detail. Neutrinos, while not exactly everyday matter, are at least no strangers in the laboratory or to astronomers. If they were the dark matter, we could see a unity in matter as we see in the chemistry of our earth: silicon and oxygen and iron are plentiful here in the rocks, though in sun and stars those atoms are found only in parts per thousand. Still, they are present, to imply a deep unity within. That unity is simply locally disturbed, but not destroyed, by those complex physicochemical processes that formed our unusual, extra-dense home planet, collecting up oxygen, even iron, but losing light, fugitive hydrogen.

Experience must one day decide. Something of the spirit of old Copernicus will hold: our earthly home seemed special, but was not. So too our kind of matter, made of nucleons and electrons, seemed prevalent, but perhaps it is not. The real unity we seek is not of the familiar alone; that seems too narrow for an unbiased

view. It is rather a unity of the demonstrable, a unity that joins all the processes we can grasp. The next decade or so may begin to bring an answer.

Freier, P., Losgren, E. J., Ney, E. P. and Oppenheimer, F., Phys. Rev., 1948, 74, 1818; Bradt, H. L. and Peters, B., Phys. Rev., 1948, 74, 1828, and 1950, 80, 943.

The two citations appeared in 1948 as adjoining papers. The FLNO group at the University of Minnesota flew a cloud chamber, and saw heavy primary tracks; the BP group at the University of Rochester flew emulsions that also showed heavy tracks. The simultaneous confirmation was not accidental. F. Oppenheimer and B. Peters had been close friends and colleagues at Berkeley; the two parallel experiments were carried on in quite friendly rivalry. The emulsion technique turned out to be more open to later development.

2. Reviewed nicely in Cosmic Pathways, (ed. Cowsik, Ramanath), Tata McGraw-Hill, New Delhi, 1986, pp. 289-310.

# The present scope of the field of terrestrial cosmogenic nuclides

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The discovery of natural radiocarbon produced by cosmic rays in the earth's atmosphere by Libby in the late forties<sup>1-3</sup> was followed by the discovery<sup>4</sup> of terrestrial cosmogenic nuclide <sup>3</sup>H in 1951. In the next few years several terrestrial cosmogenic radionuclides were detected<sup>5</sup>. The cosmogenic nuclides found a variety of new applications during 1960-1970 in meteorology, hydrology, glaciology and oceanography. The three long-lived cosmogenic nuclides produced in the atmosphere with half-lives > 100 yr, <sup>32</sup>Si, <sup>14</sup>C, <sup>26</sup>Al and <sup>10</sup>Be continued to provide invaluable data in the field of oceanography, using the methods developed in the sixties. However, in the late seventies the development of the accelerator mass spectrometry (AMS) technique made it possible to measure several long-lived radionuclides<sup>6,7</sup> with 3-6 orders of magnitude higher sensitivity. This led to an explosion in the eighties in the scope of applications of cosmogenic nuclides in both terrestrial and extraterrestrial samples. This article attempts to convey the sense of the present excitement of cosmogenic nuclides as tools in geosciences, while highlighting the modern development with perspectives of the forties and fifties when the terrestrial cosmogenic nuclides 14C, 3H, 10Be and others were discovered.

'Cosmic-ray physics, which ten years ago was a fairly specialized branch of science, has in the course of its recent development become closely linked with many other fields of research; it has become an integral part of astrophysics, radioastronomy, and solar physics; it has made important contributions to such diverse fields as geomagnetism, hydrology, and archaeology, and has begun to gain some importance in the study of meteorites and of oceanography and meteorology. It has also given rise to one of the newest and most active branches of physics, particle physics,....' Bernard Peters<sup>8</sup> said this in a paper 'Progress in cosmic-ray research since 1947' published in 1959. During that interval, Peters was himself responsible for several of the developments mentioned: in cosmic-ray physics (origin, propagation and lifetime of cosmic rays; nature of nuclear interactions of high-energy cosmic-ray particles, discovery of multiply charged nuclei in cosmic radiation, characterization of several elementary particles), and in the field of cosmic ray-produced isotopes on the earth. Peters had already carried out a tremendous amount of original work on the nature of primary cosmic radiation during his stay at the

University of Rochester<sup>9-12</sup>, when in 1949, he was invited by H. J. Bhabha to work at the Tata Institute of Fundamental Research (TIFR), Bombay. He accepted this, making his first visit to India during August-November 1950 to conduct a series of balloon flights. I was then a student of cosmic-ray physics at TIFR, working under the guidance of the well-known cosmic-ray physicist, Taylor<sup>13</sup>, and had my first opportunity to work closely with Peters during the balloon flights. Peters then joined<sup>14</sup> TIFR as professor of physics towards the end of 1951. During the next seven years of his stay in India he conducted active research in the areas of origin and composition of cosmic rays, and cosmic-ray geophysics. Besides being an important contributor to the growth of these fields, he made a long lasting impact on Indian science by conducting research and seeding academic activity par excellence, and by training scientists who later held important positions in India in the fields of education and research.

The period 1952–1954 was one of intense activity in the field of elementary particle physics at TIFR, with new information<sup>15–17</sup> emerging continuously on the types and character of elementary particles observed in nuclear emulsions exposed to cosmic radiation at high altitudes. The success of Peters' group was a direct result of his approach<sup>18</sup> to the problem, namely processing a stack of free-floating, large, thin emulsion sheets so that charged particle tracks could be traced forwards or backwards through these emulsion pellicles. The participating scientists worked round the clock with great excitement and there was an excitement even in the corridors of TIFR as fundamental information was being obtained through these simple, but well-conceived experiments.

The years 1955-1957 marked the second period of great excitement and discoveries during the short period of direction of research by Peters at TIFR. In 1955 a new activity was started by Peters<sup>19</sup>, namely the search for the cosmic-ray-produced isotopes, <sup>10</sup>Be and <sup>7</sup>Be. The isotopes of beryllium, <sup>7</sup>Be and <sup>10</sup>Be, were detected in the 1955 Bombay rains<sup>20</sup> and in 1956 in Pacific marine sediments<sup>21</sup>. The nuclide <sup>7</sup>Be was discovered by Arnold and Al-Salih<sup>22</sup> before we did in 1955 rains<sup>20</sup>, but we were then not aware of his discovery of <sup>7</sup>Be in Chicago rains during October 1953 to April 1954. The nuclide, <sup>10</sup>Be, was however discovered independently by Arnold<sup>23</sup> and by Peters' group<sup>21</sup> in 1956 in Pacific marine sediments. For accounts of discovery of <sup>10</sup>Be in Chicago and in India, reference is made to Arnold<sup>24</sup> and to Lal<sup>25</sup>. Two other nuclides, <sup>35</sup>S and <sup>33</sup>P, produced in the atmosphere by cosmic-ray spallations of argon were detected in Bombay rains<sup>26,27</sup>. The capability developed at the TIFR in the field of low-level counting and radiochemistry led to the discovery<sup>28,29</sup> of six other cosmic-rayproduced radionuclides during 1965-1966.

Peters was convinced that for any proper application of the cosmogenic nuclei in meteorology/oceanography, it was necessary to know their production rates very accurately in the atmosphere, as a function of altitude and latitude. He began on this task<sup>19</sup> in 1955 in connection with <sup>10</sup>Be. The first paper describing the most accurate production rate estimates for nuclear disintegrations and nuclides, <sup>10</sup>Be, <sup>7</sup>Be, <sup>35</sup>S, <sup>33</sup>P and <sup>32</sup>P was published<sup>30</sup> in 1958. These calculations were a substantial improvement over the earlier calculations<sup>31</sup> of Benioff for <sup>7</sup>Be which used nuclear cascade theory for determining nucleon propagation in the atmosphere. The atmosphere is equivalent to a thickness of  $\sim 12$ nuclear interaction mean-free paths, and even a 10% error in one of the nucleonic cascade parameters can lead to >100% error in nucleon flux at sea-level. The emphasis in Peters' work was on relying almost entirely on experimental data from cosmic ray and accelerator experiments. A variety of cosmic-ray data, on neutron fluxes and nuclear disintegration rates in the atmosphere, were utilized to provide an accurate relative altitude latitude source function for cosmonuclei. The absolute nuclide-production rates were later obtained using direct measurements<sup>32</sup> of production rates of nuclides in oxygen and argon targets exposed to cosmic radiation at mountain altitudes and sea level. These production rate estimates<sup>33</sup> have been found to provide fairly accurate altitude-latitude variation and are used very effectively in present day cosmogenic studies<sup>34,35</sup>.

### Cosmogenic nuclides: terrestrial and extraterrestrial

Very sensitive detection systems were developed in the fifties and sixties to measure the feeble signals due to cosmic-ray produced radionuclides<sup>1-5,33,36-38</sup>. Likewise several technical advances were made in the field of mass spectrometric detection<sup>39,40</sup>. It then seemed (incorrectly) to many that the scope of applications of cosmogenic nuclides, although constrained somewhat only by their detection sensitivities, was nearly fully realized by 1970. Cosmogenic studies continued in the seventies, but naturally at a much slower pace, but the cosmogenic nuclei remained indispensable, since in most cases they provided the only viable tracer method.

Then, in the late seventies, a new technique called accelerator mass spectrometry (AMS) appeared on the scene, which made it possible to identify several nuclides directly after accelerating them, using, essentially, techniques similar to those used by cosmic-ray physicists to study isotopes in the cosmic-ray beam<sup>41-44</sup>. It is now possible to detect ca. 10<sup>5</sup> atoms in the case of several nuclides provided the nuclide/element ratio (atom/atom) exceeds  $10^{-14}$ - $10^{-15}$ . This made a very

big difference in the sensitivity of detection for several nuclides, for example, in the case of <sup>10</sup>Be. Direct beta counting required  $> 5 \times 10^{11}$  atoms at the highest sensitivity. With AMS one can detect  $10^5-10^6$  atoms of <sup>10</sup>Be in practically any sample type, since <sup>9</sup>Be concentrations are low, usually at the ppm level in any terrestrial meteoritic sample, corresponding to a gain by a factor of  $> 1 \times 10^6$  over counting. The result was that cosmogenic <sup>10</sup>Be could be studied in all terrestrial samples, using orders of magnitude smaller samples<sup>45</sup>. The sensitivity gain was particularly important in the case of meteorities, and also in the case of other hard-to-get terrestrial samples, e.g. ice samples. What the AMS technique had done was to widen the scope of the recognized cosmogenic isotopes (by the availability of more comprehensive data base and time series in particular), and by making it possible to make new applications of cosmogenic nuclides which were not possible considering detection problems.

The possibility of studying cosmogenic effects in small extra-terrestrial samples, using the AMS technique, led to a large data base for several nuclides  $^{46-50}$  in cosmic dust grains, meteorites and lunar samples. Also, nuclides which had not been detected previously or were not easily detectable earlier were studied extensively. An interesting case is the measurement of cosmogenic  $^{129}$ I in meteorites  $^{51}$ . This radionuclide has a half-life of  $1.6 \times 10^7$  yrs, intermediate between that of  $^{53}$ Mn and  $^{40}$ K,  $3.7 \times 10^6$  yrs and  $1.3 \times 10^9$  yrs respectively, and is ideally suited for determining temporal variations in the galactic cosmic-ray intensity during the past  $5 \times 10^6 - 5 \times 10^7$  yrs. Available results so far on temporal variations in cosmic-ray flux have been discussed  $^{52,53}$ .

#### Cosmogenic terrestrial atmospheric nuclides

On the earth, the AMS technique led to extensive studies<sup>45,54,55</sup> of four radionuclides, viz. <sup>14</sup>C, <sup>10</sup>Be, <sup>36</sup>Cl and <sup>129</sup>I. All these nuclides are produced in the earth's atmosphere, and had been detected earlier. The radionuclide <sup>129</sup>I was however not studied much earlier, because of the difficulties in its detection. Due to the sensitive AMS detection, it has now become an important nuclide<sup>54</sup> for studies of hydrologic and geologic processes on time-scales up to 100 m.y. Likewise, the nuclide <sup>36</sup>Cl has become an important diagnostic tool in studies of ground waters<sup>56,57</sup>.

Studies of <sup>10</sup>Be were largely limited to dating marine sediments and manganese nodules <sup>58-61</sup>; a few measurements were however made earlier in soils <sup>62</sup>. The AMS technique made it possible to measure atmospheric <sup>10</sup>Be in soils <sup>63</sup> and in practically all the marine samples <sup>64</sup>, and in marine sediments and manganese nodules <sup>65</sup>, and also made it possible to obtain long time series of its concentrations in polar ice

cores, back to about 40 k yrs<sup>66,67</sup>. These studies have now made it possible to discuss long-term erosion-controlled budgets in soil horizons, and climate and cosmic-ray flux-induced effects in its deposition in the polar regions.

In the case of <sup>14</sup>C, a nuclide that had already been studied in practically all the dynamic reservoirs of the carbon cycle, its studies were also very dramatically affected, primarily because one can now measure 14C in samples of a few mg carbon from the biosphere/hydrosphere. The impact of AMS has been considerable in the field of oceanography. Small volumes of ocean water, about 1 litre, and a few times for aminifera shells can now be studied for their <sup>14</sup>C activities. Andree et al.68 were the first to recognize the importance of measurements of <sup>14</sup>C in benthic and planktonic foraminifera from the same sediment sample. The subsequent studies of <sup>14</sup>C in foraminifera from different locations in the world oceans are now providing a detailed insight into changes in the ocean circulation during the past 20,000 yrs<sup>69,70</sup>.

The principal applications of the atmospheric cosmogenic nuclides are listed in Table 1. Several of these 'atmospheric' cosmonuclei, in spite of their being unique tracers, have not been well exploited because of the difficulties associated with their sampling and detection. Detection and sampling, in some cases, are in fact the central problem which limit the studies of cosmogenic isotopes. It is for this reason that whichever nuclides could be studied with the AMS technique were extensively studied in the past decade. In spite of the difficulties with their measurements, concerted efforts have nevertheless been made to study several of the useful nuclides using the traditional counting methods. I would like to highlight the case of four radionuclides here to make this point: <sup>39</sup>Ar (half-life, 269 yrs), <sup>32</sup>Si (half-life  $\sim 200 \text{ yrs}$ ), <sup>33</sup>P (half-life, 25.3 days) and <sup>32</sup>P (half-life, 14.3 days). <sup>39</sup>Ar cannot be measured using the

Table 1. Principal 'atmospheric' cosmic-ray-produced isotopes and their applications\*.

Nuclide(s)	Applications				
Short-lived (1/2 hr – 2.6 yr.):  34mCl, 38Cl, 39Cl, 18F, 31Si, 38S, 24Na, 28Mg, 32P, 33P, 7Be, 35S, 22Na	Meteorology, cloud physics, atmospheric structure, large-scale air circulation and scavenging				
Others (half-life > 10 yrs): <sup>3</sup> H, <sup>32</sup> Si, <sup>39</sup> Ar, <sup>14</sup> C, <sup>36</sup> Cl, <sup>10</sup> Be and <sup>129</sup> I	Oceanography and marine sediment record: air-sea exchange, geochemical and biological cycles, chronology of sediments, palaeomagnetic reversal records and cosmic-ray prehistory				
<sup>32</sup> Si, <sup>39</sup> Ar, <sup>14</sup> C, <sup>81</sup> Kr, <sup>36</sup> Cl, <sup>81</sup> Kr and <sup>10</sup> Be	Hydrology and glaciology: chronology of ground waters, lacustrine sediments, and glaciers				
14C	Archaeology, climate history and palaeobotany				

<sup>\*</sup>Grouped in order of increasing half-lives in each category.

AMS technique. The sensitivity is not high enough yet with the AMS technique in the case of  $^{32}$ Si. The detection limits with beta counting for  $^{33}$ P and  $^{32}$ P  $\sim 5 \times 10^5$  atoms are comparable to those attained for  $^{10}$ Be,  $^{26}$ Al and  $^{36}$ Cl using AMS.

The main applications of <sup>39</sup>Ar are for studying ages of ground waters and large-scale ocean circulation<sup>33,71-74</sup>. The measurements of <sup>39</sup>Ar in ground waters and in ocean waters have steadily grown over the past but at a slow rate<sup>73,74</sup>. The principal merit of <sup>39</sup>Ar is that it is a 'conservative' nuclide for tracing motion of water, and studies so far of <sup>39</sup>Ar have documented the promise of the method. The other conservative cosmogenic nuclide for hydrological applications is <sup>81</sup>Kr (half-life, 2.1 × 10<sup>5</sup> yrs) whose detection with beta-counting methods is not feasible but possible using single-atom counting by the laser resonance ionization spectroscopy (RIS) method<sup>75,76</sup>. This nuclide has recently been used for the measurement of age of polar ice<sup>76</sup>.

The three nuclides, <sup>32</sup>Si, <sup>32</sup>P and <sup>33</sup>P are useful for studying nutrient cycles on long and short time-scales. During the GEOSECS expedition, Somayajulu and his collaborators collected several profiles of <sup>32</sup>Si in the Atlantic, Pacific and Indian oceans. Results for the Atlantic have been published; data for other oceans will soon be available. Individual measurements of ocean water samples require extraction of dissolved silicon from several tons of sea water. Somayajulu *et al.*<sup>77</sup> have employed ingenious techniques to achieve this, however, the counting of the low beta activity of the samples (of <sup>32</sup>P milked with <sup>32</sup>Si) is a slow process and limits the rate of data acquisition.

For the Atlantic Ocean, <sup>32</sup>Si results are consistent with the conventional 'vertical' and 'horizontal' silicontransport models and provide additional information on processes and rates<sup>77,78</sup>. Biological production removes dissolved silicon from surface waters; particulate siliceous shells dissolve at different depths in the ocean and lead to an in situ flux (J-flux) of dissolved silicon. Horizontal advection of waters transports dissolved (and some particulate silica) with the North Atlantic Deep Waters (NADW) to the Southern Ocean. <sup>32</sup>Si measurements are consistent with NADW transport, but showed for the first time that there was an appreciable net transport of <sup>32</sup>Si to the Southern Ocean where its column inventory is about  $4 \times that$  expected from cosmic ray production. A complete picture of silica transport in the deeper layers of the oceans can now be obtained from analysis of stable and cosmogenic silicon isotopes in the major oceans.

Studies of cosmogenic <sup>32</sup>P and <sup>33</sup>P in surface ocean waters have been carried out only recently realizing that these twin radiotracers can be effectively used for the P cycles on time-scales compatible with those involved in biogeochemical processes and in trophic

interactions within the food web. Their detection requires extraction of dissolved inorganic and organic P from several tons of sea water. Fortunately, the method used for extraction of dissolved silica works effectively for dissolved P as well. Studies of <sup>32</sup>P and <sup>33</sup>P to date<sup>79,80</sup> have demonstrated the promise of these tracers. The highest specific activities of <sup>32</sup>P and <sup>33</sup>P are found in dissolved inorganic phosphorus and in phytoplankton samples. The zooplankton samples have appreciably lower (and variable) specific activities, implying a longer turnover time of P in the zooplankton, > 30-40 days.

Attempts are now being made to quantitatively isolate different phases of dissolved P from several tons of sea water. These studies should provide a quantitative picture of dynamic interactions between the dissolved P pools and the particulate P reservoir. Studies of <sup>32</sup>P and <sup>33</sup>P have also indicated to be promise for studying time-averaged values of new production, over time periods of the order of ~1 month. The interrelationships between the P pools and new production should be an important result of these investigations.

One of the variables in studies of surface and near surface nutrient cycling is the eddy diffusivity. It has to be explicitly taken into account in any modelling of verticle circulation of P in surface waters, and for estimating upward flux of preformed nutrients. A suggestion by Lal and Krishnaswami<sup>81</sup> approaches this problem simply by studying another cosmogenic isotope <sup>7</sup>Be (half-life, 53.3 days) in the same waters where <sup>33</sup>P and <sup>32</sup>P are studied. Beryllium is biologically inactive and forms an ideal tracer for the measurement of eddy diffusivity in sea water<sup>82</sup>.

#### Cosmogenic terrestrial in situ nuclides

A new field of investigation based on cosmogenic nuclides caught roots with the emergence of the AMS technique, namely investigations of nuclides produced in situ in solids, e.g. rocks, tree rings and ice. These studies have now opened up the possibilities<sup>34</sup> of studying a wide range of geophysical processes which could not be studied earlier (Table 2).

It is interesting to note that among the various in situ nuclides in Table 2, the measurement of <sup>39</sup>Ar, <sup>3</sup>He and <sup>21</sup>Ne was made using conventional techniques: gas counting in the case of <sup>39</sup>Ar, and mass spectrometric techniques in the case of <sup>3</sup>He and <sup>21</sup>Ne (refs. 83-85). Other nuclides were detected using the AMS technique<sup>34,35,86</sup>,

In order to fully appreciate the potential of cosmogenic in situ isotopes, it is necessary to understand their production mechanisms and source functions and their expected time variability in relation

Table 2. Principal in situ terrestrial cosmic-ray-produced isotopes and their applications\*.

Samples	Isotopes	Applications				
Rocks 30Ar, 14C, 36Cl, 26Al, 10Be, 21Ne, 3He		Weathering and erosion processes, uplift rates, subsidence, glacial history, eruption ages, cosmic ray prehistory				
Sands, soils	36Ar. 14C. 36Cl. 26Al. 10Be	Transportation, burial histories; depositional and erosional rates, rates of movements of sand dunes				
Glaciers	<sup>14</sup> C, <sup>36</sup> Cl, <sup>10</sup> Be	Accumulation and ablation rates, chronology, climatic changes				
Tree rings	14C, 10Be	Temporal changes in solar activity, geomagnetic field and climate				

<sup>\*</sup>Grouped in order of increasing half-lives.

to terrestrial and solar system influences. The problem of propagation of the cosmic-ray beam through the earth's atmosphere was dealt with in great detail earlier, with a view to estimating the source strengths of cosmic-ray-produced nuclides in the atmosphere<sup>34</sup>. The composition of the secondary cosmic-ray beam undergoes a continuous change as one goes to deeper depths in the atmosphere. Within the troposphere, most nuclear disintegrations are due to neutrons. At sea level negative  $\mu$  capture becomes significant. At depths underground below sea level > 1 m rock equivalent, negative muon captures, and fast-muon interactions are responsible for nearly all the cosmogenic interactions. The composition of secondary cosmic-ray particles and the approximate rate of nuclear disintegrations are given in Table 3.

The rate of nuclear disintegrations in the atmosphere for 0–10 km altitudes for 0° latitude is shown in Figure

1, based on calculations of Lal and Peters<sup>33</sup>. The nuclide-production rates within a rock can be calculated fairly reliably from the altitude-latitude production curves in Figure 1, as long as the total sample shielding depth does not exceed  $\sim 1000 \,\mathrm{g\,cm^{-2}}$ , the total sample shielding depth being given by the sum of atmosphere pressure and the vertical shielding within the rock. When the total shielding depth exceeds 10<sup>3</sup> g cm<sup>-2</sup>, one has to carefully estimate relative neutron and muon fluxes. Some attempts have been recently made<sup>87</sup> to do this, including estimates of nuclide production due to radiogenic neutrons. In Figure 2, we reproduce the results of Lal<sup>87</sup> for production rates of <sup>3</sup>He in underground rocks, below sea level. These production rate estimates give a fair idea of the source strength of cosmic-ray-interacting particles vis-à-vis radiogenic neutrons.

The basis of applications of in situ cosmogenic isotopes in terrestrial solids is the fact that isotope production rates are extremely depth-sensitive. Thus, processes such as erosion and sedimentation can be modelled based on the observation of nuclides of different half-lives. The field grew<sup>34,35</sup> during the last decade and is rapidly undergoing changes in its scope and versatility. The in situ cosmogenic production rates of the nuclides <sup>3</sup>H, <sup>14</sup>C, <sup>10</sup>Be and <sup>26</sup>Al have been directly measured by studying their concentrations in rocks of known exposure histories<sup>86,88,89</sup>. The exposure histories of a large number of rock surfaces in the Antarctica have been studied<sup>90</sup>. The most recent application of in situ nuclides is that of <sup>14</sup>C produced in ice for determination of ablation<sup>91</sup> and accumulation rates<sup>92</sup>.

Table 3. Cosmic-ray primaries and secondaries in the atmosphere and at depths underground at 45° latitude (based on Lal and Peters<sup>33</sup>).

	Depth in the atmosphere (g cm <sup>-2</sup> )					Depth underground below sea level (kg cm <sup>-2</sup> )		
	0	200	500	1030 (sea level)		50	500	
Cosmic-ray	p	<b>e</b> , γ	<b>e</b> , γ	$\mu$	$\mu^{\pm}$	$\mu^{\pm}$	$\mu^{\pm}$	
primary and	. α.	$\mu^{\pm}$	$\mu^{\pm}$	e, y	n	n	n	
secondary	CNO	n	n	n	p	p	р	
particles*	Si, Fe	p	p	p				
Nuclear	p	n	n	n	$\mu_{c}$	$\mu^{\pm}$	$\mu^{\pm}$	
disintegration*	α	p	$\mu_{\mathbf{c}}$	$\mu_{\mathbf{c}}$	$\mu^{\pm}$	$\mu_{c}$	$\mu_{c}$	
-	CNO	y	р	p	n	n	n	
	Si, Fe	γ μ <sub>c</sub> μ±	$\frac{\ddot{\gamma}}{\mu^{\pm}}$	$\mu^{\pm}$	n p	n p	n P	
Approx. total ra	ates of nuclear disin		•					
	10-2	$5 \times 10^{-3}$	$10^{-3}$	$2 \times 10^{-5}$	10 <sup>-6</sup>	10~9	$5\times10^{-12}$	
1. $(g^{-1} \sec^{-1})$ 2. $(g^{-1} yr^{-1})$	$3 \times 10^{5}$	$1.5 \times 10^{5}$	$3 \times 10^4$	$6 \times 10^2$	30	$3 \times 10^{-2}$	$2 \times 10^{-4}$	

<sup>\*</sup>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;  $\alpha$ , <sup>4</sup>He; e, electron;  $\gamma$ , photon;  $\mu^{\pm}$ , positive and negative mu-mesons;  $\mu_c$ , nuclear capture of negative mu-meson. C, N, O, Si and Fe refer to these nuclei.

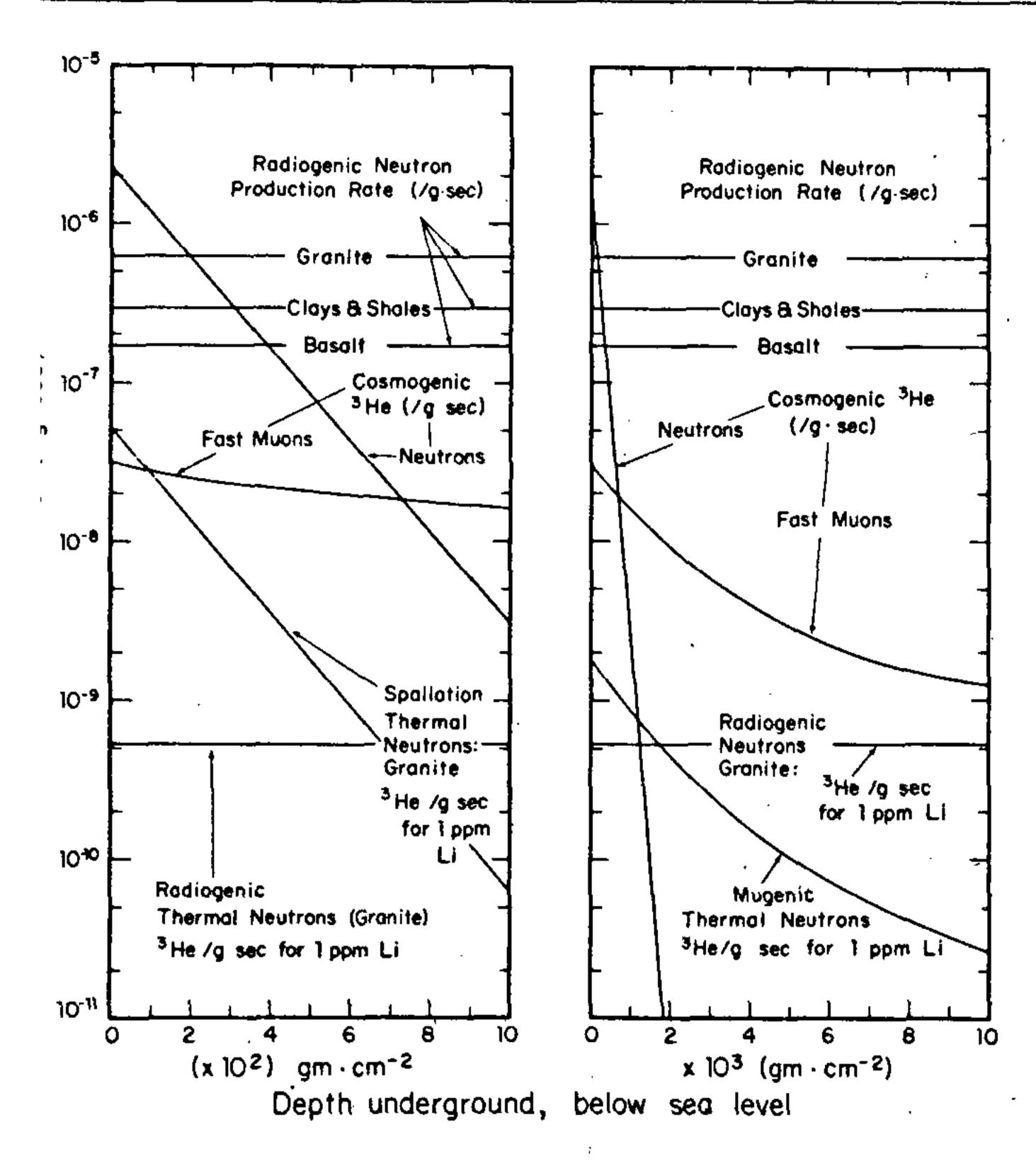
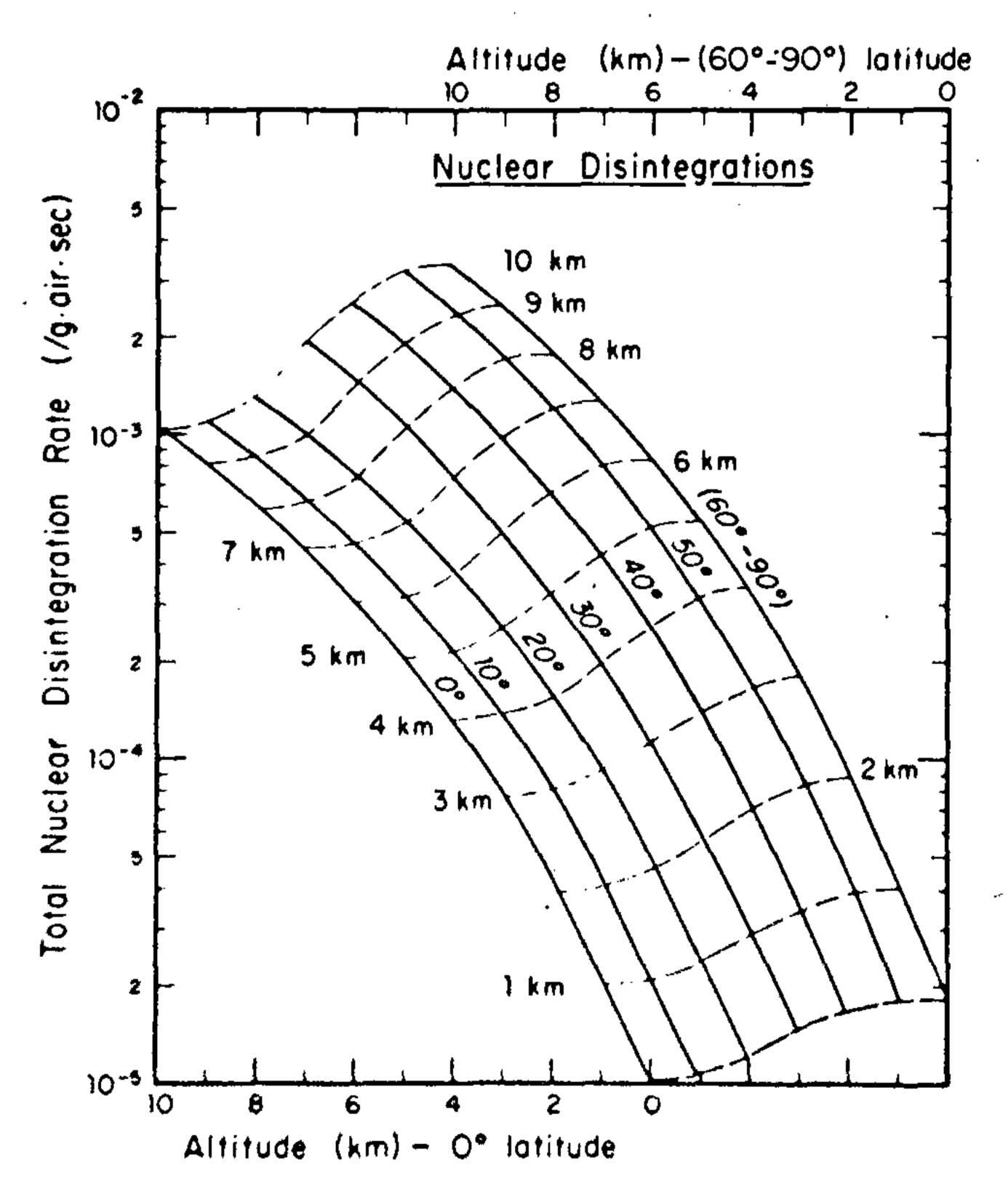


Figure 1. The total rate of nuclei disintegrations in the atmosphere with energy release > 40 MeV) plotted as a function of altitude and geomagnetic latitudes (0-90°, in steps of 10°). Curves for different atitudes have been successfully displaced along the abscissa by 1 km based on Lal and Peters<sup>33</sup>).



igure 2. Production rates of <sup>3</sup>He in rocks by cosmogenic fast eutrons and muons, and cosmogenic/radiogenic thermal neutrons in rock containing 1 ppm Li, are given for underground depths (0-10 g cm<sup>-2</sup>). The three horizontal lines in the upper part give the adiogenic production rates of neutrons in three rock types (based on .al<sup>34</sup>).

Table 4. Cosmic ray source function\*.

Global flux of hydrogen nuclei	$7.8 \times 10^{25} \text{ yr}^{-1}$			
Global mass flux of hydrogen nuclei	130 g yr <sup>-1</sup>			
Global mass flux of all nuclei	170 g yr <sup>-1</sup>			
(H, He, C, N, O, etc.)				
Mean energy of a cosmic-ray particle	$3.6 \times 10^9 \text{ eV/nucleon}$			
	$(\sim 4 \times 10^{13} \text{ K})$			
Global energy flux	$3.7 \times 10^{35}  \mathrm{eV}  \mathrm{yr}^{-1}$			
	$(1.4 \times 10^{16} \text{ cal yr}^{-1})$			
Solar energy flux	$5.2 \times 10^{24} \text{ cal yr}^{-1}$			
Heat flow	$2.4 \times 10^{20} \text{ cal yr}^{-1}$			

<sup>\*</sup>Global average flux of hydrogen nuclei =  $\sim 1$  cm<sup>-2</sup> sec<sup>-1</sup>.

#### **Conclusions**

All matter undergoes transformation continuously by a variety of nuclear mechanisms. It is difficult to find sites in the universe where nuclear transformations do not occur! As our understanding of physics improves, we learn of new ways in which matter undergoes transformation. Also, as techniques improve, we are able to measure the very minute changes brought about by these transformations.

When nuclear transformations occur at a rate which is known or can be determined, the magnitude of transformation in a sample of matter becomes a measure of the time for which matter has been subjected to transformation. In other words, a nuclear clock resides in matter. If one knows enough about nuclear processes, one can read a number of natural clocks resident in common matter, and, thereby study its history.

In this article I have dealt with nuclear transformations caused by cosmic-ray particles in extraterrestrial and terrestrial samples. I have elaborated on studies of terrestrial cosmogenic nuclides produced in the atmosphere and in situ in terrestrial solids. Recent improvements in methods of detection of nuclides at levels of  $\sim 10^5$  atoms have opened up the field of study of isotopic changes in terrestrial solids, and cosmogenic nuclides have thus begun to be useful for the study of a variety of geophysical processes.

In terrestrial samples, the isotopic changes produced by cosmic rays can now be studied up to depths of  $\sim 10^6 \, \mathrm{g \, cm^{-2}}$  below the sea level, deeper than in the case of meteorites ( $\sim 10^3 \, \mathrm{g \, cm^{-2}}$ ). The reason for this is that pi-mesons produced in the earth's atmosphere decay to 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<sup>93</sup>, and in civil engineering, to determine the overburden in different directions.

The incident flux of cosmic rays on the earth is small compared to solar-energy flux or heat flow from the interior of the earth (Table 4), but it is of significance because of its high-specific energy.

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## Emanation of radon from rock minerals

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The mechanism of emanation of radon from terrestrial minerals and rocks is briefly discussed. Employing a technique which can locate nanometre-wide holes in materials, I have observed a network of very narrow (sub-micronic) openings/gaps within an individual mineral grain and between grains of different minerals. Radon enters this network by recoil-out from the solid walls, and is then subject to rapid transport by molecular diffusion in the air in the network.

Most of the natural minerals contain small amounts of uranium and thorium, and their decay products. Amongst these, <sup>222</sup>Rn, <sup>220</sup>Rn and <sup>219</sup>Rn arise from decay of <sup>226</sup>Ra (<sup>238</sup>U series), <sup>224</sup>Ra (<sup>228</sup>Th series) and <sup>223</sup>Ra (<sup>235</sup>U series) respectively. Their half-lives are 3.8 d (<sup>222</sup>Rn), 55 s (<sup>220</sup>Rn) and 4 s (<sup>219</sup>Rn). It has been known for long that somehow a sizable fraction of these three gaseous isotopes produced inside the terrestrial minerals is able to emerge out of the minerals into the pores from where it diffuses out into the atmosphere.

The inert gaseous isotopes of Rn have found numerous applications as tracers for investigating geophysical and geochemical phenomena. In some of these applications, a knowledge of the exact nature of the mechanism by which Rn atoms emerge out of crystalline minerals is important. Attempts have therefore been made to understand this mechanism which further happens to be relevant in choosing appropriate remedial measures for controlling levels of indoor Rn, a matter of general concern these days.

The basic mechanism was worked out by Zimens in the late thirties and the early forties. But certain aspects of the phenomenon still require unravelling and details remain to be worked out. A review article by Tanner<sup>1</sup> gives a comprehensive treatment of the subject. But there has been substantial progress since then. I wish to bring that out in this article. Here I have drawn heavily from the work of Rama and Moore<sup>2-4</sup>.

#### Recoil-out

On decay, a nucleus of <sup>226</sup>Ra, <sup>224</sup>Ra or <sup>223</sup>Ra gives out an alpha particle and the residual nucleus of <sup>222</sup>Rn, <sup>220</sup>Rn or <sup>219</sup>Rn recoils with equal momentum in the opposite direction. The kinetic energy of the recoiling nucleus is in the 100 keV range, and differs slightly for the three isotopes; the respective ranges of the three recoiling isotopes in common rock minerals are about 60, 80 and 90 nm.

The recoiled Rn atoms loses its energy by atomic collisions inside the mineral, and then comes to rest within the mineral (Figure 1). There is no possibility of its coming out of the mineral since the diffusion coefficient of Rn in crystalline mineral is extremely small. However, there is a good chance for Rn atoms to be directly recoiled out of the mineral if they are generated in the skin of the mineral, i.e. within a depth equal to the recoil range from the surface (Figure 2). Considering the geometrical factors involved, one out of four such Rn atoms is expected to recoil out of the