



Naturally Occurring Radioactive Substances

A.M. Emara

Atomic Energy Authority

Natural radioactivity originates from extraterrestrial sources as well as from radioactive elements in the earth's crust. About 340 nuclides have been found in nature, of which about 70 are radioactive and are found mainly among the heavy elements. All elements having an atomic number greater than 80 possess radioactive isotopes, and all isotopes of elements heavier than number 83 are radioactive.

The radioactivity of the earth now includes the primordial radionuclides, whose half lives are sufficiently long that they have survived the interval since their creation, and the secondary radionuclides which are derived by their radioactive decay. A much larger number of radioactive isotopes than now exist, were produced when the matter of which the universe is formed first came into being several billion years ago, but most of them have decayed out of existence. The radionuclides which now exist are those that have half-lives at least comparable to the age of the universe. Radioisotopes with half-lives of less than about 10^8 years have become undetectable in the 30 or so half-lives since their creation, where as radionuclides with half-lives greater than 10^{10} years have decayed very little up to the present time.

In most places on earth the natural radioactivity varies only within relatively narrow limits, but in some localities there are wide deviations from normal levels owing to the presence of abnormally high concentrations of radioactive minerals in the local soils.

Naturally Occurring Radioactive Elements

The naturally occurring radionuclides can be divided into those that occur singly (Table 1 and Table 2) and those that are components of three chains of radioactive elements: (1) the uranium series, which originates with ^{238}U ; (2) the thorium series, which originates with ^{232}Th and (3) the actinium series, which originates with ^{235}U . In addition, there are singly occurring radionuclides

* This lecture is abstracted from a comprehensive treatmet of the subject in Merrill Elserjbud, "Environmental Radioactivity," Acad. Press 1987.

Table (1)
Singly Occurring Natural Radionuclides Produced by Cosmic Rays^a

Radionuclide	Half-life	Trophospheric concentration (pCi/kg air)	Principal radiations and energies (MeV)	Observed Concentration rainwater (pCi/liter)
³ H	12 years	3.2×1.0^{-2}	β-0.0286	--
⁷ Be	53 days	0.28	γ 0.477	18.0
¹⁰ Be	1.6×10^6 years	3.2×1.0^{-8}	β- 0.555	--
¹⁴ C	5730 years	3.4	β- 0.156	--
²² Na	15.0 hr	--	β- 1.4, γ 1.37, 2.75	0.08-0.16
³² P	14 days	6.3×1.0^{-3}	β- 1.71	"a few"
³³ P	24 days	3.4×1.0^{-3}	β- 0.246	" a few"
³⁵ S	88 days	3.5×1.0^{-3}	β-0.167	0.2 - 2.9
³⁶ Cl	3.1×10^5 years	6.8×1.0^{-9}	β-0.714	--
³⁸ S	2.87 hr	--	β- 1.1, γ 1.88	1.8 - 5.9
³⁸ Cl	37 min	--	β- 4.91, γ 1.60, 2.17	4.1 - 67.6
³⁹ Cl	55 min	--	β- 1.91, γ 0.25, 1.27, 1.52	4.5 - 22.5

^aPerkins and Nielsen (1965). Reproduced from Health Physics, vol. II, by permission of the Head Physics Society.

of both cosmic and terrestrial origin. The three families of radioactive heavy elements account for much of the background radiation to which humans are exposed. A fourth family, the neptunium series, which originated in the parent element ²⁴¹Pu, is known to have existed at one time, but this nuclide has a half-life of only 14 years and existed only briefly after its formation. Other members of that series also have relatively short half-lives. The only surviving member of the neptunium family is the nearly stable nuclide ²⁰⁹Bi, which has a half-life estimated to be about 2×10^{18} years.

Table (2)
Singly Occurring Natural Radionuclides of Terrestrial Origin^a

Radio-nuclide	Abundance (%)	Half-Life (years)	Principal radiations: energy (MeV) and yield (%)	Specific activity (elemental) (pCi/g)
⁴¹ K	0.012	1.26 x 10 ⁹	β-1.33, 89%	855
⁷ V	0.25	6 x 10 ¹⁵	γ with EC 1.46, 11%	3.0 x 10-3
¹⁰ Rb	27.9	4.8 x 10 ¹⁰	γ with β- 0.78, 30%	
¹¹⁵ In	95.8	6.0 x 10 ¹⁴	γ with EC 1.55, 70%	
²²² Te	0.87	1.2 x 10 ¹³	β- 0.28, 100%	2.4 x 10 ⁴
²²³ La	0.089	1.12 x 10 ¹¹	β- 0.48, 100%	4.98
¹⁴² Ce	11.07	> 5 x 10 ¹⁶	EC	2.11
¹⁴⁴ Nd	23.9	2.4 x 10 ¹⁵	β- 0.21, 80%	20.7
¹⁴⁷ Sm	15.1	1.05 x 10 ¹¹	γ with EC (0.81, 1.43), 70%	
¹⁴⁸ Sm	11.27	> 2 x 10 ¹⁴	(a)	5.6 x 10-3
¹⁶⁶ Sm	13.82	> 1 x 10 ¹⁵	α 1.83	0.25
¹³² Gd	0.20	1.1 x 10 ¹⁴	α 2.23	3.5 x 10 ³
¹³⁶ Dy	0.052	> 1 x 10 ¹⁸	--	1.37
¹⁷⁴ Hf	0.163	2 x 10 ¹⁵	--	1.37
¹⁷⁶ Lu	2.6	2.2 x 10 ¹⁰	α 2.1	4.3 x 10-2
¹⁸⁰ Ta	0.012	> 1 x 10 ¹²	α 2.5	1.2 x 10-6
¹⁸⁷ Re	62.9	4.3 x 10 ¹⁰	β- 0.43	1.68 x 10-3
¹⁹⁰ Pt	0.013	6.9 x 10 ¹¹	γ 0.089, 0.203, 0.306	2.4 x 10 ³
			--	0.239
			β- 0.003	2.8 x 10 ⁴
			α 3.18	0.36

^aPerkins and Nielsen (1965). Reproduced from Health Physics, vol. II, by permission of the Head Physics Society.

In nature, ²³⁵U and a few other nuclides of uranium and thorium undergo fission spontaneously or as a result of interactions with neutrons that originate from cosmic rays or other natural sources. The half-life of ²³⁵U owing to spontaneous fission is between 10¹⁵ and 10¹⁶ years, which means that decay by this process proceeds at a rate less than 10⁻⁷ of that due to a emission. Others of the heavy nuclides undergo spontaneous fission with half-lives that range from 10¹⁴ to 10²⁰ years (*Rankama, 1954*).

Many transuranic elements, such as plutonium, neptunium and americium, which now exist because they have been produced artificially (*Seaborg, 1958*), must have existed in nature at one time,

but their half-lives are so short that they disappeared long ago. However, some of the transuranic elements are produced in minute amounts by naturally occurring neutrons that result in fission of uranium isotopes. Plutonium-239 has been detected in pitchblende in a ratio to ^{238}U of 10^{-11} to 10^{-13} , and ^{237}Np has been identified in uranium minerals in a ratio to ^{238}U of 1.8×10^{-12} .

The uranium normally found in nature consists of four isotopes having mass numbers 230, 234, 235, and 238. Uranium-238 is present in the amount of 99.28% and is usually in equilibrium with ^{234}U , which is present in the amount of 0.0058%. Uranium-235, the parent isotope of the actinium series, is present in the amount of 0.71%. Uranium-230, which is also a member of the ^{238}U series, has a short half-life (20.8 days).

Uranium is found in all rocks and soils. Typical concentrations are listed in Table 3.

Uranium occurs in traces in many commercial products. Pre-World War II samples of steel analyzed by **Welford and Sutton (1957)** contained uranium in the range of 0.01 to 0.2 ppm. Surprisingly, photographic emulsions and other photographic materials contain from 0.2 to 1 ppm of uranium (**Smith and Zuzi, 1949**).

Because uranium occurs in soils and fertilizers; the element is present in food and human tissues. On average, the annual intake of uranium from all dietary sources is about 0.14 mCi. The intake of uranium from tap water is negligible by comparison, but a few exceptions have been reported, notably in France, Finland, and the Soviet Union (**Hess, et al., 1985; UNSCEAR, 1982**). The skeleton is estimated to contain about 25 mg of uranium, equivalent to about 8 pCi. The dose to the skeleton, which receives a higher dose from uranium than any other organ, is about 0.3 mrem/year (**USCEAR, 1982; ICRP, 1975; Wrenn, et al., 1985**).

Radium-226

Radium-226 and its daughter products are responsible for a major fraction of the dose received by humans from the naturally occurring

internal emitters. ^{226}Ra is an alpha emitter that decays, with a half-life of 1622 years, to ^{222}Rn , with a half-life of 3.8 days.

Table (3)
Average Uranium Concentration in Various Rocks^a

<i>Rock type</i>	<i>Uranium concentration (ppm)</i>
Acid igneous	3.0
Intermediate igneous	1.5
Basic igneous	0.6
Ultrabasic igneous	0.03
Meteorites	0.003
Phosphate rock (Fla.)	120
phosphate rock (N. Africa)	20-30
bituminous shale (Tenn.)	50-80
Normal granite	4
Limestones	1.3
Other sedimentary rocks	1.2

^aFrom *Lowder and Solon, (1956)*.

The decay of radon is followed by the successive disintegration of a number of short-lived alpha- and beta-emitting progeny. After six decay steps, in which isotopes are produced that range in half-lives from 1.6×10^{-4} sec to 36.8 min, ^{210}Pb is produced, which has a half-life of 22 years. This nuclide, decays through ^{210}Bi to produce ^{210}Po (half-life 238 days), which decays by alpha emission to stable ^{206}Pb .

Table (4)
Average Radium, Uranium, Thorium and Potassium Contents in Various Rocks^a

<i>Type of rock</i>	^{226}Ra (pCi/g)	^{238}U (pCi/g)	^{232}Th (pCi/g)	^{40}K (pCi/g)
Igneous	1.3	1.3	1.3	22
Sedimentary				
Sandstones	0.71	0.4	0.65	8.8
Shales	1.08	0.4	1.1	22
Limestones	0.32	0.4	0.14	2.2

^aAdopted from *UNSCEAR (1958)*.

Table (5)
Average Radium Content of Various Rocks and Soils^a

No. of specimens		Specimen, average value (pCi/g)
1	Quartz-mica schist	0.20
1	Quartzite	0.20
4	limestones	0.29
1	Sandstone	0.32
2	Glacial sand and rubble	0.38
4	Gravels	0.41
2	Soils	0.73
1	Kaibab limestone	0.97
5	Granites	1.02
2	Peruvian lavas	2.06

23	All specimens	0.70

^aFrom *Evans and Raitt (1935)*.

Radium, being an α emitter, does not add directly to the γ activity of the environment, but does so indirectly through its γ -emitting descendants.

Radium-226 Content of Rocks and Soils

Radium-226 is present in all rocks and soils in variable amounts. Igneous rocks tend to contain somewhat higher concentrations than sandstones and limestones. *Rankama and Sahama (1950)* give a mean concentration of 0.42 pCi/g in limestone and 1.3 pCi/g in igneous rock, as listed in Table 4. *Evans and Raitt (1935)*, who measured the ²²⁶Ra content of rock and soil specimens from the sites of cosmic-ray observations made by Millikan and co-workers, give the values shown in Table 5, which are somewhat lower than the average values given by Rankama and Sahama.

Radium-226 in Water

The radium content of public water supplies has been reviewed comprehensively by *Hess et al. (1985)*, who described the geological

and geochemical factors that influence the concentration of the two principal radium isotopes, ^{226}Ra and ^{228}Ra , which are progeny of uranium and thorium, respectively. There is more ^{232}Th than ^{238}U in nature on an activity basis, but there are geochemical factors that cause local concentrations of uranium, which often results in greater amounts of ^{226}Ra relative to ^{228}Ra .

Thus, it is generally assumed that the ratio $^{226}\text{Ra} : ^{228}\text{Ra}$ is greater than unity, although until recently most of the reported measurements have been of ^{226}Ra , with fewer measurements of ^{228}Ra .

The radium content of surface waters is low (0.1-0.5 pCi/liter) compared to most ground waters. Dissolved radium sorbs quickly to solids and does not migrate far from its place of release to ground water. It has been suggested that radium transport in ground water is even less than that of ^{222}Rn , which has a half-life of only 3.8 days (*King, et al., 1982; Kirshnas*).

Interest in the radium content of water supplies has increased during the past few years, following the publication in 1979 of a proposed EPA drinking water standard for total radium of 5 pCi/liter (*Lappenbush and Cothorn, 1985*). It has been estimated that an individual who consumes 2 liters/day of water that contains 5 pCi ^{226}Ra /liter would, at the end of a lifetime, be receiving a bone dose of 150 mrem/year. The dose would be increased by the presence of ^{228}Ra . In rare cases where the concentration of ^{226}Ra is as high as 25 pCi/liter, the bone dose can approach nearly 1 rem/year.

The EPA regulations, which limit the ^{226}Ra content of potable water to 5 pCi/liter, may require that some well water supplies be treated. A study of the effectiveness of water treatment methods for the removal of radium was undertaken by *Brinck, et al. (1978)* in areas of Iowa and Illinois where the EPA limit was exceeded. Four

different water treatment methods were studied. Reverse osmosis and sodium ion exchange process were generally about 92% effective. The removal efficiency of the lime-soda ash softening process varied from 75 to 95%. Systems designed to remove iron only were found to have removal efficiencies from 11 to 53%. A detailed analysis of the costs and effectiveness of various methods of removing radioactivity from drinking water is given by *Reid, et al. (1985)*.

Little variation is found in the ^{226}Ra content of Atlantic Ocean surface water outside Antarctica (*Broecker, et al., 1976*). Eighty samples analyzed by Broecker and co-workers averaged 0.03 pCi/liter. Deep ocean water contains somewhat greater amounts of ^{226}Ra , which were explained by the inflow of O_2 -deficient bottom water from the polar regions.

Radium-226 in Food

Radium is chemically similar to calcium and is absorbed from the soil by plants and passed up the food chain to humans. Because the radium in food originates from soil and the radium content of soil is variable, there is considerable variability in the radium content of foods. In addition, it is reasonable to expect that chemical factors such as the amount of exchangeable calcium in the soil will determine the rate at which radium will be absorbed by plants.

One of the earliest attempts to estimate the radium content of food was undertaken by *Mayneord and associates (1958, 1960)*, who made α -radiation measurements of ashed samples of foods and differentiated the α activities of the thorium and uranium series by counting the double α pulses from the decay of ^{220}Rn and ^{212}Po . These early measurements served to approximate the total ^{226}Ra and ^{228}Ra content of foods (Table 6) and were highlighted by the fact that Brazil nuts were found to be much more radioactive than other foods.

This was later investigated by *Penna Franca, et al. (1968)*, who showed that the anomaly is due to the tendency of the Brazil nut tree (*Bertholletia excelsa*) to concentrate barium, a chemical congener of radium. Penna Franca found the radium content of Brazil nuts to range between 273 and 7100 pCi/kg, with only 3 of 15 samples assaying less than 1000 pCi/kg. The radioactivity was about equally divided between ^{226}Ra and ^{228}Ra and was not related to the radium or barium content of the soil in which the tree was grown. The radium concentration of Brazil nuts is on the order of 1000 times greater the radium concentration of Brazil nuts is on the order of 1000 times greater the radium concentration in the foods that make up the average diet in the United states.

Fisenne and Kelle (1970) estimated the ^{226}Ra intake of inhabitants of New York City and San Francisco to be 1.7 and 0.8 pCi/day, respectively.

Table (6)
Alpha Radioactivity of Foods^a

<i>Foodstuff</i>	<i>Maximum a activity observed per 100 g (pCi)</i>
Brazil nuts	1400
Cereals	60
Teas	40
Liver and kidney	15
Flours	14
Peanuts and peanut butter	12
Chocolates	8
Biscuits	2
Milks (evaporated)	1-2
Fish	1-2
Cheeses and eggs	0.9
Vegetables	0.7
Meats	0.5
Fruits	0.1

^aFrom *Mayneord, et al. (1958)*.

However, this twofold difference is not reflected by differences in the ^{226}Ra content of human bone from the two cities, which were found (*Fisenne, et al, 1981*) to be 0.013 pCi $^{226}\text{Ra}/\text{g}$ of bone ash for both cities. Studies of this kind involve highly sophisticated and sometimes uncertain food and bone sampling techniques which may involve errors that can obscure differences of a factor of 2.

Radium-226 Content of Human Tissues

A number of investigators in various parts of the world have provided measurements of the ^{226}Ra content of bone, which contains about 80% of the total body radium. The median body burden is 23 pCi.

Estimates of the dose delivered by ^{226}Ra and its daughter products require knowledge of the fraction of radon retained by the tissue in which the radium is deposited, since most of the dose is due to alpha emissions from the decay of radon and its daughters. The dose estimate is also complicated by the fact that the alpha energy is deposited at loci determined by the pattern of deposition within the tissue. UNSCEAR has used a radon retention factor of 0.33 and has assumed that the radium is uniformly deposited throughout the bone. On this basis, the average annual effective dose equivalent to bone from deposited radium is about 7 mrem, using a quality factor of 10 for the dose from alpha emissions (*UNSCEAR, 1982*).

Thorium-232

The thorium content of various rocks, as reported by *Faul, (1954)*, indicates a range of 8.1 to 33 ppm for igneous rocks, with a mean value of 12 ppm. *Rankama and Sahama (1950)* reported that the concentration in sandstone is 6 ppm, intermediate between limestone and the igneous rocks. The thorium content of igneous rocks is thus about four times the uranium content, but since the specific activity of ^{232}Th is 0.11 pCi/g, compared to 0.33 pCi/g ^{238}U , the radioactivity due to the two nuclides is more nearly 1 : 1. Later it will be seen that there are areas of the world in which the ^{232}Th content of rocks and soils is very much greater than normal.

The characteristics of the thorium series are different from those of the uranium series in a number of respects :

1. ^{228}Ra has a shorter half-life than ^{226}Ra (5.8 compared to 1620 years).
2. ^{228}Ra is a β emitter that decays to α -emitting ^{228}Th , which has a half-life of 1.9 years. ^{228}Th , in turn, decays through a series of α -emitters including the noble gas ^{220}Rn (thoron), which has a half-life of only 54 sec, compared to 3.8 days for ^{222}Rn . Because of its short half-life, thoron has less opportunity than radon to diffuse from the matrix in which it is found
3. The solubility of ^{228}Ra in soil is comparable with that of ^{226}Ra , but the dose rate to an organism from assimilated ^{228}Ra , a β emitter, is time-dependent because the dose depends on ingrowth of α -emitting ^{232}Th and its short-lived descendants.
4. In the ^{238}U chain, there is no long-lived "stopping" nuclide comparable to ^{210}Pb ($T_{1/2} = 22$ years). The longest-lived nuclide beyond ^{228}Th is ^{212}Pb , with a half-life of 10.6 hr. The dosimetry and radiochemistry of the thorium series tend to be complicated by these characteristics (*Fresco, et al., 1952*).

Because of its relative insolubility and low specific activity, ^{232}Th is present in biological materials only in insignificant amounts. *Linsalata, et al., (1986a)* have reported ^{232}Th concentrations in the edible portions of washed vegetables grown in silty clay and peaty soils of New York State to range between 0001 pCi/kg (fresh weight) for carrots and 0.093 pCi/kg for a variety of squash. The mean value for 25 vegetable samples including potatoes, corn kernels, carrots, beans, and squash was 0.018 ± 0.022 pCi/kg (fresh weight). *Wrenn, et al. (1981) measured* ^{232}Th , ^{230}Th , ^{228}Th in the tissues of residents of New York and Colorado. Thorium was found to be present in the highest concentrations in pulmonary lymph nodes and lung, indicating that the principal source of human exposure is inhalation of suspended soil particles. Because thorium is removed from bone very slowly, the concentrations of both ^{230}Th (which is found in the ^{238}U decay series) and ^{232}Th were found to increase with age.

Radium-228 (Mesothorium)

Although ^{228}Ra (referred to as mesothorium in some earlier writings) frequently occurs in soil and water in approximately a 1 : 1 ratio to ^{226}Ra , there is surprisingly little information about its occurrence in foods or in human tissues. Systematic ^{228}Ra measurements in food and water have not been made on a scale comparable to those of ^{226}Ra , but such data as do exist suggest that under normal circumstances the ^{228}Ra content of food, water, and human tissues is one-half to one-fourth of the ^{226}Ra content (*UNSCEAR, 1966*).

Radon-222 and Radon-220 (Thoron)

When ^{226}Ra decays by α -emission, it transmutes to its daughter ^{222}Rn , an inert gas with a half-life of 3.8 days. Similarly, ^{224}Ra , which is a descendant of ^{232}Th , decays by α -emission to 54-sec ^{220}Rn , commonly known as thoron.

The 3.8-day radon isotope has a greater opportunity than the nuclei of shorter-lived thoron to escape to the atmosphere. The mechanisms by which radon diffuses from soil into the atmosphere have been discussed by *Tanner (1964, 1980)*.

The average radon concentration in Washington, D.C., has been shown by *Lockhart (1964)* to be more than 100 times greater than the average concentration in ice-covered Little America and more than 12 times the values observed at Kodiak, Alaska. The same investigator reported wide variability from day to day. For example, the mean daily concentrations varied more than a hundredfold in Washington, D.C., during 1957. (*Lockhart, 1958*). *Gesell, (1983)* has reviewed the reported data from various parts of the United States and estimates the annual average concentration to range from 0.016 pCi/liter in Kodiak, Alaska, to 0.75 pCi/liter in Grand Junction, Colorado. The average annual concentrations showed less variation at four more normal localities (0.22-0.30 pCi/liter).

Other reports from several countries indicate that the average concentrations of radon in outdoor air may be taken normally to be in

the range 0.1-0.5 pCi/liter. A number of investigators have observed periodicity in hour-to-hour observations of the radon and thoron content of outdoor air (*UNSCEAR, 1982*). Maximum concentrations are observed in the early hours and the lowest values are found in the late afternoon, when the concentrations are about one-third the morning maxima (*Gold, et al., 1964*).

It is likely that the variations at any given locality are dependent on meteorological factors that influence both the rate of emanation of the gases from the earth and the rate of dilution in the atmosphere (*Wilkening, 1982*). Thus, the rate of emanation from soil may increase during periods of diminishing atmospheric pressure and decrease during periods of high soil moisture, although the evidence is not consistent on this point. It is also likely that the history of an air mass for several days prior to observation influences its radon and thoron content (*Barreira, 1961*). Passage of the air over oceans, and possibly precipitation, would tend to reduce the concentration of these gases, whereas periods of temperature inversion should cause the concentrations to increase by limiting the volume of the atmosphere within which dilution can take place. The gases can be expected to be present in greater amounts over large masses of igneous rock than over large bodies of water or over sedimentary formations

Because the daughter products of radon and thoron are electrically charged when formed, they tend to attach themselves to inert dusts, which are normally present in the atmosphere. If the radioactive gases coexist with the dust in the same air mass for a sufficiently long time, the parents and their various daughters will achieve radioactive equilibrium. The growth of the ^{222}Rn daughters approaches an equilibrium in about 2 hr and beyond that time further growth would slow because of the presence of 22-year ^{210}Pb . *Blifford and associates (1952)* investigated the relationships between the concentrations of radon and its various decay products in the normal atmosphere and found, as would be expected, that the atmosphere is markedly depleted in ^{210}Pb relative to the precursors of this isotope.

This is because the inert dust of the atmosphere, the radon, and the radon daughters coexist long enough under normal circumstances for equilibria to be reached between radon and the shorter-lived daughters. Since the radionuclide with the longest half-life prior to ^{210}Pb is 26.8-min ^{214}Pb , equilibrium is reached in about 2 hr. The ^{210}Pb , which has a 22-year half-life, would take about 100 years to reach equilibrium. Various mechanisms exist for removing dust from the atmosphere, and the ratio of ^{210}Pb to its shorter-lived ancestors was shown by Blifford to be indicative of the length of time the dust resides in the atmosphere. He concluded by this method of analysis that the mean life of the atmospheric dust to which the radon daughters are attached is 15 days.

Wilkening (1964), found the concentration of ^{222}Rn daughters in the atmosphere to be depleted during passage of a thunderstorm. He attributed this to the action of electric fields, which changed from a normal value to about 1.8 to -340 V/cm. Deposition of the daughter products during rainstorms may temporarily increase the gamma background.

The thorium series below thoron (^{220}Rn) has no long-lived member. The equilibrium between thoron and its daughters will be achieved at a rate governed by the time required for the buildup of ^{212}Pb (half-life 106 hr)

The natural radioactivity of atmospheric dust, owing primarily to the attached daughters of radon, can be demonstrated readily. When air is drawn through filter media, the radon daughters attached to the filtered atmospheric dust cause both the α and β activity of the filter media to rise. Curve A of Fig. 1 (Annex II) illustrates the manner in which the increase in radioactivity occurs in the case of normal air containing 0.05 pCi/liter of radon in equilibrium with its daughter products. The rise in α activity increases for about 2 hr, at the end of which time the accumulated daughters decay at a rate compensated by the newly deposited daughters. The radioactivity of the filter media will not increase beyond this equilibrium unless the rate of air flow or the concentration of radon is increased. When air flow ceases, the α

radioactivity of the filter will diminish, as shown in curve B of fig. 7-3, with an effective half-life of about 40 min.

Indoor Radon

One of the most surprising developments during the past decade has been the finding that in many homes the concentration of ^{222}Rn (and radon daughter products) is so high as to involve risks far greater than those from many other pollution hazards that have attracted attention. The problem exists mainly in residential dwellings because the radon originates from the soil and ground water, which have their greatest effects on one or two-story buildings. The building materials themselves are a minor source of radon compared to soil and ground water, except where the materials contain relatively high concentrations of radium. This is true, for example, where gypsum board has been manufactured as a by-product of phosphate mining.

Interest in indoor radon developed worldwide in the mid-1970s, when convenient instrumentation became available. This happened to coincide with the emphasis placed on the need to conserve energy by weatherproofing homes, thereby reducing the rate of infiltration of outside air. It was widely believed that the high radon concentrations found were the result of the energy conservation programs, since the radon concentrations would be expected to be inversely proportional to the rate of ventilation which, in the U.S. homes, ranges from 0.5 to 1.5 air changes per hour. However, recent studies have shown that the ventilation rate does not have a major influence on the radon concentration in homes because the rate varies only within a factor of about 3, whereas other variables can have a more pronounced effect. **Steinhausler, (1975)** has shown that meteorological factors in particular can influence that indoor concentrations of radon and its daughters.

Radon can enter the indoor atmosphere in a number of ways, including diffusion from materials of construction or diffusion from soil through breaches in the foundation. However, there is evidence that diffusion of radon from soil is a minor source compared to the

movement of soil gases directly through the foundation as a result of slight pressure differentials that can result from barometric changes, temperature differentials, or wind velocity. Water supplies ordinarily make a small contribution to the indoor radon concentration, but can be the predominant source in areas where the radon content of ground water is unusually high. The approximate contributions of various sources to the indoor radon concentrations are given in Table 7 (Nero, 1985).

Table (7)
Approximate Contribution of Various Sources to
Indoor Radon Concentrations^a

Source	Single-famil homes (pCi/liter)	Apartments (pCi/liter)
Soil (estimates based on flux measurements)	1.5	< 1
Public water supplies	0.01 ^a	0.01 ^b
Building materials	0.05	0.1
Outdoor air	0.2	0.2

a From Nero (1985).

b Applies to 80% of the population that are served by public supplies. The contributions may average about 0.4 pCi/liter in homes using private wells, with higher concentrations in some areas.

Radon in Natural Gas

A radioactive gas that had properties similar to those of the "radium emanation" was first separated from petroleum in 1904 (Burton, 1904).

Thus, it is not surprising that radon is also present in natural gas. It is found at the wellhead in concentrations that average about 40 pCi/liter, but samples from some fields contain more than 1000 pCi/liter (Johnson, et al., 1973; Gesell, 1975). Natural gas at the wells contains from 55 to 98% methane and a much smaller percentage of other heavy hydrocarbons (ethane, propane, butane) as well as carbon dioxide, nitrogen, helium and water vapor. The gas is blended and processed to produce liquefied petroleum gas (LPG), a

product that consists mainly of propane with lesser amount of ethane. The boiling point of radon is close to that of propane and ethane, which has the effect of increasing the radon concentration in LPG while reducing the concentration in the methane-rich gas pumped into the pipelines. The long-lived daughters of ^{222}Rn (^{210}Pb and ^{210}Po) tend to accumulate on the interior surfaces of the LPG plant machinery and constitute a potential source of exposure of maintenance personnel (*Gesell, et al, 1975; Summerlin and Prichard, 1985*).

Since radon has a half-life of only 4.8 days, exposure of the consumers is influenced by storage time of LPG as well as the pipeline transit time. In 1977 it was estimated that radon from natural gas results in an average dose of about 5 mrem to about 125 million persons living in homes in which natural gas is used (*NCRP, 1977c*).

Lead-210 and Polonium-210

Lead-210 (sometimes called by its historic designation, radium D) is a 22-year β emitter separated from its antecedent ^{222}Rn by six short-lived α and β emitters. The longest-lived radionuclide between ^{222}Rn and ^{210}Pb is ^{214}Pb , which has a half-life of only 26.8 min. The ^{210}Pb is produced rapidly, but its long half-life allows very little to decay in the atmosphere before it precipitates to the earth's surface, mainly in rain or snow

The ^{210}Pb content of the atmosphere has been found to vary from 4.8×10^{-3} to 26×10^{-3} pCi/m³.

Radioactive disequilibria are found in the upper profiles of rocks and soils from which ^{222}Rn diffuses. Atmospheric transport and deposition of ^{210}Pb cause ^{210}Pb to be distributed in a more uniform pattern than the ^{226}Ra from which it is derived. It would also be expected that broad-leaved plants on which ^{210}Pb is deposited would be enriched in this radionuclide.

The $^{210}\text{Pb}/^{210}\text{Po}$ ratio depends on the length of time ^{210}Pb exists within a matrix and whether the polonium is selectively removed

from its site of production by chemical or biological mechanisms. Since ^{210}Po has a half-life of only 138 days, appreciable ingrowth in vegetation can take place during a single growing season, and additional buildup can occur during food storage, with equilibrium being reached in about 1 year. When ^{210}Pb is absorbed into the body, ingrowth of ^{210}Po can occur because ^{210}Pb has deposited in the skeleton, from which it leaves slowly, with a half-life of about 10^4 days.

Jaworowski, (1967) reported that rainwater contains from 1 to 10 pCi/liter of ^{210}Pb with a mean of about 2 pCi/liter. In an area having 1 m of rainfall per annum, this would indicate a fallout of about 2 mCi/km² per year. **UNSCEAR (1982)** estimates that the average daily intake of ^{210}Pb from dietary sources is about 3 pCi under normal circumstances. **Holtzman (1980)** found lower values for U.S. residents, a mean of 1.4 pCi, with a relatively narrow range of 1.3-1.6 pCi/day.

The concentration of ^{210}Pb in a "standard" diet was found by **Magno, et al. (1970)** to be not significantly different between locations in the United States and averaged 0.80 pCi/kg. His values were in good agreement with those reported by **Morse and Welford (1971)**, which ranged from 0.70 to 1.0 pCi ^{210}Pb /kg in eight U.S. sites. Using the ICRP model for lead transfer from the gastrointestinal tract to blood, it can be estimated that about 0.2 pCi/day of ^{210}Pb reaches the bloodstream of average inhabitants of the United States. According to Morse and Welford, air and food contributed about equally to the blood level. The ^{210}Po and ^{210}Pb contents of the standard diet are equal, indicating that in most foods there has been sufficient time for this nuclide to reach equilibrium with ^{210}Pb .

There are two notable groups in which the dose from ^{210}Po is apt to be exceptionally high: cigarette smokers and residents of the northlands who subsist on reindeer that consume lichens.

Marsden and Collins (1963) originally noted the presence of alpha activity in tobacco, following which **Radford and Hunt (1974)**

measured the concentrations of ^{210}Po in cigarettes, and **Little, et al. (1965)** demonstrated that ^{210}Po was present in the lungs of cigarette smokers. Subsequent investigators have studied this phenomenon (**Cohen, et al., 1980**) and concluded that the dose to the basal cells or the bronchial epithelium in cigarette smokers is increased by 2-12 mrem/year. Autoradiographs of the bronchial epithelium from one smoker showed the ^{210}Po to be highly localized. If the ^{210}Po remained fixed in position, the dose to the tissue surrounding the "hot spot" would be much higher than the average dose to the basal cells of the bronchial epithelium. Based on α -track measurements of autoradiographs of the lung tissue of 13 smokers, **Rajewsky and Stahlhofen, (1966)** had previously estimated that the basal cells of the subsegmental bronchi of cigarette smokers may receive as much as 86 mrem/year and the basal cells of the terminal bronchi may receive as much as 150 mrem/year. These values are higher than those reported by **Cohen, et al. (1980)**.

Compared to nonsmokers, about twice as much ^{210}Pb and ^{210}Po are found in the ribs (**Holtzman and Ilcewicz, 1966**). Ribs from smokers contained 0.28 pCi ^{210}Pb /g ash and 0.25 pCi ^{210}Po /g ash. The lungs contained 5.9 pCi ^{210}Po /kg wet. From these data, Holtzman estimated that the dose to the total skeleton is elevated by about 30% in cigarette smokers and the dose to the cells of the bone surface is increased by about 8%.

Polonium-210 is believed to enter tobacco by ingrowth of ^{210}Pb deposited on tobacco leaves from the atmosphere (**Martell, 1974**). Direct uptake of ^{210}Po from soil is probably not significant. The transfer coefficient to vegetables grown in ^{210}Po -contaminated alkaline soil has been found to range from about 6×10^{-4} to 10^{-6} (**Watters and Hansen, 1970**). **Hill (1966)** found a close correlation between the ^{210}Po and ^{137}Cs contents of human tissues from Canadian subjects, thus strengthening the suggestion that dietary habits that tend to favor broad-leaved vegetables or other foods subject to surface deposition may influence the ^{210}Po content of the tissues. Cesium-137 is known to be absorbed by humans mainly from

surface deposition on plants. Others have noted that pipe tobacco contains less ^{210}Po than cigarettes, apparently because the tobacco used for smoking pipes is not aged as long as cigarette tobacco (*Harley and Cohen, 1980*).

Potassium-40

Of the three naturally occurring potassium isotopes, only ^{40}K is unstable, having half-life of 1.3×10^9 years. It decays by β emission to ^{40}Ca , followed by K capture to an excited state of ^{40}A and γ -ray emission to the ^{60}A ground state. Potassium-40 occurs to an extent of 0.01% in natural potassium, thereby imparting a specific activity of approximately 800 pCi/g potassium. Representative values of the potassium content of rocks, as summarized by *Kohman and Saito (1954)*, indicate a wide range of values, from 0.1% for limestones through 1% for sandstones and 3.5% for granite.

The potassium content of soils of arable lands is controlled by the use of fertilizers. It is estimated that about 3000 Ci of ^{40}K is added annually to the soils of the United States in the form of fertilizer (*Guimond, 1978*).

A person who weighs 70 kg contains about 140 g of potassium, most of which is located in muscle. From the specific activity of potassium, it follows that the ^{40}K content of the human body is of the order of 0.1 μCi . This isotope delivers a dose of about 20 mrem/year to the gonads and other soft tissues and about 15 mrem/year to bone. Because of its relative abundance and its energetic β emission (1.3 MeV), ^{40}K is easily the predominant radioactive component in normal foods and human tissues. It is important to recognize that the potassium content of the body is under strict homeostatic control and is not influenced by variations in environmental levels. For this reason the dose from ^{40}K within the body is constant.

Seawater contains ^{40}K in a concentration of about 300 pCi/liter.

Rubidium-87

Of the two rubidium isotopes found in nature, ^{85}Rb and ^{87}Rb , only the latter is radioactive, with a half-life of 4.8×10^{10} years. Rubidium-87 is a pure β emitter, and it is present in elemental rubidium in the amount of 27.8%, which endows this element with a specific activity of 0.02 pCi/g. *Pertsov (1964)* quotes Vinogradov in listing the rubidium content of all but highly humic soils as about 0.01%. The ^{87}Rb content of ocean water has been reported to be 2.8 pCi/liter, with marine fish and invertebrates ranging from 0.008 to 0.08 pCi/g wet weight (*Mauchlie and Templeton, 1964*).

It is estimated (*UNSCEAR, 1982*) that the whole-body dose from ^{87}Rb is 0.6 mrem/year on average.

Natural Radioactivity in Phosphate Fertilizers

Phosphate fertilizers are used in huge amounts everywhere in the world and are essential for food production. The natural resource from which agricultural phosphorus is obtained is phosphate rock, found in sedimentary formations, usually interbedded with marine shales or limestones.

It has been known since early in this century that the phosphate rocks contain relatively high concentrations of uranium. The concentration of uranium in U.S. phosphate ores ranges from 8 to 400 ppm. The phosphate rocks from the important Florida deposits average 41 ppm, with ^{226}Ra in secular equilibrium. Phosphate rock is mined in huge quantities: it is reported that in 1974 about 26 million tons of ore were sold for fertilizer production in the United States, and that the ore contained about 1000 Ci of ^{226}Ra in equilibrium with ^{238}U (*Guimond, 1978*).

Several types of phosphate fertilizers are produced, and these are usually blended for application in the field. The radium and uranium tend to separate in the process of producing phosphoric acid, an important step in fertilizer manufacture (**Roessler, et al., 1979**). The radium passes to the gypsum produced as a by-product. A smaller fraction of the radioactivity is rejected in the process and ends up in the mounds of stored waste products.

Elevated concentrations of radon are found within structures located on land reclaimed from phosphate mines (**Guimond and Windham, 1980; Roessler, et al., 1983**). A survey of indoor radon and radon decay product exposures in buildings on or near the Florida phosphate deposits was conducted by **Guimond and Windham**, who found that 71% of homes built on reclaimed land contained more than 0.01 WL and 23% contained more than 0.05 WL. High levels were also found in structures built over mineral deposits that had not been mined, but relatively few measurements were made. Although these radon concentrations are above the average concentrations found in U.S. homes, it is noteworthy that many homes in areas of the United States contained higher concentrations than are found in the phosphate regions.

The high concentrations of ^{226}Ra also result in increased gamma-radiation exposure in homes built on reclaimed land. **Johnson and Bailey (1983)** found excess gamma dose to average 17 mrem/year, with value ranging as high as 166 mrem/year.

The radioactivity associated with land reclaimed prior to the 1940s tends to be higher than that in land reclaimed more recently, because methods have been developed for separating the phosphate-rich clay fraction of the deposit. A major fraction of the radium and uranium associated with the phosphate minerals was formerly returned to reclaimed land (**Roessler, et al., 1980**).

The contribution of uranium and radium to agricultural lands due to the application of phosphate fertilizers does not significantly affect the dose received from the general population (**Kirchmann, et al., 1980; Ryan, 1981**). However, phosphorus, in the form of mineral

phosphate rock, is sometimes added to cattle feed, and this practice can result in increased levels of uranium and radium in cow's milk (*Reid, et al., 1977*). However, continued application of phosphate fertilizers to soil over a period of many years could eventually double the radium and uranium content of the soil, which would result in a corresponding doubling of the dose to bone.

Natural Radioactivity in Building Materials

Two important by-products from the processing of phosphate rock are gypsum and calcium silicate slag, both of which are used in the building industry. Use of these products in buildings may increase the gamma radiation exposure.

The Federal Republic of Germany conducted a survey of 30,000 dwellings and found that, on average, the external radiation exposure was 33% higher within the dwellings than outdoors. Thus, although the building materials absorb the radiation that originates outside the building, exposure within the building is more than compensated by the presence of radionuclides in the materials of construction (*Kolb and Schmier, 1978*). Since most people spend 90% of their time indoors, the dose they receive from external natural radiation is increased somewhat. According to *Kolb and Schmier*, the absorbed dose to the population of the Federal Republic of Germany ranges from 40 to 80 mrad/year indoors (for continuous exposure) compared to 30-60 mrad/year for continuous outdoor exposure. Building materials can also be a source of radon, as was discussed earlier.

Natural Radioactivity in Fossil Fuels

Coal contains radionuclides of the uranium and thorium series, as well as ^{40}K . The quantity of radionuclides discharged to the atmosphere per ton of coal consumed depends on the concentration of radionuclides in the coal, the method of combustion, and the efficiency of fly ash recovery. The uranium and thorium concentrations in mined coal have been assembled from various sources by *Beck, et al. (1980)*.

The mean value for all coals sampled is 1.7 $\mu\text{g/g}$ for uranium, and 4.5 $\mu\text{g/g}$ for natural thorium, which is similar to the average concentrations found in soils and rocks. Most investigators have found that the various radionuclides of the uranium and thorium series are in secular equilibrium. *Beck, et al. (1980)* noted that there have been exceptions, but did not consider the evidence for disequilibria to be conclusive.

Because enrichment of some radionuclides occurs in the stack, the concentrations of ^{210}Pb and ^{210}Po in the fly ash are 5-10 times greater than in the original coal (*Beck, et al., 1980*). The emissions of radionuclides from a typical 1000-MW electric power plant in 1972 are given in Table 8.

Table (8)
Estimated does Equivalents to Lung of Maximum Exposed Individuals
from Model Coal-Fired Power Plant Releases^a

Nuclide	Does equivalent (mrem/year)	
	Modern plant	1972 reference plant
^{238}U - ^{234}U	0.009	0.46
^{230}Th	0.015	1.55
^{226}Ra	0.003	0.23
^{210}Pb	0.005	0.22
^{210}Po	0.075	3.10
^{232}Th	0.009	0.93
^{228}Th	0.018	1.85
^{228}Ra	0.004	0.38
Total	0.14	8.7

a From Beck, et al. (1980).

The normal levels of uranium and thorium in the environs are sufficiently high that changes due to the emissions from coal-fired power stations are barely detectable: some investigators (*Bedrosian, et al., 1970*) found no changes in the vicinity of an old, relatively poorly controlled plant, and *Beck, et al. (1980)* could find only slight changes in a carefully designed survey of several plants. It has been found, however, that the ^{226}Ra content of snow downwind of coal-

fired power plants is higher than the concentration in rainwater (*Jaworowski, et al., 1975*). The concentrations of naturally occurring radionuclides in airborne particles collected in the vicinity are elevated above background, and increase the dose to the lung, as shown in Table 7-14. It is seen that there has been major reduction in the dose equivalent received from a modern plant compared to the 1972 plant, which operated without stringent fly ash control.

The first report on radioactive emissions from coal-burning power plants (*Eisenbud and Petrow, 1964*) concluded that when the data were normalized for their radiotoxicity relative to the emissions from the first commercially operated pressurized water reactors, which had just begun operation, the dose from the fossil fuel emissions was greater than that from the nuclear reactors. However, the stringent requirements of the Federal Clean Air Act have since resulted in substantial reductions in the lung dose from the fly ash emissions, as shown in Table 7-14.

Induced Radionuclides

A number of radionuclides that exist on the surface of the earth and in the atmosphere have been produced by the interaction of cosmic rays with atmospheric nuclei. The most important of these are tritium (^3H), ^{14}C , and ^7Be , and of lesser importance are ^{10}Be , ^{22}Na , ^{32}P , ^{33}P , ^{35}S , and ^{39}Cl . The properties of these isotopes and the extent to which they have been reported in various media are listed in Table.

Carbon-14 is formed by ^{14}N capture of neutrons produced in the upper atmosphere by cosmic-ray interactions. The incident cosmic-ray neutron flux is approximately 1 neutron/sec per square centimeter of the earth's surface, and essentially all these neutrons disappear by ^{14}N capture (*Anderson, 1953*). The incident neutron flux integrated over the surface of the earth yields the natural rate of production of ^{14}C atoms, which has been estimated to be 0.038 MCi/year (*UNSCEAR, 1977*) and is believed to have been unchanged for at least 15,000 years prior to 1954, when nuclear weapons testing began to

alter the normal ^{14}C inventory to a noticeable extent (*NCRP, 1985b*).

Carbon-14 of natural origin is present in the carbon of all biota at the historically constant amount of 6 pCi/g C. After death of an organism, the ^{14}C equilibrium is no longer maintained, and the ratio of ^{14}C to ^{12}C diminishes at a rate of 50% every 5600 years, which makes it possible to use the ^{14}C content of organic materials for the purpose of measuring age (*Libby, 1952*).

The total carbon content of the body is approximately 18%, or 12.6 kg for a 70-kg man, and the ^{14}C body burden from natural sources is thus of the order of 0.1 μCi , but the dose is small owing to the soft quality of the ^{14}C β particles (0.01 MeV). It is estimated that the dose from ^{14}C is 2.4 mrem/year to the skeletal tissues of the body and 0.5 mrem/year to the gonads (*UNSCEAR, 1982*).

Tritium (^3H), a radioactive isotope of hydrogen, is formed from several interactions of cosmic rays with gases of the upper atmosphere (*Suess, 1968*). Existing in the atmosphere principally in the form of water vapour, tritium precipitates in rain and snow. Like ^{14}C , it is produced in thermonuclear explosions. The natural production rate of ^3H is estimated to be about 0.19 atom/cm² sec, corresponding to steady-state global inventory of about 26 MCi (*NCRP, 1979*).

The natural concentration of tritium in lakes, rivers, and potable waters was reported to have been 5-25 pCi/liter prior to the advent of weapons testing (*UNSCEAR, 1982*). The annual absorbed dose from tritium of natural origin is estimated to be about 0.001 mrem/year, uniformly distributed in all tissues.

The other nuclides formed from cosmic-ray interactions with the atmosphere may be potentially useful as tracers for studying atmospheric transport mechanisms, but relatively few observations have been reported.

Ureas Having Unusually High Natural Radioactivity

Except for brief mention of the occurrences of high levels of ^{226}Ra in well water, the discussion of natural radioactivity has thus far been

limited with levels to which people are normally exposed. There are, in addition, places in the world where the levels of natural radiation exposure are abnormally high.

The Mineral Springs

It has long been known that many mineral springs contain relatively high concentrations of radium and radon, and in many places in the world the radioactivity of the springs has been exploited for its alleged curative powers.

Published values of ^{226}Ra in mineral waters ranged to more than 100 pCi/liter (*United Nations, 1958*), which is several thousand times greater than the values normally reported for public water supplies. However, to illustrate the fact that the values reported for spring waters are not typical of the drinking water of the region, the ^{226}Ra concentration in tap water of Bad Gastein is reported to be 0.62 pCi/liter, compared to 100 pCi/ml for some of the local springs (*Muth, et al., 1957*).

About 5 million liters of water that contain high concentrations of radon are discharged daily from hot springs in Bad Gastein. The water is conveyed to hotels and bathhouses, where 58 Ci/year are released to the atmosphere. *Uinov, et al. (1981)* have described the manner in which radon emanations from this water result in exposure to the city residents, tourists who visit the spas, and employees of the many bathhouses. They estimate that exposure of some of the attendants is as high as 40 WLM/year.

Monazite Sands and Other Radioactive Mineral Deposits

Major anomalies in the concentrations of radioactive minerals in soil have been reported in two countries, Brazil and India.

In Brazil, the radioactive deposits are of two distinct types: the monazite sand deposits along certain beaches in the states of Espirito Santos and Eo de Janeiro and the regions of alkaline intrusives in the state of Minas Gerais (*Roser, et al., 1964; Brazilian Academy of Sciences, 1977*).

Monazite is a highly insoluble rare earth mineral that occurs in some beach sand together with the mineral ilmenite, which gives those sands a characteristic black color. The black sands are advertised for their radioactivity and are much sought by some Brazilian tourists for their perceived benefits to health. The external radiation levels on these black sands range up to 5 mR/hr, and people come from long distances to relax on the sands and enjoy the many hotels that have been constructed to care for their needs. The most active of these Brazilian vacation towns is Guarapari, which has a stable population of about 7000 people and an annual influx of 10,000 vacationers. Some of the major streets of Guarapari have radiation levels as high as 0.13 mR/h, which is more than 10 times the normal background. Similar radiation levels are found inside some of the buildings in Guarapari, many of which in parts of the village that are not built on monazite sand, are nevertheless elevated in radioactivity because the beach sands were incorporated into the building materials. **Roser and Cullen (1964)** undertook extensive external radiation measurements throughout the black sands region and concluded that almost all the approximately 60000 inhabitants of the region were exposed to abnormally high radiation levels, but that only a small number (about 6600) were exposed to more than 0.5 rem/year. The population is too small to permit meaningful epidemiologic investigations to be conducted, but cytogenetic studies have shown a higher than normal frequency of abnormalities in blood chromosomes (**Barcinski, et al. 1975**).

The principal radionuclides in monazite are from the ^{232}Th series, but there is also some uranium present and, therefore, some opportunity for ^{226}Ra uptake. However, very little food is grown in the monazite areas of Brazil, and the diets of the local inhabitants are derived principally from outside sources. The exposures in the monazite areas are due primarily to external radiation, and the internal dose is not believed to be significant (**Eisenbud, et al., 1964; Penna Franca, et al., 1970**).

In the state of Kerala, on the southwest coast of India, the

monazite deposits are more extensive than those in Brazil, and about 100,000 persons inhabit the area. The dose from external radiation is, on average, similar to the exposures reported in Brazil (500-600 mR/year), but individual exposures up to 3260 mR/year have been reported (**Sunta, et al., 1982**). The epidemiologic studies that might be possible with a population of this size have not as yet been undertaken, but some reports have been published. A higher than normal incidence of mongolism (Down's syndrome) has been reported in the high-background area (**Kochupillai, et al., 1976**), but this has been refuted (**Sundaram, 1977**). In contrast to Brazil, there have as yet been no reports of cytogenetic studies in humans. **Mistry, et al. (1970)** reported that uptake of ^{228}Ra by food grown in the monazite area is greater than the uptake reported from Brazil.

A unique anomaly located near Pocos de Caldos, also in the state of Minas Gerais, is the Morro do Ferro, a hill that rises about 250 m above the surrounding plateau. Near the summit of the hill is a near-surface ore body that contains about 30,000 tonnes of thorium and an estimated 100,000 tonnes of rare earth elements. The ambient gamma-radiation levels near the summit of the hill range from 1 to 2 mR/hr over an area of about 30,000 m² (**Eisenbud, et al., 1984**). The flora from this hill have absorbed so much ^{228}Ra that they can readily be autoradiographed.

Studies have been undertaken of the exposures of rats living underground on the Morro do Ferro. Of particular interest is the dose to these rodents due to inhalation of ^{220}Rn , which was found by **Drew and Eisenbud (1966)** to be present in the rat burrows in concentrations up to 100 pCi/ml. The dose to basal cells of the rat bronchial epithelium was estimated to be in the range of 3000 to 30,000 rem/year. Of 14 rats trapped and sacrificed for pathological study, none was observed to show any radiation effects. This is of little significance, since the Morro do Ferro is a relatively small area, and if rats were affected by this exposure they could be replenished rapidly from the surrounding normal areas. By using thermoluminescent dosimeters implanted in trapped rats that were

REFERENCES

- Anderson, E.C. (1953).** The production and distribution of natural radiocarbon. *Annu. Rev. Nucl. Sci.* **2**, 63.
- Barcinski, M.A., Abreu, M.C.A., de Almeida, J.C.C., Naya, J.M., Fonseca, L.G. and Castro, L.E. (1975).** Cytogenetic investigation in a Brazilian population living in an area of high natural radioactivity. *Am. J. Hum. Genet.* **27**, 802-806.
- Barreira, F. (1961).** Concentration of atmospheric radon and wind directions *Nature (London)* **190**, 1092.
- Blifford, I.H., Lockhart, L.b., et al. (1952).** "On the natural Radioactivity of the Air," Rep. 4036. Nav. Res. Lab., Washington, D.C.
- Brinck, W.L., Schlickelman, R.J., Bennet, D.L., Bell, C. and Markwood, I.M. (1978).** Radium removal efficiencies in water treatment processes. *J. Am. Water Works Assoc.* **70**, 31-43.
- Burton, E.F. (1904).** A radioactive gas from crude petroleum. *Philos. Mag.* **8**, 498-508.
- Cantril, S.T. and Parker, H.M. (1945).** "The Tolerant Dose," MDDC 110. USAEC, Washington, D.C.
- Catlin, R.J. (1980).** "Assessment of the Surveillance Prothium of the High-Level Waste Storage Tanks at Hanford." Rep. to U.S. Dep. Energy, Acad. Sec. Environ., Washington, D.C.
- Chamberlain, A.C. (1955).** Aspects of Travel and Deposition of Aerosol and Vapor Clouds." Rep. HP/R1261. U.K. At. Energy Auth., Res. Comp.
- Chamberlain, A.C. (1960).** Aspects of the deposition of radioactive and other gases and particles *Int. J. Air Pollut.* **3**, 57-78.
- Chamberlain, A.C. and Chadwick, R.C. (1966).** Transport of iodine from atmosphere to ground. *tellus* **18**, 226-237.
- Chang, T.Y., Cheng, W.L. and Weng, P.S. (1974).** Potassium, uranium, and thorium content of building materials of Taiwan *Health Phys.* **27**, 345, 387.
- Chepil, W.A. (1957).** Erosion of soil by wind. In "The Yearbook of Agriculture," pp 308-314. U.S. Dep. Agric., Washington, D.C.
- Clapp, C.A. (1934).** "Cataract." Lea & Febiger, Philadelphia, Pennsylvania.
- Clark, H.M. (1954).** The occurrence of an unusually high level radioactive rainout in the area of Troy, N.Y. *Science* **119**, 619-622.
- Clarke, R.H. (1974).** An analysis of the Windscale accident using the WEERIE code. *Am. Nucl. Sci. Eng.* **1**, 73.
- Claus, W.D. (1958).** "Radiation Biology and Medicine." Addison-Wesley, Reading, Massachusetts.
- Clayton, G.D., Arold, J.R. and Patty, F.A. (1955).** Determination of sources of particulate atmospheric carbon. *Science* **122**, 751-753
- Clegg, I.W. and Foley, D.D. (1958).** "uranium Ore Processing." Addison-Wesley, Reading Massachusetts.
- Clements, W. and Wilkening, M. (1974).** Atmospheric pressure effects on radon transport across the earth-air interface. *J. Geophys. Res.* **79**, 5025-5029.
- Codman, E.A. (1902).** A study of the cases of accidental x-ray burns hitherto recorded. *Philadelphia med. J.* pp. 438-442.

- Cohen, B.L. (1980).** Society's valuation of life saving in radiation protection and other contexts. *Health Phys.* **38**, 33-51.
- Cohen, B.L. (1982).** Effects of ICRP 30 and the 1980 BEIR report on hazard assessment of high level wastes. *Health Phys.* **42**, 133-143.
- Cohen, B.S., Eisenbud, M. and Hartey, N.H. (1980).** Measurement of the α -radioactivity on the mucosal surface of the human bronchial tree. *Health Phys.* **39**, 619-632.
- Colby, L.J. (1976).** Fuel reprocessing in the U.S.: A review of problems and some solutions. *Nucl. News 9La Grange Park, Ill.* Jan.
- Collins, J.T., Bell, M.J. and Hewitt, WM. (1978).** Radioactive waste source terms. In "Nuclear Power waste Technology" (A.A. Moghissi, H.W. Godbee, M.S. Ozker, and M.W. Carter, eds.), pp. 167-199. Am. Soc. Mech. Eng., New York.
- Colorado Committee for Environmental Information (1970).** "Report on the Dow Rocky Flats Fire: Implications of Plutonium Releases to the Public Health and Safety," Subcommittee on Rock Flats, Boulder, Colo., Jan. 13, 1970, Rep. HASL-235.
- Cohen, B.S., Eisenbud, M. and Hartey, N.H. (1980).** Measurement of the α -radioactivity on the mucosal surface of the human bronchial tree. *Health Phys.* **39**, 619-632.
- Drew, R.T. and Eisenbud, M. (1966).** The normal radiation dose to indigenous rodents on the Morro do Ferro, Brazil. *Health Phys.* **12**, 1267-1274.
- Eisenbud, M. (1984).** Sources of ionizing radiation exposure. *Environment* **26**(10), 1-33.
- Evans, R.D. and Raitt, R.W. (1935).** The radioactivity of the earth's crust and its influence on cosmic ray electroscopes observations made near ground level. *Phys. Rev.* **48**, 171.
- Faul, H. (1954).** "Nuclear Geology." Wiley, New York.
- Fisenne, I.M. and Keller, H.W. (1970).** "Radium-226 in the Diet of Two U.S. Cities." Rep. HASL-224. USAEC, New York.
- Fisenne, I.M., Keller, H.W. and Harley, N.H. (1981).** Worldwide Measurement of ^{226}Ra in human bone: Estimate of skeletal a dose *Health Phys.* **40**, 163-171.
- Fresco, J., Jetter, E. and Harley, J. (1952).** Radiometric properties of the thorium series. *Nucleonics* **10**, 60.
- Gesell, T.F. (1983).** Background atmospheric ^{222}Rn concentrations outdoors and indoors: A review. *Health Phys.* **45**, 289-302.
- Gold, S., Barkhau, H.W., Shleien, B. and Kahn, B. (1964).** measurement of naturally occurring radionuclides in air. In "The Natural Radiation Environment I" (J.A.S. Adams and W. M. Lowder, eds.), pp. 369-382. Univ. of Chicago Press, Chicago, Illinois.
- Guimond, R.J. (1978).** The radiological aspects of fertilizer utilization in radioactivity in consumers' products. In "Radioactivity in Consumer Products" (A.A. Moghissi, P. Paras, M.W. Carter, and R.f. Barker, eds), USNRC Rep. NUREG/CP0003, pp. 381-393. NTIS, Springfield, Virginia.
- Guimond, R.J. and Windham, S.T. (1980).** Radiological evaluation of structures constructed on phosphate-related land. In "Natural Radiation Environment III" (T.F. Gesell and W.M. Lowder, eds.),

- CONF-78042, Vol. 2, pp. 1457-1475. USDOE, Washington, D.C.
- Harley, J.H. (1978).** Radioactivity in building materials. In "Radioactivity in Consumer Products" (A.A. Moghissi, P. Paras, M.W. Carter, and R.F. Barker, eds.), Rep. NUREG/CP, 0001, pp. 332-313. USNRC, Washington, D.C.
- Harley, N.H. and Cohen, B. (1980).** Polonium-210 in tobacco. In "Radioactivity in Consumer Products" (A.A. Moghissi, P. Paras, M.W. Carter, and R.F. Barker, eds), Rep. NUREG/-CP0001, pp. 199-216. USNRC. Washington, D.C.
- Hess, C.T., Michel, J., Horton, T.R., Prichard, H.M. and Coniglio, W.A. (1985).** The occurrence of radioactivity in public water suppliers in the United States. *Health Phys.* **48**, 553-586.
- Holtzman, R.B. (1980).** Normal dietary levels of radium-226, radium-228, lead-210, and polonium-210 for men. In "The Natural Radiation Environment III" (T.F. Gesell and WM. Lowder, eds.), USAEC CONF-780422, Vol. 2, pp. 755-782. NTIS, Springfield, Virginia.
- Holtzman, R.B. and Heewicz, F.H. (1966).** Lead-210 and polonium-210 in tissues of cigarette smokers. *Science* **153**, 1259-1260.
- International Commission on Radiological Protection (1975).** "Reference Man: Anatomical Physiological and Metabolic Characteristics," ICRP Publ. No. 23. Pergamon, Oxford.
- Jaworowski, Z. (1967).** "Stable and Radioactive Lead in Environment and Human Body." Nucl. Energy Inf. Cent., Warsaw.
- Jaworowski, Z., Bilkiewicz, J., Kownacka, L. and Wlodek, S. (1975).** Artificial sources of natural radionuclides in environment. In "Natural Radiation Environment II" (J. A. S. Adams, W.M. Lowder, and T.F. Gesell, eds.), CONF-71, p. 809. USDOE, Washington, D.C.
- Johnson, R.H., Jr., Bernhardt, D.E., Nelson, N.S. and Calley, H.W., Jr. (1973).** "Assessment of Potential Radiological Health Effects from radon in Natural Gas." Rep. EPA-520/1-73-004. USEPA, Washington, D.C.
- Johnson, W.B. and Bailey, P.G. (1983).** Study of Radon daughter concentrations in Polk and Hillsborough Counties. *health Phys.* **45**, 432-434.
- King, P.T., Michel, J. and moore, W.S. (1982).** Groundwater geochemistry of Ra-228, Ra-226 and Rn-222, *Geochim. Cosmochim. Acta* **46**, 1173-1182
- Kochupillan, N., Verma, I.C., Grewal, M.S. and Ramalingaswami, V. (1976).** Down's syndrome and related abnormalities in an area of high background radiation in coasted kerala....
- Kolhman, T. and Saito, N. (1954).** Radioactivity in geology and cosmology. *Annu. Rev. Nucl. Sci.* **4**.
- Kolbo, W. and Schmier, H. (1978).** Building material induced radiation exposure of the population. In "Radioactivity in Consumer Products" (A.A. Moghissi, P. Paras, M.W. Carter, and R.F. Barker, eds.), Rep. NUREG/CP-0001, pp. 344-349. USNR, Washington, D.C.
- Lappenbusch, W.L. and Cothorn, C.R. (1985).** Regulatory development of the interim and revised regulations for radioactivity in drinking water: Past and present issues and problems. *Health Phys.* **48**, 535-551.

- Libby, W.f. (1952).** "Radiocarbon Dating." Univ. of Chicago Press, Chicago, Illinois.
- Little, J.B., (1958).** "Atmospheric Radioactivity studies at U.S. Naval Research Laboratory," Rep. 5249. Nav. Res. Lab., Washington, D.C.
- Lockhart, L.B. (1964).** Radioactivity of the radon-222 and radon-220 series in the air at ground level. In "The Natural Radiation Environment I" (J. A. S. Adams and W. M. Lowder, eds.), pp. 331-344. Univ. of Chicago Press, Chicago, Illinois.
- Magno, P.J., Groulx, P.R. and Apidianakis, J.C. (1970).** Lead-210 in air and total diets in U.S. *Health Phys.* **18**, 383-388.
- Marsden, E. and Collins, M.A. (1963).** Particle activity and free radicals from tobacco. *Nature (London)* **198**, 962.
- Martell, E.A. (1974).** Radioactivity of tobacco trichomes and insoluble cigarette smoke particles. *Nature (London)* **249**, 215-217.
- Mauchline, J. and Templeton, W.L. (1964).** Artificial and natural radioisotopes in the marine environment. *Annu. Rep. Occanogr. Mar. Biol.* **2**, 229-279.
- Mayneord, W.V., Radley, J.M. and Turner, R.C. (1958).** The alpha-ray activity of humans and their environment. *Proc. U.N. Int. Conf. Peaceful Users At Energy, 2nd. Geneva.*
- Mayneord, W.V., Turner, R.C. and Radley, J.M. (1960).** Alpha activity of certain botanical materials. *Nature (London)* **187**, 208.
- Mistry, K.B., Bharathan, K.G. and Gopal-Ayengar, A.R. (1970).** Radioactivity in the diet of population of the Kerala coast including monazite bearing high radiation areas. *Health Phys.* **19**, 353-542.
- Morse, R.S. and Welford, G.A. (1971).** Dietary intake of ^{210}Pb . *Health Phys.* **21**, 53-55.
- Muth, H., Schraub, A., Aurand, K. and Hantke, H.H. (1957).** Measurements of normal radium burdens. *Br. J. Radiol., Suppl.* **7**.
- National Council on Radiation Protection and Measurements (1979).** "Tritium in the Environment," NCRP Rep. No. 62. Bethesda, Maryland.
- National Council on Radiation Protection and Measurements (1984b).** "Exposures from the Uranium Series with Emphasis on Radon and its Daughters," NCRP Rep. 77. Bethesda, Maryland.
- National Council on Radiation Protection and Measurements (1985b).** "Carbon-14 in the Environment," NCRP Rep. No. 81. Bethesda, Maryland.
- Nero, A.V., Jr. (1985).** What we know about indoor radon. Testimony prepared for hearings on "Radon Contamination: Risk Assessment and Mitigation Research." held by the Subcommittee on Natural Resources, Agricultural Research and Environmental Committee on Science and Technology. U.S. House of Representatives (Oct. 10, 1985) USGPO, Washington, D.C.
- Penna Franca, E., Fiszman, M., Lobao, N., Costa Ribeiro, C., Trindade, H., Dos Santos, P.L. and Batista, D. (1968).** Radioactivity of Brazil nuts. *Health Phys.* **14**, 95-99.
- Penna Franca, E., Fiszman, M., Lobao, N., Trindade, H., Costa Riberiro, C., and Santos, P.L. (1970).** Radioactivity in the diet in high background areas on Brazil. *Health Phys.* **19**, 657-662.

- Rajewsky, B and Stahllofen, W. (1966).** ^{210}Po activity in the lungs of cigarette smokers. *Nature (London)* **209**, 1312-1313.
- Rankama, K. (1954).** "Isotope Geology." McGraw-Hill, New York.
- Rankama, K. and Sahama, T.G. (1950).** "Geochemistry." Univ. of Chicago Press. Chicago, Illinois.
- Reid, D.G., Sackett, W.M. and Spaulding, R.F. (1977).** Uranium and radium in livestock feed supplements. *Health Phys.* **32**, 535-540.
- Reid, G.W., Lassovszky, P. and Hathaway, S. (1985).** Treatment, waste management and cost for removal of radioactivity from drinking water. *Health Phys* **48**, 671-694.
- Roessler, C.E., Smith, Z.A., Bolch, W.F. and Prince, J.R. (1979).** Uranium and radium-226 in Florida phosphate materials *Health Phys.* **37**, 269-277.
- Roessler, C.E., Kautz, R., Bolch, W.E. Jr., and Wethington, J.A., Jr. (1980).** The effect of mining and land reclamation on the radiological characteristics of the terrestrial environment of Florida's phosphate regions. In "Natural Radiation Environment III" (T. F. Gesell and W. M. Lowder, eds.), CONF-780422, Vol. 2, pp. 1476-1493. USDOE, Washington, D.C.
- Roser, F.X and Cullen, T.L. (1964).** External radiation measurements in high background regions of Brazil. In "The Natural Radiation Environment I" (J. A. S. Adams and W. M. Lowder. eds.), pp. 825-836. Univ. of Chicago Press, Chicago, Illinois.
- Roser, F.X., Kegel, G. and Gullen, T.L. (1964).** Radiogeology of some high-background areas of Brazil. In "The Natural Radiation Environment I" (J.A.S. Adams and W.M. Lowder, eds.). pp. 855-872. Univ. of Chicago Press, Chicago, Illinois.
- Seaborg, G.T. (1958).** "The Transuranic Elements." Addison-Wesley, Reading, Massachusetts.
- Suess, H.E. (1958).** The radioactivity of the atmosphere and hydrosphere. *Annu. Rev. Nucl. Sci.* **8**, 243.
- Sundaram, K. (1977).** Down's syndrome in Kerala. *Nature (London)* **267**, 728.
- Sunta, C.M., David, M., Abani, M.C., Basu, A.S. and nambi, K.S.V. (1982).** Analysis of dosimetry data of high natural radioactivity areas of southwest coast of India. In "The Natural Radiation Environment IV" (K. G Vohra, U. S. Mishra, K. C. Pillai, and S. Sadasiva, eds.). Wiley Eastern, Bombay/New Delhi.
- Tanner, A.B. (1964).** Radon migration in the ground: A review. In "The Natural Radiation Environment I" (J. A. S. Adams and W. M. Lowder, eds.), pp. 161-190. Univ. of Chicago Press, Chicago, Illinois.
- United Nations Scientific Committee on the Effects of Atomic Radiation (1982).** 32nd Session, Suppl. No. 40 (A/37/45). United Nations, New York.
- Wei, L., et al. (1985).** Report of Third Stage (1982-1984). health survey in high background area in Yangjiang, China. *Chin. J. Radiat. Med. Prot.* **5**, 144-153.

- Welford, G.A. and Sutton, D. (1957).** "Determination of the Uranium Content of the National Bureau of Standards Iron and Steel Chemical Standards." Rep. NYOO-1755. USAEC, Washington, D.C.
- Wilkening, M.H. (1964).** Radon-daughter ions in the atmosphere. In "The Natural Radiation Environment I" (J. A. S. Adams and W. M. Lowder, eds.), p. 359-368. Univ. of Chicago Press, Chicago, Illinois.
- Wilkening, M.H. (1982).** Radon in atmospheric studies: A review. In "The Natural Radiation Environment IV" (K. G. Vohra, U. C. Mishra, K. C. Pillai, and S. Sadasivan, eds.), pp. 565-574. Wiley Eastern, Bombay/New Delhi.
- Wren, M.E., Singh, N.P., Cohen, N., Ibrahim, S.A. and Saccomanno, G. (1981).** "Thorium in Human Tissues," Rep. NUREG/CR-1227. NTIS, Springfield, Virginia.
- Wrenn, M.E., Durbin, P.W., Howard, B., Lipszstein, J., Rundo, J., Still, E.T. and Willis, D.L. (1985).** Metabolism of ingested U and Ra. *Health Phys.* **48**, 601-633.