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Lunar Radionuclide Records of Average Solar-Cosmic-Ray Fluxes
over the Last Ten Million Years

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Abstract - Because changes in solar activity can modify the fluxes of cosmic-ray particles in the solar system, the nature of the galactic and solar cosmic rays and their interactions with matter are described and used to study the ancient sun. The use of cosmogenic nuclides in meteorites and lunar samples as detectors of past cosmic-ray variations are discussed. Meteorite records of the history of the galactic cosmic rays are reviewed. The fluxes of solar protons over various time periods as determined from lunar radionuclide data are presented and examined. The intensities of solar protons emitted during 1954-1964 (11-year solar cycle number 19) were much larger than those for 1965-1975 (solar cycle 20). Average solar-proton fluxes determined for the last one to ten million years from lunar ^{26}Al and ^{53}Mn data show little variation and are similar to the fluxes for recent solar cycles. Lunar activities of ^{14}C (and preliminary results for ^{81}Kr) indicate that the average fluxes of solar protons over the last 10^4 (and 10^5) years are several times larger than those for the last $10^6 - 10^7$ years; however, cross-section measurements and other work are needed to confirm these flux variations.

INTRODUCTION

Many variations in the fluxes of cosmic-ray particles are related to solar-activity changes (see, e.g., Pomerantz and Duggal, 1974). Solar magnetic fields modulate the intensities of galactic-cosmic-ray (GCR) particles in the inner solar system, as the systematic changes of GCR fluxes over the 11-year solar cycle indicate. Eddy (1976) showed that variations of GCR-produced ^{14}C in the Earth's atmosphere correlate with sunspot numbers. However, long-term variations of GCR fluxes also can be caused by changes in their sources or in their transport to the solar system (Forman and Schaeffer, 1979). The fluxes of the solar-cosmic-ray (SCR) particles emitted from the sun are good indicators of solar activity. This paper discusses the nature of these cosmic-ray particles and their interactions with matter, emphasizing the production of nuclides by cosmic-ray-induced nuclear reactions.

The activities of various cosmogenic radionuclides in meteorites and lunar samples are very good records of average cosmic-ray fluxes in the past. Meteorites and lunar samples are better than the Earth's atmosphere as targets for cosmic-ray particles because they don't have magnetic fields which perturb the cosmic-ray fluxes. (Geomagnetic variations significantly affect the production rates of nuclides such as ^{14}C and ^{10}Be in the Earth's atmosphere.) The activities of these cosmogenic nuclides directly reflect the fluxes of the cosmic-ray particles which produce them. The half-lives of the radionuclides usually determine the time period over which the cosmic-ray fluxes are averaged. A very good method to study the past variations of solar activity is the measurement of radionuclides produced in lunar samples by solar cosmic rays. The average solar-proton fluxes deduced from such measurements are discussed below.

SOLAR COSMIC RAYS AS INDICATORS OF SOLAR ACTIVITY

The interrelations between certain solar phenomena and cosmic-ray flux variations are discussed in detail by Pomerantz and Duggal (1974). Solar cosmic rays produce a number of effects observable from the Earth's surface. The magnitudes of these effects are proportional to the fluxes of SCR particles which produce them. The SCR particles produce ionization in the 40- to 100-km level of the ionosphere which absorbs radiowaves. This phenomenon is most pronounced over the poles. At frequencies of the order of 30 MHz, extraterrestrial radio noise is absorbed by this SCR-produced ionization and is referred to as polar cap absorption (PCA). Pomerantz and Duggal (1974) list the 77 PCA events with equivalent 30 MHz absorption of ≥ 2.5 dB which were observed from 1952 to 1973. They found that both the frequency of PCA events per year and the sum of peak absorptions for a year correlate well with annual mean sunspot numbers. Only three of these PCA events occurred when the Zurich sunspot number (R_z) was less than 50 (but still above about 20); almost all large PCA events happened during periods of considerable solar activity.

Solar-cosmic-ray particles with energies above about 5 and 0.5 GeV can be detected by ionization chambers and neutron monitors, respectively, on the Earth's surface. Such ground level enhancements (GLE) have been observed regularly by ionization chambers since 1936 and by neutron monitors since the late 1940's. Almost all of the 25 GLE events listed by Pomerantz and Duggal (1974) occurred during periods when R_z was above 50; none were observed when R_z was below 18.

The fluxes of solar-cosmic-ray particles which were inferred by indirect measurements for solar cycle 19 (1954-1964) and directly measured by satellites for most of solar cycle 20 (1965-1975) also show the same trends observed for

PCA and GLE events - few occurring during periods of low solar activity. Webber (1966) noted that the number of events per year is, on average, $0.08 R_z$ and that $I(>10 \text{ MeV})$, the yearly integrated particle flux above 10 MeV in particles/cm² y, is related to the annual sunspot number by $\log_{10} I(>10 \text{ MeV}) = 7 + 0.02 R_z$.

The event-averaged integral fluxes of solar protons are tabulated by Reedy (1977) for 32 events during solar cycle 19 and 33 events during solar cycle 20. The only event in this tabulation with an omnidirectional integral flux greater than 10^7 protons/cm² for energies above 30 MeV and with a monthly averaged Zurich sunspot number below 50 was for 26 September 1963. The fluxes reported by Webber (1966) for this event and for the one on 10 September 1961 are down by factors of about 6 and 4, respectively, relative to those tabulated in Reedy (1977). I believe the Webber (1966) fluxes for these two events are better values than those adopted by Reedy (1977), and thus the integral fluxes above 30 MeV for these events both should be about 1×10^7 protons/cm². Thus, for solar cycles 19 and 20, there probably were no SCR events with $I(>30 \text{ MeV}) > 10^7$ protons/cm² and $R_z < 50$. For solar cycle 20, for which there are more and better flux data for small SCR events, the only events with $I(>30 \text{ MeV}) > 2 \times 10^6$ protons/cm² and $R_z < 42$ were three small events which occurred during 1965 and 1966 with monthly R_z 's between 12 and 18.

While there can be periods of considerable solar activity when there are no SCR events, observations during the last two solar cycles show that only a few, weak SCR events occurred during periods of "low-to-medium" solar activity ($10 < R_z < 50$) and that no SCR events with $I(>30 \text{ MeV}) > 2 \times 10^6$ protons/cm² happened during periods of "minimum" solar activity ($R_z < 10$). Almost all the major emissions of particles from the sun during the last few

decades occurred when the sun was quite active ($R_z > 50$). Thus the presence of significant fluxes of SCR particles in the past would be good evidence of considerable solar activity over the period for which the SCR fluxes were determined. (The argument that a fairly high average flux of SCR particles could be the consequence of a few "superflares" during a period of mainly low solar activity is very ad hoc, as there is not good evidence for the presence of superflares in the past; see Lingenfelter and Hudson, these proceedings.)

COSMIC-RAY INTERACTIONS WITH MATTER IN SPACE

There are two sources of energetic particles which produce nuclear reactions in most solar-system matter: galactic cosmic rays (GCR) and solar cosmic rays (SCR). The natures of these two types of cosmic rays and their interactions with matter are described in detail in several articles (e.g., Reedy and Arnold, 1972; Lal, 1972), but are reviewed here briefly. As mentioned above, the SCR originate at the sun and the GCR come from outside the solar system. Both types consist mainly of protons and alpha particles (with a proton/alpha-particle ratio of about 10) with about 1% heavier nuclei.

There are only a few GCR particles per cm^2 per second, but their mean and median energies are of the order of 10^9 eV (1 GeV). From several hundred MeV to energies of the order of thousands of GeV, the spectrum is approximated by

$$dI/dE = \text{const.} (1 + E [\text{in GeV}])^{-2.5}. \quad (1)$$

The fluxes of GCR particles, especially those with energies below about 1 GeV, are modulated by solar magnetic fields, the main temporal variation being with the 11-year solar cycle. The fluxes of GCR particles also vary

in the solar system, increasing both with distance from the sun (a few percent per AU) and with angle out of the ecliptic (McKibben, 1975).

The sun irregularly, but mainly near periods of solar maximum, emits energetic particles. Most of the particles in an SCR event at 1 AU come from a specific solar flare and are accelerated to high energies at the sun. However, there are rare occasions when the particle acceleration actually occurs in interplanetary space by two converging shock fronts (Pomerantz and Duggal, 1974; Lingenfelter and Hudson, these proceedings). The events of 17 July 1959 and 4 August 1972 involved interplanetary acceleration, but, as pointed out by Pomerantz and Duggal (1974), they are "ultimately of solar origin." During an SCR event, the peak flux above 10 MeV can reach levels of the order of 10^5 particles/cm² s. The integral flux for energies above 10 MeV averaged over a solar cycle is about 100 particles/cm² s. The energy distribution of the SCR particles varies from flare to flare, but always has a flux which rapidly decreases in intensity with increasing energy. From about 10 to 100 MeV, the spectrum is usually described by an exponential rigidity shape,

$$dI/dR = \text{const.} \exp(-R/R_0) \quad , \quad (2)$$

where R is the rigidity of the particle (defined as the momentum per unit charge, pc/ze , in units of megavolts, MV), and R_0 usually ranges from 30 to 200 MV (Reedy and Arnold, 1972). Below about 50 MeV, an energy power-law distribution,

$$dI/dE = \text{const.} E^{-\gamma} \quad , \quad (3)$$

is often used, where γ typically has values between 1 and 4 (Lal, 1972).

Energetic nuclear particles mainly interact with matter in two ways: ionization energy losses and nuclear interactions. All charged nuclear particles lose energy continuously by ionizing the atoms as they pass through matter. Low-energy protons are rapidly stopped; in lunar rocks, the range of a 50-MeV proton is 3 g/cm² (about 1 cm). High-energy particles lose energy more slowly and usually undergo nuclear interactions before stopping (a 1-GeV proton has a range of about 400 g/cm² but only about 2% of 1-GeV protons go their entire range without interacting.) Heavy nuclei lose energy much more rapidly than light nuclei. The radiation damage produced during ionization energy losses can accumulate in matter and be detected as thermoluminescence (TL). The paths traveled by individual nuclei with Z above about 20 can be etched by certain chemicals and made visible as "tracks" (Fleischer et al., 1975).

Nuclear interactions involve either nuclear reactions or the scattering of the incident particle from nuclei. Scattering reactions decrease the energy of the particle and are the only means by which neutrons are slowed. Most nuclear reactions occur when the incident particle is above a certain threshold energy and involve the formation of a residual nucleus (stable or radioactive) and certain secondary particles (such as protons, neutrons, alpha particles, pions, and gamma rays). In a nuclear reaction, the incident particle is usually absorbed by the nucleus, although high-energy particles can escape after having excited a nucleus. Nuclear-reaction mean free paths are of the order of 50 g/cm² (i.e., 1/e or 37% of the incident particles have not reacted after having traversed that thickness.) Secondary neutrons are very important for inducing nuclear reactions in extraterrestrial matter because about 10 neutrons are produced per incident primary GCR particle and they have long mean free paths while most other secondary nuclear particles have short ranges.

The relatively low-energy solar protons and alpha particles are usually stopped by ionization energy losses very near the surface of the material. The SCR particles that do induce nuclear reactions produce few secondary particles (and the product nucleus is near in mass to that of the target nucleus.) Monte Carlo calculations by Armstrong and Alsmiller (1971) indicate that nuclear reactions induced by SCR-produced secondary neutrons are negligible compared both to SCR-primary-induced reactions near the surface and to GCR-secondary-neutron-induced reactions at any depth. Because SCR secondary particles produce relatively few reactions, most calculations of production rates for SCR-induced reactions only consider the primary SCR particle fluxes and ionization-energy-loss effects (Reedy and Arnold, 1972). The interpretations of lunar SCR-produced radionuclides given below usually used the calculated production rates of Reedy and Arnold (1972). Calculations of solar-proton fluxes and nuclide production rates in lunar rocks also have been made by Yokoyama et al. (1972) and Tanaka et al. (1972); their results are in very good agreement with those of Reedy and Arnold (1972). Tanaka et al. (1972) have shown that, for ^{26}Al and ^{53}Mn , solar-alpha-particle-induced reactions are relatively unimportant.

The GCR particles producing nuclear reactions can be roughly divided into three components (Reedy and Arnold, 1972; Yokoyama et al., 1972): a high-energy component of energetic (above about 1 GeV) primary particles with a characteristic attenuation mean free path, medium-energy (between about 0.1 and 1 GeV) particles produced partially from the first component, and a low-energy group (below 100 MeV) consisting largely of secondary neutrons. The fluxes of the high-energy GCR particles decrease roughly exponentially with depth. The fluxes of secondary neutrons increase with depth near the surface, but then decrease exponentially with depth (Reedy

and Arnold, 1972). The intensity of GCR particle fluxes varies with solar activity (being highest at periods of solar minimum), but the shapes of GCR production rates versus depth do not change much over a solar cycle (Armstrong and Alsmiller, 1971).

NUCLIDES PRODUCED BY COSMIC-RAY PARTICLES

Cosmic-ray particles reacting with matter in space can produce a wide variety of product nuclei. The production rate, $R(d)$, of a given product nuclide at a location d in a piece of extraterrestrial matter is given by

$$R(d) = \sum_i N_i \sum_j \int \sigma_{ij}(E) \phi_j(E,d) dE, \quad (4)$$

where i represents all the target elements which can produce the nuclide, N_i is the target elemental abundance in the sample, j indicates the primary or secondary particles which can induce reactions, $\sigma_{ij}(E)$ is the cross section at energy E for particle j and target i producing the nuclide, and $\phi_j(E,d)$ is the flux of particle j with energy E at location d .

Over 99% of the atoms in meteorites or lunar samples have atomic number 28 (nickel) or lower, so most of the nuclides made by cosmic rays ("cosmogenic nuclides") have atomic numbers below about 28, including all radionuclides with appreciable specific activities. Some heavier cosmogenic nuclei are made in such samples, but, except for isotopes of the noble gases (He, Ne, Ar, Kr, and Xe), which are readily detected by direct measurements, relatively few of such heavier products are ever detected. Table I lists the cosmogenic radionuclides most frequently observed in meteorites or lunar samples. Stable cosmogenic noble-gas nuclides can be used to determine a sample's integral exposure to cosmic rays, provided appropriate production rates can be established. In iron meteorites, stable cosmogenic isotopes of certain lighter

elements (e.g., Cr and K) have been detected. There are only nine cosmogenic nuclides listed in Table 1 with half-lives long enough (greater than 10 years) to study cosmic-rays fluxes in the past (^3H , ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{39}Ar , ^{53}Mn , ^{59}Ni , and ^{81}Kr). Table 2 lists other long-lived nuclides produced in extraterrestrial matter; most are rarely or never seen because of the scarcity of target elements (e.g., for the uranium isotopes) or the difficulties in detecting the radionuclides (such as ^{41}Ca). Many of these radionuclides are so little studied that their half-lives are poorly known (e.g., ^{60}Fe). Hopefully, more of these nuclei listed in Table 2 will be used in future studies of the fluxes of cosmic-ray particles in the past.

Most of the useful radionuclides have been studied by measuring their characteristic emissions (e.g., alpha and beta particles, γ and X rays). Low-background counters, detecting coincidences between simultaneously emitted radiations, high-resolution spectrometers, and chemical separations are among the techniques used to detect the low levels of radionuclide activities usually present in extraterrestrial matter. Mass spectrometry has been used to detect ^{81}Kr and the stable noble gases and in studies of cosmogenic uranium isotopes. An activation technique using thermal neutrons has been well developed for measuring ^{53}Mn by converting it to short-lived ^{54}Mn (Millard, 1965; Finkel et al., 1971). New techniques are being developed to detect trace amounts of radionuclides. Particle accelerators (cyclotrons and Van de Graaffs) have been used to observe ^{10}Be , ^{14}C , ^{36}Cl , and several other radionuclides in natural samples (Raisbeck and Muller, this conference). Lasers have the capability of detecting minute amounts of any isotope (see, for example, Hurst et al., 1977) and could be used to measure very small radionuclide concentrations.

The rate of formation of a nuclide (Eqn. 4) depends largely on the cross sections for the reactions making that product. If a product is made only by high-energy particles (e.g., ^{10}Be), then it will be made in small amounts by SCR particles. A nuclide made mainly by low-energy protons (such as ^{56}Co in lunar samples) will be produced almost entirely by solar protons near the surface. Some radionuclides (like ^{39}Ar) result mainly from GCR neutron-induced reactions. However, as noted in Table 1, most radionuclides are made in significant amounts by both GCR and SCR particles.

The big difference in the depth-activity profiles for production of radionuclides by GCR and SCR particles (Reedy and Arnold, 1972) allows these two components to be resolved from experimentally determined profiles. Because the fluxes of SCR particles decrease rapidly with increasing depth, the activities for SCR-produced nuclides are high at the surface and become very small at depths of the order of 5 to 10 cm. The depth-activity profiles for a GCR-produced nuclide depend on the cross sections of the reactions producing it. A high-energy product like ^{10}Be has a GCR depth-activity which is flat from the surface to a depth of about 10 g/cm² and which decreases at greater depths, whereas a low-energy neutron-produced nuclide like ^{39}Ar has a profile in which its activity increases by about a factor of two from the surface to about 50 g/cm² and then decreases (Reedy and Arnold, 1972).

The fluxes of SCR particles as a function of depth can be accurately calculated using ionization-energy-loss relations, so depth-activity profiles of a nuclide can be well calculated if the cross sections for its formation are well known. Conversely, formation cross sections can be used to determine the incident flux of SCR particles from a measured depth-activity profile (after correcting for the GCR contributions). For the SCR-produced radionuclides discussed below, all but ^{14}C and ^{81}Kr have well known cross sections.

for their formation by SCR particles. Cross sections for the $^{16}\text{O}(p,3p)^{14}\text{C}$ have only been measured at a few energies by Tamers and Delibrias (1961), who quoted uncertainties of $\pm 25\%$. There have been no other independent measurements of the cross sections for this reaction with which to check the accuracy of the reported values. Cross sections for the formation of ^{81}Kr have only been measured for proton reactions with yttrium; cross sections for targets of Rb, Sr, and Zr are estimations only (Regnier *et al.*, 1979).

The fluxes of GCR particles as a function of depth in an extraterrestrial object are not very well known, especially for secondary neutrons. Many GCR-induced reactions have not had their cross sections measured. There are very few measured cross sections for reactions induced by neutrons with energies above 20 MeV. These uncertainties in GCR fluxes and production cross sections mean that absolute values of production rates for GCR-induced reactions are not calculated well. However, production rates calculated using the GCR model of Reedy and Arnold (1972) have reproduced well the shapes of GCR depth-activity profiles. The procedure usually used to remove the GCR component from a measured depth-activity profile is to use the relative profile calculated by Reedy and Arnold (1972) and to multiply it by a normalization constant determined from an activity measured at a depth where SCR production is not important. For the radionuclides used below to study SCR fluxes, the only one with a poorly known normalization factor for GCR production is ^{81}Kr .

COSMOGENIC NUCLIDES AS DETECTORS OF COSMIC-RAY FLUX VARIATIONS

As discussed below, many investigators have used cosmogenic nuclides in meteorites and lunar samples to study the history of cosmic rays. GCR flux variations have been looked for by comparing calculated and measured activities of cosmogenic radionuclides with different half-lives. Such comparisons can

best be made when the meteorite has been exposed to cosmic rays for a period much longer than the half-lives of the radionuclides. Although, as mentioned above, GCR production rates are not accurately calculated, a trend in the ratios of observed-to-calculated activities relative to product half-lives would be evidence for a systematic variation in the flux of GCR particles.

The duration of a sample's exposure to cosmic rays (referred to as the sample's "exposure age") can be determined from concentrations of stable cosmogenic noble-gas isotopes. The production rates for the noble-gas isotopes most frequently used to calculate exposure ages (^3He , ^{21}Ne , ^{38}Ar) have been determined empirically from meteorite radionuclide data. Some meteorites have exposure ages short enough that the activities of long-lived radionuclides like ^{26}Al and ^{53}Mn have not built up to equilibrium rates and the amount the activities are below their equilibrium values can be used to calculate the exposure age. In meteorites with long exposures, the activity of a radionuclide and a production-rate ratio for the radionuclide and a stable isotope often is used to infer the stable isotope's production rate. Pairs of radioactive and stable isotopes frequently used include ^3H - ^3He , ^{22}Na - ^{22}Ne , ^{26}Al - ^{21}Ne , ^{36}Cl - ^{36}Ar , ^{39}Ar - ^{38}Ar , ^{40}K - ^{41}K , and ^{81}Kr - ^{83}Kr . Production-rate ratios often are fairly well known from accelerator bombardments, especially for iron meteorites where iron is the main target and only high-energy GCR particles can produce the isotopes. Variations in GCR particle fluxes can be observed by comparing exposure ages determined for one meteorite using several isotope pairs. Differences in exposure age so determined would result if the average fluxes of GCR particles varied during the periods the radioactive isotopes were produced.

Because GCR particles themselves directly produce almost all the radionuclides in a sample, the activities of a radionuclide at several depths or

of several radionuclides at one depth can be used with their production cross sections to determine the flux of incident particles. Fluxes cannot be determined for the SCR particles with energies below the thresholds for the radionuclide-forming reactions. The use of two different radionuclides is limited to cases where the product half-lives are similar enough that the same SCR events produced both nuclides. Also, the cross sections for forming the two or more radionuclides should be fairly different. This approach of using activities for several radionuclides was applied by Rancitelli et al (1971), who used activities of short-lived species formed in Apollo 17 samples to determine the fluxes of the August 1972 SCR events.

The unfolding of a depth-activity profile for one radionuclide usually is used and generally is the best way to determine SCR particle fluxes. The time period is determined by the half-life of the radionuclide or the exposure age of the sample as calculated from GCR-produced isotopes or tracks. The usual approach is not to unfold the measured activity data, but to vary incident fluxes until the calculated and measured activities agree. If smoothly varying spectra for the incident fluxes are used (usually the exponential-rigidity or energy-power-law shapes described above, Eqs. 2 and 3), then the fluxes which best fit the measured depth-activity profiles generally are independent of the details of the assumed spectral shapes.

There are many sources of uncertainties in determining cosmic-ray fluxes or flux variations. When two or more radionuclides are used or compared, the relative uncertainties in cross sections or production-rate ratios must be considered in comparing the deduced fluxes or variations. Poorly known cross sections for the production of a given nuclide limit the quality of the fluxes obtained from measured activities of that nuclide (as discussed above, this is a potential problem for solar-proton fluxes deter-

mined from ^{14}C and ^{81}Kr activities). Any measurement uncertainties directly affect deduced flux results. The best SCR fluxes are obtained from samples with many activities measured for different radionuclides or depths. When depth-activity profiles are used, most of the depths should be in the top centimeter where most of the SCR-induced activities are made, although it is desirable to have at least one set of activities measured at a depth deep enough to get the GCR-production-rate normalization factor.

The models used to calculate GCR and SCR production rates are more than adequate to study cosmic-ray fluxes. The details for the complex interactions of GCR particles are sufficiently covered in the models that the shapes of the depth-activity profiles are reproduced fairly well and so the major uncertainty is getting the normalization factor for a given profile. Most models assume a simple geometry (such as a sphere or plane surface) for the irradiated samples, whereas the actual sample can have a very complicated configuration. However, sample geometries usually don't affect calculated GCR production rates. Russ and Emerson (these proceedings) have shown that considering the detailed geometries of lunar rock surfaces did not change the conclusions concerning solar-proton fluxes.

The study of cosmic-ray fluxes can be complicated by a number of factors occurring during the irradiation of a meteorite or lunar sample. Micrometeoroid impacts and ion sputtering gradually remove surface material. Lunar rocks have erosion rates (about 1 mm per million years) which seriously affect profiles of long-lived SCR-produced radionuclides like ^{53}Mn (Wahlen et al., 1972). More energetic impacts can break lunar rocks or meteoroids into a number of smaller pieces. Documentation of where a sample was taken from in a bigger piece is often poor or non-existent, and there are cases where even the identity of the parent object is unknown.

A serious concern in studies of cosmogenic nuclides in many types of meteorites or lunar samples is knowledge of the chemical abundances of important target elements. It is best if chemical abundances and cosmogenic nuclide concentrations are measured using the same piece or aliquots of one sample. The use of chemical data from another piece of the same meteorite or lunar rock can be complicated by heterogeneities in the object. Sizeable chemical variations have been observed in many classes of meteorites and types of lunar rocks (e.g., breccias made by impacts fastening together chips of varying compositions). In some cases, a sample is from a chemically homogeneous object which is so similar to other members of its class that any chemical data for that class are adequate.

Usually the time when the exposure of an object to cosmic rays was terminated is well known. However, certain meteorites are found on the Earth's surface (e.g., those being discovered in Antarctica) and the time each one fell must be inferred from the decay of short-lived radionuclides. The time when an object was first exposed to cosmic rays usually is determined from concentrations of stable cosmogenic isotopes or from non-equilibrium activities of long-lived radionuclides. However, such exposure ages assume a simple irradiation history. Sample break-up or movement in the lunar surface can seriously affect the accuracies of exposure ages.

The orbit which a meteorite had in space before hitting the Earth almost always is not known, and could include most of the solar system inside Jupiter's orbit (as could possible previous orbits). As mentioned above, the fluxes of both GCR and SCR particles vary with distance from the sun and with angle out of the ecliptic. Cressy and Rancitelli (1974) measured unusually high ^{26}Al activities in the Malakal chondrite which they felt could have resulted from a complex history which included a period of exposure

to a very high cosmic-ray flux. Usually a large portion of a meteorite is removed by ablation during passage through the Earth's atmosphere, including the surfaces with the SCR-produced isotopes. Although ablation and occasionally break-up in the Earth's atmosphere complicate the determinations of the meteorite's pre-atmospheric size and shape, concentrations of cosmogenic nuclides and tracks can be used to infer the pre-atmospheric shielding of meteorite samples (see, e.g., Bhandari et al. 1978; Fleischer et al., 1967).

Lunar samples are found in or on the loose layer of particles and rocks on the moon's surface called the regolith. A few lunar rocks have had simple histories of exposure to the cosmic rays by having been brought to the location and position from which they were recovered directly from a well-shielded depth. Many lunar rocks have been exposed to cosmic rays at several depths in the regolith or in several different positions on the moon's surface. Some lunar breccias are so weakly held together that their surface layers easily can be removed by handling (Wahlen et al., 1972).

A rock on the moon's surface can see, at most, only half of space at any moment and parts of a lunar rock's surface often have a solid angle of exposure to space even smaller than 2π steradians. However, this limited solid angle for receiving cosmic-ray particles is not a problem because most cosmic rays are quite isotropic. Some SCR particles, especially those very early in an event are anisotropic, but the majority of SCR particles are isotropic at 1 AU (Mullison and Webber, 1963). Over the long-term, the moon's rotation and variations in the direction of anisotropy from event to event help to remove any net anisotropy effects. Russ and Emerson (these proceedings) saw no anisotropy in the average fluxes of solar protons over the last 2 My.

Some lunar rocks are believed to have had been covered by a layer of dust while on the moon's surface (Hartung et al. 1977). While a relatively thick dust layer can affect the production of microcraters and heavy-nuclei tracks in a lunar rock (e.g., Zook, this conference), it would not seriously perturb solar-proton-induced reactions.

GALACTIC-COSMIC-RAY FLUX VARIATIONS

Most studies of GCR flux variations have been made using radionuclide activities in meteorites. Evans et al. (1979) measured activities of radionuclides in many meteorites which fell during the past decade. The activities of the short-lived radionuclides ^{54}Mn , ^{46}Sc , and ^{26}Al varied by factors of two or more, and the variations correlated with neutron monitor rates, and, inversely, with sunspot numbers.

Most studies of long-term GCR flux variations have used iron meteorites or metal phases of meteorites because of their chemical simplicities. Most of the radionuclides produced in iron meteorites have reaction threshold energies above several hundred MeV, so secondary particles are relatively unimportant and results from accelerator bombardments can be used quite well to predict production-rate ratios. Forman et al. (1978) examined ^{37}Ar and ^{39}Ar activities in metal phases of meteorites and noticed that ^{39}Ar activities were slightly higher than expected from the ^{37}Ar activities. They saw the slight excess in ^{39}Ar activity as evidence for long periods of reduced solar modulation during the last 500 years (e.g., the Maunder and Spörer minima). Bhandari et al. (1979) measured the ^{39}Ar in stone meteorites which fell in 1794 and 1795, but saw no enhanced activity corresponding to the Maunder minimum (although they had fairly large uncertainties in their measured activities).

Production rates for a variety of radionuclides ranging in half-life from 16 days (^{48}V) to 3.7×10^6 years (^{53}Mn) were calculated for iron meteorites and compared with experimental activities in the Aroos iron meteorite by Arnold et al. (1961). The ratios of observed to calculated activities ranged from about 0.5 to 2 (probably due to both calculational and measurement uncertainties), but did not show any systematic trend with half-lives. These and other results for radioactivities in meteorites and lunar samples have shown that the fluxes of energetic (above about 500 MeV) GCR particles have varied less than about 25-50% during the last few million years and are similar to present fluxes.

Because some of the variations in production rates of cosmogenic nuclides are due to the size and shape of the meteorite, several authors have recently developed methods for determining exposure ages which include corrections for such shielding effects. Cressy and Boyard (1976) and Herzog and Cressy (1977) have used measured $^{22}\text{Ne}/^{21}\text{Ne}$ ratios to correct exposure ages for shielding effects and have found less scatter in their results. Recently Nishiizumi (priv. comm., 1979) has studied ^{26}Al , ^{53}Mn , ^{21}Ne , and $^{22}\text{Ne}/^{21}\text{Ne}$ data for a number of stone meteorites and has concluded that, after making shielding corrections, the average flux seen by ^{26}Al in meteorites is significantly greater (~ 40%) than that for ^{53}Mn . The reactions producing these two radionuclides have low threshold energies, so this flux ratio involves lower energy particles than that for reactions in iron meteorites.

There also is evidence of GCR flux variations in the past from $^{40}\text{K}/^{41}\text{K}$ ratios in iron meteorites. Hampel and Schaeffer (1979) have determined $^{26}\text{Al}/^{21}\text{Ne}$ exposure ages for several iron meteorites and found them in agreement with $^{36}\text{Cl}/^{36}\text{Ar}$ and $^{39}\text{Ar}/^{38}\text{Ar}$ exposure ages to within about 20%. However, exposure ages determined from $^{40}\text{K}/^{41}\text{K}$ data are about 50% greater than the

others. Hampel and Schaeffer (1979) conclude that meteorite orbital changes $\sim 10^6$ to 10^7 years ago and space erosion cannot explain this difference, and therefore the flux of the cosmic rays to which iron meteorites were exposed during the past 10^6 years is $\sim 50\%$ more intense than that averaged over the last 10^9 years.

There are many things besides GCR flux changes which can cause different cosmogenic-nuclide production rates and apparent exposure ages. For example, shielding changes due to multiple collisions and other causes could alter production rates, especially in meteorites with very long exposure ages. These other sources must be considered and eliminated before concluding that GCR flux variations cause production-rate changes in meteorites. As noted above, a GCR flux change could either be solar or non-solar in origin. Yanagita and Imamura (1979) have proposed that the flux of GCR particles increased about 5×10^6 years ago due to the movement of the solar system from a high-density region of the galaxy into a low-density "interstellar tunnel."

SOLAR-COSMIC-RAY FLUX VARIATIONS

Lunar rocks are almost ideal for studying SCR particle fluxes in the past. Although erosion of the surfaces of rocks (~ 1 mm/My) affects the depth-activity profiles of radionuclides with half-lives greater than about 1 My, the direct exposure of lunar rocks to the solar cosmic rays at a fixed distance from the sun makes studies of SCR particle fluxes relatively simple. The Apollo missions brought back many lunar rocks with simple, known exposures to cosmic rays and geometries suitable for determining SCR particle fluxes. If a depth-activity profile of a radionuclide is well measured at several depths near the surface, if there are good cross sections measured for the relevant reactions, and if the GCR production rates near the surface can be predicted, then good average SCR particle fluxes can be determined.

Measurements of radionuclide activities in Apollo 11 samples showed that production by relatively low-energy (about 10 to 100 MeV) protons and alpha particles were clearly present (see, e.g., Shedlovsky et al., 1970) as shown by the profiles of activities with depth and the presence of nuclides which could have been made only by such particles (e.g., ^{56}Co by the $^{56}\text{Fe}(p,n)$ reaction). Later studies (especially those of Finkel et al., 1971, and Kohl et al., 1978) made detailed measurements of the depth-activity profiles of a number of radionuclides, showed that the excess activities near the surface were produced by SCR particles, and used long-lived products to study average solar-proton fluxes in the past.

Direct measurements of solar-proton fluxes have been made since the early 1960's, the best fluxes for intense SCR events being measured by the Solar Proton Monitor Experiment (SPME) of Bostrom et al. (1967-1973). In lunar samples, radionuclides with half-lives below about one year (e.g., ^{56}Co , ^{54}Mn , ^{37}Ar) were made almost entirely during the period when solar-proton fluxes were measured by the SPME. The SCR-produced activities of short-lived radionuclides in samples from various Apollo missions are in good agreement with those expected from the SPME-measured proton fluxes for the solar flares occurring before each mission (Reedy, 1977; Fireman, this conference). This agreement for SCR particle fluxes measured by satellites and inferred from lunar-rock radioactivities confirms the validity of using lunar samples to study the activity history of the ancient sun.

The low-energy-proton reactions which produce 2.6-y ^{22}Na and 7.3×10^5 -y ^{26}Al in lunar rocks are very similar. Because similar depth-activity profiles were measured for these radionuclides in Apollo 11 and 12 samples, Finkel et al. (1971) and others concluded that the intensity and spectral shape of solar protons averaged over the last million years were similar to

those observed recently. However, the great similarity of the ^{22}Na and ^{26}Al SCR-produced activities was largely accidental. As shown by Reedy (1977), most of the SCR-produced ^{22}Na activities in Apollo 11, 12 and 14 samples were made by a number of intense flares about a decade (four half-lives) before the Apollo missions.

Adopting the SPME- and other satellite-measured solar-proton fluxes for 1965-1972 (solar cycle 20), Reedy (1977) used lunar depth-activity profiles for ^{22}Na and 2.7-y ^{55}Fe to determine the fluxes for solar protons during solar cycle 19 (1954-1964). Only about 20% of the solar-proton-induced activities of these two radionuclides in Apollo 11, 12, and 14 samples were made during solar cycle 20. The distribution of solar protons as determined from PCA and GLE events was used and the proton intensities estimated from these indirect measurements were increased by factors of 3 to 7 to fit the ^{22}Na and ^{55}Fe activities (Reedy, 1977). The fluxes adopted for solar cycle 20 and determined for solar cycle 19 by Reedy (1977) are included in Table 3.

Depth-activity profiles for 12.3-y ^3H were measured in several lunar rocks (D'Amico et al., 1971; Niederer et al., 1975). Because of the high solar-proton fluxes during solar cycle 19, the contribution of the protons emitted from the sun prior to 1954 to ^3H production in lunar samples was relatively small, and very little can be determined quantitatively from lunar ^3H about pre-1954 solar-proton fluxes. The lunar ^3H does exclude the possibility that the solar-proton fluxes for solar cycle 18 were as high as those for solar cycle 19. A rough estimate from the ^3H data for the average fluxes of solar protons with energies above 10 MeV emitted prior to 1954 is of the order of 150 protons/cm² s (Reedy, 1977; Niederer et al., 1975).

The next longest half-life of a radionuclide regularly measured in lunar samples (see Table 1) is 269 years for ^{39}Ar . (None of the radionuclides

listed in Table 2 with half-lives above 10 years but below 269 years are produced in any significant amount by cosmic-ray particles.) This radionuclide is produced readily by GCR secondary neutrons via $^{40}\text{Ca}(n,2p)^{39}\text{Ar}$ and $^{39}\text{K}(n,p)^{39}\text{Ar}$ reactions, but only in low yields by SCR particle reactions with titanium, iron, and the minor isotopes of calcium and potassium. The ^{39}Ar depth-activity profiles measured by D'Amico et al. (1971) and Fireman et al. (1972) show indications of SCR-produced excesses in the top of several lunar rocks, but these excesses are only about 10% of the total activities in those layers and are quite uncertain. These surface layers were 5 to 8 mm thick, whereas the majority of SCR-produced ^{39}Ar would be in the top few millimeters. Good measurements taken on millimeter-thick layers from the surface of a lunar rock might be able to sufficiently identify the SCR-produced excess (estimated as being a few dpm/kg in the top millimeter) relative to the GCR-produced activity (about 8 dpm/kg). Such detailed studies of ^{39}Ar in the very surfaces of lunar rocks would be interesting because a large fraction of the ^{39}Ar was made during the Maunder and Spörer minima when solar activity was very low and very few protons probably were emitted from the sun.

Again there is a large jump to the next longest half-life of a frequently measured lunar radionuclide, 5730-y ^{14}C . There are no good radionuclides with which to fill this half-life gap, as the few candidates listed in Table 2 are often made in low yields by SCR particles relative to their GCR production (e.g., ^{32}Si) and all would be very difficult to measure in lunar samples. The activities of ^{14}C were measured for six depths in rock 12002 by Boeckl (1972) and for three depths in rock 12053 by Bequemann et al. (1972). The activities measured in the deeper samples by both groups agree well. However, the activity measured by Bequemann et al. (1972) in their top

sample (0-5 mm) is much greater than the equivalent activity for Boeckl (1972) based on data for his three top samples (0-1, 1-2, and 2-4 mm). Begemann et al. (1972) ascribed the large activity of ^{14}C in their top layer to solar-wind-implanted ^{14}C because the flux of solar protons required to fit their depth-activity profile was much greater than fluxes deduced from other radionuclides. Because of the few data points in their profile, Begemann et al. (1972) could fit their results with several flux shapes and intensities. Boeckl (1972) fitted his results with a solar-proton spectral shape of $R_0 = 100$ MV and a flux above 10 MeV of 200 protons/cm² s, 0.67 of the intensity for the same R_0 as obtained by Begemann et al. (1972).

To test the hypothesis that there was solar-wind ^{14}C implanted in lunar samples, Fireman and co-workers did some measurements of the ^{14}C released from lunar soils at different temperatures (Fireman et al., 1976) and from different size fractions (Fireman, 1978). Fireman's conclusion was that solar-wind-implanted ^{14}C was present in these soil samples.

The ^{14}C activities measured in these two lunar rocks and the soil samples can all be explained by assuming (1) that there is solar-wind-implanted ^{14}C on lunar samples (about 3×10^{-4} atoms/cm² s on 12053 but very little on 12002, probably because the very top which contained this solar-wind-implanted ^{14}C was lost in handling prior to analysis), and (2) that the solar-proton flux deduced by Boeckl (1972) is correct. The flux of solar protons which is obtained from Boeckl's ^{14}C data is essentially the same whether one includes or excludes the top sample (a good solar-proton flux can be deduced from the ^{14}C activities in the layers below 1 mm). The activities of solar-proton-produced ^{14}C in the deepest samples of 12002 and 12053 are low enough that a good normalization factor for GCR-produced ^{14}C can be obtained. As discussed above, the major source of uncertainty in these solar-proton

fluxes is the cross sections for the $^{16}\text{O}(p,3p)^{14}\text{C}$ reaction. In Table 3, the solar-proton flux for energies above 10 MeV is more uncertain than the other fluxes because the threshold energy for ^{14}C production is about 25 MeV.

Although there are no other measurements involving radionuclides with half-lives even close to that of ^{14}C , Hoyt et al. (1973) studied thermoluminescence (TL) in the top few centimeters of lunar rock 14310. They estimated that the half-life of a radiation-damage-produced trapped electron (the source of the observed TL) is about 2×10^3 years. The fluxes of solar protons which Hoyt et al. (1973) obtained from their lunar-rock TL data are given in Table 3. These fluxes have fairly large uncertainties, but are considerably lower than the fluxes deduced from lunar-rock ^{14}C activities. Additional measurements of TL and a better determination of the half-life of trapped electrons in lunar rocks might help to resolve the discrepancies between the solar-proton fluxes deduced from TL and ^{14}C data.

Again there is a quantum jump in the half-lives of radionuclides studied in lunar samples to 8×10^4 -y ^{59}Ni . Because of the relatively low nickel and cobalt contents of lunar rocks, almost all of the ^{59}Ni is produced by solar alpha particles via the $^{56}\text{Fe}(\alpha,n)^{59}\text{Ni}$ reaction (Lanzerotti et al., 1973). Unfortunately, there have been very few ^{59}Ni activities measured for lunar samples and the uncertainties in these measurements are quite large. From the few lunar ^{59}Ni measurements that exist and from satellite measurements of solar alpha particles for 1967-1969, Lanzerotti et al. (1973) concluded that long-term and current solar-alpha-particle fluxes are comparable to within a factor of four. Good measurements of a ^{59}Ni depth-activity profile in lunar rock using improved X-ray counters, accelerator ion counting, or detection of alpha particles produced by thermal-neutron-induced $^{59}\text{Ni}(n,\alpha)$ reactions would allow determination of the fluxes of solar alpha particles

over the last 10^5 years. The only complication in using ^{59}Ni is that erosion of the rock surface can affect the ^{59}Ni depth-activity profile (Lanzerotti et al, 1973).

The noble-gas isotope ^{81}Kr (2.1×10^5 y) is the next longest SCR-produced radionuclide measured in lunar samples. Marti and Lugmair (1971) measured noble-gas isotopic concentrations in samples from several depths of lunar rock 12002 and reported that the stable Kr isotopes showed no excesses near the surface due to SCR production, but that ^{81}Kr had such an excess, as might be expected for a rock with a complex exposure history. Using the Kr data for ^{81}Kr in 12002 (K. Marti, priv. comm., 1979; Yaniv et al., 1980) and the Kr-production cross sections of Regnier et al. (1979), the solar-proton fluxes given in Table 3 were obtained. No fluxes are given for energies below 60 MeV because the main reactions producing ^{81}Kr in lunar samples have threshold energies above 60 MeV.

There are many factors contributing to the uncertainties of the solar-proton fluxes determined from ^{81}Kr data. Chemical abundances of the target elements (Rb, Sr, Y, and Zr) were not measured in the samples used for Kr measurements. Measurements made with other samples of rock 12002 and similar rocks show that these elements are fairly homogeneous and the uncertainties of the elemental abundances used here are about $\pm 10\%$. Only a few cross sections have been measured for ^{81}Kr production from these target elements; most of the cross sections are estimated on the basis of nuclear systematics (Regnier et al., 1979). If the ^{81}Kr -production cross sections used by Hohenberg et al. (1978) are used, the solar-proton fluxes obtained are about 0.75 of those given here. Regnier and co-workers plan to measure additional Kr-production cross sections from these target elements soon. Another large source of uncertainty in these solar-proton fluxes is the GCR production

normalization factor for ^{81}Kr . The deepest sample in which ^{81}Kr was measured in rock 12002 (2-6 cm) appears to have considerable SCR-produced activity of ^{81}Kr . Regnier et al. (1979) compared observed and calculated Kr isotopic abundances in ten samples (none of which was deeper than 6 cm), and the average value of observed-to-calculated ^{81}Kr activities in the deeper samples was 0.97, the value adapted here, but with a 25% standard deviation. The ^{81}Kr activity in the 0-1 mm layer of rock is 1.7 times that in the 2-6 cm layer, so variations in the GCR normalization factor should not greatly change the solar-proton fluxes. An uncertainty of $\pm 25\%$ in this normalization factor affects the solar-proton flux by about $\pm 20\%$. New cross-section data and additional lunar rock analyses for ^{81}Kr will help to reduce the uncertainty (of the order of $\pm 40\%$) for the solar-proton fluxes given here.

Yaniv and Marti recently measured noble-gas concentrations in samples of lunar rock 68815, which was shielded from cosmic rays until only two million years ago when it was placed in the position it was found by the Apollo 16 astronauts. Comparisons of ^{81}Kr , made mainly over the last 3×10^5 years, and stable Kr isotopes, made over the last 2×10^6 years, allow the ratio of solar-proton fluxes over these two time periods to be fairly well determined essentially independently of the uncertainty sources listed above (because these nuclides are made from the same target elements and because only relative production ratios need to be known). Preliminary evaluation of the Kr data for 68815 (Yaniv et al., 1980) is consistent with the solar-proton flux variations determined from radionuclide activities in rock 12002 (Table 3).

The production of $3.0 \times 10^5\text{-y}$ ^{36}Cl (the next longest half-life) in lunar samples is almost entirely by GCR particles (Reedy and Arnold, 1972). Even the high solar-proton fluxes given in Table 3 for ^{14}C and ^{81}Kr would

produce only about 10% of the ^{36}Cl present in the top millimeter of a lunar rock, a value considerably less than the uncertainties in most lunar ^{36}Cl measurements. The other radionuclides in Table 2 with half-lives of the order of 10^5 years also are high-energy products and hence have low SCR/GCR production ratios, and usually are hard to measure in lunar samples.

The radionuclide, ^{26}Al (7.3×10^5 y) and ^{53}Mn (3.7×10^6 y) are almost ideal for studying solar-proton fluxes: over half of their production in the surfaces of lunar rocks is by solar protons, they are easy to measure, good cross sections exist for their production by low-energy protons, the reactions producing them have threshold energies near 10 MeV, and they have been frequently measured in deep lunar samples so the GCR-production normalization factors are well known. The main complication is the effect of erosion, especially for ^{53}Mn . Because a given depth-activity profile for ^{53}Mn can be fitted using several sets of erosion rates and solar-proton fluxes (a low erosion rate requiring fewer protons, especially at low energies), it is best to determine erosion rates independently of cosmogenic radionuclides (e.g., using heavy nuclei tracks). Erosion rates are considerably lower in igneous rocks than they are in lunar breccias (e.g., 0.5 mm/My for rock 12002 versus 2.2 mm/My for breccia 14321, Kohl et al., 1978).

Three groups of investigators have measured ^{26}Al in lunar samples and reported solar-proton fluxes. Each group used different measurement techniques and there was little agreement among the three groups on the intensity or the spectral shape of solar-proton fluxes over the mean-life of ^{26}Al . Rancitelli et al. (1972) did non-destructive γ - γ coincidence counting on a number of lunar samples and got an integral flux above 10 MeV of 60 protons/cm² s using an energy power law of the shape $E^{-3.1}$. It is not clear how or with what samples they obtained their results. Bhandari et al. (1976)

non-destructively measured positron emissions from surfaces of rock chips using β - γ coincidence counting and reported an integral flux above 10 MeV of 140 protons/cm² s with an exponential-rigidity parameter $R_0 = 150$ MV. Kohl et al. (1978) summarized their ²⁶Al measurements and those of Finkel et al. (1971) and Wahlen et al. (1972), all of which involved grinding many layers from three rocks, chemically separating aluminum, and counting the positrons in a β - γ coincidence system. They fitted their ²⁶Al and ⁵³Mn activities in the three rocks with an integral flux above 10 MeV of 70 protons/cm² s and with $R_0 = 100$ MV.

These three results differ considerably, especially at high proton energies. The integral proton fluxes above 60 MeV of Bhandari et al. (1976) and Rancitelli et al. (1972) were about 4 times and $\frac{1}{4}$ of, respectively, the value reported by Kohl et al. (1978). Because Rancitelli et al. (1972) didn't give the data used to obtain their proton fluxes, it is impossible to check their results. The SCR-produced ²⁶Al activities measured by Bhandari et al. (1976) in their Apollo 16 rocks were factors of two or more higher than those of Kohl et al. (1978) in 68815, hence their large proton fluxes. There is no obvious explanation for this discrepancy. Because the procedures used by Kohl et al. (1978) clearly determine the sample depths and chemically separate ²⁶Al from other positron emitters and because their fluxes agree with those based on ⁵³Mn activities over the last two million years from rock 68815, I prefer their flux results and list them in Table 3.

Bhandari et al. (1976) used the same measurement techniques to study ²⁶Al in four rocks with exposure ages ranging from about 0.5 to 3.7 million years. They found that the observed ²⁶Al activities scattered about the expected value and showed no trend with the samples' exposure ages. Their conclusion was that the average solar-proton fluxes during the past 0.5, 1,

and 1.5 million years have varied by less than $\pm 25\%$. The ^{26}Al and ^{53}Mn results reported by Kohl et al. (1978) for 68815 were from three faces which had different exposure angles to the sun. Russ and Emerson (these proceedings) have done production-rate calculations which considered the details of the geometry for each face and they found no indications of flux anisotropy.

The solar-proton fluxes based on ^{53}Mn activities which are given in Table 3 are those of Kohl et al. (1978), who adopted the fluxes determined from their ^{26}Al measurements and varied the erosion rates of their samples to fit the measured ^{53}Mn activities. As mentioned above, the results for rock 68815 show that the solar-proton fluxes for the last 2 My and over the mean-life of ^{26}Al (~1 My) are quite similar. The good fit to the ^{53}Mn activities of 12002 which had a low erosion rate, indicates that the fluxes of solar protons over the last five to ten million years were not greatly different than those over the last two million years.

Several other long-lived radionuclides have been studied in lunar samples. Fields et al. (1972, 1973) mass-spectrometrically studied isotopes of uranium and a few other actinides. They did not see any $8.3 \times 10^7\text{-y } ^{244}\text{Pu}$ or $2.4 \times 10^4\text{-y } ^{239}\text{Pu}$. They measured $2.3 \times 10^7\text{-y } ^{236}\text{U}$ in several samples. Most of the $^{236}\text{U}/^{238}\text{U}$ ratios were about 5×10^{-9} , near the levels expected from neutron-capture reactions with ^{235}U . However, several samples had considerably higher $^{236}\text{U}/^{238}\text{U}$ ratios which Fields et al. (1972) felt was due to high solar-proton fluxes over the mean-life of ^{236}U . The high ^{236}U concentrations were later found to be due to contamination by airborne ^{236}U (Fields et al., 1976). Concentrations of $2.1 \times 10^6\text{-y } ^{237}\text{Np}$ were measured in several lunar samples (Fields et al., 1973). Most $^{237}\text{Np}/^{238}\text{U}$ ratios were about 4×10^{-9} , an order of magnitude higher than expected from production by GCR and SCR particles (using solar-proton fluxes determined from activities

of ^{53}Mn , which has a half-life similar to that of ^{237}Np). Several samples had much higher $^{237}\text{Np}/^{238}\text{U}$ ratios, probably due to contamination (Fields et al., 1976).

Le Roulley et al. (1974) proposed to study $1.0 \times 10^8\text{-y } ^{146}\text{Sm}$ by measuring the alpha particles emitted by ^{146}Sm (2.50 MeV) and $1.05 \times 10^{11}\text{-y } ^{147}\text{Sm}$ (2.23 MeV). Samarium was chemically separated from lunar fines 10084 and its alpha activity measured for six months by Le Roulley et al. (1978). The upper limit they reported for the $^{146}\text{Sm}/^{147}\text{Sm}$ activity ratio was 30 times the value expected for production by present-day cosmic-ray fluxes. Direct measurement of ^{146}Sm , such as by mass spectrometry, probably will be needed to detect such a long-lived cosmogenic radionuclide.

The fluxes of SCR particles more than 10^7 years ago have not been determined, mainly because of difficulties in detecting activities of very long-lived cosmogenic radionuclides or in distinguishing SCR-produced stable isotopes from other sources of the same isotopes. The stable noble-gas isotopes, especially those of neon, krypton and xenon, are well suited for studies of cosmic-ray fluxes in the distant past because almost none were present when a sample formed and because isotopic ratios can be used to distinguish among the various sources (e.g., GCR, SCR, solar wind, and fission). Accurately known isotopic ratios for the various sources are needed to determine the amount present from each source (see, e.g., Hohenberg et al., 1978). Lunar rocks are of limited value in studies of very ancient SCR particle fluxes because erosion removes the surfaces where such SCR production occurred. The lunar soil retains nuclides from all sources, but gardening by micrometeoroid impacts moves around the fine material in the regolith (Langevin and Arnold, 1977). Work needs to be done on predicting and experimentally identifying SCR-produced noble gases in soils before

analyses of several soil samples (to get a statistical sampling of gardening effects) can give information on SCR particle fluxes in the distant past.

DISCUSSION

Radionuclide activities measured in meteorites and lunar samples provide evidence of some variation in the fluxes of the galactic cosmic rays and solar cosmic rays in the past. However, the variations observed in the fluxes of both types of cosmic rays over the last few decades are larger than those in the average fluxes in the distant past. In fact, the differences in the fluxes of solar protons emitted during solar cycles 19 and 20 are so great (Table 3) that the average fluxes of solar protons over the last million years are known better than "present-day" fluxes. The fluxes of both GCR and SCR particles during recent years are similar to the average fluxes over the last few million years, indicating that current solar activity is not atypical of what it has been in the past.

Changes in the average fluxes of GCR particles, especially over the last few million years, are very poorly known. Improved measurements of radionuclides in meteorites (especially iron meteorites) and calculations of their production rates would help to identify or set limits on variations in the fluxes of GCR particles. It would be interesting to measure ^{39}Ar (and ^{32}Si) in the metal phases of meteorites which fell several centuries ago and see if enhanced activities are present because of increased GCR fluxes during the Maunder and other periods of minimum solar activity.

Such studies using meteorites or lunar samples could aid studies of cosmogenic ^{10}Be and other radionuclides in ice cores or sea sediments by helping to distinguish between cosmic-ray-flux variations and effects due to the Earth's magnetic field. A major difficulty in using galactic cosmic rays to study the ancient sun is that changes in GCR particle fluxes can

occur because of non-solar effects. However, most variations due to causes other than solar activity should happen slowly, and so fairly rapid fluctuations, like those noted for tree-ring ^{14}C by Eddy (1976), are probably diagnostic of solar activity in the past.

The fluxes of solar-cosmic-ray particles are very indicative of solar activity. While the similarity of solar-proton fluxes averaged over the last few million years and observed recently is noteworthy, the evidence for considerably larger fluxes over the last 10^4 and 10^5 years is intriguing. While I believe the results given in Table 3 are generally correct, much work could be done to improve the accuracies of the fluxes given there and to add results for additional radionuclides or time periods. The discrepancies among the ^{26}Al results for various groups should be resolved. The erosion rates of lunar rocks should be determined independently of measured ^{53}Mn depth-activity profiles. Additional cross sections for protons producing certain radionuclides, especially ^{14}C and ^{81}Kr , need to be measured. Concentrations of ^{81}Kr and other noble-gas isotopes should be measured in more lunar samples and at depths where SCR production is negligible in addition to the surface layers where SCR effects prevail. Measurements of SCR-produced nuclides in rocks with known exposure ages would provide fluxes for time periods other than mean-lives of radionuclides and help to convert average integral fluxes into fluxes for "differential" time periods in the past.

Most of the above tasks could be done using existing technology. However, new methods of detecting cosmogenic nuclides, such as accelerators and lasers, would enable additional nuclides, such as those listed in Table 2, to be measured and would allow studies of much smaller samples. Many of these seldom-studied radionuclides have interesting half-lives or production

modes (e.g., solar alpha particles) with which to study the history of SCR particles. Analyses using very small samples not only would make the measurements currently made (e.g., depth-activity profiles in lunar rocks) much easier, but would allow types of materials not now studied to be accessible to investigation. To date, very few studies have been made of activities in the cosmic dust or micrometeorites present in sea sediments and ice layers. Such samples can be dated and would provide nuclides whose exposure to cosmic rays ended at a known time. At present, only meteorites which fell many years ago provide samples which have been recently shielded from cosmic rays. Sea sediments and ice cores also might contain cosmogenic radionuclides both made in cosmic dust or micrometeoroids in space over relatively long periods and made in the Earth's upper atmosphere prior to fall out (Arnold, 1979).

Cosmogenic nuclides can be used to study the history of, or processes occurring in, the samples in which they are found, in addition to being diagnostic of cosmic-ray temporal variations. Such studies are complementary, as improved understanding of such processes (e.g., lunar-rock erosion rates) aids studies of cosmic-ray flux variations and visa versa. Many meteorites and parts of the lunar regolith or of lunar breccias have samples which contain the records of solar activity back as far as 4 or 4.5 Gy ago, but a better knowledge (often gained using cosmogenic effects) is needed of their formation and evolution before the records can be deciphered.

Cosmogenic nuclides provide good evidence that the ancient sun was not perfectly constant, but that it had variations in its activity. Hopefully the fluctuations observed in cosmic-ray fluxes can be combined with other fossil records in the moon, meteorites, and the Earth to improve our knowledge of the ancient sun.

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Table 1. Radionuclides frequently observed in extraterrestrial matter, their half-lives, the target elements from which they usually are produced, and the types of cosmic-ray particles that can produce significant amounts of them in lunar samples.

<u>Radionuclide</u>	<u>Half-life (y)</u>	<u>Targets</u>	<u>Particles</u>
³ H	12.33	O, Mg, Si	GCR, SCR
¹⁰ Be	1.6 x 10 ⁶	O, Mg, Si	GCR
¹⁴ C	5730	O, Mg, Si	GCR, SCR
²² Na	2.60	Mg, Al, Si	GCR, SCR
²⁶ Al	7.3 x 10 ⁵	Al, Si	GCR, SCR
³⁶ Cl	3.0 x 10 ⁵	Ca, Fe	GCR
³⁷ Ar	0.095	Ca, Fe	GCR, SCR
³⁹ Ar	269	K, Ca, Fe	GCR
⁴⁶ Sc	0.23	Ti, Fe	GCR
⁴⁸ V	0.044	Ti, Fe	GCR, SCR
⁵³ Mn	3.7 x 10 ⁶	Fe	GCR, SCR
⁵⁴ Mn	0.86	Fe	GCR, SCR
⁵⁵ Fe	2.7	Fe	GCR, SCR
⁵⁶ Co	0.215	Fe	SCR
⁵⁹ Ni	8 x 10 ⁴	Fe, Ni	SCR, GCR
⁶⁰ Co	5.27	Co, Ni	GCR
⁸¹ Kr	2.1 x 10 ⁵	Sr, Y, Zr	GCR, SCR

Table 2. Additional long-lived radionuclides which can be or occasionally are used to study cosmic-ray fluxes in the past, in order of their half-lives.

<u>Radionuclide</u>	<u>Half-life (y)</u>	<u>Radionuclide</u>	<u>Half-life (y)</u>
^{42}Ar	33	^{233}U	1.6×10^5
^{44}Ti	47	^{60}Fe	$\sim 1 \times 10^5$
^{63}Ni	100	^{237}Np	2.1×10^6
^{32}Si	~ 370	^{129}I	1.6×10^7
^{91}Nb	~ 800	^{236}U	2.3×10^7
^{93}Mo	$\sim 3.5 \times 10^3$	^{92}Nb	3.3×10^7
^{94}Nb	2.0×10^4	^{146}Sm	1.0×10^8
^{41}Ca	1.3×10^5	^{40}K	1.28×10^9

Table 3. Average solar-proton fluxes over various time periods as determined from lunar radioactivity measurements.

Period (Data)	Fluxes (protons/cm ² s)			
	E>10 MeV	E>30 MeV	E>60 MeV	E>100 MeV
1965-1975 (SPME) ^a	89	28	8.0	--
1965-7/72 ^b (SPME) ^a	25	4.2	0.9	--
1954-1964 (²² Na, ⁵⁵ Fe) ^a	378	136	59	26
~5 x 10 ³ y (Ti) ^c	~60	~14	~6	~3
10 ⁴ y (¹⁴ C) ^d	~200	72	26	9
3 x 10 ⁵ y (⁸¹ Kr) ^e	--	--	~18	~9
10 ⁶ y (²⁶ Al) ^f	70	25	9	3
5 x 10 ⁶ y (⁵³ Mn) ^f	70	25	9	3

^aReedy (1977), SPME is the Solar Proton Monitor Experiment (Bostrom et al., 1967-1973).

^bAveraged over 11 years.

^cHoyt et al. (1973).

^dBoeckl (1972).

^eThis work, using Kr data from lunar rock 12002 (K. Marti, priv. comm., 1979; Yaniv et al., 1980).

^fKohl et al. (1978).