Chapter 12
Origin and Production of Cosmogenic Radioisotopes

12.1 Composition of Cosmic Radiation in the Earth's Atmosphere

The Earth's atmosphere is penetrated by a continuous flux of charged particles, constituting of protons and nuclei of various elements of cosmic origin. Consequently, a great variety of radioisotopes, referred to as cosmogenic, are produced due to the interaction of these particles with the atomic nuclei of elements which constitute the atmosphere. Transported by air masses, radioisotopes are abundant over the whole gaseous sphere of the Earth. Being mixed with atmospheric moisture, a proportion falls over the Earth's surface, to enter the hydrological cycle as components of surface waters, soil-ground moisture, and groundwaters. Another proportion becomes a component of ocean and inland basin waters through exchange at the surface of water reservoir. Finally, the Earth's biosphere plays an active role in exchange processes, which are of great importance for some cosmogenic isotopes.

Cosmic dust is another source of cosmogenic isotopes, as are meteorites which are continually falling on the Earth's surface. Having been in cosmic space these meteorites have been subjected to bombardment by cosmic radiation. Nuclear reactions accompanying the process produce many radioisotopes.

Cosmic radiation plays the main role in the origin of cosmogenic radioisotopes. Understanding the nature of cosmic radiation has been important in development of the Earth sciences. In particular, the solution for a number of hydrological and hydrogeological problems, related to natural water dynamics, their genesis, and age, has become possible due to investigations of abundances of cosmogenic radioisotopes in the hydrosphere, i.e., isotopes, produced by cosmic radiation.

Cosmic rays of solar and galactic origin are distinguishable. The nature of nuclear particle flux of solar origin is related to fusion reactions in the solar interior. The origin of galactic and metagalactic cosmic radiation bombarding the Earth's atmosphere is still uncertain.

In the practical studies of cosmic radiation there have been cases when particles of galactic origin with energies of 10^{19}–10^{20} eV[1] have been detected. Some idea of

[1] eV, electron-volt is a unit of energy used in nuclear physics and equal to 1.6×10^{-19} Joules.

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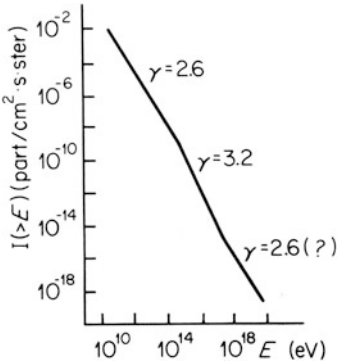


Fig. 12.1 Energy spectrum of primary cosmic radiation

the spectrum of cosmic radiation with energy greater than $E = 10^{10}$ eV is given by Fig. 12.1.

Cosmic radiation is made up of about 90% protons, about 9% helium nuclei (α-particles), and about 1% other nuclei. Table 12.1 shows specific abundances of nuclei in the solar system, cosmic space, and in cosmic radiation (Webber 1967).

Except for hydrogen and helium, the composition of primary cosmic radiation is poorly understood. The estimations of specific abundances of carbon isotopes show that in primary cosmic radiation $^{13}C/^{12}C \approx 1$. Measuring this ratio with the help of photo-emulsion techniques, a value close to unity was obtained.

As for deuterium, no sufficiently reliable measurements of its content in primary cosmic radiation have been carried out so far. Those measurements which were carried out in the upper atmosphere (for atmospheric depths 2–4 g/cm^2) gave values of $^2H/^1H = 0.05$–0.12. The large discrepancy in these data results from the fact that the measurements correspond to different energy intervals and latitudes. However, on the whole, the data are in agreement with satellite data ($^2H/^1H \leq 0.06$ or $\varepsilon = 25$–80 MeV/nucleon), where the effect of the Earth's magnetism and atmosphere are excluded (MeV = 10^6 eV).

The ratio of deuterium abundance to proton abundance in the Universe is of the order of 1.4×10^{-4} (Webber 1967), which may be accounted for by its disintegration. According to spectroscopic measurements, the ratio $^2H/^1H$ is also small in the solar atmosphere, amounting to 4×10^{-5} (Kinman 1956). Only in the atmosphere of magnetic stars does the ratio increase up to 10^{-2}. The ratio of $^2H/^1H = 10^{-5}\rho$, where ρ (particle/cm^3), is the average density of substance through which cosmic radiation has passed (Singer 1958).

While studying the helium isotope composition in cosmic radiation (Appa Rao 1962), the ratio $^3He/(^3He + ^4He) = 0.20$–$0.30$ has been obtained in the energy range of $\varepsilon = 160$–360 MeV/nucleon. Attempts to measure this ratio for higher energies have failed.

Besides nuclei, α-particles, and nuclides of various elements and their isotopes, primary cosmic radiation also contains gamma-ray protons, neutrons, electrons, and

Table 12.1 Nuclei abundances in the solar system, cosmic space, and in primary cosmic rays relative to carbon. (©IAEA, reproduced with permission of IAEA)

Element	Abundances			Element	Abundances		
	Solar system	Cosmic space	Primary cosmic rays		Solar system	Cosmic space	Primary cosmic rays
He	?	400	38	Al	0.004	0.005	0.03
Li	$\ll 0.001$	$\ll 0.001$	0.27	Si	0.063	0.13	0.11
Be	$\ll 0.001$	$\ll 0.001$	0.19	P	–	0.002	0.01
B	$\ll 0.001$	$\ll 0.001$	0.43	$16 \leq Z \leq 19$	0.050	0.02	0.02–0.05
C	1.0	1.0	1.0	Ca	–	$\ll 0.001$	0.026
N	0.16	0.27	0.46	Ti	–	0.006	0.017
O	1.7	2.3	0.61	Ci	–	0.06	0.030
F	$\ll 0.001$	$\ll 0.001$	0.09	Fe	–	0.06	0.080
Ne	?	0.80	0.18	Ni	–	0.008	0.015
Na	0.004	0.006	0.08	$Z > 20$	–	$\ll 0.001$	0.01
Mg	0.005	0.12	0.15	$Z > 30$	–	~ 0.001	< 0.004

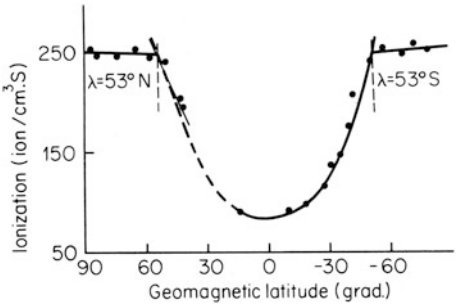


Fig. 12.2 Latitude effect of cosmic radiation for the depth of atmosphere at 50 g/cm^2

positrons. The source of gamma-ray protons and neutrons in primary cosmic radiation is supposed to be related to supernova.

Primary cosmic radiation interacts with the Earth's magnetic field, which results in a relationship between the intensity of radiation and geomagnetic latitude. All available data concerning geomagnetic effects of cosmic radiation are in good agreement with the idea that the isotropic flow of charged particles coming from the Universe is deviated by the terrestrial magnetic field. Analyzing the effect of the Earth's magnetic field on the motion of charged particles, favored and unfavored directions of charged particle motion to a given point of the Earth's surface have been found.

The interaction of the Earth's magnetic field with primary cosmic radiation results in latitude variations of its intensity I (Fig. 12.2). This effect, referred as the latitude effect, is quantitatively expressed by the ratio $[I(90°)–I(0°)]/I(90°)$.

It has been found that the intensity of cosmic radiation coming to the Earth varies with time. Four different types of these time-dependent variations are known (Fireman 1967):

1. Variations related to the 11-year cycle of the solar activity. With increasing intensity of the solar particle flux the galactic radiation intensity decreases.

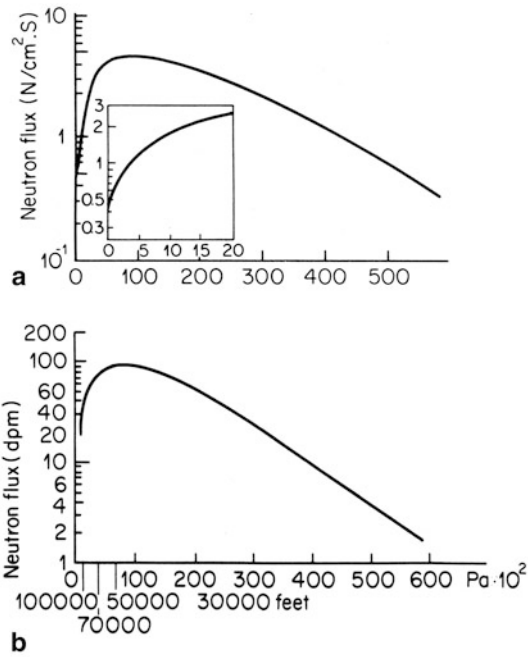
2. Heliocentric variations of galactic radiation, the radial gradient of which in the range of 1.0–1.5 a.u. of length (1 a.u. $\approx 1.5 \times 10^8$ km), is about + 9.6% per 1 a.u.
3. The secular variations of radiation were detected by measuring ^{14}C content variations in the atmosphere through different centuries and recorded in the annual rings of the trees emitting radiations at various ages.
4. The sporadic flows of nuclear particles of low energies emitted during solar flares.

The intensity of the flux of protons of solar origin, with energies greater than 10 MeV near to the Earth, is equal to 100 protons/cm^2 s during a typical solar cycle (relative to the Earth's surface) (Lal et al. 1967). This value was obtained using experimental data on the rate of ^{26}Al production during a time interval of about 10^5 year. The intensity of protons in the solar flux is higher than the galactic intensity by about one order of magnitude, i.e., the latter is characterized by a value of about 10 protons/cm^2 s, but while the solar protons have energies of about several dozens of MeV, the protons of galactic origin have energies two orders greater on average. The intensity of α-particle flux, both of solar and galactic origin, is lower than of protons by one order of magnitude. The velocity of nuclear particles of solar origin near the Earth is about 300 km/s.

12.2 Composition and Steady-state Abundances of Cosmogenic Radioisotopes in the Outer Shells of the Earth

In the course of the interaction of high-energy cosmic radiation with the atmosphere the major part of its energy is absorbed and scattered by the Earth's atmosphere. This leads to the production of secondary low-energy radiation composed of mesons, gamma-ray photons, positrons, and other particles of various energies. This secondary radiation is mainly composed of less energetic protons and neutrons which play the main role in nuclear reactions, resulting in the production of cosmogenic radioisotopes in the Earth's atmosphere. The energy threshold of these reactions ranges between 10 and 40 MeV.

The distribution of secondary neutrons and protons in the atmosphere varies both in latitude and altitude. Figure 12.3a shows the experimental data of energy distribution in the neutron flux, characterized by energies lower than 20 MeV, obtained in 1966 during an experiment in Sicily during a quiet Sun period (Boella et al. 1968). A similar thermal neutron distribution was obtained by Korff using a thermal neutron counter in the Princeton region (USA). The data of these measurements are shown in Fig. 12.3b. One can see from the figure that a total flux of neutrons at first increases with altitude and then decreases with atmospheric density due to their escaping from high atmospheric layers. With the transition in latitude from the equator to the poles the density of neutron flux increases. Figure 12.4 shows the experimental data of latitude dependency of neutron flux at an altitude of 1,000 m, obtained by Simpson (Libby 1967).



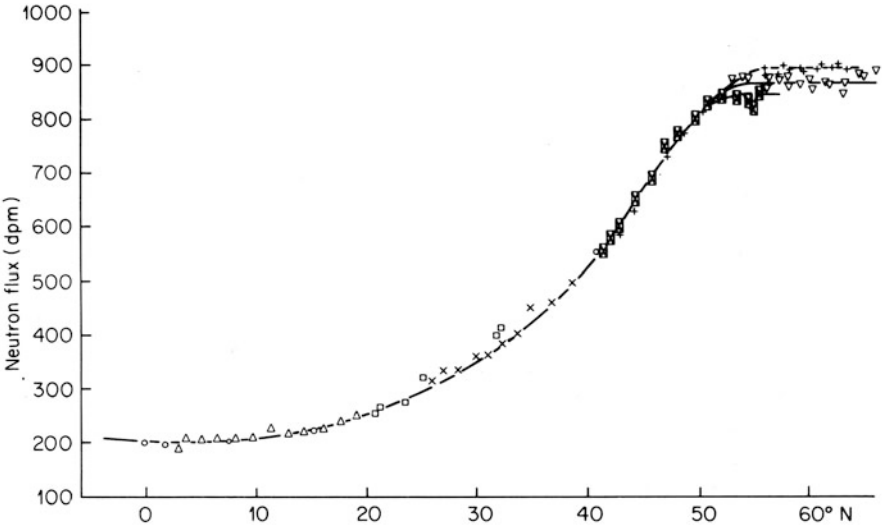


Fig. 12.3 Relation of integrated neutron flux from atmospheric depth at 0–600 mbar in Sicily for neutron energy lower than 20 MeV (**a**) and for thermal neutrons in Princeton region (**b**)

Fig. 12.4 Latitude effect in distribution of neutron flux at an altitude of 1,000 m for the North American continent. (After Libby 1967)

The effect of the production of cosmogenic isotopes in the Earth's crust is negligible even in the upper layers. Nuclear transformations occur here mainly due to the penetration of primary nucleons of light energies, thermal neutrons, and $\overline{\mu}$-mesons. The dominating component of this flux varies with depth. At a depth of several meters most nuclear transformations are provided by quick $\overline{\mu}$-mesons, and at depths where pressure is greater than 400 kg/cm^2, by the interaction with neutrino flux of both primary and secondary origin (Lal and Peters 1967).

According to Lal and Peters, at depths characterized by pressures greater than 0.7 kg/cm^2, the majority of nuclear transformations occur due to interaction with $\overline{\mu}$-mesons and 10% of them are the effect of negative meson capture.

The main proportion of cosmogenic radionuclides is formed in the atmosphere. The main components of atmospheric air are: nitrogen (78.09%), oxygen (20.95%), argon (0.93%), carbon dioxide (0.03%), and neon (0.0018%). In the course of interaction between cosmic radiation and atmospheric elements nuclear transformation occur with nuclei of neon, oxygen and argon, which play a major role in the production of cosmogenic radioisotopes. The most typical reactions are: (n, p), $(n, 2n)$, (n, α), (n, γ), (p, n), $(p, 2n)$, (p, pn), $(p, 2p)$.

Isotopes of cosmogenic origin, which are of interest for the investigations of water circulation patterns in the hydrosphere, are: ^{3}H, ^{3}He (stable), ^{7}Be, ^{10}Be, ^{14}C, ^{22}Na, ^{24}Na, ^{26}Al, ^{32}Si, ^{32}P, ^{33}P, ^{36}Cl, ^{37}Ar, ^{39}Ar, and ^{81}Kr. The irradiation of cosmic dust falling down in amounts of about 10^9 kg/year on the Earth's surface, by the cosmic radiation results in the production of such radioisotopes of great importance as ^{10}B, ^{14}C, ^{22}Na, ^{26}Al, ^{36}Cl, ^{39}Ar, ^{53}Mn, ^{59}Ni.

The rate of production of cosmogenic isotopes varies with altitude and latitude and depends on the intensity of secondary neutron and proton flux in the atmosphere. At the same time the rate of their formation remains constant over time. As a first approximation the ratio of various isotopes produced may be considered to be independent of altitude and time. According to estimations by Lal and Peters (1962) and Young et al. (1970) the rate of isotope production per gram of air, for example, at 46°N, increases exponentially at an altitude of about 21 km is three orders of magnitude higher than at the surface of the Earth. Further on it decreases with altitude due to escaping neutrons from the upper atmospheric layers. The isotope production rate increases by about one order of magnitude with transition from the equator to the poles.

Estimations of the production rate of cosmogenic radioisotopes in the atmosphere and in cosmic dust, carried out by a number of authors (Bhaudari et al. 1969; Craig and Lal 1961; Lal and Paters 1962; Lal and Venkatavaradan 1967; Lal et al. 1967; Schell 1970), used a value of average density of proton flux with energy $E > 10$ MeV at the upper boundary of the Earth's atmosphere, equal to 100 protons/cm^2 s. The latitude and altitude effects of nuclear reactions were taken into account in the course of the calculations.

In order to carry out estimations of steady-state amounts of individual isotopes on the Earth, it was assumed that the flux of cosmic radiation has remained constant over a long period of time (being not less than the half-life of a given radioisotope).

With a constant cosmic flux and a certain atmospheric composition the production rate of isotopes will also be independent of time.

The steady-state amount of an individual isotope on the Earth follows from the equation

$$nS = q\lambda,$$

where n is the production rate of a radioisotope relative to the Earth's surface (atom/cm^2 s); S is the area of the Earth's surface, cm^2; q is the steady-state amount of isotopes or atoms; λ is the decay constant of a given nucleus.

The results of the estimations carried out by the above-mentioned authors and clarified by experimental research, related to the production rates and steady-state abundances of various cosmogenic radioisotopes and also their main physical properties, are presented in Table 12.2.

The total steady-state amount of cosmogenic isotopes on the Earth depends on the balance between their production rate in the atmosphere, accumulation over their life, and reduction due to radioactive decay.

The main proportion of isotopes (except of the noble gases) is oxidized immediately after production. Among these oxides only carbon dioxide and tritium occur in the atmosphere in a free form. The other isotopes are absorbed by aerosols after a short time after of their formation. Radioisotopes, contained in aerosols, are removed from the atmosphere fairly quickly by condensation of moisture in low tropospheric layers, whereas those isotopes which remain constant gaseous components of the atmosphere (CO_2, Ar, Kr) are removed from it far more slowly by molecular exchange at the boundary between atmosphere and oceans.

The ability of cosmogenic isotopes to the study of hydrological and hydrogeological processes is restricted by the condition of correspondence between their lifetimes and the duration of a considered process, and also by the principles of their displacement in geospheres. The major proportion of cosmogenic isotopes ($\sim$70%) is formed in the upper atmospheric layers, about 30% of them in the troposphere. There sequential redistribution is caused by large-scale processes involving the motion of air masses in the troposphere and the precipitation of atmospheric moisture on the Earth's surface, together with cosmic dust or in the form of aerosols.

The absolute amount of an individual isotope, or its ratio relative to another radioactive or stable isotope within a natural reservoir, is used for dating or reconstructing events which have taken place in the past and also for elucidating the nature of prevailing physical, chemical, and biological processes. Resolution of the investigation of these processes depends largely on the information available concerning the source and rate of production of a radionuclide.

As pointed out earlier, the production rate of a cosmogenic isotope in the atmosphere depends on latitude and altitude. Comparison of the expected production rate with those amounts actually observed in the air for isotopes with adequate lifetimes provides a basis for studying the principles of large-scale circulations and tropospheric fallout. The abundance of an isotope in the hydrosphere depends on its lifetime, its biochemical role, and, finally, on the nature of oceanic circulations.

Table 12.2 Physical parameters and steady-state amounts of cosmogenic radioisotopes on the Earth

Isotope	Production reaction	Half-life	Radiation type and energy product (MeV)	Decay product	Production rate (atom/cm^2 s)	Steady-state amount on the Earth (g)
^{3}H	^{14}N(n, ^{14}C)^{3}H ^{16}O(P, ^{14}H) ^{3}H	12.32 year	β−; 0.018	^{3}He	0.25	3,500
^{3}He	N, O(^{3}He)	stable		–	0.2	3.2×10^{9}
^{7}Be	^{14}N(n, 3p5n)^{7}Be ^{14}N(p, 4p4n)^{7}Be ^{16}O(n, 5p5n)^{7}Be	53 day	γ; 0.48	^{7}Li	8.1×10^{-3}	3.2
^{10}Be	^{14}N(p, 4pn)^{10}Be ^{16}O(p, 5p2n)^{10}Be	1.6×10^{6} year	β−; 0.55	^{10}B	4.5×10^{-2}	4.8×10^{8}
^{14}C	^{14}N(n, p)^{14}C ^{16}O(p, 3p)^{14}C	5,730 year	β−; 0.156	^{14}N	2.5	7.5×10^{7}
^{22}Na	^{40}Ar(split)^{22}N	2.6 year	β+; 0.54 γ; 1.28	^{22}Ne	8.6×10^{-5}	1.9
^{24}Na	^{40}Ar(split)^{24}N	15 hour	γ; 2.75 β−; 1.4	^{24}Mg	1.2×10^{-4}	–
^{26}Al	^{26}Mg(p, n)^{26}Al ^{26}Si(p, 2p)^{26}Al	7.4×10^{5} year	β+; 2.77	^{26}Mg	1.4×10^{-4}	1.2×10^{6}
^{28}Mg	^{40}Ar (split)^{28}Mg	21.3 hour	β−; 0.42	^{28}Al	5.2×10^{-5}	–
^{32}Si	^{40}Ar (split)^{32}Si	~450 year	β−; 0.1	^{32}P	1.6×10^{-4}	1,400
^{32}P	^{40}Ar (split)^{32}P	14.3 day	β−; 1.7	^{32}S	8.1×10^{-4}	0.4
^{33}P	^{40}Ar (split)^{33}P	25 day	β−; 0.25	^{33}S	6.8×10^{-4}	0.6
^{35}S	^{40}Ar (split)^{35}S	87.4 day	β−; 1.67	^{35}Cl	1.4×10^{-3}	4.5
^{36}Cl	^{40}Ar (split)^{36}Cl	3.0×10^{5} year	β−; 0.714	^{36}Ar	1.1×10^{-3}	1.5×10^{6}
^{37}Ar	^{37}Cl (p, 2p)^{37}Ar ^{40}Ca (n, α)^{37}Ar ^{36}Ar (n, γ)^{37}Ar	35 day	K-capture	^{37}Cl	8.3×10^{-4}	–
^{39}Ar	^{40}Ar (n, 2n)^{39}Ar ^{39}K (n, p)^{39}Ar ^{38}Ar (n, γ)^{39}Ar	270 year	β−; 0.565	^{39}K	5.6×10^{-3}	–
^{53}Mn	^{53}Fe (p, 2p)^{53}Mn ^{56}Fe (p, α) ^{53}Mn	3.7×10^{6} year	K-capture	^{53}Cr	$< 10^{-7}$	–
^{59}Ni	^{59}Co (p, n)^{59}Ni ^{60}Ni (p, pn)^{59}Ni	7.5×10^{4} year	K-capture	^{59}Co	$< 10^{-7}$	–
^{81}Kr	^{82}Kr (p, n)^{81}Kr	8.1×10^{5r} year	K-capture	^{81}Br	1.5×10^{-7}–10^{-5}	–

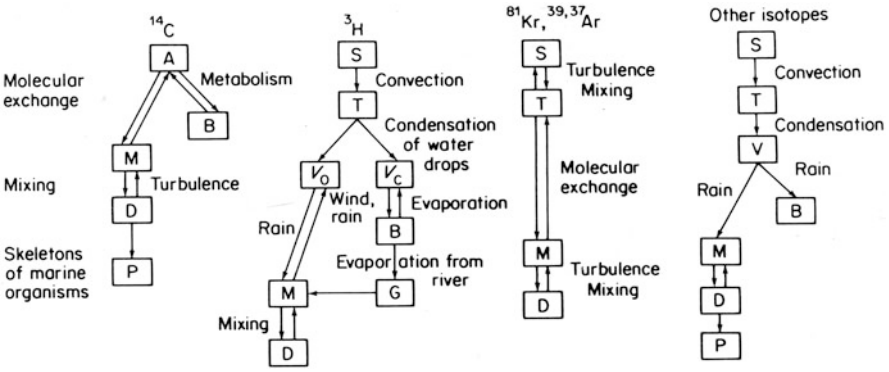


Fig. 12.5 Schematic description and migration of cosmogenic isotopes in the geosphere: (*A*) atmosphere; (*B*) biosphere; (*S*) stratosphere (90–340 g air per 1 cm^2, 1–8 year exchange time); (*T*) troposphere (720–940 g air per 1 cm^2, 30–90 day exchange time); (*V*) water vapor (3–7 g water per 1 cm^2, 4–14 day exchange time); (*M*) mixing layer (75–100 m, 20 year exchange time); (*D*) deep layer (3,500 m, 500–3,000 year exchange time); (*P*) marine sediments; (*G*) soil water. (After Lal and Peters 1962)

Experimental study of abundance of isotopes, characterized by different lifetimes and chemical properties, provide an opportunity for understanding the principles of mass-transfer of substance and, in some cases, helps in understanding the geochemistry of an element's behavior. Isotopes which have got into the biosphere, into oceanic floor sediments, and into some other objects, thus becoming isolated from the transitional dynamics in the cycle (which is characterized by a continuous process of mixing of substance), can be used to determine the time elapsed since the moment they escaped from the cycle.

12.3 Distribution of Cosmogenic Radioisotopes in the Exchange Reservoirs

While studying the nature of geophysical and geochemical processes, which effect the distribution of various isotopes, it is convenient to divide the atmosphere and upper layers of the Earth into a number of zones or reservoirs, characterized by homogeneous pressure, temperature, and character of mass-transfer. A schematic description of isotope migration in natural reservoirs is presented in Fig. 12.5 (Lal and Peters 1962).

The rate of isotope production caused by cosmic radiation in ocean waters is negligible. The detectable concentrations of all the isotopes found in these waters are the result of exchange between the atmosphere and the oceans. The upper layer of the ocean, receiving cosmogenic isotopes from the atmosphere, is characterized by vigorous movements and quick mixing. It is due to these processes that the geographical inhomogeneity of the cosmic radiation is markedly smoothened. Therefore, as

Table 12.3 Steady-state inventories and decay rates of cosmogenic radioisotopes in the exchange reservoirs relative to 1 cm^2 of the Earth's surface. (From Lal 1963. ©IAEA, reproduced with permission of IAEA)

Radioisotope	Exchange reservoir				
	Stratosphere	Troposphere	Mixed oceanic layer	Deep oceanic layer	Ocean sediments
^{3}H	6.8×10^{-2}	4×10^{-3}	0.50[a]	0.43	0
^{7}Be	0.60	0.11	0.28	3×10^{-3}	0
^{10}Be	3.7×10^{-7}	2.3×10^{-8}	8×10^{-6}	1.4×10^{-4}	0.999
^{14}C	3×10^{-3}	6×10^{-2}[b]	2.3×10^{-2}	0.91	10^{-2}
^{22}Na	0.25	1.7×10^{-2}	0.62	0.11	0
^{26}Al	1.3×10^{-6}	7.7×10^{-8}	5×10^{-5}	10^{-4}	0.999
^{32}Si	1.9×10^{-3}	1.1×10^{-4}	5×10^{-3}	0.96	4×10^{-2}
^{32}P	0.60	0.24	0.16	1.5×10^{-4}	0
^{33}P	0.64	0.16	0.19	10^{-3}	0
^{35}S	0.57	8×10^{-2}	0.34	6×10^{-3}	0
^{36}Cl	10^{-6}	6×10^{-8}	2×10^{-2}	0.98	0
^{37}Ar	0.63	0.37	0	0	0
^{39}Ar	0.16	0.83	2×10^{-4}	3×10^{-3}	0

[a] The value includes amount presents in biosphere and humus.
[b] Includes amount present in the continental waters.

a first approximation, the source function of the ocean is equal to zero at depth and constant at the surface. This approximation is accurate for ^{14}C dissolved in water and mixed together with water masses. However, there are some other isotopes, such as ^{32}Si and $^{32,33}P$, which are not displaced by water masses. The major portion of them is assimilated by fauna in the upper layers of the ocean. These isotopes enter deep parts of the ocean partly in response to water mixing and partly in the form of organic remains which interact with the environment before sedimentation at the ocean floor. Therefore, the source function of ^{32}Si and $^{32,33}P$ isotopes depends on latitude, longitude, and depth of the ocean (Lal and Peters 1962).

The use of cosmogenic isotopes in the solution of practical problems is more easily realized if the theoretical data related to their abundances and decay rates in the major reservoirs of the Earth, where their migration occurs, are available. For these calculations which were carried out by Lal (1963), the oversimplified box-model (see Fig. 12.5) was assumed. The model consists of six reservoir exchange system: (1) stratosphere (170 g/cm^2), (2) troposphere (860 g/cm^2), (3) continental reservoir (biosphere + surface waters), (4) the upper mixed ocean layer (75 m), (5) deep oceanic layer (3,500 m), and (6) ocean sediments. The calculations were carried out by using the following simplified constants: the average time of air exchange in the stratosphere is equal to 2 years and the average time of water exchange in the deep oceanic layer is equal to 1,000 years (Table 12.3).

The distribution of an isotope in individual geospheres depends both on its half-life and chemical behavior. The usefulness of an isotope in hydrological and hydrogeological process studies depends on its half-life, distribution in exchange reservoirs,

Table 12.4 Specific activity of cosmogenic radioisotopes in the oceans and atmosphere. (From Lal 1963. ©IAEA, reproduced with permission of IAEA)

Radio-isotope	Average specific activity in oceans		Radio-isotope	Average specific activity in atmosphere, disintegration $min^{-1}kg^{-1}$ air	
	Disintegrations min^{-1} t^1 water	Disintegrations min^{-1} t^1 water		Stratosphere	Troposphere
^{3}H	36	3.3×10^{-4}	^{3}H	6	7×10^{-2}
^{10}Be	10^{-3}	1.6×10^{-3}	^{7}Be	17	0.63
^{14}C	260	10	^{22}Na	5×10^{-3}	6.7×10^{-5}
^{26}Al	2×10^{-5}	2×10^{-3}	^{32}P	0.17	1.4×10^{-2}
^{32}Si	2.4×10^{-2}	8×10^{-3}	^{33}P	0.15	7.6×10^{-3}
^{36}Cl	0.55	3×10^{-5}	^{35}S	0.28	7.8×10^{-3}
^{39}Ar	2.9×10^{-3}	5×10^{-3}	^{37}Ar	0.19	2.1×10^{-2}

and the technical ability required to measure the expected activity. The specific activities of a number of cosmogenic radioisotopes in the two principal exchange reservoirs, those of the ocean and atmosphere, were evaluated by Lal (1963) and are presented in Table 12.4.

It follows from Tables 12.3 and 12.4 that the major portion of such isotopes as ^{10}B and ^{26}Al is accumulated in the ocean sediments whereas ^{3}H, and ^{7}Be and many other isotopes are absent. The greatest portions of ^{7}Be, ^{32}P, ^{33}P, ^{35}S, ^{37}Ar and ^{39}Ar are concentrated in the stratosphere and troposphere. The surface ocean layer and inland waters contain the majority of cosmogenic isotopes in significant amounts. Their concentration in ground waters depends on the conditions of interrelation between surface waters and groundwaters. Only those isotopes whose lifetimes are longer than that of infiltrated surface water recharge groundwaters. The varying proportions of concentrations of a given cosmogenic isotope in the water-bearing layer and its steady-state amount in precipitation, surface waters, or in the overlying water-bearing layer, provide a basis for studying processes of water circulation and the hydrochronology of groundwaters.

An additional source of isotopes in the atmosphere is cosmic dust, the terrestrial accretion rate of which is about 10^{8}–10^{9} kg/year. The use of some cosmogenic isotopes, being components of the cosmic dust, provides information on the accretion rate of the Earth in the past. Such isotopes are ^{53}Mn and ^{59}Ni, which are not produced in the Earth's atmosphere, and ^{26}Al, the production rate of which in the atmosphere is less than the contribution due to cosmic dust.

Changes in the isotopic composition of cosmic dust and in the outer shells of meteorites result mainly from bombardment by low-energy cosmic particles of solar origin. The outer shell of a meteorite is usually melted and ablated during movement through the atmosphere. Therefore, while studying the total flux of solar cosmic rays, and particularly low-energy protons, the most convenient object is cosmic dust accumulated in the ocean floor sediments and polar pack ices.

It should be pointed out that during the last two decades the concentration of the steady-state amounts of cosmogenic radioisotopes in nature has been broken due to the additional production of these isotopes during the course of nuclear and

thermonuclear tests in the atmosphere. An intense flux of neutrons is produced at the moment of explosion, which interact with the atmospheric constituents and result in the production of identical radioisotopes to those produced by the interaction between the atmosphere and cosmic rays (Styro 1968).

The International Atomic Energy Agency (IAEA Bulletin 1973) has reported that from 1945 to 1973, 936 nuclear tests were carried out, of which 422 took place in the atmosphere. The majority of those tests were before 1963. During the last decade, 43 tests took place in the atmosphere. The most powerful output of bomb radioisotopes in the atmosphere took place during 1958–1959, i.e., related to the most frequent and most powerful thermonuclear tests. The concentrations of some radioisotopes have increased compared with the prethermonuclear steady-state values by one or two orders of magnitude. Thus, the major portion of ^{3}H, ^{14}C, and ^{22}Na present in the atmosphere at present is of bomb origin. Steady-state level of concentration of a number of short-lived isotopes, such as ^{35}S and to a lesser extent ^{7}Be and ^{32}P have been distributed but at present their concentration has returned to normal. The distribution of ^{14}C, and to a great extent ^{36}Cl and ^{81}Kr was also distributed.

The effect of these bombs will be manifested as a distinctive ‘mark’ for a long time in those reservoirs where the processes of water mixing and dilution are slow, such as in the groundwater reservoir. These marks may serve as a good indicator of groundwater motion and also of the individual water-bearing layers between each other and with surface waters.