^{10}Be Profiles in Lunar Surface Rock 68815

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Cosmic-ray-produced ^{10}Be ($t_{1/2}$ = 1.6 × 10^6 years) activities have been measured in 14 carefully ground samples of lunar surface rock 68815. The ^{10}Be profiles from 0 to 4 mm are nearly flat for all three surface angles measured and show a very slight increase with depth from the surface to a depth of 1.5 cm. These depth profiles are in contrast to the SCR (solar cosmic ray)-produced ^{26}Al and ^{53}Mn profiles measured from these same samples. There is no sign of SCR-produced ^{10}Be in this rock. The discrepancy between the data and the Reedy-Arnold theoretical calculation for SCR ^{10}Be production (about 2 atoms/min/kg at the surface) can be explained in two ways: (1) The low-energy cross sections for proton-induced ^{10}Be production from oxygen are lower than those used in the calculations, or (2) compared to the reported fits for ^{26}Al and ^{53}Mn, the solar proton spectral shape is actually softer (exponential rigidity parameter R_0 less than 100 MV), the omnidirectional flux above 10 MeV is higher (more than 70 protons/cm^2 s), and the erosion rate is higher (greater than 1.3 mm/m.y.). ^{10}Be, as a medium- to high-energy product, is a very useful nuclide for determining the SCR spectral shape in the past.

INTRODUCTION

Variations in the flux and spectrum of solar cosmic ray (SCR) particles are related to the variations of solar activity. Knowledge of the history of solar activity is extremely important not only to understanding solar physics but also quite possibly the climatic history of the Earth (for example, the glaciation cycle). Although direct SCR measurements by satellites have been performed only for the last few decades, we do have a good way to study the past record. Cosmic rays produce radioactive and stable nuclides during interactions with lunar surface materials and meteorites. The concentrations of these cosmogenic nuclides are directly related to the average cosmic-ray intensity in the past. The nuclides of interest are produced not only by SCR but also by galactic cosmic rays (GCR). The GCR do not have a solar origin although their spectrum and flux are modulated to some extent by solar activity. The much lower energy (but higher flux) of the SCR means that their effects can only be seen in the top few millimeters of lunar materials while GCR-produced nuclides dominate at greater depths. Except for a few cases (e.g., *Nishiizumi et al.,* 1986; *Evans et al.,* 1987), the record of SCR effects is erased in meteorites during passage through the Earth's atmosphere. Details concerning these two types of cosmic rays and their interactions are given in several articles (e.g., *Reedy and Arnold,* 1972; *Reedy,* 1980; *Reedy et al.,* 1983).

Previously, we have studied the depth profiles of SCR-produced radionuclides in the upper 2 cm of lunar rocks and determined the average SCR parameters, flux, and energy spectrum over a time scale from a few months to 10 m.y. The comparison of ^{26}Al ($t_{1/2}$ = 7.05 × 10^5 years) and ^{53}Mn ($t_{1/2}$ = 3.7 × 10^6 years) depth profiles in the surface of three lunar rocks, 12002 (*Finkel et al.,* 1971), 14321 (*Wahlen et al.,* 1972), and 68815 (*Kohl et al.,* 1978), with the theoretical SCR production profiles (*Reedy and Arnold,* 1972) indicates that the flux of solar protons over the past 5 to 10 m.y. was similar to that during the past million years and that the average SCR spectrum and flux were characterized by an exponential rigidity with a spectral shape parameter R_0 = 100 MV (cf. *Reedy and Arnold,* 1972) and a flux J = 70 protons/cm^{2}s (E > 10 MeV, 4 π). These calculations assume 0.5-2.2 mm/m.y. erosion rate for the three rocks (*Kohl et al.,* 1978; *Russ and Emerson,* 1980). It is possible to fit the data also with R_0 in the range 70-150 MV (a higher value of R_0 corresponds to more higher-energy protons per lower-energy proton) with appropriate adjustments of flux J and erosion rate. [The excitation functions for producing ^{26}Al and ^{53}Mn by proton-induced reactions are quite similar (*Reedy and Arnold,* 1972)]. In this present work, we measured ^{10}Be ($t_{1/2}$ = 1.6 × 10^6 years) in rock 68815 by accelerator mass spectrometry (AMS) to investigate the SCR production of this nuclide and to verify the SCR parameters. Rock 68815 is a breccia that was collected from the top of a meter-high boulder. The ^{81}Kr-Kr exposure age of this rock is 2.04 ± 0.08 m.y. (*Drozd et al.,* 1974), and 68815 is thought to have been ejected by the South Ray crater event from a depth that cosmic-ray particles could not reach.

EXPERIMENTAL METHODS AND RESULTS

Fourteen samples were separated from aliquant samples that we had previously dissolved and used for ^{26}Al and ^{53}Mn measurements (*Kohl et al.,* 1978). The samples measured were from three different zenith angles [A 48 ± 16°, B 41 ± 17°, and C 29 ± 15° (*Russ and Emerson,* 1980)] and four different nominal depths (0-0.5, 0.5-1.0, 1.0-2.0, and 2.0-4.0 mm). A 4-8 mm and a 10-15 mm layer were also obtained from near the bottom of our specimen from beneath face A and face C. The details of the grinding procedures were described by *Kohl et al.* (1978). The sample sizes ranged from 0.6 to 2.9 g. About 700 μg of Be carrier was added to each sample dissolved. Beryllium was separated from other elements and purified by anion exchange, cation exchange, and Be-acetylacetone extraction. Finally, $Be(OH)_2$ was precipitated

TABLE 1. ^{10}Be in 68815 (dpm ^{10}Be/kg).

Depth (g/cm^2)*	Face A	Face B	Face C
0 -0.13	6.19 ± 0.21	7.28 ± 0.37	6.92 ± 0.24
0.13-0.28	6.75 ± 0.23	7.22 ± 0.65	6.47 ± 0.30
0.28-0.57	6.61 ± 0.18	7.24 ± 0.66	6.81 ± 0.20
0.57-1.03	6.81 ± 0.21	7.21 ± 0.32	7.07 ± 0.29
1.03-2.19†	7.22 ± 0.21		
2.8 -4.2†,‡	7.43 ± 0.53		

*These depths are averages over the three faces according to calculations by *Russ and Emerson* (1980). The depths of the individual faces differ no more than 15% from the averages. Values in the figures were plotted at the actual depth for each face. A density of 2.8 g/cm^3 was used in the depth calculations.

†The two deepest samples were ground from areas under both faces A and C.

‡The depth for this layer is from *Kohl et al.* (1978).

with water containing about 2% of ^{17}O so that $^{9}Be^{17}O$ would pass into the accelerator with $^{10}Be^{16}O$ (*Imamura et al.,* 1984).

The ^{10}Be measurements were carried out at the University of Tokyo's tandem Van de Graaff accelerator. The apparatus and method used for the accelerator mass spectrometry were essentially those described previously (*Imamura et al.,* 1984). We selected a 3.5 MV terminal voltage for the ^{10}Be measurements. We measured $^{10}Be/^{9}Be$ ratios in the range 1-5 × 10^{-10} with experimental errors of 3-9% (±1 σ). The $^{10}Be/^{9}Be$ measured values were normalized to ICN-UCSD ^{10}Be standard (*Nishiizumi et al.,* 1984). The ^{10}Be activities obtained from 68815 are given in Table 1.

DISCUSSION

The ^{10}Be activity depth profiles in the three faces A, B, and C of 68815 are shown in Fig. 1a. The ^{10}Be results were adjusted to saturation using the ^{81}Kr-Kr exposure age of 2.04 ± 0.08 m.y. (*Drozd et al.,* 1974). The saturation activities are used for all the following discussions. The ^{26}Al and ^{53}Mn depth profiles in the same samples are also shown in Figs. 1b and 1c. The curves shown in Figs. 1b and 1c are the Reedy-Arnold theoretical profiles for the sum of SCR and GCR production rates for each nuclide (*Reedy and Arnold,* 1972). The curves are slightly modified from the previous paper (*Kohl et al.,* 1978) by calculating the production rate on a point by point basis (*Russ and Emerson,* 1980). SCR parameters R_0 = 100 MV and J = 70 p/cm^2 s, a 2.0 m.y. exposure age, and 0.0 and 1.0 mm/m.y. erosion rates are adopted for these calculations. The ^{10}Be profiles are essentially the same for all three faces and are nearly flat except for a slight increase with increasing depth. This profile is in remarkable contrast to the ^{26}Al and ^{53}Mn profiles, which show sharp increases in activity toward the surface due to SCR production of these nuclides.

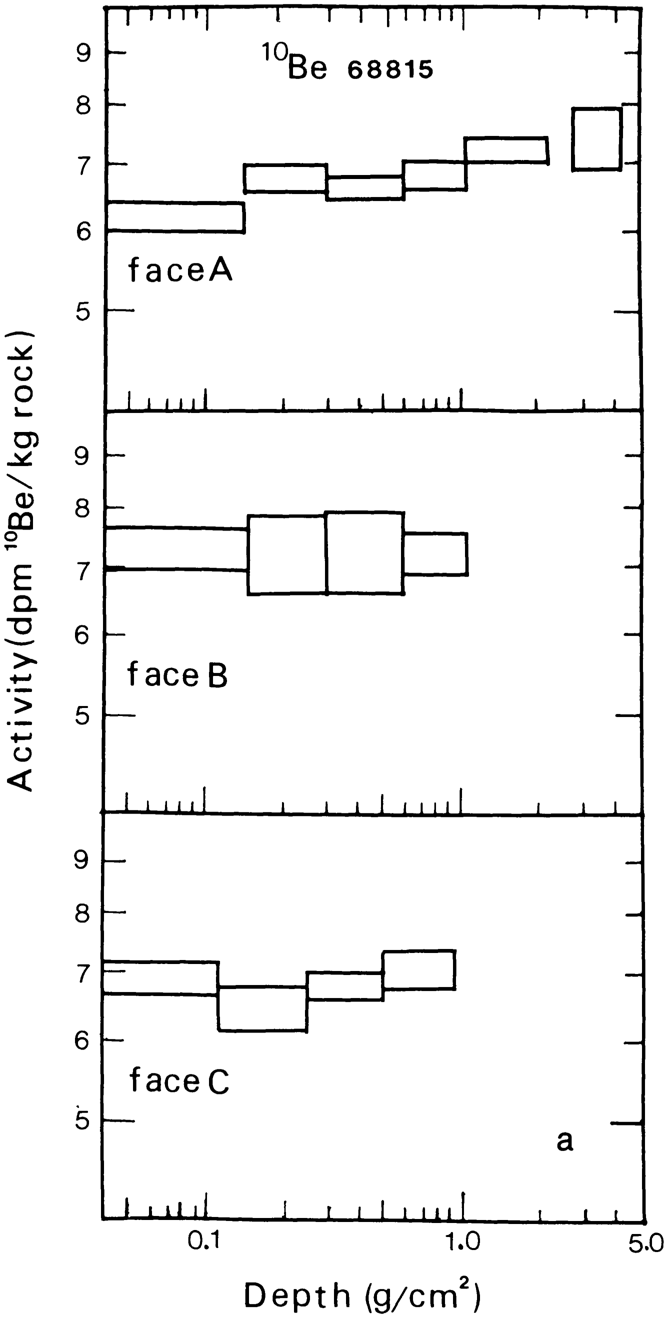
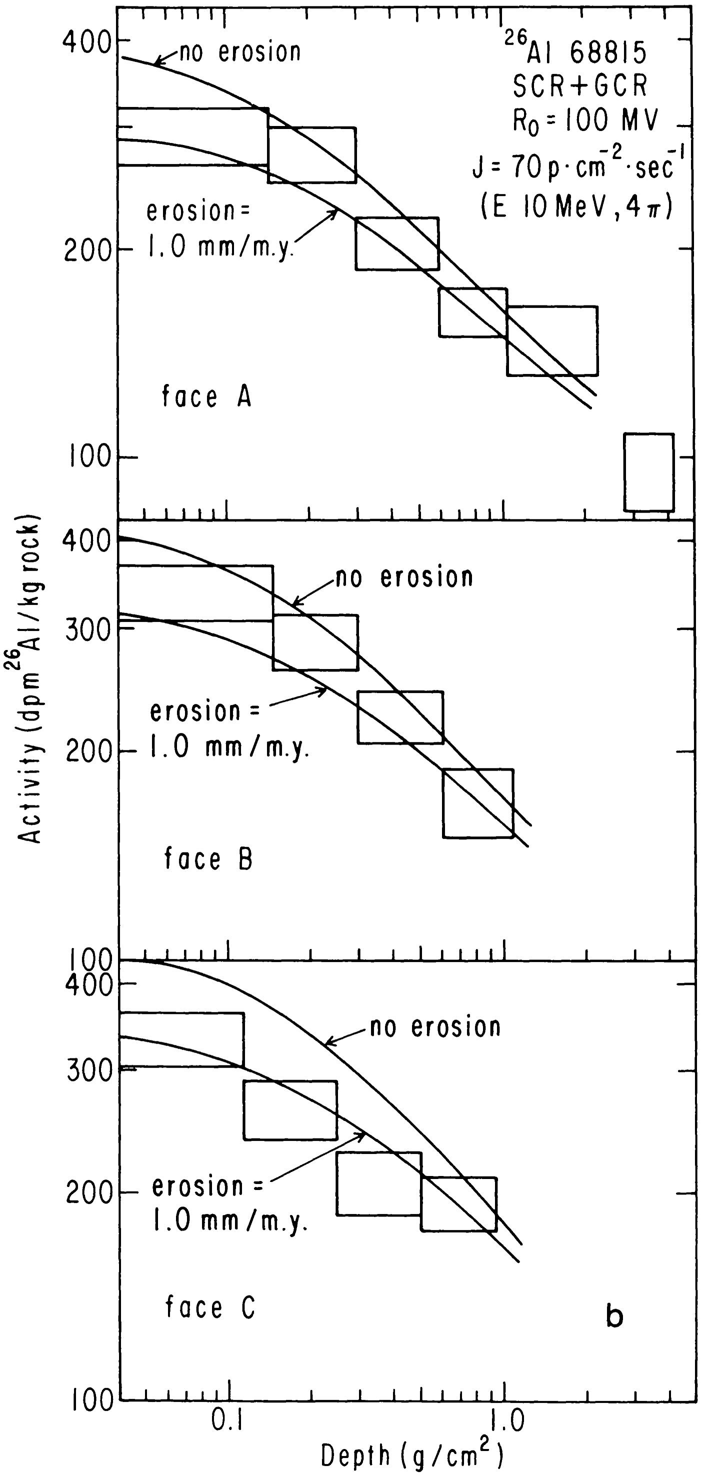
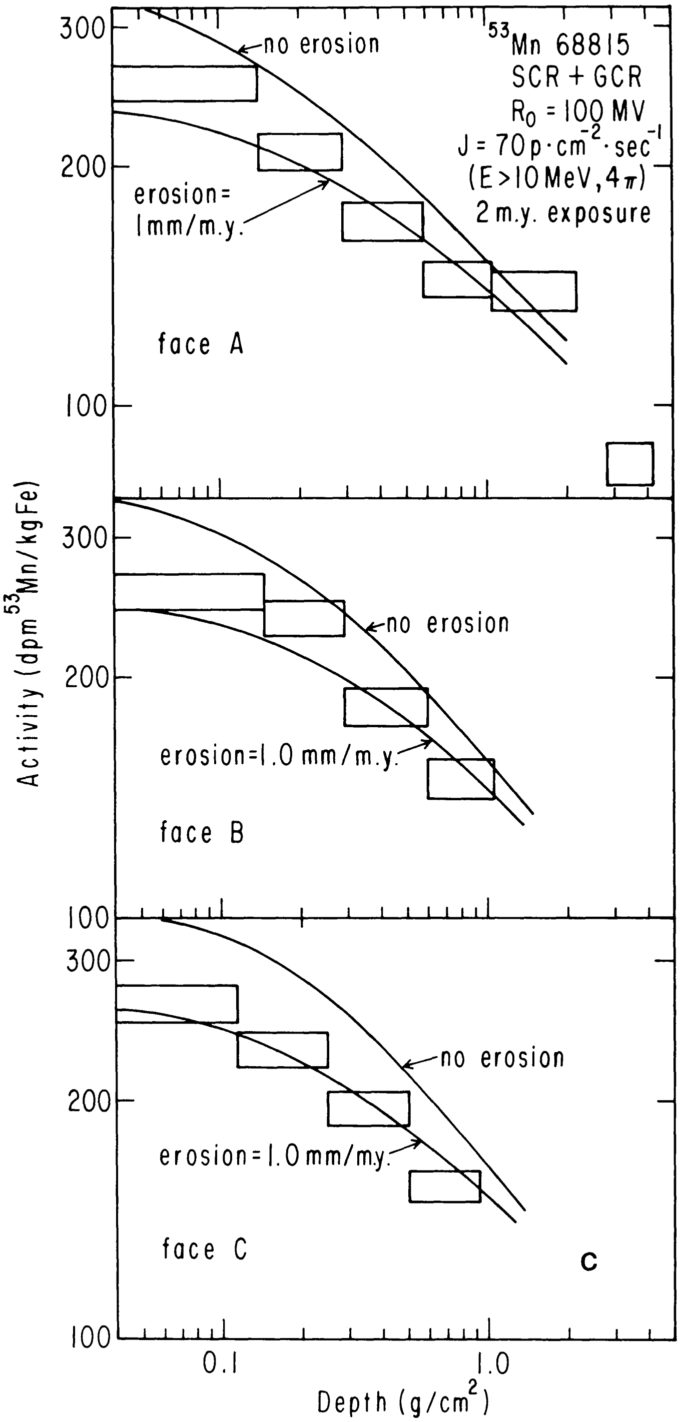


Fig. 1. Measured (a) ^{10}Be, (b) ^{26}Al, and (c) ^{53}Mn activity depth profiles in the three faces of 68815. The width of the error bars indicates the average depth interval sampled as determined from the maps made during grinding (*Russ and Emerson,* 1980). The curves shown for ^{26}Al and ^{53}Mn are the theoretical profiles of the SCR plus GCR production calculated by *Russ and Emerson* (1980) on a point by point basis for 68815 using a 2 m.y. exposure age and the Reedy-Arnold model. Figures 1b and 1c are from *Murrell* (1980).





Russ and Emerson (1980) recalculated ^{26}Al and ^{53}Mn depth profiles in 68815 using point by point mapping of all grinding faces. Even though their detailed calculation shows that the average angles of the faces from horizontal are substantially different from those used by *Kohl et al.* (1978), they obtained essentially the same conclusions with regard to the SCR parameters, R_0 and J. The more detailed calculations of *Russ and Emerson* (1980) showed no evidence of SCR anisotropy or of differential erosion for the three surfaces.

It is necessary to subtract the GCR-produced ^{10}Be from the observed ^{10}Be to see the SCR component. The expected GCR production profile using the chemical composition of 68815 was calculated based on the Reedy-Arnold model (*Reedy and Arnold*, 1972) and is shown in Figs. 2a,b. The measured ^{10}Be profile for face B is essentially the same as for face A and C, but the data contain somewhat larger errors. The original model (*Reedy and Arnold*, 1972) and the evaluated cross sections of *Tuniz et al.* (1984) were used for both GCR and SCR calculations. The Reedy-Arnold GCR profile fits the 68815 data well without any of the normalization that was required for both the ^{26}Al and ^{53}Mn GCR production profiles (*Nishiizumi et al.,* 1983). The Reedy-Arnold GCR ^{10}Be profile using the new cross sections also fits the ^{10}Be results for the Apollo 15 drill core (*Nishiizumi et al.,* 1984). However, the Reedy-Arnold GCR profile for ^{10}Be appears to increase with depth slower than the measured data, suggesting that the Reedy-Arnold GCR model might be slightly inaccurate for the production rate versus depth profile near the surface, at least for high-energy products. As pure GCR production profiles are hard to find (almost all nuclides have significant SCR components near the surface), it is difficult to test the Reedy-Arnold model at such shallow depths.

We would expect to see excess ^{10}Be due to SCR production, if present, in near-surface samples. Figures 2a and 2b, however, show no indication of SCR-produced ^{10}Be in this rock. The Reedy-Arnold SCR model predicts a ^{10}Be SCR production rate of about 2 atoms/min/kg in the surface layer using a SCR flux with R_0 = 100 MV and J(>10 MeV) = 70 p/cm^2 s (see Fig. 3a), the parameters obtained from ^{26}Al and ^{53}Mn profiles in this and other lunar surface rocks (*Kohl et al.,* 1978). Taking values at the limits of the errors, minimum values for the deeper samples and maximum for the near surface samples, we obtained a value for SCR-produced ^{10}Be of less than 1 dpm/kg.

The use of an exponential-rigidity spectral shape for the solar protons needs to be justified here much more than in previous works because much of the SCR production of ^{10}Be occurs for proton energies above 100 MeV, whereas most of the production of ^{53}Mn and ^{26}Al occurs at much lower energies. For example, only about 4% of ^{53}Mn or ^{26}Al but 55% of ^{10}Be

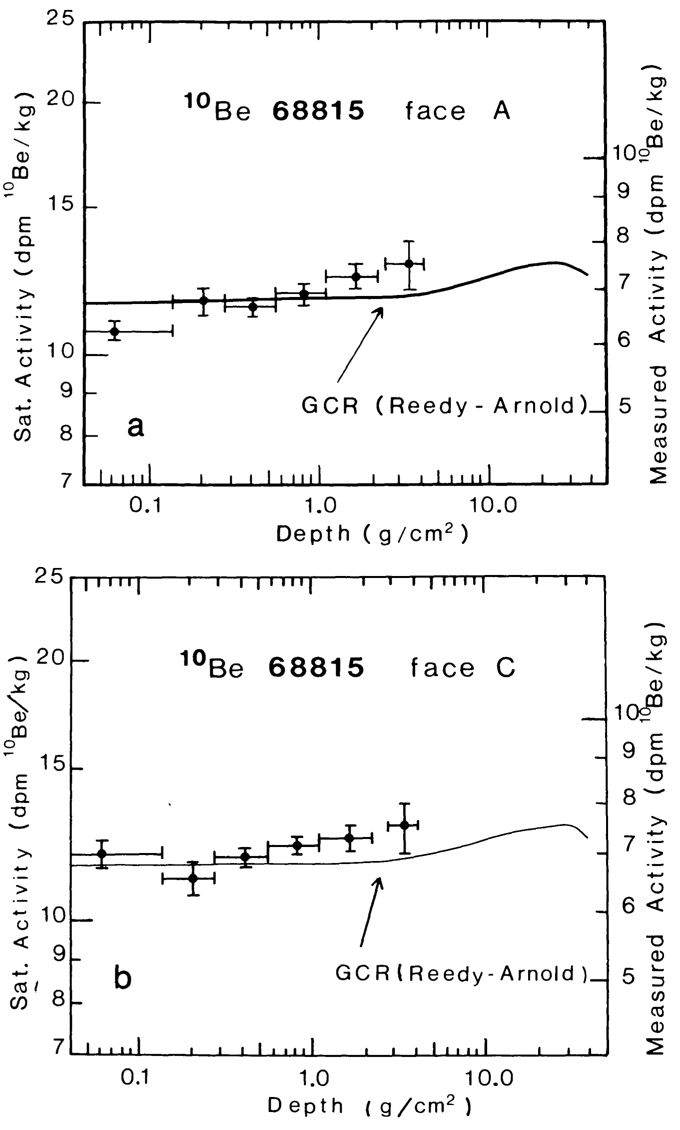


Fig. 2. ^{10}Be activity depth profiles in (**a**) face A and (**b**) face C of 68815. Values are plotted as measured activities on the right-hand scale and as saturation activities on the left-hand scale. The ^{81}Kr-Kr exposure age of 2.04 ± 0.08 m.y. (*Drozd et al.,* 1974) was used to calculate the saturation activities. The curves are the unnormalized GCR production profiles calculated using the Reedy-Arnold model (*Reedy and Arnold,* 1972) and the revised cross sections (*Tuniz et al.,* 1984). They include no corrections for erosion or surface inclination. The two deepest points were ground from under both faces A and C and are shown in both (**a**) and (**b**).

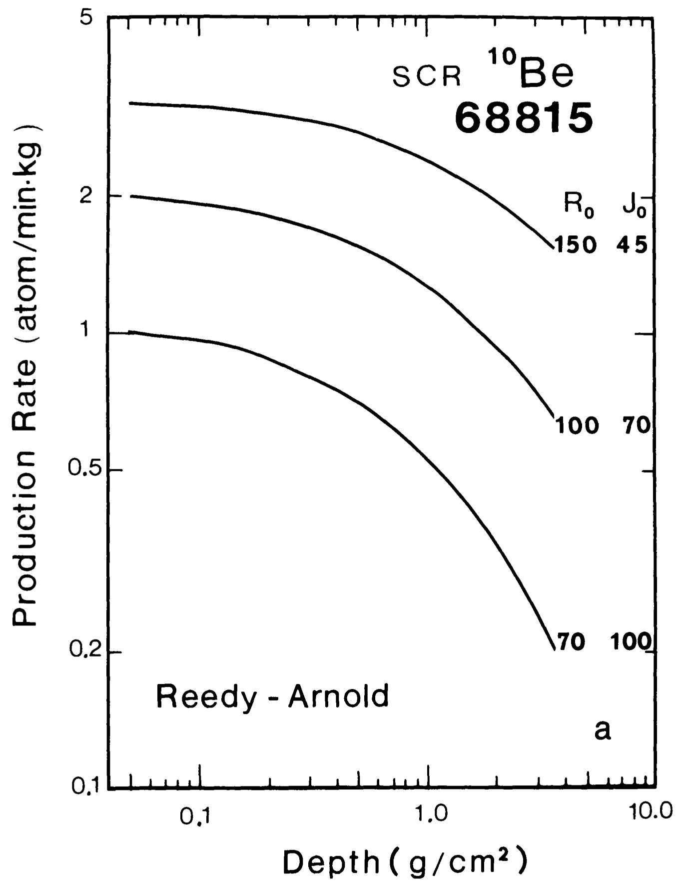
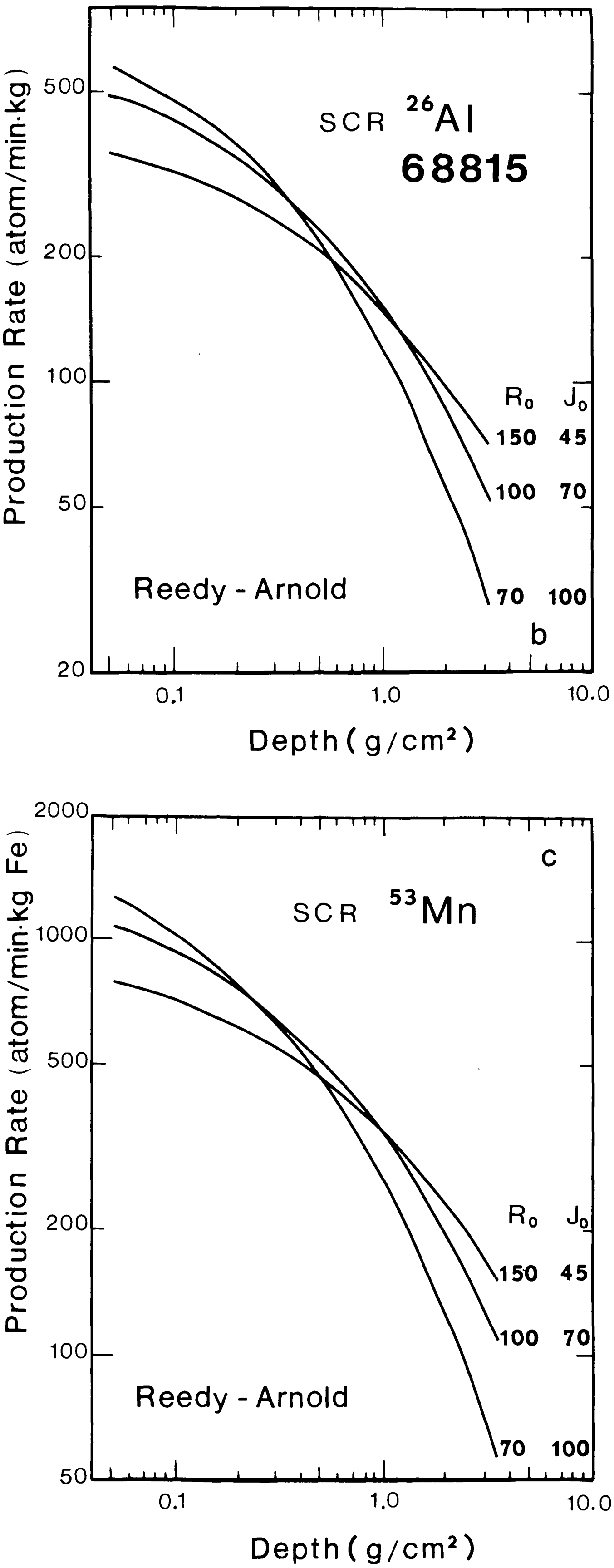


Fig. 3. SCR production profiles for (**a**) ^{10}Be, (**b**) ^{26}Al, and (**c**) ^{53}Mn, calculated using 68815 chemical composition. The depth profiles were calculated using the Reedy-Arnold model (*Reedy and Arnold,* 1972) and the cross sections of *Tuniz et al.* (1984) for ^{10}Be. They show expected saturation levels for each nuclide for three sets of SCR parameters (R_0 = 150 MV, J = 45 p/cm^2 s, R_0 = 100, J = 70, and R_0 = 70, J = 100). Erosion was assumed to be 0 for these calculations.



are made at energies above 100 MeV by primary protons with a spectral shape of R_0 = 100 MV. Below the lunar surface, these fractions for production by protons with E >100 MeV increase as the lower energy protons are preferentially stopped. While some investigators use a power law in energy for the spectral shape of solar protons over limited energy intervals below 100 MeV, all spectral observations for proton energies above 100 MeV show a curvature down relative to a pure power law (see, e.g., *McGuire and von Rosenvinge,* 1984). *Freier and Webber* (1963) used several techniques to measure the fluxes of solar protons up to energies of a few GeV and concluded that an exponential rigidity was a good fit to solar proton spectra at high energies. However, most of their spectra were based on indirect observations using balloons. *McGuire and von Rosenvinge* (1984) used instruments on the IMP-7 and IMP-8 satellites to measure proton fluxes for many energies up to 400 MeV during several solar flares. While they preferred a modified Bessel function of the second kind to fit the proton's energy spectra, in the figures of their paper an exponential rigidity function appears to give equally good fits for energies from 10 to 400 MeV. As less than 1% of the ^{10}Be is made by protons with E >400 MeV, the study by *McGuire and von Rosenvinge* (1984) confirms the validity of our using an exponential rigidity for the spectral shape of solar protons for calculating and comparing the SCR productions of ^{10}Be, ^{26}Al, and ^{53}Mn.

The Reedy-Arnold model is a well-developed method for calculating cosmic-ray interactions in bodies of various sizes for both SCR and GCR. However, the model contains some uncertainties with regard to estimating the GCR flux at near-surface depths. If the calculated Reedy-Arnold GCR ^{10}Be production rates are overestimated near the surface, the flat profile we measured may be due to a real SCR component in the surface layers. However, SCR ^{10}Be production rates decrease drastically with increasing depth below a few g/cm^2 regardless of the SCR parameters (see Fig. 3a). The observed ^{10}Be activities at a few g/cm^2 and below are almost entirely produced by GCR interactions and the measured values at these depths are in agreement with the theoretical values and the Apollo 15 drill cores. The GCR production rate would have to decrease 15-20% from 1 g/cm^2 to the surface to match the measured profile if the SCR production rate is 2 atoms ^{10}Be/min/kg in this region. There is no theoretical or experimental support for such an abrupt change for the penetrating GCR particles, and this explanation is unlikely. The discrepancy between the Reedy-Arnold model for SCR ^{10}Be production and the data can be explained as discussed below.

1. The ^{10}Be proton-induced cross sections that are used for Reedy-Arnold SCR calculations may be too high, especially for the low-energy region. Although the SCR spectrum varies from flare to flare, SCR particle intensity decreases exponentially with increasing energy (*Reedy and Arnold,* 1972). The low-energy proton-induced cross sections, especially below 100 MeV, are therefore very important for total SCR production. However, there are no cross section measurements for ^{10}Be production below 135 MeV for protons on any target element. *Reedy and Arnold* (1972) estimated the ^{10}Be cross sections from nuclear systematics and comparisons with the measured

^{7}Be cross sections at lower energies. The original Reedy-Arnold calculation for R_0 = 100 MV, J = 70 protons/cm^2 s, predicted about 4 dpm ^{10}Be/kg produced by SCR in the surface layer. The new calculation, which uses new and lower-proton cross sections (*Tuniz et al.,* 1984), predicts 2 dpm ^{10}Be/kg at the top layer of 68815, still more than we find experimentally. The target element responsible for the majority of ^{10}Be produced by SCR protons is oxygen. The elemental abundance of oxygen in 68815 is 44.8% (*Apollo 16 Preliminary Science Report,* 1972; *Wänke et al.,* 1974). ^{10}Be is also produced by proton interactions with Mg (3.85% in 68815), Al (14.2%), and Si (21.8%). However, the threshold energies for these nuclear reactions are higher than those interactions of O, and the elemental abundances of Mg, Al, and Si are also lower than oxygen. The threshold energy for the ^{10}Be-producing reaction with O is 34 MeV. There are only two cross section measurements for ^{10}Be production from O available below 500 MeV proton energy. *Yiou et al.* (1969) reported the cross section to be 0.37 ± 0.12 mb at 135 MeV. *Amin et al.* (1972) reported the cross section to be 0.59 ± 0.04 mb at 135 MeV. [The result by *Amin et al.* (1972) has been corrected for the revised half-life of ^{10}Be.] At 135 MeV, the higher cross section was used for the Reedy-Arnold calculations because, as noted in *Tuniz et al.* (1984), the *Yiou et al.* (1969) cross sections are consistently lower than other measurements at 600 MeV and higher energies. There are no other cross section measurements below 500 MeV proton energy except on boron and carbon, which are not abundant elements in lunar rocks. This matter could be resolved by AMS measurements of the low-energy cross sections below 100 MeV for ^{10}Be production from O and other elements. Lower cross sections, especially below ~300 MeV, could decrease the calculated SCR production rates by factors of 2 or more.

2. The second possibility is that the average SCR flux and mean rigidity over the last two million years differed from the adopted parameters R_0 = 100 MV and J(>10 MeV) = 70 p/cm^2 s. The Reedy-Arnold SCR production rates of ^{10}Be, ^{26}Al, and ^{53}Mn with three different rigidities (R_0 = 70, 100, and 150 MV) are shown in Figs. 3a-c. Although the depth profiles of both ^{26}Al and ^{53}Mn at near-surface depths are very insensitive to changes in R_0, different SCR fluxes and erosion rates would be required to fit those profiles to the data. To fit the observed ^{26}Al and ^{53}Mn profiles in lunar surface rocks, different proton fluxes and rock erosion rates must be chosen for each rigidity. If we use a lower R_0, a higher SCR flux and larger erosion rate would be required. It is known that R_0 = 100 MV, J = 70 p/cm^2 s, and an erosion rate of 1.3 mm/My (*Kohl et al.,* 1978) are not unique parameters to fit the ^{26}Al and ^{53}Mn profiles in 68815 (*Russ and Emerson,* 1980). For example, R_0 = 70 MV, J(>10 MeV) = 150 p/cm^2 s, and an erosion rate of 3 mm/m.y. (which was the erosion rate reported from track data for 68815 by *Blanford et al.,* 1975) can also fit the measured ^{26}Al and ^{53}Mn activities in 68815.

The SCR ^{10}Be depth profile is very different from both the ^{26}Al and ^{53}Mn profiles. As shown in Fig. 3a, the ^{10}Be production rates change from 1-3 dpm/kg at near-surface depths depending on the rigidity used. The production rate for ^{10}Be in the top of rock 68815 using R_0 = 70 MV and J(> 10 MeV) = 150 p/cm^2 s is 1.0, only 60% of that calculated for the other set of spectral and flux parameters. Even though the SCR production rate of ^{10}Be is lower than the GCR production, the amount of ^{10}Be activity produced by SCR is very sensitive to changes in R_0. The very low SCR production of ^{10}Be observed in 68815 could indicate that the mean SCR rigidity over the last two million years was lower than the 100 MV adopted by *Kohl et al.* (1978). However, lowering the R_0 conflicts with the conclusion by *Bhandari et al.* (1976). They proposed a higher rigidity (R_0 = 150 MV) and a higher flux [J(> 10 MeV) = 140 p/cm^2 s] based on their nondestructive ^{26}Al measurements in Apollo 16 rocks. Their SCR parameters do not fit the observed ^{10}Be depth profiles in 68815 nor the ^{26}Al and ^{53}Mn profiles in 68815 and the other lunar surface rocks. A higher R_0, such as 150 MV, is most unlikely unless the ^{10}Be cross sections are more than factor of 5 smaller than the values adopted by *Reedy and Arnold* (1972) for their calculations. It should be noted that measurements of ^{26}Al in five pieces from the top 4.4 cm of lunar rock 74275 by *Fruchter et al.* (1982) gave results in good agreement with those reported in *Kohl et al.* (1978) and not with those of *Bhandari et al.* (1976). *Reedy* (1980), using ^{81}Kr data in 12002 (*Yaniv et al.,* 1980), found a somewhat higher R_0 for the period of 3 × 10^5 years, but this is not necessarily a contradiction because the main reactions producing ^{81}Kr have threshold energies above 60 MeV and the chemical abundances of the target elements were not well measured in the sample. Also, ^{81}Kr, because of its half-life of 2.1 × 10^5 years, integrates solar protons for a much shorter period than the other radionuclides. Also, unpublished ^{81}Kr measurements (K. Marti, personal communication) in 68815 support a lower R_0.

In general, SCR production rates of high-energy products such as ^{10}Be and ^{36}Cl are potentially very useful for obtaining the SCR spectrum. ^{10}Be has a distinct advantage over ^{36}Cl since ^{36}Cl is produced in both high-energy and low-energy reactions. The comparison of ^{53}Mn and ^{26}Al profiles made by *Kohl et al.* (1978) is very useful for detecting variations in cosmic rays with time, since the half-lives differ by a factor of ~5 while the excitation functions are similar. Since both ^{53}Mn and ^{26}Al are low-energy products and their profiles are steep near the surface, they are also sensitive to the erosion rate. The comparison of these two nuclides with ^{10}Be is useful in a complementary way. Because the excitation functions are quite different, only a narrow range of R_0 values can satisfy the constraints imposed by the three profiles, even though the ^{10}Be production by SCR can only be given as an upper limit. If we accept the published proton cross sections as representative, this fixes R_0 close to 70 MV. New low-energy cross section data would be most desirable, but unless they are lowered by a factor of 2 or more this conclusion will remain valid.

Previous ^{10}Be measurements, which used decay counting techniques (*Finkel et al.,* 1971; *Wahlen et al.,* 1972) to study lunar surface rocks 12002, 14310, and 14321, give results that are in good agreement with the ^{10}Be activity found in 68815 by AMS measurements. The ^{10}Be activities in the above rocks also show no increase of ^{10}Be at the surface and therefore no evidence of SCR production. Since substantially all the ^{10}Be in these rocks was produced by GCR and since they have different exposure ages, we conclude that no significant changes in the GCR flux were observed during at least the last few million years.

SUMMARY

Cosmogenic ^{10}Be activities were measured in lunar surface rock 68815. Four different depths were sampled for three different angles. The ^{10}Be profiles are flat or increase slightly with depth for all three faces and show no sign of SCR-produced ^{10}Be. The extremely low SCR production of ^{10}Be compared to the calculations of the Reedy-Arnold model suggests that either (1) low-energy proton-induced cross sections for ^{10}Be production are lower than expected, or (2) the SCR rigidity R_0 is lower than 100 MV averaged over the last few million years.

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