IN SITU–PRODUCED COSMOGENIC ISOTOPES IN TERRESTRIAL ROCKS

D. Lal

Scripps Institution of Oceanography, Geological Research Division,
La Jolla, California 92093 and Physical Research Laboratory,
Ahmedabad 380009, India

> *There is something fascinating about science. One gets such wholesale returns of conjecture out of such a trifling investment of fact.*
>
> Mark Twain, *Life on the Mississippi*

In the late forties, Libby and his collaborators detected the naturally occurring radioactive isotope ^{14}C, produced by cosmic radiation in the Earth's atmosphere (Libby 1946, Anderson et al 1947). This marked the onset of the search for cosmic-ray-produced (cosmogenic) isotopic changes in terrestrial and extraterrestrial samples and in the cosmic rays themselves. During the four decades since Libby's discovery, some four dozen cosmogenic stable and radioactive isotopes in extraterrestrial samples (Reedy et al 1983) and two dozen in terrestrial samples (Lal & Peters 1967) have been discovered. These isotopes have found applications in cosmic-ray physics (in the study of acceleration and propagation of galactic and solar cosmic rays), in solar system astrophysics (in determining the evolutionary history of meteorites, the lunar surface, and interplanetary dust particles), and in the Earth sciences (in archaeology, meteorology, glaciology and oceanography). For reviews on these topics, reference is made to Simpson (1983); Reedy et al (1983); and Lal & Peters (1967), Oeschger et al (1970), and Faure (1986), respectively. The process of discovery continues, propelled by advances in the techniques used to measure these isotopes. Even now, examples of new isotopic effects are being discovered, and new applications are being made of the various isotopic effects. When one looks at the history of the development of the terrestrial cosmogenic field, one observes that it grew in waves: After an idea is implemented, the field

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matures soon thereafter, followed by a quiescent period until the next technical advance produces a quantum jump in detection capability.

But why is technology the limiting factor? And wherein lie the potentialities of the fields of cosmic-ray physics and of cosmogenic isotopes in the solar system? Cosmic rays were discovered 75 years ago by Victor F. Hess during a series of 10 manned balloon flights, made during day and night, up to altitudes of 5350 m. The ionization chambers used by him established that the radiation was of extraterrestrial origin. H. C. van de Hulst once remarked that the word cosmic rays, at the time the name was given, was a curious double confession of ignorance—cosmic meaning that we do not know from where they were coming and ray meaning that we did not know what they were! The rapid development of a variety of cosmic-ray detectors during the 1930s and 1940s removed much of the mystery of the "rays," but we are still struggling to discover their origin. New techniques that have increased our knowledge of their composition and energy spectrum have also improved our understanding of galactic astronomy. Carl Anderson once noted that when we measure something that could not be measured before, or learn how to measure it more accurately, we almost always find out something interesting. The continuing discoveries in the field of cosmic rays are apt illustrations of this hypothesis.

The interconnection between cosmic rays and geophysics was established with the discovery of cosmogenic ^{14}C on Earth. Improvements in techniques added new isotopes to the list of useful tracers in Earth sciences, and similar developments occurred in the study of meteorites. However, in the latter case, the improvements came more rapidly because of the generally higher nuclear reaction rates and longer effective exposure times.

Implicit in the above is the single fact that makes cosmic rays a very powerful tool in astronomy, astrophysics, and geophysics. During its lifetime, the Galaxy has been continuously filled with high-energy cosmic rays, up to energies of $\sim 10^{20}$ eV. The long history of cosmic-ray effects, going back to close to the time of formation of the Galaxy, and the significant depths in matter down to which cosmic-ray effects can be investigated give the field of cosmogenic isotopes its versatility. In the case of extraterrestrial objects without an appreciable atmosphere, the nuclear effects of cosmic rays are discernible up to depths of $\sim 10^3$ g cm^{-2}. On Earth, the effects extend deeper, up to 10^6 g cm^{-2}, owing to decay of charged pi-mesons in the Earth's atmosphere, giving rise to the penetrating muons. The penetrating component has been used as cosmic "X rays" in a variety of applications: for example, to determine the geometry of pyramids or, in civil engineering, to determine the overburden in different directions. We

discuss in this article the intensity of nuclear disintegrations caused by this component in rocks at depths down to 10^4 g cm^{-2} below sea level.

Before getting on with the topic of the present paper, it seems appropriate to comment briefly on another aspect of the process of discovery. A generally valid statement that can be made here is that no ideas were ever entirely new! They all had been given an expression to earlier. Ideas evolve; most ideas do not gel when they first appear on the scene. Even feasible ones do not catch on immediately. Let us cite some examples relevant to the subject of this review:

1. The possibility of production of ^{14}C in the Earth's atmosphere by cosmic-ray neutrons was first suggested by Montgomery & Montgomery (1939), soon after discovery of cosmic-ray neutrons (Locker 1933, Rumbaugh & Locker 1936). Korff (1940) pointed out that most of the slow cosmic-ray neutrons would lead to production of ^{14}C because of the rather high thermal neutron cross section for the ^{14}N $(n, p)^{14}$C reaction. The search for radiocarbon in nature was made by Libby and his collaborators in 1946. This soon culminated in the detection of ^{14}C in nature (Anderson et al 1947). Soon thereafter, Arnold & Libby (1949) demonstrated the feasibility of using terrestrial cosmogenic ^{14}C for dating archaeological samples.

2. The production of isotopes by nuclear interactions of cosmic rays was first discussed by Grosse (1934), five years prior to the realization of cosmic-ray neutron production of ^{14}C on Earth. For an excellent historical account of development of the cosmogenic isotope field, see Faure (1986).

3. The accelerator mass spectrometry (AMS) technique as we know it was developed a decade ago; it has lowered the detection limit for several long-lived isotopes (e.g. ^{14}C, ^{36}Cl, ^{10}Be, ^{26}Al) by a factor of a million. The first application of this technique had, however, been made earlier by Alvarez to determine if ^{3}He or ^{3}H was stable (Alvarez & Cornog 1939). His colleague, Richard Muller, elaborated on this idea and made the first proposal for AMS studies of ^{14}C (Muller 1977).

4. Studies of ^{10}Be, ^{26}Al, ^{36}Cl, ^{3}He, and other isotopes produced by cosmic rays in rocks have now opened up the scope of terrestrial cosmogenic isotopes to studies of geophysical processes during the Pleistocene; these developments are discussed later in this paper. Their usefulness had, however, been pointed out long ago, and attempts had been made to detect them (see, for example, Davis & Schaeffer 1955, Tanaka et al 1968, Takagi et al 1974, Srinivasan 1976).

5. Only recently was it realized that cosmic rays are the principal agency for producing Li, Be, and B in our Galaxy, after two decades of studies of cosmic-ray spallations in nature. The "galactogenesis" hypothesis was first

proposed by Reeves et al (1970), following a suggestion by B. Peters (Audouze & Vauclair 1980, Ch. 7). For a recent treatment of the subject, see Walker et al (1985).

An idea that is feasible often gets stalled. There was no great urgency to develop the AMS technique during the 1950s and 1960s because the counting techniques available then appeared quite adequate! The beta activity of small solid samples could easily be measured at levels of disintegrations per hour (Lal & Schink 1960). The gas proportional counting technique had then been considerably improved for ^{14}C counting (Oeschger et al 1972). For samples of nominally 1 cm^3 STP volume, Davis (1968) had demonstrated measurement sensitivity of 1 disintegration per day for ^{37}Ar. A number of ingenious low-level counting systems, employing coincidence-anticoincidence schemes, were available for X-ray and gamma-ray emitters. (For a comprehensive review of the state-of-the-art counting techniques in the 1970s, sec Oeschger & Wahlen 1975.)

The principal motivation to develop the AMS technique was technological, possibly to use particle accelerators that had otherwise outlived their usefulness! As mentioned earlier, the idea of using in situ isotopes preceded technological developments. When the AMS method worked for ^{14}C (Nelson et al 1977, Bennett et al 1977), there was then sufficient motivation to develop it for ^{10}Be, ^{26}Al, etc, and successes in their detection and new applications led to rapid flowering of the terrestrial "atmospheric" and "in situ" cosmogenic isotope studies using the AMS technique. The initial developments occurred· contemporaneously in Canada, the USA, and France, and the technique spread in the 1980s to Israel, Switzerland, and Italy. For a comprehensive discussion of early AMS studies, reference is made to Hedges (1979), Litherland (1984), Raisbeck & Yiou (1984), Brown (1984), Suter et al (1984), and other articles in the *Proceedings of the Third International Symposium on AMS* (Wölfli et al 1984). For recent studies, see Lal (1987a), Elmore (1987), and Elmore & Phillips (1987).

IN SITU VS THE ATMOSPHERIC COSMOGENIC ISOTOPES

The AMS technique has widened the scope of applications of both "atmospheric" and "in situ" cosmogenic isotopes produced on the Earth. The principal applications of the former are given in Table 1. We shall from here on confine our attention to the latter. The atmospheric isotopes in fact often become a hindrance to the study of the in situ isotopes. The former are transported through the upper layers of the Earth by geophysical/ geochemical processes and in some cases (e.g. ^{10}Be) can be incorpo-

Table 1 Principal "atmospheric" cosmic-ray-produced isotopes and their applications[a]

^{14}C	$^{22}Na, ^{35}S, ^7Be, ^{33}P$ $^{32}P, ^{28}Mg, ^{24}Na$ $^{38}S, ^{31}Si, ^{18}F$ $^{39}Cl, ^{38}Cl, ^{34m}Cl$	$^{10}Be, ^{36}Cl, ^{14}C$ $^{32}Si, ^{39}Ar, ^3H$	$^{10}Be, ^{81}Kr, ^{14}C$ $^{39}Ar, ^{32}Si, ^{36}Cl$
Archaeology and paleobotany	Meteorology, cloud physics, atmospheric structure, and large-scale air circulation and scavenging	Hydrology and glaciology. Chronology of ground waters, lacustrine sediments, and glaciers	Oceanography and marine sediment record. Air-sea exchange, geochemical and biological cycles, chronology of sediments, paleomagnetic reversal records, cosmic-ray prehistory

[a] In some cases, an appreciable "artificial" production of radionuclides has significantly changed the global inventories (e.g. ^3H, ^{14}C, and ^{36}Cl).

rated into the rocks to overwhelm the amount that is directly produced in situ. Cosmic-ray fluxes in the troposphere are lower than the mean fluxes in the stratosphere by factors of up to 500. Consequently, we confront two problems: (a) low isotope concentrations, and (b) the possibility of an appreciable amount of the isotope produced in the atmosphere being incorporated into the rock by chemical or physical processes. This effect is clearly applicable only for those isotopes that can be scavenged from the atmosphere by wet precipitations. For convenience, the "atmospheric" component is termed the "garden variety" (Nishiizumi et al 1986).

Special precautions have to be taken to insure that an in situ measurement is not affected by the garden variety isotope (Lal & Arnold 1985). Further, one has to work at a high level of sensitivity to insure that sources of contamination are minimized and that there exist no other unrecognized (nuclear) sources of production of the isotope. For example, ^{10}Be can be produced in situ in rocks by radiogenic nuclear reactions (Middleton & Klein 1987); the term radiogenic refers to as due to alpha particles from nuclear transmutations or from their interactions, e.g. neutrons. The carrier beryllium salt obtained commercially has been found to contain an appreciable amount of ^{10}Be (Middleton et al 1984).

Terrestrial in situ cosmogenic studies began two decades after the cosmogenic meteorite (in situ) studies, primarily because the former studies are more difficult. The terrestrial cosmogenic studies require sensitivities 3–4 orders of magnitude higher. But there are fruits to be gathered from such studies because one learns about geophysical events that cannot be studied by other means! And the principal promise of the cosmogenic terrestrial in situ studies is that they will allow us to examine recent Earth history—the geological and geophysical events during the Pleistocene.

IN SITU ISOTOPES DETECTED IN ROCKS SO FAR

We have listed in Table 2 the cosmogenic in situ isotopes detected in rocks to date. The number that can be detected is much greater than the number on this list, and it is also much more than the number of isotopes produced in the atmosphere because of the presence of higher mass number target elements in rocks. A list of often-studied isotopes with half-lives exceeding two weeks is presented in Table 3 (Lal 1987a). Even considering the small terrestrial in situ isotope production rates in rocks exposed at sea level and

Table 2   In situ terrestrial cosmogenic isotopes detected so far

Isotope	Half-life (yr)	Technique used[a]	Reference(s)[b]
Radioactive			
^{10}Be	1.6×10^6	AMS	1, 2, 3,
^{26}Al	7.1×10^5	AMS	1, 2, 3, 4, 5
^{36}Cl	3.0×10^5	C, AMS	6, 7, 8
^{37}Ar	35 days	C	9, 10
^{39}Ar	269	C	9, 10
^{41}Ca	1.0×10^5	AMS	11, 12
^{92}Nb, ^{94}Nb	$3.6 \times 10^7, 2.0 \times 10^4$	C	13, 14
^{129}I	1.6×10^5	C, AMS	15, 16
Stable			
^{3}He	Stable	MS	17, 18
^{21}Ne	Stable	MS	19
^{126}Xe	Stable	MS	20

[a] Abbreviations: C = counting, MS = mass spectrometry, AMS = accelerator mass spectrometry.

[b] References: (1) Jha & Lal (1982), (2) Yiou et al (1984), (3) Nishiizumi et al (1986), (4) Tanaka et al (1968), (5) Hampel et al (1975), (6) Davis & Schaeffer (1955), (7) Kubik et al (1984), (8) Phillips et al (1986), (9) R. Davis (private communication, 1984), (10) Lal et al (1986), (11) Raisbeck & Yiou (1979), (12) Henning et al (1987), (13) Apt et al (1974), (14) Clayton & Morgan (1977), (15) Takagi et al (1974), (16) Fehn et al (1986), (17) Craig & Poreda (1986), (18) Kurz (1986a), (19) Marti & Craig (1987), (20) Srinivasan (1976).

Table 3 Often-studied cosmogenic nuclides with half-lives exceeding two weeks

Isotope(s)	Half-life (yr) (S = stable)	Main targets[a]
^{3}H[b]	12.3	O, Mg, Si, Fe (N, O)
^{3}He, ^{4}Hc	S	O, Mg, Si, Fe (N, O)
^{7}Be[b]	53 days	O, Mg, Si, Fe (N, O)
^{10}Be[b]	1.6×10^{6}	O, Mg, Si, Fe (N, O)
^{14}C[b]	5730	O, Mg, Si, Fe (N)
^{20}Ne, ^{21}Ne, ^{22}Ne	S	Mg, Al, Si, Fe
^{22}Na[b]	2.6	Mg, Al, Si, Fe (Ar)
^{26}Al	7.1×10^{5}	Si, Al, Fe (Ar)
^{32}Si[b]	100–200	(Ar)
^{35}S[b]	87 days	Fe, Ca, K, Cl (Ar)
^{36}Cl[b]	3.0×10^{5}	Fe, Ca, K, Cl (Ar)
^{36}Ar, ^{38}Ar	S	Fe, Ca, K
^{37}Ar[b]	35 days	Fe, Ca, K (Ar)
^{39}Ar[b]	269	Fe, Ca, K (Ar)
^{40}K	1.3×10^{9}	Fe
^{39}K, ^{41}K	S	Fe
^{41}Ca	1.0×10^{5}	Ca, Fe
^{46}Sc	84 days	Fe
^{48}V	16 days	Fe, Ti
^{53}Mn	3.7×10^{6}	Fe
^{54}Mn	312 days	Fe
^{55}Fe	2.7	Fe
^{56}Co	79 days	Fe
^{59}Ni	7.6×10^{4}	Ni, Fe
^{60}Fe	1.5×10^{6}	Ni
^{60}Co	5.27	Co, Ni
^{81}Kr[b]	2.1×10^{5}	Rb, Sr, Zr (Kr)
^{78}Kr, ^{80}Kr, ^{82}Kr, ^{83}Kr	S	Rb, Sr, Zr
^{129}I[b]	1.6×10^{7}	Te, Ba, La, Ce (Xe)
$^{124-132}$Xe	S	Te, Ba, La, Ce, I

[a] Elements from which most production occurs; those in parentheses are for the Earth's atmosphere.
[b] Atmospheric cosmogenic isotopes.

at depths underground ($<10^{4}$ g cm^{-2}), most of these isotopes can be measured, and it is primarily the promise of the applications that decides whether or not attempts are being made today to look for an isotope.

IN SITU ISOTOPE SOURCE FUNCTIONS

In order to fully exploit the potential of cosmogenic in situ isotopes, it is necessary to understand their production mechanisms and source func-

tions and their expected time variability in relation to terrestrial and solar system influences. The problem of propagation of the cosmic rays through the Earth's atmosphere was dealt with in great detail earlier (Lal & Peters 1967) to estimate the source strengths of cosmic-ray-produced isotopes in the atmosphere. The composition of the secondary cosmic rays also undergoes a continuous change as one goes to deeper depths in the atmosphere. This is illustrated in Table 4. In the troposphere, most nuclear disintegrations are due to neutrons. At sea level, negative muon captures become significant. At depths underground below sea level, exceeding ~ 1-m rock equivalent, negative muon captures, fast muon disintegrations, and the secondary neutrons are responsible for the cosmogenic products. Reference is made to an earlier publication on the subject (Lal & Peters 1967) and a recent update on production rates underground (Lal 1987b). Table 4 provides a qualitative guide to the types of nuclear reactions that must be considered for estimating the production rate of an isotope from a target.

Isotope production, of course, depends on the composition of the target. In the case of the atmosphere, which can be regarded to be composed of only N, O, Ar, and Kr for the present purposes, life is simple. Atmospheric isotopes are produced primarily either by high-energy spallation or by neutron capture. The neutron capture reactions yield ^{14}C and ^{81}Kr; the remainder of the two dozen or so atmospheric isotopes (Table 1) are all produced by high-energy spallation, principally by neutrons. In the case of ^{39}Cl, an appreciable contribution arises from μ^- capture in ^{40}Ar (Winsberg 1956). For all practical purposes, one could ignore μ^- reactions for all other isotopes produced in the atmosphere. However, the situation is more complex when it comes to in situ production in rocks. Several isotopes can be produced effectively in spallation reactions as well as by thermal neutrons and slow muons, as discussed below.

ISOTOPE PRODUCTION BY FAST NUCLEONS, SLOW NEUTRONS, AND MUONS

The production rates of thermal and fast neutrons in the atmosphere have been estimated fairly accurately by Lal & Peters (1967), who based their rates largely on experimental measurements of (a) slow and fast neutrons in the atmosphere, (b) nuclear disintegrations in nuclear emulsions and cloud chambers, (c) energy spectra of protons and the penetrating component in the atmosphere, and (d) energy spectra of various charged particles at production in nuclear disintegrations at high altitudes in the atmosphere. These data allow one to determine the flux and energy spectrum of the nucleons and mu-mesons within the atmosphere at all latitudes

Table 4 Cosmic-ray primary and secondary particles in the atmosphere and at depths underground at 45° latitude (based on Lal & Peters 1967)

	Depth in the Atmosphere (g cm^{-2})				Depth Underground Below Sea-level (kg cm^{-2})		
	0	200	500	1030 (sea level)	1	50	500
Cosmic ray primary and secondary particles[a]	p	e, γ	e, γ	μ	$\mu^{\pm}$	$\mu^{\pm}$	$\mu^{\pm}$
	α	$\mu^{\pm}$	$\mu^{\pm}$	e, γ	n	n	n
	CNO	n	n	n	p	p	p
	Si, Fe	p	p	p			
Nuclear disintegrations[a]	p	n	n	n	μ_c	$\mu^{\pm}$	$\mu^{\pm}$
	α	p	μ_c	μ_c	$\mu^{\pm}$	μ_c	μ_c
	CNO	γ	p	p	n	n	n
	Si, Fe	μ	γ	$\mu^{\pm}$	p	p	p
		$\mu^{\pm}$	$\mu^{\pm}$				
Approximate Total Rates of Nuclear Disintegrations							
(1) /g s	10^{-2}	5×10^{-3}	10^{-3}	2×10^{-5}	10^{-6}	10^{-9}	5×10^{-12}
(2) /g yr	3×10^5	1.5×10^5	3×10^4	6×10^2	30	3×10^{-2}	2×10^{-4}

[a] Particles contributing to total cosmic-ray fluxes and nuclear disintegrations are shown in decreasing order of importance. For the present purposes, we define nuclear interactions as those involving nuclear excitation of >10 MeV. Symbols are p = proton, n = neutron, α = ^{4}He, e = electron, γ = photon, $\mu^{\pm}$ = positive and negative mu-mesons, μ_c = nuclear capture of negative mu-meson. C, N, O, Si, and Fe refer to these nuclei.

and altitudes. Direct measurements of isotope production rates in targets exposed at different altitudes (cf. Lal et al 1960, Rama & Honda 1961) and the observed fallout of long- and short-lived isotopes are in good agreement with the family of curves obtained for the rate of production of nuclear disintegrations in the atmosphere (Figure 1). The energy spectrum at production, based on observations of nuclear disintegrations in nuclear emulsions exposed at 68,000 feet over England, is shown in Figure 2 (Powell et al 1959). Based on these data, Rossi (1952) calculated the expected energy spectrum of neutrons and protons in the troposphere at 700 g cm^{-2} depth. He deduced the differential energy spectrum to be of the form

$$\frac{dN}{dE} = \text{constant} \times \frac{1}{(50+E)^2},$$ (1)

where E is the kinetic energy.
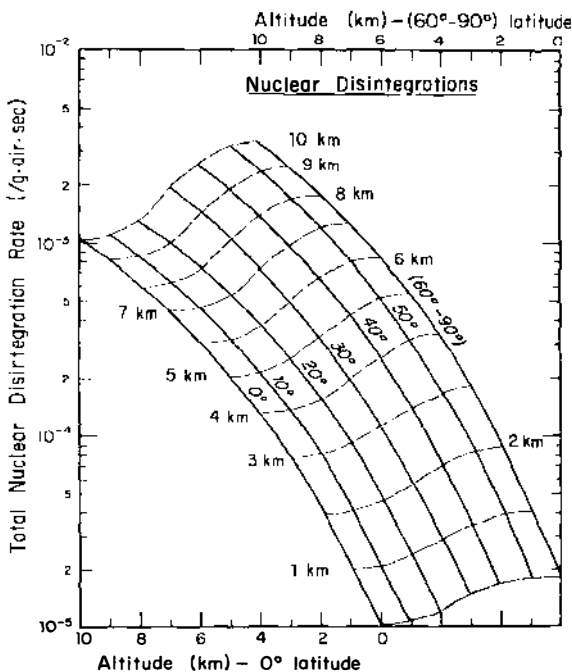
Figure 1   The total rate of nuclear disintegrations in the atmosphere (with energy release >40 MeV) plotted as a function of altitude and geomagnetic latitudes (0–90°, in steps of 10°). Curves for different latitudes have been successively displaced along the abscissa by 1 km (based on Lal & Peters 1967).
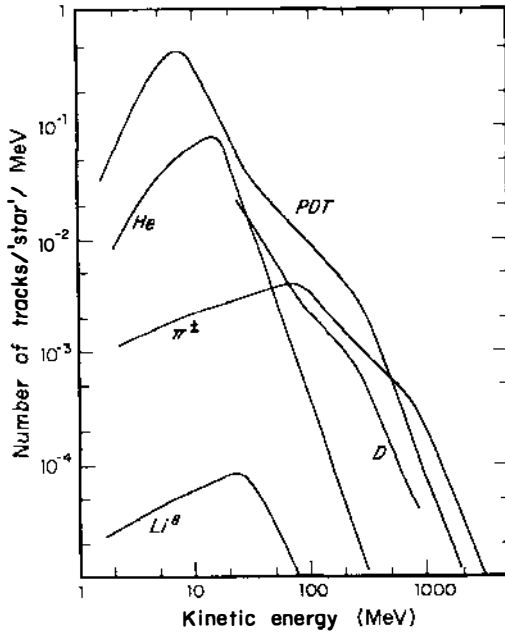
Figure 2   Kinetic energy spectrum of different classes of secondary particles emitted in nuclear disintegrations in "nuclear emulsions" exposed at 68,000 ft over England (based on Powell et al 1959). The curve labeled PDT refers to the total spectrum of protons, deuterons, and tritons emitted.

There exists a more direct way of obtaining the neutron energy spectrum in the troposphere—by combining data on the measured proton energy spectrum with the ratio of neutron- to proton-produced nuclear disintegrations of different sizes (Lal 1958). Using this technique, Lal (1958) has deduced the derived absolute neutron energy spectrum at $\geq 45°$ latitude, 680 g cm^{-2} depth (Figure 3). The spectrum is of the form

$$\frac{dN}{dE} = \text{constant} \times \frac{1}{(60+E)^{2.45}} \tag{2}$$

for $40 < E < 500$ MeV (Figure 3) and is in good agreement with the predictions of Rossi (1952), based on the energy spectrum of nucleons at production (Figure 2). Rossi's predicted proton energy spectrum also agrees well with the measured proton spectrum.

Further, since the relative size distributions of nuclear disintegrations of different sizes as observed in nuclear emulsions and cloud chambers have been measured in the atmosphere at several locations, it is possible to deduce energy spectra of neutrons at different locations in the atmo-
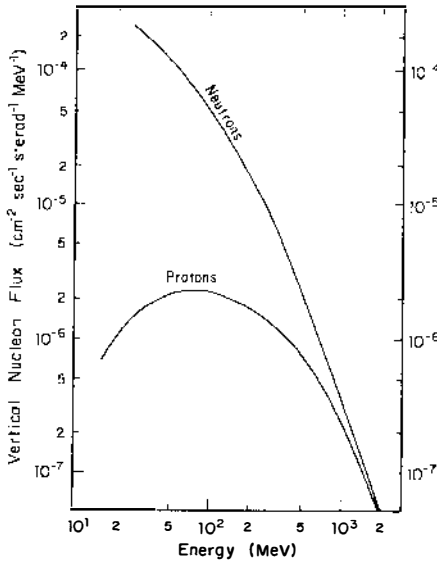
Figure 3 The omni-directional flux of neutrons and protons at latitude $\lambda = 50°$ and depth 680 g cm^{-2} (based on Lal 1958).

sphere from the known spectrum at any point (Lal 1958). Reference is made to a compilation of cosmic-ray data in the atmosphere (Allkofer & Grieder 1984). Thus we have a fair understanding of the behavior of the nucleonic and the penetrating muons in the atmosphere. The isotope production rates within a rock can therefore be calculated fairly reliably from the altitude-latitude isotope production curves given by Lal & Peters (1967), as long as the total sample shielding depth does not exceed ~ 1000 g cm^{-2}. (The total sample shielding depth is given by the sum of atmospheric pressure and the vertical shielding within the rock.) When the total shielding depth exceeds 10^3 g cm^{-2}, one has to carefully estimate relative neutron and muon fluxes. The energy spectrum of fast muons changes appreciably within the troposphere. The stopping muon flux at shallow depths underground (below sea level) does not show the usual exponential absorption, up to depths of about 2 kg cm^{-2}. Within the troposphere, a simple absorption behavior is found, and the stopping flux of negative muons, S_{μ^-}, can be well represented by the relation (Rossi 1948, Winsberg 1956, Lal 1958)

$$S_{\mu^-} (\text{g}^{-1}\text{s}^{-1}) = 5.1 \times 10^{-6}\exp(-X/L), \tag{3}$$

where X is the atmospheric depth (g cm^{-2}) and L is the absorption mean free path for slow muons. The value of L is determined (Rossi 1948, Lal 1958) to be 247 g cm^{-2} for $160 < X < 1030$ g cm^{-2}, the depth range of

validity of Equation (1). Equation (3) gives the rate of stopping of negative muons in matter; the rate of interaction would depend on the composition of the target. In light nuclei, an appreciable fraction of negative muons can undergo decay in orbit in the K-shell before being captured by the nucleus. The capture lifetimes have been measured in different elements. A great deal of work has been reported in the literature on the muonic capture probabilities in binary and more complex compounds (cf. Eckhause et al 1966). The capture rates in an element are expected to depend on the nature of the compound and thus deviate from the expected probabilities calculated on the basis of geometrical calculations. We note, however, that generally the magnitude of the deviation is small (von Egidy & Hartmann 1982). For the present purposes we therefore assume that μ^- capture in the K-shell of an atom depends on the geometric cross sections of the nucleus and the lifetime for capture of μ^- by the nucleus, τ_c. The mean lifetime of muon for decay, τ_d, is 2.2 μs. The probability that a muon captured by an atom will interact (i.e. be captured by the nucleus) rather than decay in the K-shell of the nucleus is then given by τ_m/τ_c, where τ_m (the mean lifetime of μ^- for decay or capture) is a measured quantity and is related to decay mean lifetime, τ_d, by the equation

$$(1/\tau_m) = (1/\tau_c) + (1/\tau_d). \tag{4}$$

In Figure 4 we show the measured values of τ_m for different elements as a function of the atomic number Z. The data are based on measurements by Eckhause et al (1966), who have also reviewed all earlier published data. The data in Figure 4 do not fall along a smooth curve. Capture probabilities are not direct functions of Z, but rather of the effective charge. Also, hyperfine effects are expected for some nuclei (cf Eckhause et al 1966, and references therein). Again, for our present purposes, we choose the smoothened capture probabilities; at a later date, when refinements are needed, we can reopen this issue. The rock-averaged μ^- capture probabilities are given in Table 5.

PRODUCTION RATES OF ISOTOPES

As discussed, there are three principal mechanisms by which the isotopes are produced in terrestrial rocks: (a) by high-energy spallation by nucleons, (b) by neutron capture reactions, and (c) by muon-induced nuclear disintegrations. The latter include nuclear capture of μ^- and coulombic interactions of fast muons. Within the troposphere, nuclear reactions of protons and alpha particles contribute to less than 10% of the nuclear reactions (Figure 3); photons are an order of magnitude less significant. Here, fast neutrons are responsible for most of the nuclear interactions
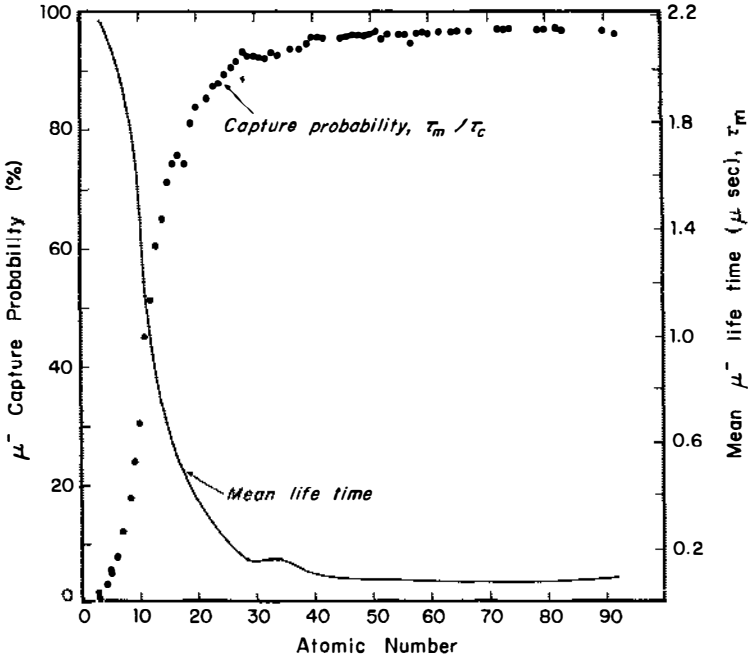
Figure 4   The calculated percentages of stopping negative muons captured by the nucleus
are plotted as a function of the atomic number Z. The values are based on the measured
mean lifetimes τ_m of negative mu-mesons brought to rest in different elements, shown by the
smooth line (data from Eckhause et al 1966). Note that in some cases, τ_m values have been
measured in isotopes (e.g. ^{6}Li, ^{7}Li, ^{10}B, and ^{11}B).

(Table 4). In nucleon or fast-muon-induced spallation reactions, the pro-
duct nuclei are lighter than or equal to the target. The product closest to
the target has either a lower A or Z value. In the case of negative muon
capture, the heaviest product has the same A value as the target but its Z
value is lower by unity. Neutron interactions can lead to a product with
an A value higher by unity than the target; this is in fact the most frequent
mode for thermal neutron capture reactions.

Thus, whereas generally a product can form from the target (Z, A) by
any of the three nuclear actions, in the troposphere (and at depths under-
ground) a product with $A = A+1$ can form only from neutron capture
because proton- and alpha-induced reactions are not important. Negative
mu-captures lead to low nuclear excitation (a few tens of MeV), since the
major part of the available energy (107 MeV) is liberated by the capture
of μ^- with a proton, leading to a nucleus of $(Z-1, A)$ and emission of a
neutrino. The neutrino takes away most of the energy. Fast muon inter-

Table 5 Some useful physical parameters for common rock types[a]

Rock type	(ρ) Assumed density (g cm^{-3})	(R_σ) Geometric cross section relative to air nuclei	(f_c) Fraction of stopping negative muons captured	(σ_m) Macroscopic thermal neutron absorption cross section (mole barns g^{-1})	(q_n) Radiogenic neutron production rate (10^{-6} cm^{-3} s^{-1})	$(\bar{\sigma})$ Mean thermal neutron cross section per atom (barns)
Granite	2.6	1.23	0.395	0.0120	1.6	0.61
Basalt	2.9	1.27	0.422	0.0084	0.44	0.76
Ultramafic	3.0	1.22	0.369	0.0073	5.7×10^{-4}	0.44
Shale	2.4	1.12	0.383	0.0163	0.76	0.73
Sandstone	2.3	1.22	0.378	0.0102	8.3×10^{-2}	0.47
Limestone	2.7	1.16	0.352	0.0067	▬	0.35

[a] Neutron parameters σ_m and q_n are based on calculations of Andrews & Kay (1982) and J. N. Andrews et al (preprint).

actions lead to much higher nuclear excitation (George 1952), depending on the kinetic energy of the muon.

The above considerations have to be taken into account in deciding which reactions would be principal contributors to a product in rocks. In Table 6 we have listed some isotopes for which thermal neutron and muon reactions are significant as examples of reactions that have been considered.

The isotope production rates $P \, (\text{g}^{-1} \, \text{s}^{-1})$ can be conveniently written as

$$P_{\text{sp}} = \sum_i q_i Y_i, \tag{5}$$

$$P_{\text{n}} = F_{\text{n}} \frac{\sigma_i n_i}{\sum_j \sigma_j n_j} = F_{\text{n}} \frac{\sigma_i n_i}{\sigma_{\text{m}}}, \tag{6}$$

$$P_{\mu_c^-} = \sum_i J_i(\mu^-) \cdot Y_i, \tag{7}$$

where the subscripts sp, n, and μ_c^- refer to the three reaction types. The rate of nuclear disintegration by fast nucleons or muons is designated as $q \, (\text{g}^{-1} \, \text{s}^{-1})$; $J(\mu^-)$ is the capture rate of μ^- mesons $(\text{g}^{-1} \, \text{s}^{-1})$; Y is the yield of the isotope per disintegration; σ is the neutron capture cross section (barns); and n is the concentration of target atoms (mole g^{-1}). The subscript i refers to the target element or the atomic species leading to the production of the isotope; j refers to different elements in the target. The macroscopic thermal neutron absorption cross-section, σ_{m}, is thus the summation of $\sigma_j n_j$ (mole barns g^{-1}). F_{n} is the production rate of neutrons in the nuclear disintegrations (n $\text{g}^{-1} \, \text{s}^{-1}$).

Table 6 Selected isotopes with an appreciable production in terrestrial rocks[a] due to neutron capture, mu-meson, and low-energy alpha particle reactions

| Isotope(s) | Thermal neutrons | | Capture of μ^- | Low-energy α-particles | |
	Target(s)	Reaction(s)	Principal target(s)	Target(s)	Reaction(s)
^{3}He	^{6}Li	(n, α)	—	^{11}B	(α, t)
^{10}Be	—	—	^{10}B, C, N, O	^{7}Li	(α, p)
^{14}C	^{14}N; ^{17}O	$(n, p); (n, \alpha)$	N, O	^{11}B	(α, p)
^{21}Ne, ^{22}Ne	—	—	Na, Mg, Al	^{17}O, ^{18}O, ^{19}F	(α, n)
^{26}Al	—	—	Si, S	^{23}Na; ^{25}Mg	$(\alpha, n); (\alpha, t)$
^{36}Cl	^{35}Cl; ^{39}K	$(n, \gamma); (n, \alpha)$	K, Ca, Sc	^{33}S	(α, p)
^{129}I	^{128}Te	(n, γ)	^{130}Te, Ba	—	—

[a] Contingent, of course, on the presence of the target element in the rock.

The rates of nuclear disintegrations produced by fast nucleons and fast muons have been determined for the atmosphere (Figure 1) and for rocks exposed below sea level (Lal 1987b). For rocks exposed within the troposphere, nucleon spallation rates can be estimated fairly accurately by scaling the rates with essentially the same absorption mean free path as in the atmosphere. This rate is latitude dependent, with the value changing from 200 g cm^{-2} at the equator to 150 g cm^{-2} at 50–90° latitude (Lal & Peters 1967).

Thermal neutron fluxes T_n (cm^{-2} s^{-1}) within a rock can be estimated quite accurately from (6) with the following relation for T_n:

$$F_n = P_{sp} \bar{Y}_n, \tag{8}$$

$$T_n = (F_n/\sigma_m), \tag{9}$$

where $\bar{Y}_n$ is the average yield of neutrons in the spallation of target elements in the rock, and ρ is the mean density of the rock. Estimated values of σ_m for different rock types are given in Table 5. We estimate that for a typical rock composition, 3.2 neutrons are produced; the corresponding yield for air nuclei is 2.9 (Lal 1987b). The value of $J_i(\mu^-)$ is given by

$$J_i(\mu^-) = S(\mu^-) \cdot f_i p_i, \tag{10}$$

where $S(\mu^-)$ is the μ^- stopping rate (g^{-1} s^{-1}), and f_i is the fraction stopped by the target nucleus i; this quantity can be deduced by approximating a geometric capture cross section. The value of p_i, the probability of capture of μ^- by the nucleus i, is given by direct measurements of τ_m (Figure 4). The mean fractions of stopping μ^- mesons that are captured, $f_c = \Sigma f_i p_i$, for different rock types are listed in Table 5.

By far the largest uncertainties in estimating isotope production rates at present are in evaluating the various yield factors in Equations (5), (7), and (8). At first sight it may appear that this should be a fairly easy task, since extensive data are available for most isotopes of interest for different meteorite types. This is not true for several reasons. The composition and energy spectrum of the nuclear interacting particles in the meteorites differ considerably from those in the atmosphere (cf. Lal & Peters 1967). First, most of the pions (π mesons) decay in the atmosphere while they interact in the meteorite; pions are produced efficiently in nucleon interactions (Figure 2). Second, the path length in the atmosphere (1030 g cm^{-2}) is considerably larger than that in the case of meteorites (typically < 100 g cm^{-2}). Finally, the nucleonic cascade in meteorites is not in equilibrium. In the atmosphere, for depths exceeding 200 g cm^{-2}, an equilibrium is reached between fast and slow neutrons. In the atmosphere, the spectrum of fast neutrons that produce most of the nuclear disintegrations is much

softer compared with that in meteorites. The energy spectrum of tropospheric neutrons for $40 < E < 500$ MeV was stated to be of the form

$$\frac{dN}{dE} = \text{constant} \times (60+E)^{-2.5}, \tag{11}$$

where E is expressed in MeV (Lal 1958). The primary cosmic-ray proton spectrum for energies above 500 MeV is well expressed by the relation

$$\frac{dN}{dE} = \text{constant} \times (\alpha+E)^{-2.5}, \tag{12}$$

with a value of 1000 MeV for α. The energy spectrum becomes softer as nucleonic cascade develops within the meteorite. The value of α continuously decreases. For energies exceeding 100 MeV, the nucleonic spectra in meteorites are well represented by Equation (12) with 250 MeV $< \alpha < 600$ MeV for a 5–100 cm range of meteorite radii (Reedy & Arnold 1972, Bhattacharya et al 1980, Bhandari & Potdar 1982, Reedy 1985). Thus, the atmospheric neutron spectra are much softer than the nucleonic spectra in meteorites. (Nucleon spectral shapes in meteorites lie in between the neutron and proton curves in Figure 3.) A greater proportion of the fast nucleons in the atmosphere are neutrons. Even in meteorites, isotope production rates based on proton excitation functions are often found to be grossly in error. The case of ^{10}Be production in chondrites is a good example (Tuniz et al 1984).

Neutron cross-section data are, however, very limited to date. Considering their importance in in-situ terrestrial cosmogenic studies, these should certainly be determined in the near future.

The yields of several isotopes (^{14}C, ^{10}Be, ^{26}Al, ^{36}Cl, ^{37}Ar, ^{39}Ar, and ^{129}I) are now being determined in irradiations of suitable targets with beam-stop neutrons arising from proton bombardments of several targets and with slow and fast muons. This work is being carried out jointly between scientists of the University of California, San Diego, Los Alamos National Laboratory, San Jose State University, University of Pennsylvania, Brookhaven National Laboratory, and University of Arizona. In some cases, one can use natural production of isotopes in well-documented rock samples to obtain isotope production rates. An example of this is the recent study by Nishiizumi et al (1987) of ^{10}Be and ^{26}Al in glacially polished rocks exposed since the retreat of the Wisconsin glacial era.

It is also feasible to measure isotope production rates directly by exposing suitable targets to cosmic rays. Production rates of several isotopes can be determined in this manner. This method was first adopted by Lal et al (1960) to determine absolute isotope production rates and has since

been successfully employed by several investigators (Rama & Honda 1961, Bhandari et al 1969, Yokoyama et al 1977, Reyss et al 1981).

A large number of in situ cosmogenic isotopes promise to find useful applications in geophysical problems. Table 3 lists isotopes with half-lives exceeding two weeks. Several of these isotopes have multiple production mechanisms, and some examples of these are given in Table 6. Thermal neutron cross sections are available in most cases. The yields in μ^- capture can be estimated reasonably well from available data (cf. Charalambus 1971, and references therein). At present, the principal uncertainties in estimating reaction yields are for fast neutron and muon interactions; cross-section data are limited for neutrons and not available for fast muons. Some attempts have been made earlier to obtain the best-guess estimates by combining information on the experimental data on isotope production in the atmosphere, meteorites, and lunar samples; on observations of the nature of nuclear disintegrations in visual detectors; and on reaction systematics (Rudstam 1955, Lal 1958, Arnold et al 1961).

Production rates of ^{3}He in rocks up to underground depths of 10^4 g cm^{-2} have recently been estimated (Lal 1987b). These estimates include production by fast muons and thermal neutrons. The surface production rates of a dozen long lived isotopes in rocks exposed in the troposphere have been estimated by N. Bhandari & D. Lal (in preparation).

In rocks shielded by more than 3 kg cm^{-2} (including the path length in the atmosphere), the radiogenic neutron production in typical rock types becomes an important source of thermal neutrons (Lal 1963, 1987b). The production rates of ^{3}He by neutron capture in ^{6}Li and ^{10}B (Lal 1987b) and of a number of radioactive isotopes (^{3}H, ^{14}C, ^{36}Cl, ^{37}Ar, ^{39}Ar, ^{81}Kr, ^{85}Kr, and ^{129}I) due to radiogenic reactions have been estimated by Zito et al (1980) and Andrews et al (1987). Production of some of these isotopes in spontaneous and neutron-induced fission of uranium have also been considered by these authors.

In Figure 5, we reproduce the calculations of Lal (1987b) for rates of nuclear disintegrations due to nucleons and muons in standard rock (density = 3.09 g cm^{-3}, $Z^2/A = 6.3$) exposed at sea level (latitudes $\geq 45°$) as a function of depth (0–10 kg cm^{-2}). The corresponding estimates (Lal 1987b) for ^{3}He production, including cosmogenic and radiogenic thermal neutrons, are shown in Figure 6.

TEMPORAL VARIATIONS IN ISOTOPE PRODUCTION RATES IN ROCKS

Temporal changes in the terrestrial cosmic-ray flux are expected to arise due to (a) changes in the Earth's dipole field, (b) solar modulation of
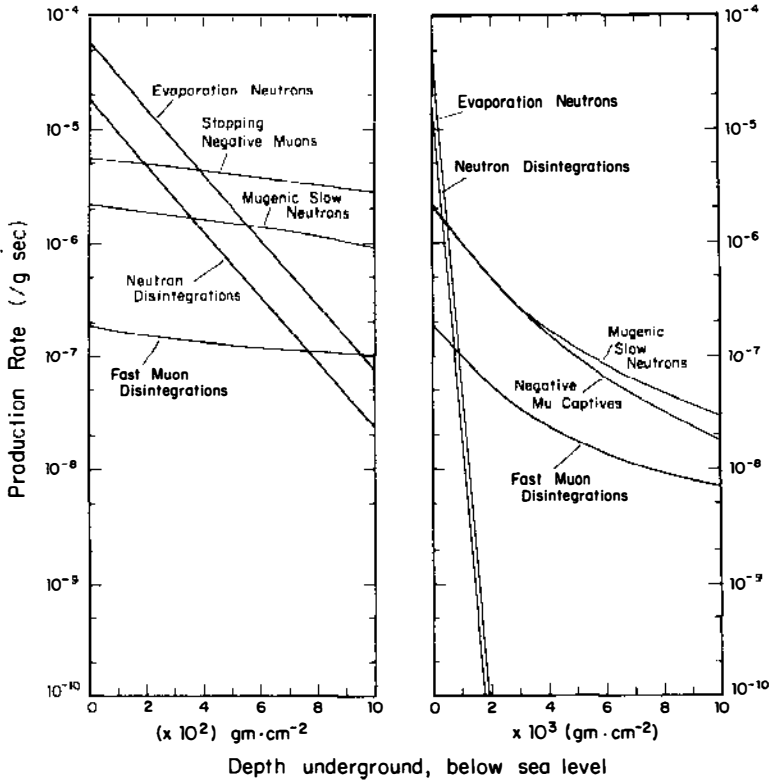
Figure 5 The rates of occurrence of nuclear disintegrations due to nucleons and muons, and production rates of evaporation neutrons in fast-neutron-produced disintegrations, in standard rock (density = 3.09 g cm^{-3}, Z^2/A = 6.3) as a function of depth underground (below sea level) for geomagnetic latitudes $\geq 45°$. The left side shows on an enlarged scale the production rates for 0–1 kg cm^{-2} depth (0–3.3 m rock equivalent) (based on Lal 1987b).

cosmic-ray flux, and (c) sporadic acceleration of cosmic rays in solar flares. In in-situ studies, we are concerned primarily with the first two. Solar flare cosmic radiation has a very soft spectrum, i.e. dN/dE = constant $\times E^{-\gamma}$ with $3 < \gamma < 6$ (Lal 1972, Reedy et al 1983). This radiation produces significant in situ isotopic changes in cosmic dust grains, outer layers of meteorites, and the lunar regolith. Since most of the solar cosmic-ray flux is confined to particles below a few hundred MeV kinetic energies, the secondary particle flux in the troposphere is not increased generally. A few exceptions are the unusually hard spectrum and/or high fluence events such as the flare events during 23 February 1956, 14 July 1959, and August 1972 (see Reedy 1977). Solar cosmic-ray production rates in the
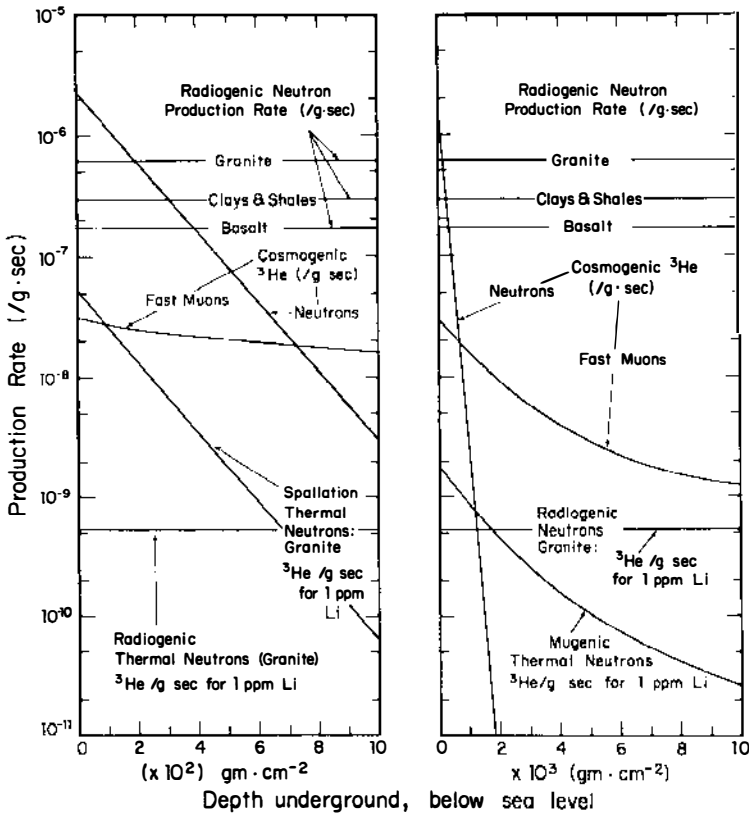
Figure 6 Production rates of ^{3}He in rocks by cosmogenic fast neutrons and muons, and cosmogenic/radiogenic thermal neutrons in a rock containing 1 ppm Li, are given for underground depths (0–10 kg cm^{-2}). The three horizontal lines in the upper part give the radiogenic production rates of neutrons in three rock types (based on Lal 1987b).

atmosphere for ^{3}H, ^{14}C, and ^{10}Be have been estimated (Lingenfelter 1963, Lal & Peters 1967, Lal 1987c).

Changes in the dipole field can appreciably change the depth altitude production rates in the atmosphere. The minimum energy of particles incident at the top of the atmosphere at any given latitude depends on the Earth's dipole field. Since the altitude-latitude production rates are known for the present dipole field, they can be easily calculated for any specified dipole field strength. The procedure has been discussed earlier (Lal & Peters 1967) and applied for the case of ^{14}C and ^{10}Be (Lal & Venkatavaradan 1970, O'Brien 1979, Castagnoli & Lal 1980, Lal 1987c). The minimum cutoff rigidity R_c (momentum to charge ratio) at any latitude λ is given by the relation

$$R_c(\lambda) = 14.9 \, (M/M_0) \cos^4 \lambda \, (GV), \tag{13}$$

where M_0 is the present dipole field intensity and M is the new dipole field intensity. Since production rates as in Figure 1 are known for all latitudes and M_0, the latitude-altitude curves for any given M value can be constructed for values of $M/M_0 < 1$. For values higher than 1, the R_c values at low latitudes are higher than those at $0°$ for $M/M_0 = 1$. Suitable corrections must then be made for the reduced flux at $0°$, according to the procedure adopted by Castagnoli & Lal (1980).

Changes in cosmic-ray flux incident at the top of the atmosphere due to changes in the level of solar modulation can now be modeled quite satisfactorily. Detailed calculations have been made for ^{14}C, ^{3}H, and ^{10}Be (Lal & Peters 1962, Lingenfelter & Flamm 1964, O'Brien 1979, Castagnoli & Lal 1980, Lal 1987c). As our understanding improves of the character of solar modulation, which is to a first approximation related to solar activity (indexed by a number of indices, such as sunspot number and geomagnetic indices K_p), we can improve on the expected variations in isotope production rates due to changes in solar activity.

In Figure 7 we show the measured changes in the differential primary cosmic-ray proton flux at the top of the atmosphere during different years. For each of the curves we have also specified the value of the modulation parameter ϕ, which appears as the single parameter in the force field approximate solution for the inward transport of cosmic rays through the solar plasma in interplanetary space (Garcia-Munoz et al 1975, Castagnoli & Lal 1980, Lal 1987c). The higher the value of ϕ, the lower the incident cosmic-ray flux; there exists a reasonable correlation between ϕ and sunspot number (see also Lingenfelter & Ramaty 1970). However, according to our present understanding, ϕ is not related in a straightforward manner with sunspot number (Garcia-Munoz et al 1977).

Extensive continuous data exist for counting rates of low-energy neutrons at several neutron monitor stations, mostly since 1955. These and the latitude-altitude ^{14}C production rates calculated by Lingenfelter (1963) allow one to obtain changes in isotope production rates in rocks exposed in the troposphere [see also the discussions by Lal (1987c) for ^{14}C and ^{10}Be solar modulation-dependent production rates in the atmosphere].

Observational data pertinent to temporal variations in cosmic-ray flux are (a) ^{14}C in tree rings, and (b) ^{10}Be in ice and in marine deposits. The tree ring ^{14}C data now go back to 10,100 yr B.P. (Becker & Kromer 1986, Stuiver et al 1986), and the ^{10}Be ice data go back to 140,000 yr B.P. (Yiou et al 1985). The ^{10}Be data in marine sediments go back to ~ 10 m.y. B.P. (Inoue & Tanaka 1979, Raisbeck & Yiou 1984), and in manganese crusts they also go back to ~ 10 m.y. B.P. (Ku et al 1982). The records of
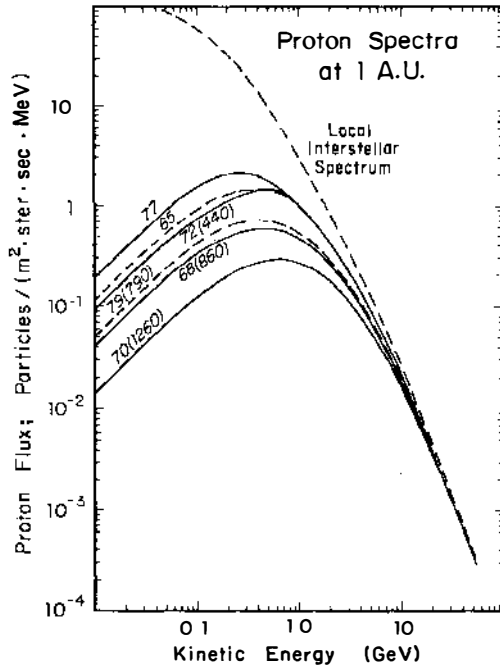
Figure 7 Measured differential kinetic energy spectra of protons near the Earth during different years of observation. The last two digits of the year (A.D.) are marked on the curves; the numbers within parentheses give the effective value of the modulation parameter ϕ. For sources of data, calculated ϕ values, and the predicted local interstellar spectrum, reference is made to Garcia-Munoz et al (1975).

atmospheric ^{14}C and ^{10}Be have to be deconvoluted suitably to obtain variations in the cosmic-ray flux; geophysical and geochemical processes introduce appreciable modulations in the record. Reference is made to Beer et al (1984) and Lal (1985) for discussions of the complexities of the ^{14}C record in tree rings, to Lal (1987d) for a discussion of the ^{10}Be record in ice, and to Somayajulu (1977) for a review of the ^{10}Be record in marine sediments. Our present techniques do not allow determination of changes in the terrestrial cosmic-ray flux due to dipole field from these records. The solar modulation of cosmic-ray flux is, however, seen in the ^{14}C tree ring record, and the long-term record of ^{10}Be in marine deposits does show that, averaged over periods of the order of 10,000 yr, the gross cosmic-ray flux incident in the Earth has not changed by more than 30–50% during the past 10 m.y.

It is to be expected that our techniques for the deconvolution of the record of atmospheric isotopes will improve substantially in the near future

to allow estimation of changes in the terrestrial cosmic-ray flux due to the three causes discussed above. However, the studies of in situ isotopes in rocks and trees from different latitudes show greater promise (Lal et al 1985).

APPLICATIONS OF IN SITU COSMOGENIC ISOTOPES

At sea level the production rates of ^{14}C, ^{10}Be, and ^{26}Al in suitable targets are of the order of 10-50 atoms g^{-1} yr^{-1}. The production rates at 2 and 5 km altitude are higher by factors of ~ 4 and ~ 30, respectively. If the effective cosmic-ray irradiation time is of the order of 10^3 yr at sea level, one would expect detectable amounts using AMS ($\sim 10^6$ atoms) in samples of a few tens of gram weight. This rough calculation forms the basis for the wide range of applications of the in situ method. The potential applications of in situ isotopes were pointed out earlier by Jha & Lal (1982) and Lal (1986a) for a variety of geophysical processes, e.g. erosion and tectonic movements.

Closed System

Besides the primary question of detection, there are several other questions that decide the usefulness of an in situ isotope as a geophysical tracer. These questions are the same that have been asked earlier in absolute age-dating techniques and in studies of cosmogenic isotopes in meteorites. The central requirement is that the rock/mineral system being dated should be a "closed system" with respect to its losing an isotope to or gaining an isotope from the environment. In age dating of rocks, the location or disposition of the rock is not important; however, the in situ cosmogenic isotope amount produced depends sensitively on the irradiation site and geometry. One has, therefore, to confine studies to rocks/sediments where sufficient geological information is available to make plausible models of the past geometric configurations.

To avoid possible interferences from atmospheric ^{10}Be adsorbed in rock matrix via seeping rainwater, studies of in situ ^{10}Be and ^{26}Al can be done in quartz mineral grains. The idea (Lal & Arnold 1985) has been tested and has passed stringent tests (Nishiizumi et al 1987). Quartz is a ubiquitous mineral in terrestrial settings; it serves as an ideal target for in situ production of three long-lived isotopes: ^{14}C, ^{10}Be, and ^{26}Al. Other minerals should likewise be explored for in situ applications.

The applications of the in situ cosmogenic production can be conveniently divided under two headings: (a) samples whose past evolution geometry is well known a priori, and (b) samples whose past evolution

geometry is known qualitatively but the time constants are not known. The expected isotope concentrations due to in situ production can be determined if both the cosmic-ray intensity $I(t)$ and the sample geometry evolution $G(t)$ are known. Therefore, if only one of these is known, the other can be modeled; the degree of resolution on the parameter evaluated would depend on the number of isotopes studied. This, in short, is the basis of application of the in situ isotope method. We now discuss the two sample types and illustrate some of their applications.

Well-Characterized Samples

Examples of samples in this class are trees, monuments, and glacial polished-rock surfaces. In the case of trees, the bombardment geometry changes with time in a precisely known manner (i.e. it can be fully characterized from the tree rings). In the other two cases, the geometry remains fixed, since the objects were exposed in that orientation. Such samples clearly would yield information on the changes in cosmic-ray flux, which in turn could be due to any of the following three causes:

1. Changes in the geomagnetic dipole field of the Earth.
2. Movement of the sample due to change in the altitude and/or latitude.
3. Changes in the primary cosmic-ray flux incident at the top of the atmosphere due to solar modulation.

These questions have been discussed, with special reference to (1) and (3), by Lal et al (1985).

Noncharacterized Samples

A variety of geological samples fall in this category. Some of the samples (for example, sands) will have had a very complex "irradiation" past, but a number of samples can be assigned a qualitative irradiation history based on geophysical/geological evidence. Examples of the latter are accreting sediments and eroding rock surfaces. The goal of in situ isotopes in such cases would be to study either of the following: (a) rates of accumulation of sediments, or (b) rates of erosion of rock surfaces.

The cosmic-ray intensity variations cannot be deduced generally from such samples. To illustrate the use of noncharacterized samples, we consider the expected concentration of an isotope C in a rock surface eroding at a constant rate ε at a fixed location. Let us further assume that the rock formed from igneous activity at some time T in the past; in this case, the initial isotope concentration is zero. After T years of irradiation, we would expect the following depth profile for the concentration $C(x)$ of the isotope (Lal & Arnold 1985):

$$C(X) = \frac{P_0 e^{-\mu X}}{\lambda + \varepsilon \rho \mu} (1 - e^{-\lambda T}), \tag{14}$$

where P_0 is the isotope production rate at the rock surface, λ is the disintegration constant of the isotope, ρ is the mean density of the rock, μ is the inverse of the mean absorption length for cosmic-ray particles in the rock, and the units of X, μ, and ε are g cm^{-2}, cm^2 g^{-1}, and cm s^{-1}, respectively.

There are several interesting aspects of Equation (14), which is a special solution for the expected in situ isotope concentration in a rock as a function of depth (Lal & Arnold 1985, Lal 1986b): It assumes constant isotope production and erosion rates and no preirradiation history of the rock. To bring out certain interesting geophysical aspects of in situ irradiation, we further assume that several meters of rock have been eroded away. The isotope concentrations in the rock are then in a quasi-steady state, and the surface concentration C_s becomes

$$C_s(X = 0) = \frac{P_0}{\lambda + \varepsilon \rho \mu}, \tag{15}$$

with the concentration at depth X decreasing exponentially, as in Equation (14).

The resulting surface isotope concentration depends on the isotope half-life, the mean erosion rate, and of course the isotope production rate. We may define two irradiation time periods: the equivalent time T_{eq} and the effective time T_{eff}. The former is the time it would take to result in a certain isotope production if the rock mass was irradiated in a fixed geometry, and the latter is the effective time corresponding to the ratio of the concentration to production rate. We obtain

$$T_{eq} = \frac{1}{\lambda} \ln \left(1 + \frac{\lambda}{\mu \rho \varepsilon} \right), \tag{16}$$

$$T_{eff} = \frac{1}{\lambda + \mu \rho \varepsilon}. \tag{17}$$

The effect of erosion is to set a new equilibrium value, in effect lowering the mean lifetime of the isotope by a factor $1/(1 + \mu \rho \varepsilon)$. This is equivalent to saying that the erosion mean lifetime is $(1/\mu \rho \varepsilon)$. The characteristic mean erosion period T_{ero} is defined by the ray attenuation length $(1/\mu)$, the density ρ, and the erosion rate ε:

$$T_{ero} = \frac{1}{\mu \rho \varepsilon}. \tag{18}$$

If the mean rock density is taken to be 3.5 g cm^{-3} and $1/\mu = 150$ g cm^{-2}, we obtain

$$T_{\text{ero}}(\text{yr}) = 43/\varepsilon, \qquad (19)$$

where ε is expressed in cm yr^{-1} (Lal 1986b). The mean erosion period T_{ero} becomes comparable to 10^6 yr (the order of half-lives of ^{10}Be and ^{26}Al) when $\varepsilon \sim 5 \times 10^{-5}$ cm yr^{-1}. For smaller erosion rates, the effective irradiation time will be determined by the isotope half-life, and for higher erosion rates, by the erosion rate.

From a study of concentrations of isotopes of different half-lives, an exposed surface can be characterized and the erosion rate determined. The erosion time scales accessible are simple functions of the isotope half-life. If different isotope pairs yield similar erosion rates, one can determine the validity of the steady-state uniform erosion rate model adopted. Deviations between results for different isotope pairs can conversely be used to model time variations in the erosion rate $\varepsilon(t)$.

About two-dozen rock surfaces have been studied for erosion by our group (Nishiizumi et al 1986; K. Nishiizumi, D. Lal, J. R. Arnold, J. Klein & R. Middleton, unpublished results). The in situ ^{10}Be and ^{26}Al erosion rates fall in the range of 10^{-2}–10^{-5} cm yr^{-1}, corresponding to effective irradiation periods of 5×10^3–10^6 yr.

A very interesting application of the in situ irradiation method is to the determination of episodic surface losses. After a discrete loss of surface up to a certain depth, (say X) following a steady-state situation, all isotope concentrations still follow the exponential decrease with depth, but the absolute isotope concentrations depart considerably from the steady-state situation, depending on the isotope half-life and the value of X. From a comparison of surface isotope concentrations, it should be possible to decide whether the horizon has suffered episodic losses.

In principle, from in situ studies one should be able to measure rates of uplift (which change the surface production rate). However, in practice this may be a difficult task, especially for rapidly eroding rocks. It can easily be shown that if the erosion rate is lower than 10^{-4} cm yr^{-1}, it should be possible to determine uplift rates exceeding 1 cm yr^{-1}.

Finally, can one date a surface using the in situ method? Let us reconsider the case of an igneous rock formation T years ago, for which Equation (14) applies. If now two in situ isotopes are studied in which, at least for one, the term within parentheses departs significantly from unity (i.e. $\lambda \sim 1/T$), both the date of eruption and the erosion rate can be determined.

The principal applications of different in situ radioisotopes in recent geophysical problems are shown schematically in Table 7.

Table 7   Principal in situ cosmic-ray-produced isotopes and their geophysical applications

Rocks	Sands Soils	Glaciers	Trees
^{3}He, ^{21}Ne, ^{10}Be, ^{26}Al, ^{36}Cl, ^{14}C	^{10}Be, ^{26}Al, ^{14}C, ^{39}Ar	^{10}Be, ^{14}C	^{10}Be, ^{14}C
Weathering and erosion processes, uplift rates, subsidence, glacial history, eruption ages, cosmic-ray prehistory	Transportation, burial histories; depositional and erosional rates, rates of movements of sand dunes	Accumulation and ablation rates, chronology, climatic changes	Temporal changes in solar activity, geomagnetic field, and climate

DISCUSSION

We have discussed the potential of the newly emerging field of terrestrial cosmogenic in situ isotopes and isotope production mechanisms and rates. The AMS technique, originally motivated by the necessity to measure the long-lived atmospheric cosmogenic isotopes, has now made this field tractable and attractive. These developments have in turn led to a resurgence in the use of ultra-sensitive counters and mass spectrometers for studying in situ cosmogenic short-lived and stable isotopes, respectively, that cannot be studied using AMS (for example, ^{39}Ar, ^{3}He, and ^{21}Ne).

The experimental field of in situ isotopes using AMS is only a few years old. In the initial stages it becomes necessary to establish the technique from the study of selected samples and also to determine the cosmic-ray production rates experimentally. Work is now in progress in these directions. The study of glacial polished-rock samples from the Sierra Nevada ranges (Nishiizumi et al 1987) seems to solve these problems. These data show that (*a*) quartz is an ideal closed-system mineral, and that (*b*) the amounts of ^{10}Be and ^{26}Al are in good agreement with expectations for cosmic-ray irradiation for 12,000 yr. Accelerator irradiations are now being carried out (Klein et al 1987) to determine the production rates of these isotopes by neutron irradiation and in μ^--capture in SiO_2.

Use of the stable isotopes ^{3}He and ^{21}Ne seems attractive for the study of erosion and other geophysical processes (Table 7). The stable isotopes

are useful for determining very slow erosion rates and thus supplement erosion studies based on ^{10}Be and ^{26}Al in quartz. Additionally, they can be used to characterize multiple exposure histories. The effective irradiation period for a stable isotope would of course be expected to be greater than that for a radioactive isotope. However, if $\lambda \ll (1/T_{eff})$ and T_{eff}(stable isotope) $> T_{eff}$(radioactive isotope), then clearly the sample has received an earlier irradiation several half-lives ago. A particular advantage of using in situ ^{3}He may lie in the possibility of making a large number of measurements conveniently. AMS studies of ^{10}Be and ^{26}Al are time consuming and expensive; an annual output of 50 samples per year for one project is large. In the case of ^{3}He, the corresponding yield could be an order of magnitude higher, judging from the performance of two groups (Craig & Poreda 1986, Kurz 1986a,b). Attempts have been made to estimate ^{3}He production rates in a radiocarbon-dated Hawaiian lava flow (Kurz 1986b). More work in this direction is needed, including simultaneous studies of ^{3}He and other radioactive isotopes (e.g. ^{10}Be and ^{26}Al).

Studies of in situ ^{129}I, ^{41}Ca, ^{14}C, and ^{39}Ar are just beginning. In the cases of ^{41}Ca and ^{14}C, techniques for their detection have yet to be perfected. It is certainly advantageous to study several in situ isotopes in the same sample, but this may not be practical. However, the potentials of these isotopes in various studies (cf. Table 7) are enormous. Thus, the applications of these isotopes would depend largely on the advances that would be made in the near future in high sensitivity and rapid analyses of several isotopes in a sample.

With an increase in the sensitivity and ease of detection of isotopes, we are sure to see new uses of particular in situ isotopes for the study of specific problems. Raisbeck & Yiou (1979) discussed the possible applications of ^{41}Ca in the dating of $CaCO_3$ sediments and animal remains. Phillips et al (1986) discussed the possibility of age dating young volcanic rocks using in situ ^{36}Cl. Age determinations are possible if two or more isotopes are used (Lal 1987a); unless the magnitude of erosion is known a priori, one has to determine both the erosion rate and the time of exposure of the surface.

An interesting recent application of in situ ^{10}Be was made by Lal et al (1987) in placing a lower limit to the amount of cosmogenic ^{3}He in alluvial diamonds from Zaire. Their results showed that a minimum of 29% of the ^{3}He in the alluvial diamonds was cosmogenic. They surmised that before concluding that the high ^{3}He/^{4}He ratios observed in some diamonds, corresponding to the ratios in solar wind, existed in the mantle at some time (Ozima et al 1983), it was necessary to study diamonds collected by underground mining methods. Such diamonds would be free from cosmogenic in situ ^{3}He. The high ^{3}He/^{4}He ratios reported by Ozima and his

collaborators are all for undocumented diamonds, and thus an appreciable amount of cosmogenic ^{3}He cannot be ruled out in these diamonds.

During the next decade we should be able to obtain a wealth of geophysical data and information about changes in the cosmic-ray intensity in the past 10^2–10^6 yr due to changes in the geomagnetic field intensity and in solar activity (cf. Lal et al 1985). Cosmic rays are ideal probes for sensing changes in the dipole field of the Earth. Studies of isotopes deep underground should provide valuable information on the long-term secular variations in the flux of energetic cosmic-ray particles ($> 10^4$–10^5 GeV).

I have discussed the principal applications of in situ isotopes as they appear to me from the present-day perspective. The history of science is replete with examples of rapid and drastic changes in outlook in a field. It may be interesting to recall here the statement ascribed to F. W. Aston, the father of mass spectrometry. In 1935 Aston reportedly said that the job of mass spectrometry was done and that the field of research would die away (Cadogan 1986). This example suggests that I may have too narrowly constrained the potential applications of in situ cosmogenic isotopes.

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