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COSMIC-RAY PRODUCED RADIONUCLIDES IN THE ENVIRONMENT*

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Abstract—Radionuclides are continually produced by interactions of cosmic rays with the atmosphere, hydrosphere and lithosphere. Most of the studies of these radionuclides are concerned with those produced in the atmosphere or in meteorites. These radionuclides result mainly from spallation reactions with atmosphere constituents or with iron in the case of meteorites; however, ^{14}C is produced by the thermal neutron capture of nitrogen. The production rates of these radionuclides in the atmosphere are strongly dependent on both altitude and geomagnetic latitude. The uses of the radionuclides ^{14}C and tritium in radioactive dating, atmospheric and ocean transport, and other studies are well known. While the radionuclides ^{14}C, ^{7}Be, ^{10}Be and tritium are produced from the light element constituents of the atmosphere, numerous radionuclides are produced by spallation reactions on atmospheric argon. To date the radionuclides ^{22}Na, ^{24}Na, ^{32}Si, ^{32}P, ^{33}P, ^{35}S, ^{38}S, ^{36}Cl, ^{38}Cl and ^{39}Cl have been reported. The meteoritic dust which is continually showered into the atmosphere contains spallation products of iron and other cosmic ray induced radionuclides. The concentrations of most of the radionuclides are extremely low and in some cases their artificial production by weapons testing has greatly exceeded their natural production. The biosphere is continually exposed to the wide spectrum of cosmic-ray produced radionuclides and some of these which are concentrated by biological processes can serve as tracers in studies of the biological processes in nature.

INTRODUCTION

RADIONUCLIDES are produced continuously in the atmosphere by cosmic rays. The production rates of these radionuclides vary appreciably with both altitude and latitude but are relatively constant with time. The half-lives of the cosmic-ray produced radionuclides cover a range from minutes (or less) to several million years, and more than a dozen have been observed. The discovery and use of these radionuclides as tracers in geophysical and other processes was made possible by development of very sensitive analytical methods. Several reviews of the cosmic-ray produced radionuclides discuss their discovery, their theoretical and observed production rates, their distribution, and their use as tracers in studying and dating geophysical and other processes.[1–5] From the nature of cosmic-ray interactions with atmospheric constituents it is known that several radionuclides are being produced which have not yet been observed. During the past 2 years, three cosmic-ray produced radionuclides were observed for the first time[6,7] and it is expected that several others will be found as needs for their measurement arise.

Almost all of these radionuclides are produced in the atmosphere; however, some cosmic rays reach the earth and cause radionuclide production in the earth's surface. In addition, approximately 10^4 tons of extraterrestrial dust which may have been subjected to cosmic radiation in space for aeons showers into the atmosphere daily. This material contains numerous radionuclides but their total contribution is extremely small and measured values for the radionuclides thus deposited on the earth have not yet been reported.

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PRODUCTION AND DISTRIBUTION OF COSMIC-RAY PRODUCED RADIONUCLIDES

The primary interactions of high energy cosmic rays with the atmosphere produce numerous secondary neutrons and protons of sufficient energies to produce spallation, or other nuclear reactions with atoms in the atmosphere. The radionuclides which are thus produced in the atmosphere and have been discovered are listed in Table 1. The production of these radionuclides results mainly from the interactions of the secondary protons and neutrons with nitrogen, oxygen and argon nuclei in the atmosphere. The formation of most of these radionuclides in the atmosphere involves the emission of only a few protons, neutrons or alpha particles; the threshold for such reactions is in the range of 20–40 MeV. Among the secondary nucleons in this energy range the neutrons greatly exceed the protons (because of ionization losses) and production of radionuclides is proportional to the intensity of neutrons below a few hundred MeV. Since neutrons are thermalized locally the radionuclide formation rate is also proportional to the thermal neutron flux, except near the top of the atmosphere where neutrons escape.

Although all of the radionuclides in Table 1 can be formed by spallation, other mechanisms are important in the formation of ^{3}H, ^{14}C, ^{36}Cl and ^{39}Cl. The formation of ^{14}C is mainly due to the reaction $^{14}N(n, p)^{14}C$ by neutrons with energies below 2 MeV. Tritium may be formed by the reaction $^{14}N(n, t)^{12}C$ with a threshold of only 4.4 MeV. ^{36}Cl may be produced by thermal neutron capture of ^{35}Cl and is produced in that manner in the ocean and on land. The production of ^{39}Cl from atmospheric argon can occur with slow μ^- meson capture, $^{40}A(\mu^-, n)^{39}Cl$, and this reaction becomes important near sea level.[8] Several of the radionuclides in Table 1 are also produced during nuclear weapons testing. The radionuclides T, ^{7}Be, ^{14}C, ^{22}Na, ^{32}P, ^{33}P, ^{35}S and ^{36}Cl are known to be produced during weapon testing. For some of these, contributions from weapon testing currently exceed that due to cosmic ray production.

The relative production rates of radionuclides in the atmosphere by spallation or neutron capture depend on both altitude and latitude. The rate of occurrence of nuclear disintegrations (which is proportional to radionuclide production rates except as mentioned above) has been calculated for the different regions of the atmosphere by LAL *et al.*[2,9] In the equilibrium region of the atmosphere (at below altitudes equivalent to 200 g/cm^2) the radionuclide production rate increases with altitude but above this region the relationship is disturbed due to the escape of locally produced neutrons. This production rate per gram of air increases by about 2 to 3 orders of magnitude between sea level and the top of the atmosphere. While the radionuclide production

Table 1. Cosmic-ray produced radionuclides

Radionuclide	Half-life	Radiation	Reference
^{10}Be	2.7×10^6 y	β^- 555 KeV	(16)
^{36}Cl	3.1×10^5 y	β^- 714	(21)
^{14}C	5568 y	β^- 156	(1)
^{32}Si	~500 y	β^- 100	(19)
^{3}H	12.3 y	β^- 18	(35)
^{22}Na	2.6 y	β^+, γ 1.28	(28)
^{35}S	88 d	β^- 167	(26)
^{7}Be	53 d	β^-, γ 480	(27)
^{33}P	25 d	β^- 246	(24)
^{32}P	14.3 d	β^- 1.724 MeV	(23)
^{24}Na	15.1 hr	β^-, 1.38 + 2.76	(6)
^{38}S	2.9 hr	β^-, γ* 1.6 + 2.16	(7)
^{39}Cl	55 min	β^-, γ 0.246 + 1.266	(8)
^{38}Cl	37 min	β^-, γ 1.6 + 2.16	(7)

* Radiation due to the ^{38}Cl daughter.

rate is relatively constant with latitude in the troposphere it increases by about an order of magnitude in moving from the geomagnetic equator to the poles. Of the radionuclide production from cosmic ray spallation reactions, about 30 per cent takes place in the troposphere and 70 per cent in the stratosphere.

If the absolute production rate of a given radionuclide is known at a given altitude and latitude its production rate at other locations in the atmosphere can be determined from the relationships derived by LAL *et al.*[2] The absolute production rates for ^{7}Be from oxygen and of ^{32}P, ^{33}P and ^{35}S from argon were determined by LAL *et al.*[10] by exposing water and argon to cosmic rays on a mountain in Colorado at a geomagnetic latitude of $\lambda = 51°$ and a pressure of 685 g/cm^2. The ^{7}Be, ^{32}P, ^{33}P and ^{35}S were chemically separated and their disintegration rates measured. Measurements of the production rates of other radionuclides have been determined either by indirect methods including irradiation with artifically accelerated particles or by calculations based on semiempirical spallation yield curves.[11]

A variation in the cosmic-ray flux to the earth and in the radionuclide production rate is caused by solar activity. The sunspot cycle with its 11-y period results in a variation from the average radionuclide production rate of about ± 5 per cent during average solar cycles; however, compared to the quiet solar period in 1954, the global isotope production during the unusually high solar cycle of 1958 was reduced by 24 per cent. The reduction in isotope production which occurs during periods of strong solar activity is due to impaired transmission in interplanetary space of galactic particles. This reduction may be partially compensated for by production from particles coming from the sun. However, because of the uncertainty in the energy spectrum and flux of these particles a good estimate of their contribution to isotope production has not been made.

BEHAVIOR OF THE COSMIC-RAY PRODUCED RADIONUCLIDES

Following formation all of the radionuclides except ^{14}C have similar behavior. The newly formed isotopes become oxidized and attach themselves to particles of the aerosol. Most aerosol particles, particularly in the stratosphere, are extremely small and follow the motion of the surrounding air. Radionuclides produced high in the stratosphere require a year or more to reach the troposphere. Radionuclides produced in the troposphere or which enter from above reach the earth's surface in a comparatively short time. They are subjected to a rapid mixing in the troposphere which brings them in contact with water droplets, or may allow their direct deposition as dry fallout.

DISTRIBUTION AND OBSERVATIONS OF THE COSMIC-RAY PRODUCED RADIONUCLIDES

The production rates, atmospheric concentrations, and the materials in which the radionuclides have been observed are summarized in Table 2. The production rates and tropospheric concentrations serve as an index of the usefulness of these radionuclides in the various geophysical and biological processes. The half-lives and chemical characteristics of the radionuclides determine the various reservoirs on earth in which the radionuclides are deposited. For example, more than 90 per cent of the world's inventory[5] of the long-lived radionuclides ^{10}Be, ^{36}Cl, ^{14}C and ^{32}Si is located in the deep ocean layer or in ocean bottom sediments, while a major portion of the other shorter-lived radionuclides ^{22}Na, ^{35}S, ^{7}Be, ^{33}P, ^{32}P, ^{24}Na, ^{38}S, ^{39}Cl and ^{38}Cl is located in the upper atmosphere. Over 90 per cent of the tritium is located in the oceans and continental hydrosphere.

A comparison of the calculated and observed concentrations in the atmosphere of the intermediate half-lived radionuclides has provided a basis for studying the nature of large scale atmospheric circulation and tropospheric scavenging processes.[2,12] The radionuclides ^{22}Na, ^{35}S, ^{7}Be, ^{33}P and ^{32}P have yielded much useful information in this regard, and the very short-lived radionuclides ^{24}Na, ^{38}S, ^{38}Cl and ^{39}Cl show promise in studying the very short-term atmospheric scavenging processes. For example, the ratio of two or more of these could be used in studying the washout efficiency of the atmospheric aerosol by rain or snow. The more easily measured radionuclides ^{7}Be and ^{22}Na have provided information on the fallout patterns of the cosmic-ray produced radionuclide

Table 2. Production rates, tropospheric concentration, and detection of cosmic-ray produced radionuclides

Radionuclide	Half-life	Atmospheric production rate[2] (atoms/cm.^{2}-sec)	Troposphere concentration[5] (dpm/kg air)	Detected and measured in
^{10}Be	2.7×10^6 y	4.5×10^{-2}	7×10^{-8}	Deep sea sediments
^{36}Cl	3.1×10^5 y	1.1×10^{-3}	1.5×10^{-8}	Rock and rain†
^{14}C	5568 y	1.8	7.6*	Organic material and $^{14}CO_2$
^{32}Si	~500 y	1.6×10^{-4}	1.2×10^{-6}	Marine sponges and sea water
^{3}H	12.3 y	0.25	7×10^{-2}	HTO, HT
^{22}Na	2.6 y	5.6×10^{-5}	6.7×10^{-5}	Rain, air, biological material
^{35}S	88 d	1.4×10^{-3}	7.8×10^{-3}	Rain, air, biological material
^{7}Be	53 d	8.1×10^{-2}	0.63	Rain, air
^{33}P	25 d	6.8×10^{-4}	7.6×10^{-3}	Rain, air, biological material
^{32}P	14.3 d	8.1×10^{-4}	1.4×10^{-2}	Rain, air, biological material
^{24}Na	15.1 hr			Rain
^{38}S	2.9 hr			Rain
^{39}Cl	55 min	1.6×10^{-3}		Rain
^{38}Cl	37 min			Rain

* Including amount present in biosphere and humans.

† The observed concentration in rain was due to nuclear weapons testing.

group. The radionuclides ^{14}C and ^{32}Si are providing information on the ocean's circulation process[13] while the radionuclides ^{10}Be and ^{32}Si are useful in studying the chronology of the ocean sediments. Because of the artificial production of several of these radionuclides by nuclear testing, caution must be exercised in the interpretation of their measurements.

The fallout patterns of the short-lived cosmic-ray produced radionuclides have been established by studying the more easily measured radionuclides ^{32}P and ^{7}Be. Measurements of these radionuclides in rain have shown that their fallout rate is proportional to their tropospheric production rate and the annual precipitation rate. For these short-lived radionuclides (53 d ^{7}Be and 14.3 d ^{32}P) the contribution from stratospheric influx is negligible even at the middle latitudes where it plays a major role in the injection of nuclear bomb debris into the troposphere. The ratios of the 14.3 d ^{32}P to the 53 d ^{7}Be in tropospheric air indicate an average residence time in the troposphere of about 40 days which is in reasonable agreement with the value of 30 days obtained by STUART *et al.*[14] for the residence time of fission products in the troposphere.

^{14}C *and tritium*

^{14}C and tritium have the highest production rates of the cosmic-ray produced radionuclides, and were the first to be observed. Because of their abundance and usefulness a much larger effort has been expended in their study than in that of the other radionuclides. The ^{14}C in the atmosphere is in the form of $^{14}CO_2$. It is diluted with stable CO_2 and has a specific activity of about 15 dpm/g of carbon. It has been studied in all of the principal geophysical reservoirs and can be used in age dating for periods of 50,000 y without enrichment and up to 70,000 y following heavy isotope enrichment. Tritium is in the form of HTO and its natural concentration in the atmosphere or rain is on the order of a few T.U. (a Tritium Unit equals one tritium atom per 10^{18} atoms of protium). Tritium has also been extensively studied in the various geophysical reservoirs. Prior to

the 1961 atomic test series the atmospheric ^{14}C was three and tritium several hundredfold above normal cosmic-ray production levels due to nuclear testing.

^{10}Be

^{10}Be has a half-life of 2.7×10^6 y and its decay is by emission of a 0.55-MeV beta particle. ^{10}Be has only been observed in the core material from the ocean floor and at concentrations of a few dpm/kg. Its production by cosmic rays was predicted by PETERS[15] and it was first observed somewhat later by ARNOLD[16] in ocean floor cores. The stable beryllium content of the cores is only a few ppm and it can therefore be separated in high specific activity for counting. Measurements by ARNOLD[16] KHARKAR *et al.*[17] and others have demonstrated its usefulness in dating of ocean sediments.

^{32}Si

^{32}Si has a half-life of about 500 y[18] and decays by emission of 100-keV maximum beta particles. It was first reported by LAL *et al.*[19] in a sponge taken from shallow water in the Gulf of California. The ^{32}Si was determined by chemically isolating and measuring its daughter ^{32}P. Subsequent studies by KHARKAR *et al.*[17] have found concentrations of ^{32}Si ranging from 6 to 70 dpm/kg of silicon in samples of sponge and plankton taken from around the world. Measurements of the dissolved ^{32}Si in sea water were made by SCHENK,[20] and involved separation of silicon from several tons of sea water. These measurements showed concentrations ranging from 210 dpm of ^{32}Si/kg of silicon at the surface to 6 dpm at 2500 m.

^{36}Cl

^{36}Cl has a 3.1×10^5 year half-life and decays by emission of a 714-keV (maximum) beta particle. Its production by cosmic rays in the atmosphere has not been observed; however, it has been observed in phonolite rock at Cripple Creek, Colorado, by DAVIS and SCHAEFFER[21] and it was later found in rainwater by SCHAEFFER *et al.*[22] at a concentration of about 3000 times that due to cosmic production. Its presence in this rainwater was therefore attributed to nuclear testing.

^{32}P, ^{33}P

The phosphorous isotopes ^{32}P (14.3 d) and ^{33}P (25 d) decay by beta particle emission with maximum energies of 1.7 and 0.25 MeV, respectively. ^{32}P was first observed by MARQUEZ *et al.*[23] who separated it from rainwater. ^{33}P was found later in rainwater by LAL *et al.*[24]. They observed the ^{32}P and ^{33}P at concentrations of a few dpm/liter. Both of these radionuclides as well as ^{7}Be and ^{35}S are produced artificially during atomic weapons testing and were observed in the 1961 and 1962 test series by DREVINSKY *et al.*[25] on high altitude air filters.

^{35}S

^{35}S has a half-life of 88 days and decays by emission of 167-keV (maximum) beta particles. It was observed by GOEL[26] who separated it from 100 to 300-liter rain samples and found concentrations of 0.5–6.5 dpm/liter. It has served as a tracer in studies of atmospheric circulation and is produced artificially during weapons testing (DREVINSKY *et al.*[25]).

^{7}Be

^{7}Be has a half-life of 53 days and decays by K-capture with the emission of a 480-keV gamma ray. It was first observed by ARNOLD and AL-SALIH[27] who chemically separated it from rainwater. Its average concentration in rainwater is about 40 dpm/liter and is measured with relative ease. It is easily measured on air filters and has been used in numerous atmospheric circulation studies.

^{22}Na

^{22}Na has a 2.6-year half-life and decays by emission of a positron and a 1.28-MeV gamma ray in cascade. It was first observed as a cosmic-ray produced radionuclide by MARQUEZ[28] who found it in rainwater in a concentration of a 0.017 dpm/liter. In spite of its low concentration, it has been useful in atmospheric circulation studies.[12] The present ^{22}Na concentration in the atmosphere is several times that due to the natural cosmic-ray production rate, and this is due to the past nuclear weapons tests. Its air concentration at ground level during a 2.5-y period was recently reported by PERKINS *et al.*[29]

^{24}Na

^{24}Na has a half-life of 15 h and decays by emission of a 1.7 MeV (maximum) beta particle and two gamma rays in cascade. It was first observed by RODEL[6] who isolated it from 200 to 300-liter samples of rainwater and observed concentrations of a 0.18 to 0.35 dpm/liter. It has since been observed by PERKINS[30] by direct gamma–gamma coincidence counting of samples as small as 7 liters. Sample preparation simply involved evaporation to dryness and counting the residue.

^{39}Cl

^{39}Cl has a half-life of 55 min and decays by emission of a 1.28 MeV beta particle and two gamma rays (0.246 and 1.226 MeV) in cascade. It was first observed by WINSBERG[8] in concentrations ranging up to 100 dpm/liter.

^{38}Cl and ^{38}S

^{38}Cl (37.3 min) and ^{38}S (2.9 h) were observed for the first time during 1964 by PERKINS *et al.*[7] ^{38}Cl decays by emission of a beta particle plus two gamma rays (1.60 and 2.16 MeV) in cascade, while ^{38}S decays to ^{38}Cl and the ^{38}Cl gamma rays can be used for its measurement. The radionuclides were measured both with and without chemical separation of the chlorine isotopes by multidimensional gamma-ray spectrometry.[31] ^{38}Cl was observed at concentrations of 9–150 dpm/liter, while ^{38}S was observed at concentrations of 4–13 dpm/liter.

Another source of cosmic-ray produced radionuclides is that contained in the extraterrestrial dust which is continually showered on the earth. LAL[5] has pointed out that it may be possible to measure the rate of accretion of this material by examining the activity of ^{53}Mn ($\sim 10^6$ y) and ^{26}Al (7.4×10^5 y) in slowly accumulating ocean bottom sediments. Satellite and rocket data have shown that the present flux of meteoric dust is $\sim 10^4$ tons/day which results in several times more ^{26}Al from this source than from the predicted cosmic ray production in the atmosphere.[5] Meteoritic dust contains numerous radionuclides [32] produced by spallation and activation of the stony and iron constituents, but the concentrations are extremely low and they have not as yet found application in tracing the geophysical processes on earth.

APPLICATIONS OF COSMIC-RAY PRODUCED RADIONUCLIDES IN STUDYING THE BIOSPHERE

Some of the cosmic-ray produced radionuclides are extremely useful as tracers in the biosphere. ^{14}C and tritium are well known for their applications in age dating and studying biological processes. The behavior of the other radionuclides has received relatively little attention in regard to their accumulation and movement through the biosphere. The radionuclide ^{32}Si has been determined in ocean sponge and plankton and these measurements show a tenfold higher specific activity of ^{32}Si in plankton than sponge.[17] Recent developments employing multidimensional gamma-ray spectrometry[31] have allowed a detailed study of the movement of the radionuclide ^{22}Na through the food chain to man[33]. The relative importance of various foods in channeling this radionuclide to man is indicated in Table 3. Due to the past atomic tests, the present concentration of ^{22}Na in the atmosphere is about an order of magnitude higher than the predicted cosmic-ray production and this, of course, simplifies its measurement. The high ^{22}Na concentrations in the Alaskan animal samples are apparently due to their diet which consists largely of lichen. This plant collects large amounts of fallout radionuclides which are then in a position to move through the food chain via the caribou and reindeer to man.

The role of Alaskan lichens in collecting fallout is presently being used to study the natural biological behavior of other radionuclides. The radionuclides ^{35}S, ^{7}Be, ^{32}P and ^{33}P are expected to be present in relatively high concentrations on the lichens and their behavior in the food chain is under investigation in our laboratory.[34]

The cosmic-ray produced radionuclides, although present in very low concentrations in the atmosphere, are produced at a nearly steady rate and provide a natural group of tracers for studying both geophysical and biological processes. The natural concentration processes which operate in certain biological systems provide high concentration factors so

Table 3. ^{22}Na *in foods, bioassay samples and people (dpm/kg)*

Western United States				Alaska			
Sample	Location	Date obtained	^{22}Na	Sample	Location	Date obtained	^{22}Na
Elk	Quinault, Wash.	5/64	74.4	Caribou meat	Near Kotzebue, Alaska	7/63	180
Bass	Columbia River	4/64	8.3	Caribou meat		7/63	89.1
Milk	Washington	3/64	6.2	Moose meat		3/64	58.7
Wheat	Washington	8/63	3.31	Moose meat		3/64	80.6
Beef	Washington	5/64	2.47	Caribou meat		8/5/61	9.13
Beef	Washington	3/64	0.52	Caribou meat		10/9/61	23.0
Corn	Oregon	4/64	~0.04	Reindeer meat		8/63	15.6
Lettuce	California	3/64	0.16	Urine*	Anaktuvuk Pass, Alaska	1/64	126
Carrots	California	4/64	~0.07	Man (Eskimo)†			3427†
Potatoes	Idaho	4/64	0.68				
Green Beans	Oregon	4/64	~0.20				
Urine Sample‡	Washington		1.45				
Man†	Washington	11/63	39.5†				

* Composite from two Alaskan Eskimos.

† These values are estimated whole body burdens for sodium-22 based on an 11-day biological half-life.

‡ Richland, Washington, composite from several individuals.

that the movement of some of these radionuclides through the biosphere can be traced with relative ease.

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