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LA-UR87-1397

CONF-8704112--1

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

LA-UR--87-1397

DE87 009008

TITLE: Predicting the Production Rates of Cosmogenic
Nuclides in Extraterrestrial Matter.

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SUBMITTED TO: Proceedings of the 4th International Symposium on
Accelerator Mass Spectrometry,
Niagara-on-the-Lake, Ontario, 27-30 April 1987.

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PREDICTING THE PRODUCTION RATES OF COSMOGENIC NUCLIDES IN EXTRATERRESTRIAL MATTER

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The production rates of nuclides made by the galactic and solar cosmic rays are important in the interpretations of measurements made with lunar samples, meteorites, and cosmic spherules. Production rates of cosmogenic nuclides have been predicted by a variety of methods that are reviewed in this paper, ranging from systematic studies of one or a group of meteorites to purely theoretical calculations. Production rates can vary with the chemical composition and the preatmospheric depth of the sample and with the size and shape of the object. While the production systematics for cosmogenic nuclides are fairly well known, our ability to predict their production rates can be improved, with a corresponding increase in the scientific return. Additional detailed studies of cosmogenic nuclides in extraterrestrial objects are needed, especially for fairly small and very large objects. Nuclides made in simulation experiments and cross sections for many major nuclear reactions should be measured. Such studies are especially needed for the long-lived radionuclides that have only recently become readily measurable by accelerator mass spectrometry.

1. Introduction

The inner solar system not only contains the terrestrial planets and their moons, but also a number of smaller objects and a variety of radiations. Some of these objects reach the surface of the earth without being completely destroyed, such as meteorites, cosmic dust, and cosmic spherules. In addition, lunar samples have been returned to earth by the Apollo and Luna missions. There are a great variety of radiations, including some of low energies (such as the solar wind) or low fluxes (such as gamma rays). Two types of radiation, the galactic cosmic rays and solar-flare-associated energetic particles (often called solar cosmic rays), have high enough energies and are intense enough that they can induce nuclear reactions and make detectable amounts of certain products, such as radionuclides, that can be identified as having been made by cosmic-ray particles [1].

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After the discovery of cosmic-ray-produced (cosmogenic) nuclides about 40 years ago, a wide variety of studies soon were being done with them (see historical review in ref. [2]). These studies ranged from the nature of the cosmic-ray interactions with matter to studies of the histories of the cosmic rays and of the objects themselves (see, for example, the review in ref. [1]). The initial work on cosmogenic nuclides came soon after the first high-energy (above ~ 1 GeV) accelerators were built, and studies of cosmogenic nuclides made by the GeV protons in the galactic cosmic rays (GCR) with meteorites complimented work being done on high-energy nuclear reactions at these accelerators (e.g., the work on spallation systematics with iron by ref. [3]). Long-lived cosmogenic radionuclides showed that the intensities of GCR particles had not varied much in the past. The lengths of time that meteorites had been exposed to cosmic rays (called exposure ages) were determined to have been ~ 10 Ma (10^7 years) for stony meteorites and ~ 100 Ma to 1 Ga for iron meteorites, much less than their formation ages 4.5 Ga ago.

Very soon after the first observations of cosmogenic nuclides, several systematic studies were done with meteoritic samples that helped to establish some initial production rates of cosmogenic nuclides in meteorites. Several models were soon developed for the prediction of the production rates of cosmogenic nuclides (e.g., [4, 5]). Thick-target experiments at high-energy accelerators were also done to help in predicting production rates of nuclides made by the GCR (e.g., [6, 7]). With the return of samples from the moon, the study of solar cosmic rays (SCR) and their effects in extraterrestrial matter progressed rapidly (e.g., [8, 9]). Some production rates of cosmogenic nuclides have been predicted using purely theoretical calculations (for example, [10, 11]). Many models for the prediction of these production rates used a theoretical approach that was normalized to experimental data for extraterrestrial samples (such as [4, 12]), and some models require a variety of basic data, such as cross sections for nuclear reactions (e.g., [8]).

This review of the predictions of cosmogenic-nuclide production rates will consider only the galactic cosmic rays (GCR) and solar cosmic rays (SCR). These two types of cosmic rays differ in many ways [1, 8], especially their energies ($\sim 1 - 10$ GeV for the GCR versus $\sim 10 - 100$ MeV for the SCR) and fluxes (~ 3 particles $\text{cm}^{-2} \text{s}^{-1}$ for the GCR in contrast to ~ 100 particles $\text{cm}^{-2} \text{s}^{-1}$ above 10 MeV for the SCR averaged over long time periods). The high energy GCR particles produce a cascade of secondary particles, especially neutrons, while the solar particles usually are stopped by ionization energy losses before they can induce nuclear reactions. The targets for the cosmic rays discussed here will mainly be meteorites,

lunar samples, cosmic dust, and cosmic spherules. Only passing reference will be made to the production of cosmogenic nuclides in the earth's atmosphere and in terrestrial rocks. Because of the limitations of space for this article, only a few references will be given below, and many excellent works involving cross sections, cosmogenic-nuclide production rates, measurements of cosmogenic nuclides, and related work unfortunately can not be discussed or cited.

2. Solar Cosmic Rays

2.1 Nature of SCR particles and their interactions

The energetic particles from the sun that occasionally pass through the inner solar system have energies of $\sim 1 - 50$ MeV but usually not extending much above ~ 100 MeV [1, 8]. These energetic solar particle events mainly occur when the sun is relatively active and not during periods of low solar activity [1, 8, 9]. The fluxes of particles (which are $\sim 98\%$ protons, plus some alpha particles and a few heavier nuclei [13]) in these events vary from barely detectable to up to $\sim 10^6$ protons $\text{cm}^{-2} \text{ s}^{-1}$ at the peak of the largest events. Although first detected by ground-level instruments in 1942, the detailed study of SCR particles began in the 1960s and was aided by studies of SCR-produced nuclides in lunar samples (cf., [1, 9]). Our main source of knowledge about these particles before the 1960s has been from lunar samples. As intense particle fluxes in some rare SCR events are very hazardous to men and electronics in space, the studies of ancient SCR particles from their fossil records in lunar samples have been very valuable in understanding these energetic particles. However, additional work is needed by doing additional measurements of SCR-produced nuclides in lunar samples and in getting cross sections to interpret many of these measurements, such as for ^{14}C [14]. Studies of solar alpha particles over the last $\sim 10^5$ years using ^{59}Ni in lunar samples also could be done.

The energy spectra of the SCR particles decrease rapidly with increasing energy, having shapes roughly like a power law in energy, $E^{-\gamma}$, with γ typically in the range of 2-4. The spectrum of SCR particles varies considerably from event to event and even within an event. The observed spectra are usually flatter than a power law at lower energies and steeper at higher energies, and the spectral shape of the flux, J , is better described as an exponential rigidity [8],

$$\frac{dJ}{dR} = k \exp\left(\frac{-R}{R_0}\right), \quad (1)$$

where the parameter R_o describes the shape. Typical values of R_o range from $\sim 35 - 150$ MV [8, 9]. Over the last $\sim 10^6$ years, the average omnidirectional fluxes of solar protons above 10, 30, 60, and 100 MeV have been about 70, 25, 9, and 3 protons $\text{cm}^{-2} \text{s}^{-1}$, respectively [1].

Because the ranges of most SCR particles are much less than their nuclear interaction lengths, most SCR particles are stopped by ionization energy losses before they can react. Their ranges are short, $\sim 1 \text{ g cm}^{-2}$ in silicate material, so the few reactions that they produce are mainly in the top centimeter of an extraterrestrial target [8, 10]. The nuclear reactions induced by SCR particles tend to be low-energy ones like $^{56}\text{Fe}(p,n)^{56}\text{Co}$ and $^{28}\text{Si}(p,2pn)^{26}\text{Al}$, and few high-energy reactions, such as those that produce ^{10}Be from oxygen and heavier elements, are induced [8]. Relatively few secondary particles, such as neutrons, are produced in these reactions [10], so almost all nuclides are made by the primary SCR particles. SCR-produced nuclides were readily observed in lunar samples because little material had been lost by handling or by micrometeorite erosion on the lunar surface (which has rates of $\sim 1 \text{ mm Ma}^{-1}$). The outer layers of meteorites that have the SCR-produced nuclides are usually removed by ablation. Only very recently have meteorites been found that have large concentrations of SCR-produced nuclides, such as ALHA77005 [15] and Salem [16]. Concentrations of SCR-produced radionuclides should be very high in very small objects in space, such as those recovered in deep-sea sediments [17, 18].

2.2 Calculations of the production rates of SCR-produced nuclides

One of the first published results for the production rates of SCR-produced nuclides in lunar samples was the Monte Carlo calculations of Armstrong and Alsmiller [10]. These purely theoretical calculations started with the intranuclear cascade induced by the primary particle and followed the subsequent evaporation of particles from the excited nucleus to get the residual nucleus. All of the emitted secondary particles were tracked until they escaped from the moon or were removed by nuclear reactions or stopping. The results reproduced the lunar-sample results fairly well. A deficiency of this approach is that the code directly calculates the production of all residual nuclei without the use of any experimental cross sections. While such intranuclear-cascade/evaporation calculations usually work fairly well in reproducing experimental cross sections, they often fail for certain products. These calculations also ignored all incident solar protons with energies below 30 MeV, and such protons can be very important in the top few millimeters. Another limitation of this work was that only a few production profiles were reported, so that the interpretation of other nuclides made by solar protons was not possible. The calculations in ref. [10] did confirm

the expectation that secondary particles were relatively unimportant for the production of nuclides by the SCR particles.

The other type of model, the one that has almost always been used for calculating the production rates of nuclides by SCR particles, ignores secondary particles and only follows the primary particles. The first published paper using this approach for lunar samples was by Reedy and Arnold [8]. Using the incident flux and spectrum of SCR particles and the well-known relations for the slowing down of charged nuclei in matter, this model calculates the fluxes of SCR particles as a function of depth inside a target. These depth-dependent SCR-particle fluxes are calculated well by this approach. The production rate of a nuclide is then calculated by integrating over energy the product of these particle fluxes and the cross sections for the reaction making that nuclide. Often a nuclide can be made in appreciable yield from several different target elements, and this integration must be done for each element. The main limitation of this approach is the need for a detailed excitation function (cross sections as a function of energy) for each important reaction [8, 14]. Using known excitation functions and satellite-measured fluxes of solar protons, this model has reproduced well the activities of 78-day ^{56}Co measured in several lunar rocks [9]. This type of model has also been applied to several solar-proton-produced nuclides in meteorites by Michel and coworkers [19].

The excitation functions for the production of long-lived ^{26}Al by the reactions of protons with aluminum and silicon are shown in Fig. 1. Such excitation functions, especially for energies below 100 MeV, are needed for calculating production rates by SCR particles. Because of the rapid decrease in SCR-particle fluxes with increasing energy, the most important cross sections for nuclide production by SCR particles are those nearest the reaction thresholds. These excitation functions have been fairly well measured (especially by ref. [20]), although there are no measured cross sections between 52 and 300 MeV (which is not a serious problem, as the shape there can be fairly well estimated from other similar excitation functions). There is also a disagreement between the measurements by the Orsay [21, 22] and Bordeaux [23, 24] groups for $\text{Si}(p,x)^{26}\text{Al}$ reactions at and above 600 MeV, but the uncertainties at such energies only would affect calculations for the production of ^{26}Al by high-energy GCR particles. It would be nice to have several cross sections below 25 MeV for the $^{29}\text{Si}(p,\alpha)^{26}\text{Al}$ reaction (which is crudely estimated in the adopted curve on Fig. 1).

Production rates calculated as a function of sample depth and meteoroid radius for the production of ^{26}Al by solar protons are shown in Fig. 2. These production rates were calculated using the model of Reedy

and Arnold [8] after it was modified for spherical meteoroids and with the evaluated excitation functions shown in Fig. 1. The ranges of approximate ^{26}Al GCR production rates are indicated by the X on the left axis and the "(GCR)" near the right axis and show that solar-proton production of ^{26}Al in meteorites is important in all small meteoroids (radii less than $\sim 15 \text{ g cm}^{-2}$) and in the outer few centimeters of larger meteoroids. Similar production profiles for ^{26}Al and several other solar-proton-produced radionuclides were reported by ref. [19] for several types of meteorites. For all nuclides made by such low-energy reactions, the production rates drop very rapidly from the surface of the extraterrestrial object, as shown in Fig. 2. The observation of such high concentrations for nuclides made by low-energy proton-induced reactions is a strong indication that the sample had been very close to the surface. Rates for the SCR production of several nuclides in very small objects (such as indicated by the O in Fig. 2) have also been calculated (e.g., [18, 19]).

Some cosmogenic nuclides can only be made by solar particles with fairly high energies, such as ^{10}Be from oxygen and heavier target elements. Excitation functions for the production of ^{10}Be were recently presented in ref. [25], and their proton cross sections at lower energies are much less than those used previously by Reedy and Arnold [8]. The ^{10}Be solar-proton production rates in lunar rock 68815 are shown in Fig. 3 for several solar-proton spectral shapes and a flux of $70 \text{ protons cm}^{-2} \text{ s}^{-1}$ above 10 MeV. The average spectral shape of solar protons for the last few million years have been determined by several groups to be about $R_o = 100 \text{ MV}$, although another group has reported higher values for R_o (cf., [1]). The production rate of ^{10}Be in lunar rocks by GCR particles is $\sim 10 \text{ atoms min}^{-1} \text{ kg}^{-1}$, so solar-proton-produced ^{10}Be at the very surface would be hard to detect unless the precision of the analyses was less than $\sim 10\%$, especially if the correct solar-proton spectral shape is $R_o = 100 \text{ MV}$ or less. The use of ^{10}Be along with a radionuclide made by low-energy reactions (such as ^{26}Al or ^{53}Mn) could be very useful in restricting the possible spectral shapes of the solar proton. One limitation at present in calculations for solar-proton-produced ^{10}Be is that there are no measured cross sections for energies below 135 MeV (cf., [25]). Some ^{10}Be production cross sections should be measured at several lower proton energies from oxygen and the next most important target group (magnesium, aluminum, and silicon).

Many cross sections used for calculating production rates by solar proton were reported in ref. [8], although many are now out of date because of more recent measurements. Many excitation functions for targets with atomic numbers from 22 to 28 have been measured by Michel and coworkers (cf., references

in ref. [19]). However, there is still a need for additional cross sections, such as mentioned above for ^{10}Be . Cross sections for the $^{16}\text{O}(p,3p)^{14}\text{C}$ reaction have only been measured once, and some additional cross sections for this and other ^{14}C -producing reactions should be measured to confirm those used in determining the fluxes of solar protons over the last ~ 10 ka from lunar ^{14}C profiles [14]. The status of other excitation functions for several other solar-proton-produced radionuclides was also reviewed in ref. [14].

The average fluxes of solar protons over several time periods have been determined from measurements of activity-versus-depth profiles of ^{14}C , ^{81}Kr , ^{26}Al , and ^{53}Mn in the top centimeter of lunar rocks. As mentioned above and discussed in refs. [1, 14], there are some disagreements among the measurements. Now that the measurement of some of these long-lived radionuclides by accelerator mass spectrometry (AMS) is well established, it would be a good time to repeat these lunar measurements, especially with a finer depth grid. The inclusion of ^{10}Be with ^{26}Al or ^{53}Mn measured in these samples would help in interpreting the data for the last million years. For example, a different set of spectral shapes and lunar-rock erosion rates often could fit the measurements about as well as the adopted values. The average fluxes of solar alpha particles over the last ~ 100 ka could be studied by the ^{59}Ni made in the top few millimeters of lunar rocks by the $^{56}\text{Fe}(\alpha,n)^{59}\text{Ni}$ reaction. The SCR effects in meteorites should be further studied, especially since the recent discoveries of high concentrations of solar-proton radionuclides in the Salem [16] and ALHA77005 [15] meteorites. As the average fluxes of SCR particles should vary inversely with the square of distance from the sun [1], solar-proton-produced radionuclides could be useful in determining orbits of meteorites.

3. Galactic cosmic rays

3.1 The nature of GCR particles and their interactions

The major differences between the SCR and GCR are their energies and fluxes. The average energy of a GCR proton is ~ 2 GeV, and the energies range from a few MeV up to $\sim 10^{20}$ eV. Above ~ 10 GeV, the flux of GCR particles decreases with increasing energy roughly as $E^{-2.65}$. The flux of GCR particles near the earth is ~ 3 particles $\text{cm}^{-2} \text{ s}^{-1}$. The spectra and fluxes of GCR particles are modified from those in the local interstellar space by the solar wind and its magnetic field as it expands from the sun. During a typical 11-year solar cycle, the GCR particle fluxes are the greatest at the time of minimum solar

activity and the least at solar maximum. The GCR particles with energies below a few GeV nucleon⁻¹ are most affected by solar modulation [1], and the integral flux above 1 GeV nucleon⁻¹ varies by a factor of 2 between solar minimum and solar maximum [18]. The particles in the GCR are $\sim 87\%$ protons, $\sim 12\%$ alpha particles, and $\sim 1\%$ heavier nuclei [26].

As the ranges of GCR protons and alpha particles are much longer than their interaction lengths, almost all GCR particles induce nuclear reactions before they are stopped in matter. These high-energy reactions usually result in many secondary particles, especially pions and neutrons. On average, each GCR particle makes ~ 7 neutrons [8, 11] in its cascade of particles. Most of the secondary charged particles in this cascade are stopped by ionization energy losses, but the neutrons travel until they scatter from or react with a nucleus or they escape from the object. Below ~ 100 MeV, the dominant particles in the GCR cascade in all but the smallest objects are neutrons [8, 12]. The fluxes of GCR particles with energies above 1 GeV nucleon⁻¹ decrease exponentially in a large object, while the fluxes of the secondary neutrons first build up then decrease exponentially with depth [8].

Because the types of the particles and their energies in this cascade are sensitive to the size of the object and the depth inside it, the production rate of a cosmogenic nuclide can vary considerably in extraterrestrial matter. This effect of depth, size, and shape on production rates is referred to as “shielding.” In predicting the production rate of a nuclide by GCR particles, shielding needs to be considered along with the chemical composition of the sample. The effects of shielding vary considerably with the types of reactions that produce a cosmogenic nuclide, with the extremes being nuclides made only by high-energy reactions (e.g., ¹⁰Be) and those made by neutron-capture reactions (such as ⁵⁹Ni and ⁶⁰Co in large objects). The production-rates-versus-depth profiles shown in Fig. 4 for the moon illustrate these two extremes and two intermediate cases for the variation of shielding effects with the nature of the nuclear reactions producing a cosmogenic nuclide.

3.2 Different approaches to predicting production rates by GCR particles

Unlike the case for SCR particles, where only a couple of approaches have been used to predict the production rates of cosmogenic nuclides, there have been many different methods used for predicting GCR-particle interactions in extraterrestrial matter. This large number of approaches is partially because GCR-produced nuclides have been studied for about twice as long as SCR-produced nuclides but is mainly due to the variety of extraterrestrial material in which GCR-produced nuclides have been studied (stony

meteorites, iron meteorites, lunar samples, and cosmic spherules) and the diversity in types and energies of GCR particles inside these objects. Four approaches will be discussed in more detail in sections below: use of cosmogenic-nuclide systematics measured in extraterrestrial samples, theoretical calculations, models using inferred particle spectra and reaction cross sections, and laboratory simulation irradiations. Several other approaches will be briefly described in the next two paragraphs. These various approaches have their advantages and their limitations. Most models have fairly well reproduced the measured concentrations and profiles for cosmogenic nuclides in extraterrestrial matter.

One of the simpler approaches to predicting production rates has been to determine the production ratio of two cosmogenic nuclides and to get the rate for making one of these nuclides from this ratio and a known rate for the other nuclide. For many nuclides made by high-energy reactions (e.g., ^{36}Cl , ^{37}Ar , and ^{39}Ar in the metal phases of meteorites), the cross section ratios measured at an accelerator with GeV protons can be used to predict production ratios. The cross sections for the production of most nuclides made by high-energy spallation reactions systematically vary with ΔA , the difference in the mass of the target to that of the product. For $\Delta A > 5$, the yield of all species with mass A , $Y(A)$, can be expressed

$$Y(A) = c(\Delta A)^{-k}, \quad (2)$$

where the power k typically varies between 2 and 3 for GCR-particle reactions in iron meteorites [3]. To get the relative rate for a specific isotope, the isobaric yield of that isotope for that mass needs to be known [3]. This expression does not work well for products very close in mass to the target or with masses below about 15 (e.g., ^{10}Be).

One of the oldest models is that used by Signer and Nier [4] to parameterize their measurements for the distribution of noble-gas isotopes in large pieces of iron meteorites. This model uses an equation that considers only the exponential decay of primary GCR particles and the growth and subsequent exponential decay of secondaries along the path of the incident primary particle. This equation can be integrated for simple geometries. This approach does not consider energy losses or wide-angle production or scattering of the particles. This model works best for cosmogenic nuclides made by high-energy reactions, such as most nuclides in iron meteorites, where the model's limitations are least important. It has recently been applied to ^{10}Be and noble-gas isotopes in the Knyahinya chondrite [27].

3.3 Systematic measurements of nuclides in extraterrestrial samples

Soon after the discovery of cosmogenic nuclides in meteorites in the early 1950s, some systematic studies were done [2]. Several cosmogenic nuclides were measured in samples from a large number of meteorites. These studies showed that, to a first approximation, the production rates of most cosmogenic radionuclides did not vary considerably in most meteoritic samples. For example, the spread in activities of cosmogenic radionuclides was typically within $\sim 20\%$ of the mean activity. Several isotopic ratios were shown to correlate well with other ratios, such as the $^{22}\text{Ne}/^{21}\text{Ne}$ ratio versus the $^3\text{He}/^{21}\text{Ne}$ ratio. More recently, experimental trends of meteoritic measurements (e.g., [28]) showed that the $^{22}\text{Ne}/^{21}\text{Ne}$ ratio was a fairly good indicator of shielding for several radionuclides.

Some additional studies concentrated only on a number of samples from known locations of one meteorite, such as cores from the Keyes, Jilin, and St. Severin chondrites (e.g., [25]) or slabs from several iron meteorites (for example, [4]) and the Knyahinya chondrite [27]. These measurements for chondrites showed that the activity of a radionuclide as a function of depth in most meteorites increased with increasing depth near the surface, then remained fairly constant in the central parts. Iron meteorites, which are usually much larger than stony meteorites, have production profiles that often decrease towards the center (e.g., [4]). The trend lines, such as a radionuclide activity versus the $^{22}\text{Ne}/^{21}\text{Ne}$ ratio, for these cores or slabs sometimes have different slopes than did the same trend line for samples from different meteorites. Several studies used samples with different chemical compositions from one meteorite (such as mineral separates) or a group of meteorites to determine production rates from individual target elements, such as rates for producing ^{26}Al from aluminum, silicon, and other elements.

Many of the results from these systematic studies, such as the trend lines, have been used in predicting production rates in meteorites. These measurements also have been valuable in developing and testing models for the production rates of cosmogenic nuclides (e.g., [4, 12]). Often the calculated production rates for a nuclide from such models should be normalized to experimentally determined production rates. Additional systematic studies are being done, especially for long-lived radionuclides that have only recently been made easier to analyze by AMS. Some detailed studies of many cosmogenic nuclides in several samples from a meteorite also are being done, especially for meteorites with a wide range of preatmospheric sizes (e.g., [29]).

3.4 Theoretical calculations

The production rates for several cosmogenic nuclides have been calculated by computer codes that use only some basic nuclear data, such as scattering, capture, and reaction cross sections, and are not normalized to observations. As discussed above in the section on SCR calculations, Monte Carlo calculations have been done for cosmogenic nuclides in lunar samples by Armstrong and Alsmiller [10]. For the GCR, they published production-rate-versus-depth profiles for ^{26}Al and ^{22}Na and the neutron fluxes as a function of depth. The calculations followed the neutrons until they reached thermal (technically < 0.4 eV) energies. The calculated ^{26}Al and ^{22}Na production rates and the thermal-neutron fluxes were consistent with lunar measurements [10]. As with the SCR Monte Carlo calculations, these GCR calculations [10] show the basic processes involved but are of limited use in detailed studies of cosmogenic nuclides in lunar samples.

The one case where essentially pure theoretical calculations have been very useful in studies of cosmogenic nuclides is for nuclide production by the capture of neutrons with low (thermal and epithermal) energies. The radionuclides that can be made in appreciable amounts by neutron-capture reactions include 5.7-year ^{60}Co , 75-ka ^{59}Ni , 100-ka ^{41}Ca , and (occasionally) 300-ka ^{36}Cl . Several results for such neutron-transport calculations have been published for the moon [10, 11] and meteorites (e.g., [30]). These calculations start with the chemical composition and the corresponding nuclear properties of the object and with the fluxes of energetic ($\sim 1 - 10$ MeV) neutrons made in the GCR-particle cascade as a function of energy and depth. The equilibrium distribution of neutrons is then calculated. The production profile in Fig. 4 for ^{60}Co in the moon was calculated by a neutron-transport code [11]. Although ^{36}Cl is usually produced in much greater quantities by spallation reactions than by neutron-capture reactions [30], the latter source can be important in big objects with samples having high chlorine contents, as was the case for the Apollo 15 drill core [31].

As shown in Fig. 4, the production-rate-versus-depth profiles for neutron-capture products are very different than those for nuclides made by higher-energy reactions, being very low at the surface and peaking with much higher production rates at fairly great depths. In meteorites, production of nuclides by thermal neutrons is negligible until the preatmospheric radius is greater than ~ 75 g cm $^{-2}$, and the presence of neutron-capture-produced nuclides means that the meteorite was large in space [30]. These big changes in the production rates of neutron-capture-produced nuclides allow us to sometimes use such nuclides to infer a sample's preatmospheric depth. Unfortunately, neutron-capture-produced nuclides have seldom been

used in meteorite studies, mainly because the relatively short half-life of readily measurable ^{60}Co and the weak radiations emitted by the decay of long-lived ^{59}Ni and ^{41}Ca . If AMS techniques can be developed for the low ratios of these long-lived radionuclides to the stable isotopes of the same element, then these products with their very different production profiles could be used to help unfold the exposure histories of meteorites.

3.5 Inferred particle fluxes and reaction cross sections

This approach to calculating production rates is similar to the main one used for SCR-produced nuclides, except that the particle fluxes are not calculated by a simple relation like ionization energy loss but must be inferred by other means. When Arnold, Honda, and Lal [5] used this approach in predicting the production rates of cosmogenic nuclides in iron meteorites, they used particle fluxes derived from several sources. For the primary GCR particles, they used available experimental data. For secondary particles above 100 MeV, they used data from nuclear emulsions that had been exposed at high altitude and other measurements in the earth's atmosphere. The flux as a function of energy used for all particles with energies above 100 MeV was

$$\frac{dJ}{dE} = c(\alpha + E)^{-2.5}, \quad (3)$$

where α was 1.0, 0.4, and 0.2 GeV for the primary GCR particles and for depths of 10 and 100 g cm $^{-2}$, respectively, and c was normalized to experimental cosmic-ray fluxes above 3 GeV. Below 100 MeV, they considered only neutrons and used a spectrum based on measurements made in the earth's atmosphere. This spectrum was normalized to agree with (3) at 100 MeV. The cross sections above 100 MeV were based on spallation systematics. The cross sections below 100 MeV assumed that the incident particle was a neutron. Experimental cross sections were used whenever possible. The production rates were calculated by integrating over energy the product of the fluxes and cross sections and generally reproduced the trends for cosmogenic nuclides measured in the Aroos iron meteorite [5].

The same approach was used by Reedy and Arnold [8] for the moon, and they used many of the features of the above model. The main differences were that the flux above 100 MeV, (3), was allowed to continuously vary with depth in the moon, and the flux shape below 100 MeV was made a function of α . The Reedy-Arnold spectral shape below 100 MeV was based on the one from ref. [5] for deep in the moon and on activities of cosmogenic nuclides in lunar samples and theoretical calculations [10] for near the lunar

surface. The GCR-particle fluxes above 1 GeV as a function of depth in the moon were calculated from known interaction lengths and parameters for the incident GCR protons and alpha particles. This model thus has only one free parameter, α , and a curve for α versus depth was derived from several sources. Many excitation functions were constructed using experimental cross sections or estimations based on nuclear systematics. This model only considers particles with energies down to ~ 1 MeV and can not be used for calculating neutron-capture rates. It has reproduced the activity-versus-depth profiles measured in lunar samples quite well, although occasionally absolute values are off by up to $\sim 40\%$, such as for ^{53}Mn . It is believed that these few poorly calculated absolute production rates are a consequence of using incorrect cross sections, especially using proton-induced ones at energies below ~ 100 MeV where neutron-induced cross sections should be used.

The flexibility of this approach has been illustrated by the fact that new production rates are frequently being calculated using new cross sections. Beside the problems with the lack of cross sections for neutron-induced reactions, the only other deficiency has been the model's failure below depths of $\sim 350 \text{ g cm}^{-2}$. Down to that depth (few lunar cores go much deeper), the Reedy-Arnold model [8] has worked quite well, especially if good measurements can be found to normalize the calculated production profiles at one depth. The availability of new cross sections have almost always resulted in better agreement between the calculated production rates and measurements of cosmogenic nuclides in lunar samples and meteorites (for example, ^{10}Be in St. Severin [25]). There are still many reactions for which experimental cross sections are needed, both for protons with energies above ~ 100 MeV and for neutrons at lower energies. The model has been surprising successful even though it only has one free parameter, α .

The Reedy-Arnold model was extended to meteorites by ref. [12]. Curves for α as a function of depth were derived for two meteorites with different preatmospheric radii (St. Severin and Jilin), and an expression was developed that could calculate α for any radius and depth [12]. The ^{26}Al production rates shown in Fig. 5 for L chondrites were calculated with this model. The model fails for very large (radii more than $\sim 200 \text{ g cm}^{-2}$) meteorites, most likely because the derivation of α for such radii was not correct. It also calculates production rates for fairly small meteorites (radii less than $\sim 30 \text{ g cm}^{-2}$) that are too high, probably because the model has too many neutrons for such small objects. An improved model for fairly small meteorites will probably need to consider protons and neutrons separately, as the flux of neutrons increases fast with radius in such small bodies. As mentioned above, this model does work

well for very small objects, such as those that possibly are the parent bodies for cosmic spherules [17], because secondary-particle fluxes are negligible and only the well-known primary-GCR-particle fluxes and proton-induced cross sections are needed [18].

3.6 Simulation irradiations at accelerators

Many thick targets have been irradiated with high-energy particles at accelerators to simulate the cosmic-ray bombardment of extraterrestrial matter (see review of thick target experiments in ref. [2]). In these irradiations the distributions of various products are measured at several locations inside the target. These product distributions varied with the nature of the reactions producing them. For example, in Honda's bombardment of a thick iron target with 3-GeV protons [6], the activities of high-energy products, like ^{22}Na , decrease rapidly with depth and had very little lateral spread, whereas radionuclides made by low-energy reactions, such as ^{55}Fe , built up at first with depth and the lateral spread was broad. These radioactivities map the distributions of the primary and secondary cosmic-ray particles in large objects in space. Honda developed relations to convert these thick-target measurements into production profiles expected in iron meteorites. A more detailed set of expressions was developed by Kohman and Bender [7] for converting thick-target results into production profiles in iron meteorites. Their production profiles for fairly low energy reactions look similar to that for ^{26}Al in Fig. 5. The production rates from these models for high-energy products show very little increase near the surface (somewhat like the ^{10}Be curve in Fig. 4).

The production profiles inferred from thick-target irradiations were among the first predicted for meteorites and are still useful in interpreting cosmogenic-nuclide measurements. However, they are limited to nuclides that have been measured in the bombarded thick targets or for other nuclides made by similar reactions (such as using ^{54}Mn to model production of ^{53}Mn). Most thick targets were iron, and only a few groups irradiated stony targets (see compilation of thick-target experiments in ref. [2]). Some of the production profiles from these thick-target models are not very good, such as those for large stony objects reported by ref. [32] (as discussed by ref. [30]). Some of the targets were too small to contain the entire cascade of secondary particles, and thus the particle distribution inside the target was distorted [33]. Almost all of these thick-target bombardments used high-energy protons [2]; however, $\sim 12\%$ of the particles in the GCR are alpha particles [26], which interact differently in thick targets [33]. As with most

results obtained prior to AMS, certain long-lived radionuclides, for example ^{10}Be , were seldom measured in these thick-target experiments.

Another limitation of most thick-target experiments is that they are not applicable to fairly small objects. The full cascade of secondary particles usually is generated in a thick target, but only the initial stages of such cascades will have developed in small objects. Thus the models like that of Kohman and Bender [7] are probably not valid for such small objects. Recently, several isotopic irradiations with 600-MeV protons have been done of small stony spheres with radii from 5 to 25 cm [34]. Even in a sphere of radius 5 cm, there are a number of secondary neutrons, and the measurements are in fairly good agreement with Monte Carlo calculations [34]. Additional isotropic irradiations of small “thick targets” are being planned, including spheres with several different chemical compositions and possibly of an ellipsoidal target.

4. Summary and conclusions

As surveyed above, there are many methods used to predict the production rates of cosmogenic nuclides by cosmic-ray particles in extraterrestrial matter. All models have their strengths and their weaknesses. Most approaches work well, although the agreement with experimental measurements of cosmogenic nuclides in real samples is the ultimate test of any model. Only a very few models are so theoretical that they don't need the results of measurements of naturally or artificially irradiated samples. These measurements, especially of the long-lived radionuclides now being readily measured by AMS, are very important in improving our predictions of production rates. As additional radionuclides, such as ^{41}Ca and ^{59}Ni , become measurable by AMS, studies of those nuclides in both artificially irradiated targets and in extraterrestrial materials will be needed to help in predicting their production rates. Some of the methods and data used for extraterrestrial samples can be applied to cosmogenic radionuclides made in the earth's atmosphere and surface. For example, cross sections for the production of nuclides like ^{26}Al and ^{10}Be can be used with fluxes of terrestrial neutrons to calculate production rates.

For the nuclides made by the protons and alpha particles in the solar cosmic rays, the most important need is for thin-target cross sections at several energies, especially near the energy of the reaction threshold. Cross sections are needed for several reactions, such as for ^{10}Be and ^{14}C from oxygen. Additional measurements of long-lived radionuclides in lunar samples also should be done to confirm and extend existing studies of the fluxes of SCR particles in the past [14]. Very little work has been done on the fluxes of solar

alpha particles in the past using ^{59}Ni in the top few millimeters of lunar samples. Such studies probably would be one of the first projects to do when the AMS techniques for Ni are fairly well established because the nickel contents of lunar samples are low, so the ^{59}Ni /stable-Ni ratios won't be too low.

For nuclides produced by the galactic cosmic rays, many types of measurements are needed. Systematic studies of cosmogenic-nuclide concentrations in extraterrestrial samples are important in predicting production rates. Most models need such measurements to determine their parameters or for normalizations of the absolute activities. Many cross sections are needed for the production of these nuclides by energetic protons and especially neutrons with energies up to ~ 100 MeV (and more in several cases, such as ^{10}Be [25]). Laboratory simulations using thick targets or isotropically irradiated targets, especially for long-lived radionuclides, are still valuable in helping to predict production rates. These simulations are useful in testing Monte Carlo calculations, such as was done for the 5-cm sphere [34]. These Monte Carlo calculations, such as those with the High Energy Transport Code used by ref. [34], are being improved, and soon incident high-energy alpha particles will be included in such calculations.

A wide range of problems can be studied with these predicted production rates and measurements of long-lived cosmogenic radionuclides in extraterrestrial samples. The lengths of time that the thousands of meteorites now being found in Antarctica have been on or in the ice, their terrestrial ages, are important for many studies of these interesting and often unique (for example, those from the moon) meteorites. Better exposure ages are needed to help determine the origins of meteorites. Especially important will be the ability to distinguish between simple irradiation histories and those complex irradiations that occurred both before and after the meteorite was broken apart in space by a collision. Such studies of the irradiation histories of meteorites will help to determine where the parent bodies were in the solar system. More studies of the cosmic spherules found in deep-sea sediments [17] will help to determine the origins of these interesting objects, such as whether they were small objects in space or are just fragments of larger meteorites that broke apart while passing through the earth's atmosphere. Some meteorites or cosmic spherules may have had unusual orbits around the sun, such as small perihelions or high inclinations (a region of the solar system that has not been explored yet by satellites).

One great power of AMS is its ability to measure the concentrations of long-lived radionuclides in small samples, which allows us to measure many different cosmogenic nuclides in a sample. The more nuclides and other cosmic-ray products (such as tracks of heavy cosmic-ray nuclei) measured, the better our ability

to interpret all aspects of the sample's history [1]. For example, short-lived radionuclides can establish the production rates if the object had a short or complex irradiation history. Radionuclides like ^{81}Kr , ^{36}Cl , and ^{26}Al can be used to determine terrestrial ages of Antarctic meteorites. Long-lived radionuclides can occasionally show that the object had an unusual exposure history. Extending the range of half-lives available, such as with 16-Ma ^{129}I , 32-Ma ^{92g}Nb , or 103-Ma ^{146}Sm , would be very useful in studies of most extraterrestrial materials, as these half-lives are similar to many exposure ages. Much of the more exciting work that will be done during the next few years with extraterrestrial materials will involve measurements of long-lived radionuclides done with AMS.

Acknowledgements

This paper benefited from nearly countless past collaborations and discussions with many of my colleagues, especially Peter Englert, Jim Arnold, D. Lal, David Elmore, and Kuni Nishiizumi. This work was supported by NASA purchase order T-294M and done under the auspices of the U.S. Department of Energy.

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Figure captions

Fig. 1. Cross sections as a function of energy for the production of 0.7-Ma ^{26}Al by proton reactions with aluminum and silicon. Symbols are the measured cross sections by the indicated authors. (The Bordeaux group's measurements are reported in refs. [23, 24] and references therein. The other published results are in refs. [20, 21, 22].) Only some of the uncertainties for the measurements are shown. The two curves are the evaluated cross sections used for the production rates calculated with the models of refs. [8, 12].

Fig. 2. The rates calculated for the solar-proton production of 0.7-Ma ^{26}Al as a function of preatmospheric depth inside spherical meteoroids of various radii. A solar-proton spectrum with $R_p = 100$ MV and a flux above 10 MeV of 70 protons $\text{cm}^{-2} \text{s}^{-1}$ were used. The chemical composition was that of an L chondrite. The symbols O and X on the left axis are the production rates calculated for a very small object of this composition by solar protons and GCR protons, respectively. The symbol (GCR) is the GCR production rate of ^{26}Al observed inside typical L chondrites, which have radii greater than $\sim 40 \text{ g cm}^{-2}$.

Fig. 3. The rates calculated for the solar-proton production of 1.6-Ma ^{10}Be as a function of depth in lunar rock 68815 for several exponential-rigidity spectral shapes. For all spectral shapes, the omnidirectional flux above 10 MeV was 70 protons $\text{cm}^{-2} \text{s}^{-1}$. Also shown are the ^{10}Be production rates of ref. [8] calculated using an earlier set of cross sections; they are a factor of 2 higher than the present calculated production rates done with the ^{10}Be production cross sections given in ref. [25]. Only very hard spectra (high values of R_p) yield significant amounts of ^{10}Be compared to the $\sim 10 \text{ atoms min}^{-1} \text{kg}^{-1}$ produced by GCR particles at these depths.

Fig. 4. The rates calculated for the GCR production of four radionuclides as a function of depth in the moon. These production profiles are examples for cosmogenic nuclides made mainly by high-energy reactions (^{10}Be), both low- and high-energy reactions (^{26}Al), low-energy (~ 10 MeV) neutron reactions (^{39}Ar), and by the capture of thermal neutrons (^{60}Co).

Fig. 5. The rates calculated for the GCR production of 0.7-Ma ^{26}Al as a function of preatmospheric depth inside spherical meteoroids of various radii [12]. The chemical composition was that of an L chondrite. The symbol X on the left axis is the production rate calculated for a very small object of this composition by GCR protons [18].

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5