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Cover illustration: Simulated biome patterns at current climate (300 ppm CO_2). The changes due to greenhouse gases and climate change depicted in Figures 20-2 and 20-3 are shifts in these patterns (see Chapter 20, pages 358 and 360). [Note the color classification of biomes has been labled A to O in Chapter 20, page 358]

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28 Radionuclides

Chester R. Richmond and Andrea A. Richmond

28.1 Introduction

"Every falling raindrop and snowflake carries some radioactive material to earth, while every leaf and blade of grass is covered with an invisible film of radioactive material." (Rutherford, 1905).

Humans and their environment are made from both radioactive and stable elements. The atoms, radioactive and non radioactive, that make people had their origin in the stars. Humans are children of the stars and all life evolved in the presence of cosmic and terrestrial radiation. The Earth is thought to be about 4 500 million years old. It is believed that human beings first appeared about 4 million years ago — recent developments on the evolutionary scale. Except for primordial hydrogen, all the stable elements from which humans are made were originally produced elsewhere in the universe (which is somewhere between 8 000 million and 15 000 million years old). Humans are indeed children of the stars. Mankind evolved in the presence of radiation in the body, in food, in soil and in air. Each inhabitant of planet Earth is also radioactive because of radionuclides in the body. It is ironic that so many people fear radiation as something completely foreign and unnatural.

All energy used by Earth's inhabitants comes directly or indirectly from two nuclear sources; the sun and heat produced by radioactive elements and their progeny incorporated into the Earth during its creation some 4 500 million years ago. About 2 000 million years ago, during pre-Cambrian times, deposits of Uranium ore in western Africa produced a natural nuclear fission reactor. Evidence of this prehistoric reactor was found during mining operations at the Okla site in Gabon during 1972. Analysis of the core samples showed unusual ratios of $^{238}U/^{235}U$.

Naturally occurring (primordial) radionuclides are non-homogeneously distributed around the earth as are the stable nuclides. Thus, elevated levels of radionuclides from the ^{238}U and ^{232}Th decay series exist in locations such as Brazil, India, China and France. Man appeared on earth several million years ago. Primordial radionuclides, including ^{40}K, ^{87}Rb and cosmogenic ^{3}H, ^{7}Be, ^{14}C and ^{22}Na enter the body from air, food and water. Body burdens of these radionuclides depend on location and diet.

Early in the twentieth century, scientists produced artificial radionuclides in accelerators and other machines for use in research, medicine and other applications. Subsequent discovery of nuclear fission in the late 1930s produced nuclear energy

Box 28-1 Definitions

Radionuclide is a radioactive isotope of an element. Isotopes of a given element have the same atomic number but different masses. For example, ^{235}U and ^{238}U are radionuclides of uranium which has an atomic number of 93.

Half-life of a radionuclide is the rate at which its atoms decay. For example, ^{131}I decays with an 8 day half-life. Half the atoms in a sample will decay every 8 days. After 8 days, half of the original number of atoms will have decayed and 50% remain. After two half-lives, 16 days, another 50% decay and 25% of the oroginal number of atoms remain.

Activity, A, is the number of radioactive decays per unit time occurring in a given source. Unit is becquerel (Bq), one disintegrating atom per second. The old unit is the curie (Ci), 3.7×10^{10} disintegrating atom per second. One Ci $=3.7 \times 10^{10}$ Bq.

Absorbed Dose, D, is the energy absorbed from the radiation per unit mass of exposed tissue. Unit is the gray (Gy), 1 joule per kilogram. The old unit is the rad, 100 ergs per gram. One Gy = 100 rad.

Equivalent Dose, H, is the absorbed dose weighted for the effectiveness of the radiation for causing biological damage. Previously called the Dose Equivalent. Unit is the Sievert (Sv). One H (Sv) = Q x D (Gy) where Q is the radiation-weighting factor. The old unit is the rem. H (rem) = Q x D (rads). Q ranges from 1 to 20 depending on the radiation type and energy of the radation. One Sv = 100 rem.

Effective Dose, E, is the sum of the weighted equivalent doses in all tissues and organs of the body. Effective dose is calculated for the whole body in which the dose-equivalent for various organs are weighted to account for different sensitivities of the organs to radiation: $E = \Sigma\, w_T\, H_{T,R}$ where w_T is the tissue-weighting factor for organ or tissue T; and $H_{T,R}$ is the equivalent dose in tissue or organ T due to a given radation R. Previously called the Effective Dose Equivalent. Unit is the Sievert (Sv). The old unit is the rem. One Sv = 100 rem.

Collective Dose, the sum total of all individual radiation doses to a specified group or population. Unit is the person Sievert.

Committed Dose. The time integration of dose rate, ususally for a 50 year period for adults and 70 years for children. The committed dose may be expressed in terms of the collective dose or the dose to an individual and may be expressed in terms of the various dose quantities such as absorbed dose and effective dose.

Adapted from NCRP 87 with permission.

radiation dose to men is generally higher than that to women because potassium concentration is directly proportional to lean body mass. Potassium, a physiologically important cation, is also under homeostatic control (NCRP, 1987).

Some radionuclides in the body arise from Thorium-232 (^{232}Th, 14.05×10^9 years physical half-life) and its decay products. Others, such as radon and its daughter products, are continually produced as part of the uranium decay series. Uranium-238 (^{238}U) has a physical half-life of 4.47×10^9 years. About 15 million ^{40}K atoms, 30 000 radon, polonium, bismuth and lead atoms and 7 000 uranium atoms disintegrate each hour within the body (NRPB, 1986). It is not surprising that it is difficult to measure

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with its attendant radioactive fission products. Man then learned to mine radioactive ore and to fission isolated radionuclides in nuclear reactors.

Man also fashioned new weapons from enriched U and Pu from nuclear reactors. Enormous secret research and production facilities developed around the military applications of U an Pu in several countries. Weapons production and testing and the use of U in reactors, produced wide-scale redistribution of radioactive materials. Predominant among the released materials are the high-yield fission products of Cs, Sr and I. Large areas suffering radioactive contamination from accidents and operational releases in several countries are the legacy of the nuclear weapons programs. These include sites in the USA, UK, the former Soviet Union, France, China, Australia and the Pacific Atolls.

Accidents involving nuclear weapons produced environmental contamination in Palomares, Spain and Thule, Greenland. Other releases, notably ^{238}Pu from power sources and several reactors, came from accidents in the USA and USSR space programs. Nuclear power station accidents, particularly the power station at Chernobyl, added radioactive contaminants to large areas in Europe. Consequently, public health officials are interested in the allowable levels in food and water and the dose per unit intake for radionuclides following contamination from accidents. On a much smaller scale the use of radioactive materials in consumer products has contributed to radioactive contamination.

Clean-up of contaminated areas and environmental restoration are now major activities in several countries. In addition to the high-yield fission products, radioactive actinides are of concern because of their larger biological effects. Major uncertainties exist as to where contaminated material should be disposed of or stored. Ocean disposal is not an option in many cases. Environmental clean-up options include in-situ modification and a means of protecting disposal areas from man and weathering. Burning of some radionuclides in nuclear reactors and accelerators is also possible as a means of recycling. This may be of particular importance in reducing the nuclear weapon stockpiles of U and Pu.

Until recently exposure to radon in homes was not considered as a potential health problem. Many countries now evaluate the health effects of it and attribute a large fraction of the naturally occurring radiation dose to man from exposure to radon decay products.

28.2 Primordial and Cosmogenic Radionuclides

Some radionuclides in people and the environment are primordial in origin and have physical half-lives measured in billions of years. For example, Potassium-40 (^{40}K) has a physical half-life of 1.28×10^9 years. About a quarter of the Earth's original ^{40}K remains today. A small fraction, 0.0118 %, of all potassium isotopes are ^{40}K. Thus, the potassium in each person contains ^{40}K that delivers an annual radiation dose equivalent of approximately 0.25 mSv (25 mrem) (see Box 28-1 for definitions). The

small amounts of radioactive contaminants in human subjects because of the "natural" body radioactivity.

Some of the radionuclides in the body and in the environment, such as Hydrogen-3 (^{3}H), Beryllium-7 (^{7}Be), Carbon-14 (^{14}C) and Sodium-22 (^{22}Na) are cosmogenic in origin and continuously replenished. The radiation dose (annual effective dose equivalent) to a person from the most important cosmogenic radionuclide, ^{14}C, is about 0.01 mSv (1 mrem). Globally, people's exposure is similar because CO_2 is uniformly distributed at ground level. Until recently (about 100 years ago) humankind was ignorant of this internal radioactive burden. Radioactivity was not discovered until 1896 by Henri Becquerel and the term isotope was not coined until 1913 by Frederick Soddy.

All energy used by Earth's inhabitants comes directly or indirectly from two nuclear sources; the sun and heat produced by radioactive elements and their progeny incorporated into the earth during its creation some 4 500 million years ago. Radioactive elements incorporated into the earth at its formation produce enormous amounts of heat. At times this heating is dramatically expressed as major volcanic releases. More often the release is continuous and less dramatic, although still very spectacular as seen in the geysers in the Yellowstone National Park in the USA.

Suboceanic ridges and terrestrial volcanoes release hot materials into the oceans forming new land masses. The Earth's crust is continually recycled through the geologic process of subduction on a timescale that is very long compared to the human life span.

Rutherford believed that heat produced from alpha particles in the earth kept the temperature constant. He believed that Lord Kelvin's estimate of 100 million years for the age of the earth was in error because Kelvin believed the earth was cooling off. Rutherford actually knew what an alpha particle was. Kelvin never changed his belief on this subject (Morris, 1981).

Primordial radionuclides, including ^{40}K and Rubidium-87 (^{87}Rb) and cosmogenic ^{3}H, ^{7}Be, ^{14}C and ^{22}Na enter the human body from air, food and water. Body burdens of these radionuclides depend both on location and diet. Naturally occurring (primordial) radionuclides are distributed around the earth, sometimes non-homogeneously, as are the stable nuclides. Thus, elevated levels of radionuclides from primordial ^{238}U and ^{232}Th and their decay series exist at locations in India, China, Brazil, Egypt and France. Residents of these areas receive several or many times the out-of-doors dose rate of 0.45–1.3 mSv (45–130 mrem) received by people in other parts of the world.

28.3 Uranium

There are 14 uranium radioisotopes. However, naturally occurring uranium normally contains a mixture of three radioisotopes of which ^{238}U is the most abundant (99.276%) with much smaller amounts of ^{235}U (0.718%) and ^{234}U (0.0056 %). Thus, the ^{235}U content of uranium must be enriched to higher concentrations before it can

be used for power generation (3–4%) or in nuclear weapons (90%). Uranium-238 has a long physical half-life, 4.47×10^9 years, about the same as the age of the Earth. Its decay series radionuclides are in secular equilibrium on a world-wide basis. However, some disequilibrium is caused by long-term geologic processes and human actions such as mining. The same is true for thorium and phosphate ores (NCRP, 1987).

Uranium oxide was discovered in the uranium ore pitchblende by the German Martin Klaproth in 1789. The French chemist, E. Peligot first obtained elemental uranium in 1841. Thorium was first identified by the Swede Berzelius in 1828. The French husband and wife team, Pierre and Marie Curie, discovered radium and polonium in 1898.

About 2 000 million years ago, during pre-Cambrian times, deposits of uranium ore in western Africa produced several natural nuclear fission reactors. Evidence of these prehistoric reactors became apparent during mining operations by the French at the Oklo site in Gabon, Africa during 1972 (Cowan, 1975). The Oklo system sustained criticality for perhaps 0.6×10^6 years. Analysis of the core samples showed unusual ratios of $^{235}U/^{238}U$ in which ^{235}U was depleted. These provided much information about the long-term migration of heavy elements such as plutonium.

The world's oceans contain more than 3.9 Pg (3.9 Gtonnes) of uranium equivalent to 740 EBq (20 GCi) (Oliver, 1980) (see Box 28-2 for an explanation of multiplication factors and prefixes). Average uranium concentrations in USA surface, domestic and ground water are 37, 74 and 111 Bq L^{-1} (1, 2 and 3 nCi L^{-1}), respectively. The average uranium concentration in the earth's crust is 4×10^{-4}% making it more abundant than mercury, antimony or silver. Average concentration of ^{238}U in USA soils is about 37 Bq g^{-1} (1 nCi g^{-1}), about the same concentration as that for radium. Commercial uranium ore bodies have concentrations in hundreds of Bq g^{-1}. The average ^{238}U concentration in a person is about 1.11 mBq (30 fCi).

The average concentration of Radium-226 (^{226}Ra, 1.3×10^3 years physical half-life), in equilibrium with its parent ^{238}U, in surface soil and rocks is about 37 Bq (1 nCi) per gram earth. At this concentration, the content per square mile of ordinary land to a depth of 30.5 cm, is 2 gram radium containing 74 GBq (2 Ci) and 5.45 Mg (5.45 tonnes) of uranium (Evans, 1981).

Data have been reported on the content of naturally occurring alpha-emitting radionuclides in vegetation and food plants (Drury et al., 1983). Some foods have very high radionuclide levels; Brazil nuts have 3–130 Bq kg^{-1} (0.08–3.5 nCi kg^{-1}) ^{226}Ra.

28.4 Plutonium

Plutonium was discovered by Seaborg and coworkers in 1941. It can be found in natural ore bodies in trace amounts. Only limited quantities were available until after World War II. Several countries built nuclear weapons from enriched uranium and plutonium produced in nuclear reactors. Enormous research and production facilities developed around the secret military applications of uranium and plutonium. Weap-

Box 28-2 Multiplication factors and prefixes

Multiplication Factor	Prefix	Symbol
10^{18}	exa	E
10^{15}	peta	P
10^{12}	tera	T
10^{9}	giga	G
10^{6}	mega	M
10^{3}	kilo	k
10^{-3}	milli	m
10^{-6}	micro	μ
10^{-9}	nano	n
10^{-12}	pico	p
10^{-15}	femto	f
10^{-18}	atto	a

Examples: 1 nBq is 10^{-9} Becquerel and 5 PBq is 5×10^{15} Becquerels

ons production and testing and the use of uranium in reactors resulted in redistribution of radioactive materials. Predominant among the high yield fission products released are radionuclides of cesium, strontium and iodine. Large areas of radioactive contamination from accidents and operational releases in several countries including the USA, USSR, China and UK are the legacy of the nuclear weapons programs. These include sites in the USA, UK, the former Soviet Union, France, China, Australia and the Pacific Atolls.

28.5 Radiocesium

Cesium-137 (^{137}Cs) is a major contributor to the radiation dose received by people as it has been released to the environment in large quantities from nuclear weapons tests, Chernobyl, fuel reprocessing, waste storage facilities and medical sources. Concern about ^{137}Cs and ^{90}Sr building up in people led to the limited test ban treaty on testing of nuclear weapons in the atmosphere in September 1963. These, and other radionuclides including plutonium, were present in people throughout the world from nuclear weapons testing.

Table 28-1 shows the contribution to radiation dose likely to have been received by people in the USA (mean dose equivalent commitment) to the year 2000 from nuclear testing from the year 1970. The majority of the dose comes from ^{90}Sr, ^{137}Cs, $^{239,240}Pu$ and ^{14}C. Most of the radiation dose has been delivered, except for ^{14}C. The

Table 28-1 Mean dose equivalent commitments to the year 2000 in the USA from nuclear testing since 1970

Source	Dose equivalent commitment	
	(μSv)	(mrem)
External (whole body)	750	75
Internal		
^{90}Sr (marrow)	450	45
^{90}Sr (endosteal)	650	65
^{137}Cs (whole body)	150	15
$^{239,240}Pu$ (lung)	400	40
$^{239,240}Pu$ (bone)	20	2
^{3}H (whole body)	20	2
^{14}C (whole body)	80	8
TOTAL	252	25.2

Adapted from NCRP 87 with permission

total dose commitment from ^{14}C to be delivered over many generations is 1.4 mSv (140 mrem) (NCRP, 1987). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) calculated the effective dose equivalent commitment for individuals in the northern hemisphere from weapons testing. The dose to the year 2000 was 2.1 mSv (210 mrem) (UNSCEAR, 1982).

One of the best understood cases of environmental contamination is fallout ^{137}Cs. It has been measured in many plants, animals and foods for many years and can be detected by *in vivo* measuring techniques. The levels in man from dietary intake can be predicted with some confidence. Peak body burdens of about 500 Bq (13.5 nCi) were reached in the Northern hemisphere in 1964 following the limited test treaty ban in 1963. Members of some population groups reached levels of 50 kBq (1.35 μCi).

A more difficult situation exists for many actinide radionuclides, particularly plutonium-239 (^{239}Pu). Estimates of plutonium concentrations in environmental media and selected human tissues are available from several countries. About 12 PBq (0.32 MCi) were globally distributed by atmospheric weapons tests. About 9.8 PBq (0.26 MCi) fell onto the northern hemisphere. By 1970–71, about 0.59 PBq (15.9 kCi) of ^{239}Pu had been deposited on the conterminous USA and about 0.11 PBq (2.97 kCi) on the Australian continent (about the same land area). On average, a resident of the northern hemisphere acquired a burden of 0.15 Bq (4.1 pCi) by 1964. For comparison, the average resident of the northern hemisphere contains about 20 Bq (0.54 nCi) of naturally occurring alpha activity. In the USA approximately 220 PBq (5.95 MCi) are found in the upper 5 cm (2 in) of soil.

On a much smaller scale, the use of radioactive materials in consumer products are a potential source of contamination. In some instances these radionuclides are disposed of in local landfills, for example smoke detectors that typically contain 37 kBq (1.0 μCi) of Americum-241 (^{241}Am). Polonium-210 (^{210}Po) found in cigarettes at levels around 10 mBq g^{-1} (0.27 pCi), has caused some concern because of the high incidence of lung cancer in many countries. Domestic water supplies also contain radionuclides.

28.6 Human Exploitation of Radionuclides

Early in the twentieth century scientists produced artificial radionuclides in accelerators and other machines for use in research, medicine and other applications. Subsequent discovery of nuclear fission in the late 1930s produced nuclear energy with its attendant radioactive fission products. Man then learned to mine uranium ore, separate ^{235}U, enrich it and fission the enriched material in nuclear reactors to produce electric power, propel ships and produce radionuclides for consumer products and for diagnostic and therapeutic medical applications.

Researchers throughout the world learned much about the working of the human body by using radionuclides as tracers for stable elements. The naturally occurring radionuclides (e.g. radium and thorium) were used first, then radionuclides produced in accelerators and other machines (e.g. iron, phosphorus). After nuclear fission was discovered in 1938, fission products were used (e.g. strontium, cesium, iodine, gadolinium, gallium, technetium and molybdenum).

28.7 Sources of Radioactive material

28.7.1 Natural High Radiation Areas

The levels of contemporary, naturally occurring uranium, thorium and radium in soil, water and plants vary depending on location. The best documented locations with soil concentrations much higher than average are in Brazil (Minas Gerais State), India (Kerala State) and the People's Republic of China (Guangdong Province). The Kerala coast in India has sands with high thorium concentrations. Studies of the high levels of thorium at Minas Gerais provide useful information on the transport of actinide elements. Plants that grow in this area absorb so much radioactivity that they can be autoradiographed simply by placing the leaves on X-ray film (Eisenbud, 1978).

Certain lichens concentrate radionuclides such as radium, polonium, uranium and plutonium and so present specific problems for population groups in far Northern lati-

Table 28-2 Release of uranium and thorium from coal combustion (tonnes)

Element	USA, 1982	World, 1982	World, 1937–2040
U	4.7	3 300	751 200
^{235}U	0.034	23	5 300
Th	11.6	8 100	1 800 000

tudes. Animals, such as reindeer, eat the lichens, incorporate the radionuclides into their tissues and, in turn, are consumed by people.

Much can be learned from these areas about the possible effects of low level radiation on human subjects. The radiation doses are elevated relative to naturally occurring levels, yet they are very low compared to doses received by some population groups that have been studied, such as the Japanese who survived the atom bomb attacks on Japan during World War II. Data from the bomb survivors are not relevant to most real world environmental exposures as their health effects are estimated from extrapolations of high dose and high dose-rate data. The data from naturally occurring 'high background' areas provide a valuable insight as to the equilibrium levels of radionuclides reached in human subjects. This information is important and relevant to many current situations that involve exposure of populations to low dose and low dose-rate radiation. People in the Guangdong Province reach equilibrium levels of thorium in their bodies that are about three times that in the soil.

28.7.2 Coal Combustion

Combustion of fossil fuels releases large quantities of uranium and thorium. According to the USA Environmental Protection Agency (EPA), coal averages about 1.3×10^{-4}% uranium and about 3.2×10^{-4}% thorium. The uranium content ranges from less than 10^{-4}% to 10^{-3}%. The estimated annual release of ^{238}U from a 1 GWe coal-fired power plant in the USA, assuming all the uranium is in the ash and 1% of ash is released, is 296 MBq (8 mCi). This is about 4.7 Mg (4.7 tonnes) of uranium containing 34 kg (75 pounds) of ^{235}U. The annual input to the plant in the coal would be 2.32 Mg (2.32 tonnes) of uranium in 3.68 Tg (3.68 Mtonnes) of coal. The same power station would release 11.6 Mg (11.6 tonnes) of thorium each year. Other radionuclides including radium, radon, polonium, bismuth, lead and potassium are also released from coal combustion (Gabbard, 1993).

Table 28-2 shows the estimated releases of uranium, thorium and ^{235}U from a typical 1 GWe coal-fired, electricity generating station in the USA, and the total for the world, for 1982. Also shown is the estimated release of these materials from worldwide coal combustion for the period 1937–2040. Combustion redistributes huge

quantities of uranium, thorium and other radionuclides and elements from the pre-combustion concentration in coal.

28.7.3 Nuclear Fuel Cycle

Releases of radioactive materials from the nuclear fuel cycle also occur. These are small under normal operating conditions but can be significant in the event of large accidents. For normal operations the radiation dose to the public is less than that from coal combustion. One of the major problems associated with various stages of the nuclear fuel cycle, i.e. waste disposal, involves technical and social acceptance problems. Decontamination and decommissioning of nuclear power plants at the end of their operational lives also contributes to occupational exposure (see Chapter 50).

28.8 Accidental releases

28.8.1 Nuclear Weapons Accidents

Accidents involving nuclear weapons have produced environmental contamination in Palomares, Spain and Thule, Greenland. Other releases, notably ^{238}Pu from power sources and uranium with its fission products from several reactors, came from accidents in the USA and USSR space programs. In January 1966, a USA B-52 bomber carrying four nuclear bombs and a USA KC-135 tanker collided and were destroyed in the air during a refueling operation over Spain's southeastern coast. One bomb was recovered intact from the Mediterranean Sea, about 8 km offshore, 80 days after the accident. Another was recovered intact from the dry Almanzora river bed just east of Palomares. The primary parachutes did not open for the other two weapons. They underwent high explosive detonation upon impact. One weapon landed approximately 1.7 km west of the village and the second landed at the eastern edge of Palomares. The population at the time was estimated to be about 1500.

The impact, high explosive detonation and burning of weapon number two produced a dust cloud bearing plutonium which was blown by a 30-knot westerly wind across cultivated fields and the northern edge of the village. The cloud traveled down a small valley in a northeasterly direction. The end of the valley nearest the impact point was used for dry farming but, because of inadequate rainfall, had not been cultivated for many years. Part of the valley nearest the village was irrigated and under cultivation at the time of the accident. A plutonium-bearing cloud produced by the weapon that fell in the eastern edge of the village traveled away from Palomares across the irrigated fields used for growing tomatoes, beans and alfalfa. At the time of the accident the last tomato crop of the season was ready for harvest (Iranzo et al., 1987).

Following the accident, a survey of the surface distribution of plutonium was conducted to determine the extent of ground contamination. The isopleths describing the surface plutonium contamination within weeks of the accident were used as the basis for the clean-up operation. About 226 ha (558 acres) were contaminated with plutonium. Some vegetation and soil from the most heavily contaminated areas was physically removed, placed in 4 810 metal 208-liter (55 USA gallon) drums and transported by ship for burial in the USA. Recent resurveys of some areas still show residual contamination of soil and some plants. Only rarely is plutonium found in the edible parts of plants.

Medical studies of Palomares residents continue in an attempt to estimate the radiation dose to those exposed to plutonium. None of 800 people measured by in vivo chest counting have had plutonium burdens above the minimum detectable activity level for the counting system i.e. 814 Bq (22 nCi). Some residents have internally deposited plutonium as measured by urinary excretion. The 50-year cumulative effective dose equivalent calculated from urinary excretion for 45 people who were present at the time of the accident ranged from 0.02–0.2 Sv (2–20 rem). The mean was 79.4 mSv (7940 mrem).

The crash of a B-52 bomber on the ice of North Star Bay about 12 km from Thule, Greenland, occurred in January 1968. A fire made it necessary for the crew to bail out and the plane hit the ice with a velocity of about 500 knots at a 15 degree attitude. The fuel ignited on impact and the four plutonium-bearing weapons exploded, spreading debris and flaming fuel along a path about 700 meters long. A blackened area about 130 m wide along this path was formed by combustion products trapped in refrozen ice and snow. All but about 1% of the plutonium within the defined contaminated zone was contained in the black crusted ice and snow of this area. Road graders windowed the black material and mechanized loaders loaded it in large wooden boxes for removal from the area. Eventually, 67 94.5-kL (25,000 USA gallons) fuel containers were filled with this material and shipped to the USA for final disposal (Jordan, 1971).

Danish scientists thoroughly investigated the levels of plutonium in the environment and concluded from their findings that the environmental impact was negligible. No significant exposure to people or damage to the environment resulted from the accident.

28.8.2 Chernobyl

The nuclear power station accident at Chernobyl in the Soviet Union on April 26, 1986 created an immediate demand for information. World governments were extremely concerned for the health and safety of citizens because of atmospheric fallout and ingestion of plant and animal products containing radionuclides from Chernobyl. Earlier but much less severe accidents occurred at Windscale in the United Kingdom (1957) and Three Mile Island in the USA (1979). About 0.74 PBq (20 kCi) Iodine-131 (^{131}I), the major contaminant, were released from Windscale. Smaller quantities of ra-

diocesium and radiostrontium were released. Contaminated cow's milk was destroyed to prevent excessive radiation dose to the thyroid gland. Noble gases were released in small quantities from Three Mile Island. Radiation doses to small numbers of people were trivial.

The Chernobyl accident raised many economic considerations and, in retrospect, some decisions such as large scale destruction of food and animals were later viewed as being unsound. Previously documented information was used to advantage in some cases but ignored in many others. It came as a surprise to many that cesium levels in goat and sheep milk were higher than those in cow milk in a given location. The high level of contamination in reindeer also surprised some. These situations were predictable because of information collected decades ago from studies on the uptake, distribution, retention and loss of radionuclides from nuclear weapons fallout (Ng and Hoffman, 1984). Neither should the high levels of contamination in mushrooms or in animals, including man, have been surprising (Greuter, 1971). Following the Chernobyl accident, people were contaminated from eating mushrooms that preferentially concentrated ^{134}Cs and ^{137}Cs. Actually, these data were reported in studies of nuclear weapons fallout long before the Chernobyl accident.

Large uncertainties are found when attempting to estimate the amount of radiocesium from Chernobyl that would find its way to people (Richmond, et al., 1988). Many parameters are required for the various models used to move the material from source through the environment, including animals that are eaten as food, to man. The results indicate that the amount actually detected in people was between one and two orders of magnitude higher than that predicted by the models. Much is known about cesium because of its ubiquitous presence in nuclear fallout and its ease of measurement in man. This is not the case for other radionuclides.

Nuclear power station accidents, particularly Chernobyl, have released large quantities of radioactive contaminants to large areas of Europe. Consequently, public health officials are interested in the allowable levels in food and water and the dose per unit intake for radionuclides following contamination from accidents.

The Chernobyl accident was one of several factors that dictated the need for information on radiation dose per unit intake and as a function of age. Secondary radiation limits for intake of radionuclides were developed earlier for radiation workers but were not available for members of the general public. This problem was resolved with the publication of two reports on age dependent doses to the public from intake of radionuclides (ICRP, 1989,1993). These reports provide dose coefficients (equivalent dose in Sv per Bq) for five age groups and adults following acute intake. Dose is calculated for 70 years following intake for all age groups except adults. For adults, a 50 year calculation is used. National authorities now have the tools necessary to calculate the dose in Sieverts following a known or estimated intake of radioactivity measured in Becquerels.

28.8.3 Radionuclide Thermoelectric Generators

Both the USA and the former Soviet Union used radionuclide thermoelectric generators (RTG) as power sources for space systems. Shortly after launch in April 1964, a USA satellite, Snap 9A, reentered the atmosphere and burned up at high altitude over the Mozambique channel. 0.63 PBq (17 kCi) of ^{238}Pu reentered the southern hemisphere and was subsequently measured for many years in both hemispheres where it greatly perturbed the global ^{238}Pu/^{239}Pu ratios resulting from nuclear weapons testing. In addition, 481 GBq (13 Ci) of ^{239}Pu was released from this source (Richmond, 1989).

Other accidents have occurred with no significant releases of radioactivity. However, the April 1970 USA mission to the moon (Apollo13) experienced a failure mode that caused the radionuclide thermoelectric generator (RTG) to return to Earth with the crew in the lunar expedition module. The 1.65 PBq (44.59 kCi) ^{238}Pu RTG attached to the lunar landing vehicle was deliberately disposed of in the Pacific Ocean near the Tonga Trench. The RTG also contained 1.22 TBq (33 Ci) ^{239}Pu (Dobry, 1980).

Several large RTGs were launched by the USA National Aeronautical and Space Agency. These missions, Ulysses and Galileo, used three RTGs containing almost 14.8 PBq (400 kCi) ^{238}Pu; two RTGs containing a total of 9.8 PBq (265 kCi) aboard Galileo and a single RTG on Ulysses. Voyagers I and II launched in 1990 also carried ^{238}Pu fueled RTGs. Several RTGs containing ^{238}Pu were left on the moon from the Apollo series.

28.8.4 Space Reactors

In April 1988 the USSR satellite Kosmos 1900 entered a decaying orbit that, without orbital corrections, would cause it to enter Earth's atmosphere. Kosmos 1900 was a Radar Ocean Reconnaissance Satellite (RORSAT) whose orbit covered the Earth between 65 degrees south and north latitude. The Soviet Union launched Kosmos 1900 on 12 December, 1987 and lost radio contact with the craft in April. It fell from orbit, together with two other satellites in the Kosmos RORSAT series. Kosmos 954, powered by 50 kg (110 pounds) of uranium, reentered the atmosphere in January 1978 and spread radioactive contamination across parts of Canada. Kosmos 1402 dropped from orbit over the Indian Ocean on 23 January, 1983. The detection of uranium isotopes in the USA confirmed the destruction of Kosmos 1402 (Tracy et al., 1984).

Current predictions are that the amount of radioactive materials used in space applications will continue to increase. By 1988, eight countries (Britain, China, France, India, Israel, Japan, Russia and the USA) had the capability to launch rockets powerful enough to put satellites in space. Most, if not all, these countries have the capability to use radionuclides as power sources in these satellites.

28.9 Cold War Leftovers

Huge quantities of highly enriched (in ^{235}U) uranium remain from the cold war nuclear weapons production. Because of its long physical half-life, 703.8 $\times 10^6$ years, ^{235}U will be present in concentrated form for many years. In June 1994 the USA Department of Energy (DOE) released revised information on ^{235}U production for nuclear weapons. Of the 0.994 Gg (994 tonnes) of weapons grade uranium produced, 0.259 Gg (259 tonnes) remained in the stockpile, mostly in Oak Ridge, Tennessee. DOE estimates the Russian stockpile at 0.5 Gg (500 tonnes). The International Atomic Energy Agency estimates that a crude nuclear device can be made from 2 kg of highly enriched uranium.

The DOE also disclosed that the USA had 22 229 warheads in 1961 and, at the peak in 1960, the USA arsenal contained nuclear weapons with the equivalent of an estimated 20.49 Gt of TNT. This has subsequently decreased by about 90% (Lippman and Smith, 1994). In November 1994, the USA Department of Energy successfully moved about 0.5 Mg (0.5 tonnes) of highly enriched weapons grade uranium from Kazahkstan to the USA for storage in Oak Ridge. A continuing global problem will be the safe storage and disposition of hundreds of tons of plutonium and highly enriched uranium that pose potential threats to human beings and their environment.

28.10 Environmental Clean-Up and Restoration

Clean-up of contaminated areas and environmental restoration are now major activities in several countries. In addition to the high yield fission products, radioactive actinides are of concern because of their larger biological effects. Major uncertainties exist as to where contaminated material should be disposed of or stored. Ocean disposal is not an option in many cases. Environmental clean-up options include in-situ modification and means of protecting disposal areas from man and weathering. Burning of some radionuclides in atomic reactors and accelerators is also possible as a means of recycling. This may be of particular importance in reducing the nuclear weapon stockpiles of uranium and plutonium.

Large quantities of fission products were released into the Techa River and from the Kyshtym nuclear complex in the Cheliabinsk Province of the former Soviet Union. Disposal into the ground may have been a common practice. Recent reports suggest that as many as 111 EBq (3 GCi) of radioactive materials were pumped directly into the ground at several sites.

Huge costs are projected to clean-up contamination from nuclear facilities in several countries. Risk ranking of individual sites must be accomplished to allow prioritization of resources as the contamination is so extensive. Among unanswered questions are "how clean is clean?" and "where to put the contaminated material?"

Legislation against ocean dumping complicates the problems and reduces the remediation options. Some proponents of ocean deposition of nuclear wastes have argued for the need of a Cousteau for the land.

28.11 Radiation Dose to People

Until recently, exposure to radon in homes was not considered as a potential health problem. Many countries now evaluate the health effects of radon in homes and attribute a large fraction of the naturally occurring radiation dose to man from exposure to radon decay products. Maps of natural radiation in western Europe show the wide variation in ambient levels and therefore the radiation dose to people from radon (NRPB, 1993).

The natural radiation dose to man results from three sources; air, ground and within the body. Air, food and water contribute to the amount in the body. For many years the average annual effective dose equivalent (EDE) to individuals in the USA was taken to be about 1 mSv (100 mrem) per year, about one-third resulting from internal sources. Polonium-210 (^{210}Po) and ^{40}K dominate the total dose equivalent from radionuclides in the body. The ^{210}Po in the body is a function of intake and therefore very variable. The average annual dose equivalent is about 0.14 mSv (14 mrem) to soft tissue and about 0.70 mSv (70 mrem) to bone.

As worry about the contribution from radon decay products increased, revisions were made to the recommended EDE values for "natural background" radiation. The total EDE for an individual was increased by the USA National Council on Radiation Protection and Measurements (NCRP) (NCRP, 1987) to about 3 mSv (300 mrem) per year, mainly from the addition of 2 mSv (200 mrem) from radon daughters irradiating specific tissues within the respiratory tract with alpha particles. If the 3 mSv (300 mrem) is applied to each member of the USA population, the collective effective dose equivalent from natural sources becomes 78×10^4 person Sieverts (78 million person rem).

All three isotopes of the noble gas radon (^{219}Rn, ^{220}Rn and ^{222}Rn) are radioactive and occur in nature as members of the primordial actinium, thorium and uranium series, respectively. Radon-222 and its short-lived daughter products, ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po, are intermediate decay products of primordial ^{238}U. These daughters irradiate the bronchial epithelium of the respiratory tract (NCRP, 1988).

Radon was the first known association of a radioactive material with human cancer. Miners removed silver from pitchblende ore in the Joachimstahl and Schneeberg area of Europe (Erzgebirge mountains) early in the fifteenth century. Prevalent lung disease in the miners was written about by the early physicians Agricola and Paracelsus in the 1500s. In Schneeberg, much of the disease was identified as lung cancer by German investigators Härtung and Hesse in 1879. Physicists were also very interested in radium and thorium emanations. Ernest Rutherford made the following statement by in 1907:

> *"We must bear in mind that all of us are continuously inhaling the radium and thorium emanations and their products and ionized air...Some have considered that possibly the presence of radioactive matter and ionized air in the atmosphere may play some part in physiological processes..."* (Rutherford, 1907).

Much controversy exists concerning the effects to health from radon in homes and the levels that should be allowed in dwellings and buildings. Surveys have been made of radon levels in homes in many countries. Much uncertainty and misinformation exists as to the possible health effects associated with specific exposure levels. National regulatory agencies have established allowable levels that vary considerable from country to country. The costs associated with determining the exposure level and remediation can be high. Much of the health effects data is from uranium miners and is, according to some, of limited value for calculating radiation dose to people from radon in homes.

Radon concentrations in nature are very variable. In some British caves, concentrations range from 454–8 868 Bq m^{-3} (0.012 μCi–0.240 μCi m^{-3}). In one system levels reached 155 kBq m^{-3} (4.2 μCi m^{-3}). Results indicated that the potential dose from one 4-hour trip could exceed the average national annual background for Rn in the United Kingdom of 1.25 mSv (125 mrem) (Hyland and Gunn, 1994).

28.12 References

Cowan, G.A. (1975) A natural fission reactor. *Scientific American*, 235: 36–47.

Dobry, T.J. (1980) Transuranic elements in space nuclear power systems. In: *Transuranic Elements in the Environment*. Hanson, W.C. (Ed.). DOE/TIC–22800. United States Department of Energy Technical Information Center, USA. pp. 83–85.

Drury, J.S., Baldauf, M.F., Daniel, E.W., Fore, C.S. and Uziel, M.S. (1983) *Radioactivity in Food Crops*. Report ORNL–5963. Oak Ridge National Laboratory, Oak Ridge, TN, USA.

Eisenbud, M. (1978) *Environmental Technology and Health*. New York: New York University Press.

Evans, R.D. (1981) *Uranium Ore Residues: Potential Hazards and Deposition. Hearings before the Procurement and Military Nuclear Systems Committee of the House Committee on Armed Services*. United States Government Printing Office, Washington DC, USA.

Gabbard, A. (1993) Coal combustion: nuclear resource or danger. *Oak Ridge National Laboratory REVIEW*, 26: 24–33.

Grueter, H. (1971) Radioactive fission product ^{137}Cs in mushrooms in West Germany during 1963–1970. *Health Physics*, 20: 655–656.

ICRP (1989) Age-dependent doses to members of the public from intake of radionuclides. Part 1. (Publication 56). *Annals of the International Commission on Radiological Protection*, 20(2): viii + 122 pps.

ICRP (1993) Age-dependent doses to members of the public from intake of radionuclides. Part 2: ingestion dose coefficients. (Publication 67). *Annals of the International Commission on Radiological Protection*, 23(3/4):viii + 167.

Iranzo, E., Salvador, S. and Iranzo, C.E. (1987) Air concentrations of ^{239}Pu and ^{240}Pu and potential radiation doses to persons living near Pu-contaminated areas in Palomares, Spain. *Health Physics*, 52: 453–461.

Jordan, H.S. (1971) *Distribution of Plutonium fromAaccidents and Field Experiments*. Report LA–4756. Los Alamos Scientific Laboratory, Los Alamos, New Mexico, USA.

Morris, R. (1981) *Times Arrow's*. New York: Simon and Schuster.

NCRP (1987) *Ionization Radiation Exposure of the Population of the United States*. NCRP Report No. 93. National Council on Radiation Protection and Measurements, Bethesda, MD, USA.

NCRP (1988) *Measurement of Radon and Radon Daughters in Air*. Report No. 97. National Council on Radiation Protection and Measurements, Bethesda, MD, USA.

NRPB (1986) *Radon*. At a Glance Series. National Radiological Protection Board, Harwell, UK.

NRPB (1993) *Maps of Western Europe*. At a Glance Series. National Radiological Protection Board, Harwell, UK.

Ng, Y.C. and Hoffman, F.O. (1984) Selection of terrestrial transfer factors for radiological assessment models and regulatory guides. In: *Proceedings of a Seminar on the Environmental Transfer to Man of Radionuclides Released from Nuclear Installations (Volume II)*.1984. Publication V/7400/84–EN. Commission of the European Communities, Luxembourg. pp. 841–855.

Oliver, B.M. (1980) Some energy facts and fallacies. *Perspectives in Biology and Medicine*, 23: 335

Richmond, C.R. (1989) Transfer of radionuclides to animals – an historical perspective of work done in the United States. *Science of the Total Environment*, 85: 1–15.

Richmond, C.R., Hoffman, F.O., Blaylock, B.G., Eckerman, K.F., Lesslie, P.A., Miller, C.W., Ng, Y.C. and Till, J.E. (1988) *The Potential Use of Chernobyl Fallout Data to Test and Evaluate the Predictions of Environmental Radiological Assessment Models*. Report ORNL/TM–6466. Oak Ridge National Laboratory, Oak Ridge, TN, USA.

Rutherford, E. (1905) Some cosmical aspects of radioactivity. *Harper's Magazine*, 390–396.

Rutherford, E. (1907) Radium – the cause of the Earth's heat. *Journal of the Astronomical Society of Canada*, 145–165.

Tracy, B.L., Prantl, F.A. and Quinn, J.M. (1984) Health impact of radioactive debris from the satellite Cosmos 954. *Health Physics*, 47: 225–233.

UNSCEAR (1982) *Ionizing Radiation: Sources and Biological Effects. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation.* UN Publication No. E.82.–IX.8.–06300P. United Nations, New York, USA.