton spectral parameters and 4π integral fluxes ($p/cm^2/s$) above 4 energies (in MeV) determined from spacecraft data (SEL, IMP-8, and SPME) and lunar samples using different isotopes

Time range	Data source	Reference	R_0 (MV)	$E > 10$	$E > 30$	$E > 60$	$E > 100$
1954–1964	^{22}Na, ^{55}Fe	Reedy (1977)	100	378	136	59	26
10^4 yr	^{14}C	This work	110–115	103	42	17	7
10^5 yr	^{41}Ca[1]	Klein et al. (1990)	70	120	28	7	1.5
		Fink et al. (1998)	80	200	56	16	4
$3' \ 10^5$ yr	^{81}Kr[1]	Reedy and Marti (1991)	~85	—	—	14	4
$5' \ 10^5$ yr	^{36}Cl[1]	Nishiizumi et al. (1995)	~75	100	26	7	2
10^6 yr	^{26}Al[2]	Kohl et al. (1978)	100	70	25	9	3
10^6 yr	^{10}Be, ^{26}Al[2]	Nishiizumi et al. (1995)	75	100	26	7	2
	^{10}Be, ^{26}Al	Michel et al. (1996)	125	55	24	11	5
		Fink et al. (1998)	100	89	32	12	4
$2' \ 10^6$ yr	^{10}Be, ^{26}Al[2]	Nishiizumi et al. (1998)	>70	—	~35	~8	~2
$5' \ 10^7$ yr	^{53}Mn	Kohl et al. (1978)	100	70	25	9	3
$\sim 2' \ 10^6$ yr[3]	$^{21,22}Ne$, ^{38}Ar	Rao et al. (1994)	80–90	58–87	~22	~7	~2

[1] The values obtained for ^{41}Ca, ^{36}Cl, and ^{81}Kr (in italics) are uncertain due to few cross-section measurements.
[2] The determinations for ^{26}Al before 1996 are based on old cross sections and could change using recently-measured cross sections (Michel et al., 1996; Sisterson et al., 1997c).
[3] The determinations based on stable nuclides depends on the erosion rate model used by Rao et al. (1994), who assumed an erosion rate of 1–2 mm/Ma.

68815 (Nishiizumi et al., 1988) were essentially correct. The latest cross sections for ^{10}Be and ^{26}Al were used by Fink et al. (1998) in their analysis of measurements in 74275. Also recently, Michel et al. (1996) estimated J_{10} of 55 protons/cm^2/s for $R_0 = 125$ MV based on their ^{10}Be and ^{26}Al cross sections and reported profiles. The studies by Fink et al. (1998) and Nishiizumi et al. (1995) give J_{10} values within ~15% of our estimate from ^{14}C, but not for the same R_0. Several radionuclides, including ^{14}C, are produced by reactions with threshold energies of 30–60 MeV, thus we must assume that fluxes for lower energies down to 10 MeV depend on the spectral shape parameter R_0 in the same way as Eqn. 1.

Hence, we reached the result that radioisotopes having a half-lives longer than 10^6 yr generally have lower J_{10} and lower R_0 than our new estimates for ^{14}C in this work. This is true if we compare the results of Kohl et al. (1978), Rao et al. (1994), Michel et al. (1996) and Fink et al. (1998) with our work. In addition, all of the various J_{30} and J_{60} fluxes in Table 6 for nuclides integrating over ~1–5 Myr have much less scatter than J_{10} and are all are consistently lower than J_{30} and J_{60} for ^{14}C.

3.5.2. *Comparison to results for nuclides of shorter half-life (3×10^5 yr)*

The radionuclide ^{81}Kr measured in 68815 for $E > 60$MeV, (Reedy and Marti, 1991) and new ^{41}Ca (Fink et al., 1998) data on 74275 appear to be in better agreement with our ^{14}C data, although both these papers indicate this estimate is uncertain due to uncertainties in the relevant cross sections. A study by Klein et al. (1990) of ^{41}Ca in rock 74275 resulted in an estimate of solar-proton flux of 120 protons/cm^2/s but with a low R_0 of about 70 MV. Recent calculations fitting the ^{41}Ca measurements on 74275 suggest a J_{10}, of ~200 protons/cm^2/s and R_0 of about 80 MV give a good fit to the observations (Fink et al., 1998). A lack of cross sections prevented Nishiizumi et al. (1989) from deducing solar-proton fluxes from their ^{36}Cl measurements. A preliminary set of cross sections were used for ^{36}Cl in rock 64455 by Nishiizumi et al. (1995). However, good solar proton fluxes derived from ^{36}Cl measurements can now be calculated using the recently reported new cross section measurements (Sisterson et al., 1997d). The short-lived radioisotopes ^{22}Na and ^{55}Fe show higher estimated solar-proton fluxes, reflecting the very high solar activity in the decade immediately before the recovery of the Apollo samples (Reedy, 1977). We also note that all values of J_{10} and R_0 estimated for all radionuclides are within the range of spacecraft measurements, which depend strongly on the solar cycle and show variations from 63 p/cm^2/s ($R_0 = 40$ MV) to 312 p/cm^2/s ($R_0 \sim 70$ MV), as discussed by Reedy (1977, 1996) and Goswami et al. (1988).

3.5.3. *Other studies involving ^{14}C*

Boeckl (1972) estimated a very high flux using limited cross sections, and he did not account for any implanted component. The renormalized early cross sections for $O(p,x)^{14}C$ (Audouze et al, 1967) are similar to those used here (Sisterson et al., 1991a,b, 1992, 1994, 1996). We conclude the much higher fluxes which Boeckl (1972) reported were due to an underestimation of the GCR contributions to the ^{14}C activities and an attempt to fit the high surface values without considering an implanted component (see Jull et al., 1995).

Rao et al. (1994) produced a summary of solar-proton fluxes and other model parameters for a series of best fits to radionuclide data for noble gases, ^{14}C, ^{26}Al, ^{53}Mn, and ^{81}Kr. To obtain the lowest estimates of solar-proton flux, they used several assumptions in their normalizations of the GCR component. They concluded that discrepancies between SCR parametrizations, deduced from ^{14}C and ^{81}Kr data, could be minimized but not eliminated. The highest values of J_{10} were found from ^{81}Kr, but Rao et al. (1994) still required a 20–30% higher flux but similar R_0 to explain ^{14}C, compared to the best range of values discussed by Rao et al. (1994) for Ne and Ar for erosion rates of 1–3 mm/Ma. Erosion does not affect ^{14}C.

4. LIMITS TO GIANT SOLAR PARTICLE EVENTS

For the time integrated by ^{14}C, of ~30,000 years, our R_0 = 110–115 MV mean solar proton flux >10 MeV of 98–108 protons/cm^2/s gives us a mean annual flux of 3.1–3.4 × 10^9 protons/cm^2/yr. Short-lived nuclides such as ^{22}Na are responsive to individual solar-particle events, SPEs (Reedy, 1977). We can use ^{14}C to estimate some limits to giant SPEs over the time scale of up to 30,000 years.

Let us define an equation for production by solar protons of ^{14}C so that

$$^{14}\mathrm{C(atoms)} = P_{av}(1 - e^{-\lambda t1})/\lambda + P_G e^{-\lambda t2} \qquad (2)$$

where P_{av} is the long-term average production rate over time t_1, and P_G is the production by a giant solar particle event occurring at time t_2 in the past. If we integrate over the mean life, τ, of ^{14}C using the first part of Eqn. 2, then we obtain 2.7 ± 0.1 × 10^{13} protons/cm^2 required to induce the observed activity of ^{14}C in lunar rocks. Let us consider the effects of a giant SPE of 10^{13} protons/cm^2, which would raise solar-proton production of ^{14}C. Using Eqn. 2, such an event 10^4 years ago would still affect the ^{14}C measured today in a lunar rock, but only about 30% of the originally-induced ^{14}C would remain. Thus, the present level of a SCR ^{14}C would be increased by 37%, for a giant SPE event of 10^{13} protons/cm^2 which occurred 100 years ago, but a similar event 10,000 years ago would leave an excess of only 11% SCR-produced ^{14}C today. This gives us a useful tool to estimate limits for the size and number of giant events. Our measurement of a possible increase of up to 25% compared to long-lived nuclides would require only one giant event of this magnitude in the last 10,000 yr. Rapid fluctuations of <100 yr in the tree-ring records of terrestrial ^{14}C over the last 7000 years are less than 3%, implying that the largest SPE during this period was less than 3 × 10^{11} protons/cm^2 (Lingenfelter and Hudson, 1980). Using the different radionuclide histories and the ^{14}C record in tree rings, it is possible to construct limits to giant solar-particle events (Reedy, 1996). Over the last 20 ka, there cannot have been more than one event of >5 × 10^{13} protons/cm^2. The longer-lived radionuclides integrate a longer time period, so the limit to one large SPE is higher. An alternative interpretation is that these limits represent a limit in integral flux over this time for a number of smaller events.

5. CONCLUSIONS

Results of ^{14}C depth profiles in cores from Apollo 15 and from the Apollo 16 rock 68815 display the expected high values near the very surface due to SCR effects with a steep drop in activity to depths of ~5–10 g/cm^2, a gradual rise to about 36 g/cm^2, and then a slow exponential decrease at increasing depth due to GCR. More detailed sampling of the GCR depth distribution is needed, but does not affect the interpretations of the SCR component. We interpret the SCR profiles as being due to fluxes of solar protons similar or up to 25% higher than those determined for other long-lived radionuclides, at comparable values of rigidity, R_0. We can explain the ^{14}C activities as due to an enhanced solar-proton flux over the time-scale integrated by ^{14}C. This is true if we compare our results for 68815 with those of Kohl et al. (1978) for ^{26}Al and ^{53}Mn, the Rao et al. (1994) estimates for several nuclides; the calculations of Michel et al. (1996) for ^{10}Be and ^{26}Al, and the measurements on 74275 by Fink et al. (1998), who measured ^{10}Be, ^{26}Al, and ^{41}Ca. However, estimates based on ^{41}Ca (Fink et al, 1998) and ^{81}Kr (Reedy and Marti, 1981) appear to be similar to ^{14}C, and recent studies by Fink et al. (1998) and Nishiizumi et al. (1995) on ^{10}Be and ^{26}Al give J_{10} values within ~15% of our estimate from ^{14}C, but not for the same R_0. Nishiizumi et al. (1995) determined fluxes close to ours, but they were fitted with a lower R_0. It is important when making these comparisons to take into account that fluxes determined from different nuclides need to be compared for the same energy range (E) and rigidity R_0. Using the same data, one can usually fit a range of different combinations of higher J_{10} and lower R_0 to the same depth profiles

Results on Apollo 17 core 76001 show evidence in the ^{14}C profile for a recent disturbance, so that little SCR ^{14}C was detected. These results are similar to those obtained by Nishiizumi et al. (1990) for ^{36}Cl in this core and are in contrast to the profiles of longer-lived radionuclides.

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REFERENCES

Armstrong T. W. and Alsmiller R. G., Jr. (1971) Calculation of cosmogenic radionuclides in the Moon and comparison with Apollo measurements. *Proc. 2nd Lunar Sci. Conf.*, 1729–1745.

Audouze J., Epherre M., and Reeves H. (1967) Survey of experimental cross sections for proton-induced spallation reactions with He^4, C^{12}, N^{14}, and O^{16}. In *High-Energy Nuclear Reactions in Astrophysics* (ed. B. S. P. Shen), pp. 255–271.

Begemann F., Vilcsek E., Rieder R., Born W., and Wänke H. (1970) Cosmic-ray produced radioisotopes in lunar samples from the Sea of Tranquillity (Apollo 11). *Proc. Apollo 11 Lunar Sci. Conf.*, 995–1005.

Begemann F., Born W., Palme H., Vilcsek E., and Wänke H.(1972) Cosmic-ray produced radioisotopes in Apollo 12 and Apollo 14 samples. *Proc. Lunar Sci. Conf. 3rd*, 1693–1702.

Bodemann R. et al. (1993) Production of residual nuclei by proton-induced reactions on carbon, nitrogen, oxygen, magnesium, aluminum, and silicon. *Nucl. Instrum. Methods* **B82,** 9–31.

Boeckl R. S. (1972) A depth profile of ^{14}C in lunar rock 12002. *Earth Planet. Sci. Lett*. **16,** 269–272.

Born W. (1973) ^{14}C in Meteoriten und Mondproben und ihre Deutung durch Vergleich mit berechneten Tiefenprofilen. Doctoral thesis, Universität Mainz.

Carrier W. D. III, Olhoeft G. R., and Mendell W. (1991) Physical properties of the lunar surface. In *The Lunar Sourcebook* (ed. G. Heiken et al.), pp. 475–594. Cambridge.

Donahue D. J., Jull A. J. T., and Linick T. W. (1990a) Isotope-ratio and background corrections for accelerator mass spectrometry radiocarbon measurements. *Radiocarbon* **32,** 135–142.

Donahue D. J., Jull A. J. T., and Toolin L. J. (1990b) Radiocarbon measurements at the University of Arizona AMS facility. *Nucl. Instrum. Methods* **B52,** 224–228.

Evans J. C., Fruchter J. S., Reeves J. H., Rancitelli L. A., and Perkins R. W. (1980) Recent depositional history of Apollo 16 and 17 cores. *Proc. 11th Lunar Planet. Sci. Conf.,* 1497–1509.

Fink D., Klein J., Middleton R., Vogt S., Herzog G. F., and Reedy R. C. (1998) ^{41}Ca, ^{26}Al, and ^{10}Be in lunar basalt 74275 and ^{10}Be in the double drive tube 74002/74001. *Geochim. Cosmochim. Acta,* in press.

Fireman E. L., DeFelice J., and D'Amico J. (1976) The abundances of ^{3}H and ^{14}C in the solar wind. *Earth Planet. Sci. Lett*. **32,** 185–190.

Fireman E. L., DeFelice J., and D'Amico J. (1977) ^{14}C in lunar soil: Temperature-release and grain-size dependence. *Proc. 8th Lunar Sci. Conf,* 3749–3754.

Fireman E. L. (1978) Carbon-14 in lunar soil and in meteorites. *Proc. 9th Lunar Planet. Sci. Conf.,* 1647–1654.

Fruchter J. S., Rancitelli L. A., and Perkins R. W. (1976) Recent and long-term mixing of the lunar regolith based on ^{22}Na and ^{26}Al measurements in Apollo 15, 16, and 17 deep drill stems and drive tubes. *Proc. 7th Lunar Sci. Conf.,* 27–39.

Fruchter J. S., Evans J. C., Reeves J. H., and Perkins R. W. (1982) Measurement of ^{26}Al in Apollo 15 core 15008 and ^{22}Na in Apollo 17 rock 74275. *Lunar Planet. Sci.* **13,** 243–244.

Gault D. E., Hörz F., Brownlee D. E., and Hartung J. B. (1974) Mixing of the lunar regolith. *Proc. 5th Lunar Sci. Conf.,* 2365–2386.

Goswami J. N., McGuire R. E., Reedy R. C., Lal D., and Jha R. (1988) Solar flare protons and alpha particles during the last three solar cycles. *J. Geophys. Res.* **93,** 7195–7205.

Herzog G. F. (1994) Applications of accelerator mass spectrometry in extraterrestrial materials. *Nucl Instrum. Methods Phys. Res*. **B92,** 492–499.

Imamura M. et al. (1990) Measurement of production cross sections of ^{14}C and ^{26}Al with high-energy neutrons up to $E_n = 38$ MeV by accelerator mass spectrometry. *Nucl. Instrum. Methods Phys. Res.* **B52,** 595–600.

Jull A. J. T. and Cloudt S. (1996) Evidence for recent gardening or disturbance in lunar core 76001 from solar cosmic ray records of ^{14}C. *Lunar. Planet. Sci*. **27,** 627–628.

Jull A. J. T., Donahue D. J., and Linick T. W. (1989a) Carbon-14 activities in recently-fallen meteorites and Antarctic meteorites. *Geochim. Cosmochim. Acta* **53,** 1295–1300.

Jull A. J. T., Englert P. A. J., Donahue D. J., Reedy R. C., and Lal D. (1989b) Cosmogenic nuclide production rates: Carbon-14 from neutron spallation. *Lunar Planet. Sci.* **20,** 490–491.

Jull A. J. T., Donahue D. J., and Reedy R. C. (1991) Carbon-14 in Apollo 15 cores. *Lunar Planet. Sci.* **22,** 665–666.

Jull A. J. T., Donahue D. J., and Reedy R. C. (1992) ^{14}C depth profile in lunar rock 68815. *Lunar Planet. Sci..* **23,** 639–640.

Jull A. J. T., Donahue D. J., Cielaszyk E., and Wlotzka F. (1993) Carbon-14 terrestrial ages and weathering of twenty-seven meteorites from the southern high plains and adjacent areas (USA). *Meteoritics* **28,** 188–195.

Jull A. J. T., Donahue D. J., Reedy R. C., and Masarik J. (1994) A carbon-14 depth profile in the L5 chondrite Knyahinya. *Meteoritics* **29,** 649–738.

Jull A. J. T., Lal D., and Donahue D. J. (1995) Evidence for a noncosmogenic implanted ^{14}C component in lunar samples. *Earth Planet. Sci. Lett.* **136,** 693–702.

Keays R. R., Ganapathy R., Laul J. C., Anders E., Herzog G. F. and Jeffrey P. M. (1970) Trace elements and radioactivity in lunar rocks: Implications for the meteorite infall, solar-wind flux and formation conditions of the moon. *Science,* **167,** 490–493.

Klein J. et al. (1990) Average SCR flux over the last 10^5 years: Inference from ^{41}Ca in lunar rock 74275. *Lunar Planet. Sci.* **21,** 635–636.

Kohl C. P., Murrell M. T., Russ G. P. III, and Arnold J. R. (1978) Evidence for the constancy of the solar cosmic ray flux over the past ten million years: ^{53}Mn and ^{26}Al measurements. *Proc. 9th Lunar Planet. Sci. Conf. 9th,* 2299–2310.

Langevin Y., Arnold J. R., and Nishiizumi K. (1982) Lunar surface gardening processes: Comparisons of model calculations with radionuclide data. *J. Geophys. Res*. **87,** 6681–6691.

Lanzerotti L. J., Reedy R. C., and Arnold J. R. (1973) Alpha particles in solar cosmic rays over the last 80,000 years. *Science* **179,** 1232–1234.

Lingenfelter R. E. and Hudson H. S. (1980) Solar particle fluxes and the ancient sun. In *The Ancient Sun: Fossil record in the Earth, Moon and Meteorites* (ed. R. O. Pepin et al.), pp. 69–79. Pergamon.

Masarik J. and Reedy R. C. (1994) Effects of bulk composition on nuclide production processes in meteorites. *Geochim. Cosmochim. Acta* **58,** 5307–5317.

Michel R., Leya I., and Borges L. (1996) Production of cosmogenic nuclides in meteoroids: Accelerator experiments and model calculations to decipher the cosmic ray record in extraterrestrial matter. *Nucl. Instrum. Methods Phys. Res*. **B113,** 434–444.

Muehlberger W. R. et al. (1973) Preliminary geologic investigation of the Apollo 17 landing site. Apollo 17 Prelim. Sci. Rept. NASA Spec. Publ. 330, 6-1–6-91.

Nagle J. S. (1979) Drive tube 76001 - continuous accumulation with complications? *Proc. 10th Lunar Sci. Conf.,* 1385–1399.

Nishiizumi K., Elmore D., Ma X. Z., and Arnold J. R. (1984a) ^{10}Be and ^{36}Cl depth profiles in an Apollo 15 drill core. *Earth Planet. Sci. Lett.* **70,** 157–163.

Nishiizumi K., Klein J., Middleton R., and Arnold J. R. (1984b) ^{26}Al depth profile in Apollo 15 drill core. *Earth Planet. Sci. Lett.* **70,** 164–168.

Nishiizumi K. et al. (1988) ^{10}Be profiles in lunar surface rock 68815. *Proc. 18th Lunar Planet. Sci. Conf.,* 79–85.

Nishiizumi K., Kubik P. W., Elmore D., Reedy R. C., and Arnold J. R. (1989) Cosmogenic ^{36}Cl production rates in meteorites and the lunar surface. *Proc. 19th Lunar Planet. Sci. Conf.,* 305–312.

Nishiizumi K., Arnold J. R., Kubik P. W., and Sharma P. (1990) New results on history of gardening in lunar cores 15008 and 76001 using cosmogenic radionuclides. *Lunar Planet. Sci*. **21,** 895–896.

Nishiizumi K. et al. (1995) Final results of cosmogenic nuclides in lunar rock 64455. *Lunar Planet. Sci*. **26,** 1055–1056.

Nishiizumi K., Caffee M. W., and Arnold J. R. (1997) ^{10}Be from the active sun. *Lunar Planet. Sci*. **28,** 1027–1028.

Rao M. N., Garrison D. H., Bogard D. D., and Reedy R. C. (1994) Determination of the flux and energy distribution of energetic solar protons in the past 2 Myr using lunar rock 68815. *Geochim. Cosmochim. Acta* **58,** 4231–4245.

Reedy R. C. (1977) Solar proton fluxes since 1956. *Proc. 8th Lunar Sci. Conf.,* 825–839.

Reedy R. C. (1980) Lunar radionuclide records of average solar-cosmic-ray fluxes over the last ten million years. In *The Ancient Sun: Fossil record in the Earth, Moon, and Meteorites* (eds. R. O. Pepin et al.), pp. 365–386. Pergamon.

Reedy R. C. (1996) Constraints on solar particle events from comparisons of recent events and million-year averages. In *Solar Drivers of Interplanetary and Terrestrial Disturbances* (ed. K. S. Balasubramaniam et al.); *Astron Soc. Pacific Conf. Ser*. **95,** 429–436.

Reedy R. C. and Arnold J. R. (1972) Interactions of solar and galactic cosmic-ray particles with the Moon. *J. Geophys. Res*. **77,** 537–555.

Reedy R. C. and Marti K. (1991) Solar-cosmic-ray fluxes during the last ten million years. In *The Sun in Time* (ed. C. P. Sonett et al.), pp. 260–287, Univ. Arizona Press.

Reedy R. C., Arnold J. R., and Lal D. (1983) Cosmic-ray record in solar system matter. *Annu. Rev. Nucl. Part. Sci*. **33,** 505–537.

Schiekel Th. et al. (1996) Nuclide production by proton-induced reactions on elements ($6 < Z < 29$) in the energy range from 200 to 400 MeV. *Nucl. Instrum. Methods Phys. Res*. **B114,** 91–119.

Sisterson J. M., Roman H., Vogel J. S., Southon J. R., and Reedy R. C. (1991a) Determination of solar-proton fluxes using carbon-14 in lunar rocks. *Lunar Planet. Sci.* **22,** 1267–1268.

Sisterson J. M., Jull A. J. T., Donahue D. J., Koehler A. M., Reedy R. C., and Englert P. A. J. (1991b) Cross sections for production of carbon-14 from oxygen and silicon: Implications for cosmogenic production rates. *Meteoritics* **26,** 395–396.

Sisterson J. M. et al. (1992) Cross section measurements for the production of carbon-14 and beryllium-10: Improved estimates for cosmogenic nuclide production rates. *Lunar Planet. Sci*. **23,** 1305–1306.

Sisterson J. M. et al. (1994) Revised solar cosmic ray fluxes estimated using measured depth profiles of ^{14}C in lunar rocks: The importance of good cross section measurements. *Nucl. Instrum. Methods Phys. Res*. **B92,** 510–512

Sisterson J. M. et al. (1996) Revised solar cosmic ray fluxes estimated using measured depth profiles of ^{14}C in lunar rocks: The importance of good ^{14}C cross section determinations. *Lunar Planet. Sci*. **27,** 1209–1210.

Sisterson J. M. et al. (1997a) Measurement of proton production cross sections of ^{10}Be and ^{26}Al from elements found in lunar rocks. *Nucl. Instrum. Methods Phys. Res*. **B123,** 324–329.

Sisterson J. M. et al. (1997b) Measuring excitation functions needed to interpret cosmogenic nuclide production in lunar rocks. In *Conf. Applications of Accelerators in Research and Industry* (eds. J. L. Duggan and I. L. Morgan); *AIP Conf. Proc*. **392,** 811–814.

Sisterson J. M., Kim K., Caffee M. W., and Reedy R. C. (1997c) ^{10}Be and ^{26}Al production in lunar rock 68815: Revised production rates using new cross section measurements. *Lunar Planet Sci*. **28,** 1327–1328.

Sisterson J. M., Nishiizumi K., Caffee M. W., Imamura M., and Reedy R. C. (1997d) Revised ^{36}Cl production rates in lunar rock 64455 using new cross section measurements. *Lunar Planet. Sci*. **28,** 1329–1330.

Tamers M. A. and Delibrias G. (1961) Sections efficaces de l'oxygéne 16 pour la production de carbone 14 par des protons de hautes énergies. *Compt. Rend*. **253,** 1202–1203.

Tuniz C. et al. (1984) Beryllium-10 contents of core samples from the St. Severin meteorite. *Geochim. Cosmochim. Acta* **48,** 1867–1872.

Vogt S., Herzog G. F., and Reedy R. C. (1990) Cosmogenic nuclides in extraterrestrial materials. *Rev. Geophys.* **28,** 253–275.

Wieler R. et al. (1996) Exposure history of the Torino meteorite. *Meteorit. Planet. Sci.* **31,** 265–272.