

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

### 6.1 OVERVIEW

All organisms (e.g., bacteria, plants, or animals, including humans) are exposed everyday to varying amounts of ionizing radiation. Figure 6-1 shows average contributions from various sources of radiation to which the average U.S. citizen is exposed during his or her lifetime. Approximately 82% of the radiation dose is from natural sources: 55% from radon (see Figure 1-3), 8% from cosmic radiation (from the sun and stars), another 8% from terrestrial sources (radioactive material in rocks and soil), and 11% from internal sources (radioactive materials, primarily potassium-40, from food and water consumed in the daily diet).

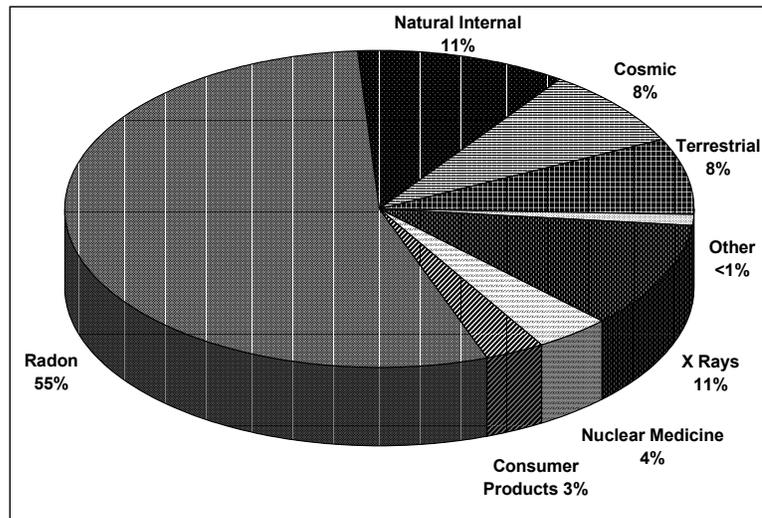


Figure 6-1. Radiation Exposure to the Average U.S. Citizen adapted from (NCRP1987a)

The remaining 18% of the dose comes from anthropogenic (man-made) sources such as medical x ray exposure (11%), nuclear medicine procedures exposure (4%), consumer products (3%), and other sources (<1%). These other sources include occupational exposure, nuclear fallout, and the nuclear fuel cycle. The total average annual effective dose equivalent for the population of the United States, natural and anthropogenic, is approximately 360 mrem (3.6 mSv) and is described further in Chapter 1 of this profile (BEIR V 1990).

The majority of exposure to radiation comes from natural sources. With the exception of indoor radon exposure (and to some extent exposure from terrestrial sources), exposure to natural radiation is only moderately controllable. Controllability in relation to radon refers to mitigation of radon concentrations in buildings and homes. The average annual effective dose equivalent from all natural sources combined is approximately 3 mSv (300 mrem). Of this amount, approximately 98 mrem (98 mSv) is due to

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background radiation; this includes cosmic rays, 29 mrem (0.29 mSv); terrestrial gamma rays, 29 mrem (0.29 mSv); and naturally existing radionuclides within the body, 40 mrem (0.40 mSv). Individual doses from natural sources may be much greater. The magnitude of natural exposures depends upon numerous factors such as geographic location, height above sea level, and the construction and ventilation of buildings. For instance, the average annual radiation dose received by a person living in Boston, Massachusetts, is approximately 300 mrem (3 mSv), while people living in Denver, Colorado, and Kerala, India, receive average annual doses of approximately 600 mrem and 1500 mrem, respectively. The difference in these doses is due mainly to greater concentrations of radioactive materials found in the soils of the Colorado and Kerala areas and to a smaller extent the increase in cosmic radiation at higher altitudes in these areas (BEIR V 1990; Eisenbud 1987; Harvard Medical School 1996; UNSCEAR 1993).

Commonly used terms and scientific unit symbols and abbreviations used in this chapter are defined in Table 6-1 and Table 6-2, respectively. These and other terms may also be found in the glossary in Chapter 9 or in the index at the end of this toxicologic profile. The following sections discuss natural external exposures (Sections 6.2 and 6.3), natural internal exposures (Section 6.4), and internal and external man-made/industrial exposures (Sections 6.5, 6.6, and 6.7).

## 6.2 COSMIC RADIATION EXPOSURE

Cosmic radiation contributes an estimated 8% to the average population radiation dose. It is primarily composed of galactic radiation originating outside the solar system in addition to a varying degree of solar radiation. The primary cosmic rays that arrive in the upper atmosphere are high-energy subatomic particles—primarily protons, but also nuclei and electrons—moving almost at the speed of light; these primary rays create secondary rays that bathe the atmosphere in radiation. Austrian physicist Victor Hess discovered cosmic rays in 1912 when he and two assistants flew a balloon to an altitude of 16,000 ft (4,877 meters). Hess proved that the source of a mysterious radiation previously detected over the ocean, where terrestrial radiation levels were expected to be very low, was actually coming from outside the atmosphere; he also found that the rate of decline in radiation as the balloon ascended over land was slower than would be expected if the radiation emanated from the earth. The difference is caused by the cosmic rays. Only a small fraction of cosmic radiation originates from the sun; however, the proportion of cosmic radiation contributed by the sun increases during periods of increased sunspot and solar flare activity, which run in

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**Table 6-1. Common Terms and Abbreviations**

becquerel (Bq)	SI unit for quantity of radioactive material; 1 Bq equals that quantity of radioactive material in which there is 1 transformation or disintegration per second (dps).
curie (Ci)	Conventional unit for quantity of radioactive material. One Ci is the quantity of any radionuclide in which there are 37 billion transformations or disintegrations in 1 second. This is the activity of 1 gram of <sup>226</sup> Ra.
rad	The unit of absorbed dose equal to 0.01 Joule/kg in any medium.
gray (Gy)	SI unit of absorbed dose.
rem	Conventional unit for dose equivalent. The dose equivalent in rem is numerically equal to the absorbed dose in rad multiplied by the quality factor.
roentgen (R)	A unit of x ray and gamma ray exposure. It is measured by the amount of ionization in air produced by x ray and gamma radiation. One R equals 2.58x10 <sup>-4</sup> coulomb per kg of air.
sievert (Sv)	The SI unit of dose equivalent. It is equal to the dose in grays times a quality factor; 1 Sv equals 100 rem.
quality factor (Q)	The linear-energy-transfer-dependent factor by which absorbed doses are multiplied to obtain (for radiation protection purposes) a quantity that expresses the effectiveness of the absorbed dose on a common scale for all ionizing radiation.

**Table 6-2. Scientific Units**

Prefix (symbol)	Power of 10	Decimal Equivalent
atto	10 <sup>-18</sup>	0.000000000000000001
femto (f)	10 <sup>-15</sup>	0.000000000000001
pico (p)	10 <sup>-12</sup>	0.000000000001
nano (n)	10 <sup>-9</sup>	0.00000001
micro (μ)	10 <sup>-6</sup>	0.000001
milli (m)	10 <sup>-3</sup>	0.001
centi (c)	10 <sup>-2</sup>	0.01
deci (d)	10 <sup>-1</sup>	0.1
kilo (k)	10 <sup>3</sup>	1,000
mega (M)	10 <sup>6</sup>	1,000,000
giga (G)	10 <sup>9</sup>	1,000,000,000
tera (T)	10 <sup>12</sup>	1,000,000,000,000
peta (P)	10 <sup>15</sup>	1,000,000,000,000,000
exa (E)	10 <sup>18</sup>	1,000,000,000,000,000,000

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11-year cycles. Cosmic rays bombard the periphery of the earth's atmosphere at a rate of  $2 \times 10^{18}$  particles per second, at a density of about 4 rays/cm<sup>2</sup>-sec, and at an energy flux of 2,000 MeV/cm<sup>2</sup>•sec. These rays, referred to as "primary cosmic rays," are deflected and slowed by particles in the earth's atmosphere, creating "secondary cosmic rays" that often reach and even penetrate the earth's surface. The interaction of cosmic rays with the atmosphere leads to the production of several cosmogenic radionuclides, notably carbon-14 (<sup>14</sup>C), tritium (<sup>3</sup>H) and beryllium-7 (<sup>7</sup>Be). Because of the shielding effect of the atmosphere and the earth's geomagnetic fields, which tend to deflect charged cosmic ray particles towards the magnetic poles, the cosmic ray dose rate increases with altitude and latitude. The average annual dose from cosmic radiation in the United States is 29 mrem (0.29 mSv), but this value doubles for every 6,000-foot (1,828 meters) increase in altitude. Thus, the dose from cosmic rays received in Denver, Colorado, and Leadville, Colorado (altitudes of 1,600 m and 3,200 m, respectively), is approximately two and four times that received at sea level, respectively (Eisenbud 1987; Korff 1964; NASA 1995; Shapiro 1990; UNSCEAR 1993). At altitudes of 30,000 to 40,000 feet (9144 to 12192 meters), where most jet aircraft fly, the cosmic ray dose rate is about 1 mrem per hour (0.01 mSv/hr).

### 6.3 TERRESTRIAL RADIATION EXPOSURE

Cosmic radiation contributes approximately the same amount of background radiation as terrestrial radiation (8%), which is emitted by naturally occurring radioactive materials found in the earth's crust, such as <sup>40</sup>K, uranium and its progeny, and thorium and its progeny (see Figure 6-1). Uranium, for example, is found in all types of soil and rock at concentrations ranging from 0.003 ppm in meteorites to 120 ppm in phosphate rock from Florida. Exposure to radioactive materials in the soil and earthen products occurs continuously since we are surrounded by these sources. The radiation dose varies tremendously and is affected by such factors as geographic location, concentration of natural radioactive materials in the soil and building materials, and the types of materials used in building structures.

Some communities situated on soil with high concentrations of granite or mineral sand receive doses many times the average. Examples include coastal areas in Espiritos Santos and Rio de Janeiro in Brazil; Kerala, on the southwest coast of India; and the Guangdong province in China. In Brazil, the black sand beaches are composed of monazite, a rare earth mineral containing 9% radioactive thorium. External radiation dose rates from these sands may be as high as 5 mrem/hr (0.05 mSv/hr); permanent residents experience an average annual dose equivalent of approximately 500 mrem (5 mSv). In Kerala, on the

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west coast of India, residents receive 1,300–1,500 mrem (13–15 mSv) annually, due to the presence of monazite sand. Some dose rates are as high as 3,000 mrem/yr (Eisenbud and Gesell 1997).

Apart from radiation exposures due to living in close proximity to the earth's crust, people are also exposed to additional radiation when earth crust products (oil, coal, coal ash, minerals) are extracted, refined, and used. The naturally occurring radioactive materials in these products are concentrated into what is called technologically enhanced naturally-occurring radioactive materials (TENORM). In general, the hazards of exposure to TENORM during the extraction and processing of earth materials are relatively small compared to the hazards of exposure to other chemicals. As a result, radiation exposure from these sources, with the exception of uranium mining, milling, and processing, is not routinely monitored (Eisenbud 1987; UNSCEAR 1993). The radiation hazards associated with the mining of coal, oil, natural gas, phosphate rock products, and sand are discussed below.

Radon exposure makes up the largest fraction of total radiation dose and contributes to both internal and external radiation exposures. The following subsections discuss various radioactive materials primarily associated with external radiation exposures from various terrestrial activities. Radon is inherent in these terrestrial sources, as well, and is discussed because of its association, but with the understanding that it is a major internal radiation exposure source.

### 6.3.1 Coal Production

Exposure to radionuclides occurs during the mining and use of coal and coal ash. The methods of coal usage vary considerably among countries; on average worldwide, about 40% of coal is burned in electric power stations, 10% in dwellings, and 50% in other industries. Based on samples from 15 countries, the average concentrations of  $^{40}\text{K}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$  in coal are 50, 20, and 20 Bq/kg (1.35, 0.54, and 0.54 nCi/kg), respectively. These concentrations may vary considerably, depending upon the mine location. For example, concentrations of these radionuclides in China are 104, 36, and 30 Bq/kg (2.81, 0.97, and 0.81 nCi/kg), respectively. Coal mine exhaust typically contains radon; the estimated annual per person dose from radon in coal mine dust is 0.1–2 nSv. The average annual per person doses of radiation from coal-fired power plants and from domestic cooking with coal are about 0.2 mrem (2  $\mu\text{Sv}$ ) and 0.04–0.8 mrem (0.4–8  $\mu\text{Sv}$ ), respectively.

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About 280 million tons of coal ash are produced by power plants each year. Potential uses for the ash include fertilizers and building materials for roads and dwellings. Most U.S. power plants recover fly ash exhaust using scrubbers, electrostatic precipitators, or bag houses. The radioactive content of coal tends to concentrate in the ash, resulting in 5- to 10-fold increases in the concentration of lead-210 ( $^{210}\text{Pb}$ ) and polonium-210 ( $^{210}\text{Po}$ ) as compared to unburned coal. When fly ash is used in building materials, the degree of external exposure to radiation and the inhalation of radon gas increases directly with the amount of ash incorporated into these materials, and, for radon, the porosity of the materials. The average annual exposure associated with living in concrete and wooden houses is 7 mrem (70  $\mu\text{Sv}$ ) and 3 mrem (30  $\mu\text{Sv}$ ), respectively. An EPA report published in 1979 estimated that the exposure to radioactive materials emitted from all 250 coal-fired power plants resulted in an additional 1.5 cancers per year (Eisenbud 1987; EPA 1979 as cited by Eisenbud; UNSCEAR 1982, 1988).

### 6.3.2 Crude Oil and Natural Gas Production

About  $3 \times 10^{12}$  kg of crude oil and  $10^{12}$  m<sup>3</sup> of natural gas are produced worldwide annually. Oil-fired power plants use about 15% of all oil. Gas-fired power plants are estimated to use about 15% of all gas. Radon is present in natural gas; concentrations of radon in gas at well heads average approximately 40 pCi/L (1.5 Bq/L). The processing and blending of liquefied petroleum gas (LPG) tends to enhance radon concentrations, and the long-lived radon daughters ( $^{210}\text{Pb}$  and  $^{210}\text{Po}$ ) tend to accumulate on LPG processing machinery, resulting in low level exposure to maintenance workers. The annual per person doses from crude oil and gas are estimated to be 0.001 mrem (10 nSv) and 0.0001 mrem (1 nSv), respectively. The estimated doses are small and result from inhalation of radioactive particles and radon gas (Eisenbud 1987; UNSCEAR 1993).

### 6.3.3 Phosphate Rock Products

Phosphate rock, the precursor of all phosphorous products including fertilizer, is mined at a rate of 130 million tons per year worldwide. The worldwide use of fertilizer, estimated to be 30 million tons, constitutes the greatest source of  $^{40}\text{K}$  and  $^{226}\text{Ra}$  mobility. In the United States, the application rate for fertilizers ranges from 30 kg of phosphate per hectare (barley) to 150 kg/hectare (potatoes and tobacco) for commercial agricultural application, and possibly less for residential applications. Concentrations of  $^{40}\text{K}$  and  $^{232}\text{Th}$  in phosphate rock are similar to those in soil (a few grams per hundred grams of soil and a few grams per million grams of soil, respectively). Levels of  $^{238}\text{U}$  and its transformation products are

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much higher in phosphate rock than in soil. Concentrations of  $^{238}\text{U}$  in phosphate deposits are typically about 1,500 Bq/kg (40.5 nCi/kg). The practice of using phosphate fertilizers has resulted in uranium concentrations in food at levels up to 8 ng/g (0.005 pCi/g). Exposure to the general public occurs near areas of mining and processing through waste effluent. Several end-products of phosphate processing, phosphogypsum and calcium silicate, are used for fertilizer, for back-fill and road-base material, in additives to concrete, in mine reclamation, and in the recovery of sulphur. Phosphogypsum is also used as a substitute for natural gypsum in the manufacture of cement, wallboard and plaster. The primary radioactive material in phosphogypsum is  $^{226}\text{Ra}$ , which is found at concentrations of 900 Bq/kg (24.3 nCi/kg).

Exposure to phosphate-borne radioactivity also occurs as a result of discharges into surface waters. The primary pathway of exposure to radioactivity in humans is through the consumption of fish and shellfish. Elevated concentrations of radon have been detected in structures built over reclaimed phosphate mines, and over unmined mineral deposits. Maximum annual individual doses near phosphate facilities range from 4 mrem (40  $\mu\text{Sv}$ ) in the Netherlands to 600 mrem (6 mSv) in the United States. The average annual per person dose of  $^{40}\text{K}$  derived from fertilizers is approximately 0.2 mrem (2  $\mu\text{Sv}$ ); while the average per person dose from potassium in the body is about 20 mrem (0.2 mSv) per year, maximum annual individual doses, from consumption of seafood, have been estimated at 15 mrem (150  $\mu\text{Sv}$ ), with  $^{210}\text{Po}$  as the main contributor. The annual per person radiation dose from  $^{226}\text{Ra}$ -laden phosphogypsum in building materials is estimated to be about 1 mrem (0.01 mSv) (Eisenbud 1987; Shapiro 1990; UNSCEAR 1982, 1988, 1993). The use and disposal of phosphogypsum is regulated by the EPA, and these regulations are intended to ensure that the public is not exposed to unsafe levels of radionuclides from this material.

#### 6.3.4 Sand

Mineral sands, defined as sands with a specific gravity of more than 2.9, originate from eroded rock. These sands are mined in Australia, Bangladesh, Indonesia, Malaysia, Thailand, and Vietnam. The heavy mineral is extracted from the sand and is processed into, among other items, paint pigment, titanium metals, catalysts, and structural materials. The sand itself is used as abrasive material for sandblasting. The principal radioactive components are  $^{232}\text{Th}$  and  $^{238}\text{U}$ . Exposure is mainly external through minerals spilled at the processing plants. Although information on exposures is scant, annual levels are estimated to be in the low  $\mu\text{Sv}$  range (UNSCEAR 1993).

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**6.3.5 Hot Springs and Caves**

Geothermal energy, produced in Iceland, Italy, Japan, New Zealand, Russia, and the United States, is produced from steam or water from high-temperature areas within the earth's crust. Mineral springs and spas, which are found in South America, Europe, Japan, and the United States, are also clustered around these high-temperature areas of the earth's crust. The primary radionuclides in this source are those of the uranium transformation chain. Of these,  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  are considered to be the most important to public health.

The diffusion of radon from ordinary rock and soils and from radon-rich water can cause notably elevated radon concentrations in tunnels, caves, and spas. In Bad Gastein, Austria, approximately 5 million gallons of mineral water are distributed to hotels and spas daily, allowing the release of about 58 Ci (2 Mbq) of radon per year into the environment. In comparison with levels in outdoor air, the concentrations of radon and its transformation products in confined air spaces such as mines and caves are elevated. The average annual per person radiation dose from this source is estimated to be 0.0001 mrem (1 nSv); however, the doses received by those intentionally spending time in these environs (e.g., tourists, workers, miners) are much greater than this amount (Eisenbud 1987; IARC 1988; UNSCEAR 1993).

**6.4 NATURAL INTERNAL EXPOSURE**

Natural internal radionuclides contribute an estimated 11% to the average population radiation dose. Radioactive materials enter the body by inhalation, ingestion, or dermal absorption. Although radioactive materials may also enter the body through punctures (either wounds or injections), this route of exposure will not be addressed in this toxicological profile.

The effects induced by internally deposited nuclides or external radiation are classified as either "acute" (early-occurring effects of radiation, which appear within days or weeks after exposure) or "latent" (chronic or late-occurring effects of radiation, which appear months or years after exposure). Acute effects are not expected for natural sources of radiation because they are not capable of producing high dose rates in a short period of time. However, they may cause latent effects. The most common latent radiation effect is an increased probability of certain types of cancer. More information on the health effects of ionizing radiation can be found in Chapter 3 of this toxicological profile.

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**6.4.1 Inhalation**

The sources of inhaled radioactive materials include debris from atmospheric nuclear testing; nuclear reactor and medical gaseous waste; radioactive materials manufacturing; diagnostic medical radionuclide use; coal- and gas-burning power plants; airborne soil; and naturally emanating gases. The radionuclides (and their average concentrations) commonly found in the atmosphere include:  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  ( $270 \text{ pCi/m}^3$  [ $10 \text{ MBq/m}^3$ ] each);  $^{210}\text{Pb}$  ( $0.01 \text{ pCi/m}^3$ );  $^{210}\text{Po}$  ( $0.001 \text{ pCi/m}^3$ );  $^{238}\text{U}$  ( $12 \times 10^{-5} \text{ pCi/m}^3$ );  $^{232}\text{Th}$  ( $3 \times 10^{-5} \text{ pCi/m}^3$ );  $^{230}\text{Th}$  ( $4.5 \times 10^{-5} \text{ pCi/m}^3$ ); and  $^{228}\text{Th}$  ( $3 \times 10^{-5} \text{ pCi/m}^3$ ),  $^{14}\text{C}$  and  $^3\text{H}$ . In addition, smokers are exposed to radiation from the radionuclide  $^{210}\text{Po}$ , which is found in tobacco; the resulting dose to the bronchial epithelium can be as high as 20 mrem (0.2 mSv) per year (NCRP 1984; Shapiro 1990; UNSCEAR 1993).

The largest dose of radiation from natural sources comes from the inhalation of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  (thoron) gases. These colorless and odorless gases, which are in the uranium and thorium transformation chains, respectively, are continuously released from the soil. Worldwide, the total emanation rate of radon is estimated to be 50 Ci/sec (2 TBq/sec); the total atmospheric content is estimated to be 25 MCi (1 EBq). The main factors controlling the rate of radon release and subsequent exposure are: ground porosity, ground cover, temperature, meteorological conditions, and the type of construction and ventilation properties of dwellings. The rate of radon emanation from soil is thought to increase with diminished atmospheric pressure and to decrease during periods of, or in areas of, elevated moisture, while the atmospheric concentration of radon tends to increase during temperature inversions and as the humidity decreases.

The health hazards of radon exposure were first recognized in the 1930s when radium miners in Scheen-burg, Germany, and Joachimstal, Czechoslovakia, were found to have a high incidence of lung cancer. Over half of all miner deaths were attributed to lung cancer, and most of the miners were less than 50 years of age when they died. In the general U.S. population, the EPA estimated that radon exposure accounts for approximately 10% (17,000) of all lung cancers, while smoking accounts for approximately 85% (144,500) of all lung cancers. The average annual effective dose equivalent from radon is about 200 mrem (2 mSv), but individual doses may be much higher. It is estimated that 1–3% of all homes have radon levels in excess of 8 pCi/L, which is twice the EPA recommended residential limit of 4 pCi/L. Approximately 50,000 to 100,000 homes in the U.S. have radon concentrations exceeding 20 pCi/L, which results in exposures equal to or exceeding the limit for occupational exposure. The  $^{220}\text{Rn}$  doses are considerably lower than those of  $^{222}\text{Rn}$ , due to its short half-life (55 sec for  $^{220}\text{Rn}$  versus 3.8 days for

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$^{222}\text{Rn}$ ). Both  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$  have several short-lived progeny in their transformation chains (see Table 6-3), and radiation from these daughter products constitutes the hazard from radon. Thus, in assessing the effects associated with radon exposure, one must consider the simultaneous and cumulative effect of the entire radon series (BEIR V 1990; Eisenbud 1987; LBL 1993; NCI 1996; Shapiro 1990; UNSCEAR 1993).

**Table 6-3. Radioactive Properties of  $^{222}\text{Rn}$  Radon and its Daughter Products**

Radionuclide	Half-life	Radiation energies (MeV)		
		$\alpha$	$\beta$	$\gamma$
$^{222}\text{Rn}$	3.8 days	5.49	—	—
$^{218}\text{Polonium}$	3.1 minutes	6.00	—	—
$^{214}\text{Lead}$	26.8 minutes	—	0.67 0.73	0.30 0.35
$^{214}\text{Bismuth}$	19.9 minutes	—	1.51 1.54 3.27	0.61 1.12 1.76
$^{214}\text{Polonium}$	164 $\mu\text{seconds}$	7.60	—	0.8
$^{210}\text{Lead}$	22.3 years	—	0.016 0.06	0.05
$^{210}\text{Bismuth}$	5 days	—	1.16	—
$^{210}\text{Polonium}$	138 days	5.31	—	—
$^{206}\text{Lead}$	No half-life; stable element			

Source: adapted from Schleien 1992 (includes radiations over 10% intensity)

Coal mine exhaust and the combustion products from the use of coal and oil typically contain radon and daughter products, which contribute doses of 0.001 mrem (10 nSv) or less. The average annual per person doses from radiation from coal- and oil-fired power plants are about 0.2 mrem (2  $\mu\text{Sv}$ ) and 0.001 mrem (10 nSv), respectively. The average annual per person dose from radiation from domestic cooking and heating with coal is about 0.04–0.8 mrem (0.4–8  $\mu\text{Sv}$ ); this dose originates primarily from radon and its daughter products (UNSCEAR 1993).

Several radioactive by-products of the nuclear power industry may be inhaled and result in internal exposure. During uranium fuel fabrication, uranium hexafluoride gas is enriched to increase the percentage of  $^{235}\text{U}$  and then converted into uranium oxide or metal. Depending upon the type of reactor

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fuel or nuclear weapons material being produced, uranium must be enriched to a minimum of 3%  $^{235}\text{U}$  for fuel and 93.5% for nuclear weapons. Emissions from fabrication facilities usually consist of the long-lived isotopes  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ , and the short-lived isotopes  $^{234}\text{Th}$ , and protactinium-234m ( $^{234\text{m}}\text{Pa}$ ). The major route of exposure from this source is inhalation. More information about uranium is available in the ATSDR *Toxicological Profile for Uranium* (ATSDR 1999).

### 6.4.2 Ingestion

The sources of radionuclides that contribute to radiation exposure by ingestion include nuclear weapons testing, the accidental or intentional release of radioactivity from nuclear reactors, the release of medical or experimental radionuclides into sanitary sewers, and naturally occurring radionuclides (which normally represent the source of highest oral dose). For most radionuclides present at waste sites containing low levels of radioactive nuclides, oral exposure is not a major route of exposure. There is a small probability of radionuclide ingestion because of the potential for surface water and groundwater contamination and uptake by plants and animals following erosion of ground cover from a contaminated site.

Among the naturally occurring radionuclides, uranium,  $^{40}\text{K}$ , and  $^{226}\text{Ra}$  are found in soils and fertilizers; as a result, they are incorporated into foods consumed by animals and humans. The practice of using phosphate fertilizers has resulted in uranium concentrations in food at levels up to 8 ng/g, resulting in an estimated average annual intake of uranium from dietary sources of 10 Bq; as a result, the average skeletal content of uranium is estimated to be 25  $\mu\text{g}$ , which is equivalent to approximately 17 pCi (Eisenbud 1987; UNSCEAR 1993).

The most important radionuclides that are ingested are  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and the transformation products of  $^{226}\text{Ra}$ . However, wherever it is found, all potassium is radioactive because its natural isotope,  $^{40}\text{K}$ , is radioactive. The body content of potassium is under strict homeostatic control and is maintained at a relatively constant level of about 140 g/70 kg. This amount of potassium contains approximately 0.1  $\mu\text{Ci}$  (4,000 Bq) of  $^{40}\text{K}$ . Because the body controls the potassium balance, environmental variations have little effect on the  $^{40}\text{K}$  content in the body (Eisenbud 1987; Shapiro 1990). This natural  $^{40}\text{K}$  delivers a dose of 20 mrem/year (0.2 mSv/year) to the gonads and other soft tissues and 15 mrem/year (0.15 mSv/year) to bone.

FDA has developed guidelines for radionuclide levels in food for individuals from 3 months to adult that are summarized in Chapter 7 (Regulations), Table 7-4, FDA Derived Intervention Levels (FDA 1998).

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**6.4.3 Dermal**

For the purposes of this profile, dermal exposure to radionuclides refers to exposures from a radionuclide placed in direct contact with skin surface. Dermal exposure is typically a minor route of internal and external exposure. In general, depending on the specific physical properties of the radionuclide that may reside on the skin, the percutaneous absorption of radionuclides from particles is negligible, especially if the skin is thoroughly washed immediately after exposure. The long-term biological effects of dermally absorbed radionuclides are limited to the level of the epidermis and dermis (and its vasculature). More soluble forms of the radionuclides may result in a small percentage of the nuclide being absorbed if it is not removed from the skin's surface, for example tritium, as tritiated water or vapor, is readily absorbed into the body through the skin. Generally, the skin is an effective barrier against absorption of radionuclides (except for tritiated water) into the body. The dermal exposure pathway is, therefore, a minor route of exposure at low-level radioactive waste sites.

**6.5 X RAY AND NUCLEAR MEDICINE EXPOSURES**

Radioactive materials and other sources of ionizing radiation are widely used in the diagnosis and treatment of some diseases in human and veterinary medicine (NCRP 1989a; 1996). They represent 15% of the average population dose, 11% for x rays and 4% for nuclear medicine. In 1989, the estimated number of x ray machines used in the US was 109,000 for medical diagnosis, 143,000 for dental diagnosis, and 1,300 for therapy: 3 million diagnostic examinations were made which produced a collective US dose of 92,000 man•Sv (9,200,000 man-rem). Typical effective dose equivalents for various procedures are 0.14 mSv (14 mrem) for a chest x ray, 1.0 mSv (100 mrem) for mammography, and 7.2 mSv (720 mrem) for an upper GI tract evaluation. Due to the usefulness of nuclear medicine, radioactive drugs and diagnostic compounds have become significant contributors to internal radiation dose from man-made sources today. The average U.S. nuclear medicine examination gives an effective dose equivalent of 5 mSv (500 mrem) with individual procedures delivering 2.5 mSv (250 mrem) for a thyroid uptake, 6.3 mSv (630 mrem) for a bone scan, and 14 mSv (1,400 mrem) for a cardiovascular screen. Each year in the US, the collective effective dose from nuclear medicine procedures is 32,000 man•Sv (3,200,000 man-rem). Therapeutic doses are much larger to the individual but many fewer individuals are exposed. After many millions of diagnostic radionuclide procedures, we have found no increase in cancers from these procedures.

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The common sources of radiation exposure associated with radiotherapy and diagnosis include x rays, thallium-201 ( $^{201}\text{Tl}$ ), technetium-99m ( $^{99\text{m}}\text{Tc}$ ),  $^{125}\text{I}$ , and  $^{131}\text{I}$ . More exposures are related to diagnosis than to therapy, and the average number of treatments per person increases as the level of health care improves. Also, the average dose per individual treatment tends to decrease as techniques and equipment improve. Overall, x ray treatments deliver a higher average per person dose in industrialized nations (average of 0.3–2.2 mSv) than in countries with less developed health care (average exposure 0.02–0.2 mSv). On an individual basis, the average dose increases with age, from 52 mrem/year in adolescents to 151 mrem/year in persons over 65 years of age. Exposures are usually lower for examinations of the extremities and skull and higher for examination of the gastrointestinal (GI) tract. In the United States, the average annual dose to the bone marrow from this source increased from 83 mrem in 1964 to 103 mrem in 1970. A person receiving a full set of dental x rays would add approximately 40 mrem to his or her annual dose. On the other hand, the average annual dose per patient from the diagnostic use of radionuclides is lower in industrialized nations, largely because of greater use of  $^{99\text{m}}\text{Tc}$ . This radionuclide is preferred over  $^{131}\text{I}$  because its shorter half-life (6 hours versus 8 days) gives a much lower patient dose. There is currently no radioiodine in the atmosphere due to atmospheric testing because of the 8-day half-life of  $^{131}\text{I}$ . The shorter half-life and higher cost of  $^{99\text{m}}\text{Tc}$  make it more available in developed than in developing nations, where  $^{131}\text{I}$  has frequent use. While the average dose (per individual) in patients undergoing radiotherapy is much greater than in patients undergoing diagnosis, the exposure group is much smaller, resulting in a lower overall population-at-risk. Unfortunately, serious exposures resulting from failures of equipment, procedures, or personnel errors (usually a result of not following procedures) sometimes occur, with several hundred failures out of several hundred million procedures per year worldwide.

There are several emerging trends in diagnostic nuclear medicine. Some of these trends include: the introduction of radiolabeled monoclonal antibodies for imaging and treatment; the emergence of new compounds used in positron emission tomography (PET) and single-photon emission computed tomography (SPECT) studies; and the use of computed x ray tomography (CAT scans). Radiolabeled monoclonal antibodies have proven useful in the localization of tumors and metastases. Common radionuclides associated with these antibodies include indium-111 ( $^{111}\text{In}$ ),  $^{131}\text{I}$ , and  $^{99\text{m}}\text{Tc}$ . SPECT is used for tumor localization, brain and cardiac studies, and bone or abdominal imaging. PET, which uses nuclides such as  $^{11}\text{C}$  and  $^{18}\text{F}$ , can gather anatomical and physiological information that would otherwise be difficult to collect. Whole-body imaging using radiolabeled compounds (e.g., anticancer drugs) is becoming a common PET application (DOE 1996; Eisenbud 1987; Shapiro 1990; UNSCEAR 1993).

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

The use of radiopharmaceuticals has stabilized in industrialized countries but is increasing in developing countries. Long-lived radionuclides are used more frequently in developing countries, while industrialized countries tend to use short-lived radionuclides. This results in increased exposures per examination among developing country patients compared to those of industrialized nations. For example, a typical thyroid scintigraphy with  $^{99m}\text{Tc}$  can give an effective dose of less than 0.1 rem (1 mSv), while the same procedure using  $^{131}\text{I}$  gives 10 rem (100 mSv); however,  $^{99m}\text{Tc}$  is less readily available in developing countries. Although the average per patient dose equivalent is lower in developed (2–5 mSv [200–500 mrem]) than in less developed countries (20 mSv [2,000 mrem]), an apparently larger fraction of individuals in developed nations receive nuclear medicine treatment, so the average per capita annual dose from radiopharmaceuticals in developed countries (0.07 mSv [7 mrem]) is an order of magnitude more than that of developing nations (0.004 mSv [4mrem]) (UNSCEAR 1993).

Radionuclides are frequently produced and used in industry, medicine, and research. The number of users and frequency of radionuclide use are both steadily increasing. The number of establishments in Japan that generate and/or use radionuclides has increased from 100 in 1960 to 5,000 in 1990. The public may be exposed to radionuclides from these sources as a result of routine use or from being near someone who has recently received a nuclear medicine procedure, as well as improper handling, use, or disposal. In Japan, the usage of  $^{14}\text{C}$ ,  $^{125}\text{I}$ ,  $^3\text{H}$ , and  $^{131}\text{I}$  has been estimated to be 5.2, 6.1, 14, and 34 GBq (0.14, 0.16, 0.38, and 0.92 Ci) per million persons, respectively. In contrast, the production of  $^{14}\text{C}$  in the United States and Britain has been estimated to be 30 and 55 GBq (0.81 and 1.49 Ci) per million, respectively. The annual global production and usage of  $^{14}\text{C}$  has been estimated to be 30 GBq (810 mCi) per million persons, or a total of 0.05 PBq (1.5 kCi). The total amount of  $^{131}\text{I}$  produced in Sweden for medical purposes was estimated to be 0.9 TBq (110 GBq [2.97 Ci] per million) in 1986, while the amount of  $^{131}\text{I}$  discharged from Australian hospitals in 1988 and 1989 was estimated to be 2.9 TBq (190 GBq [5.13 Ci] per million) (UNSCEAR 1993). Information about some radionuclides used for medical applications is shown in Table 6-4.

$^3\text{H}$  and noble gases are released to the air, while  $^{14}\text{C}$  release is through airborne and fluid effluents. The isotopes  $^{131}\text{I}$  and  $^{125}\text{I}$  are primarily released through liquid effluent. The annual collective dose from medical and radiopharmaceutical wastes to local populations is thought to be in the range of 10,000 man•rem (100 man•Sv). This level of exposure is relatively unimportant compared to that from other sources (Eisenbud 1987; UNSCEAR 1993).

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine**

Radionuclide	Preparation	Use	Properties
<sup>99m</sup> Tc Albumin	Reduce pertechnetate <sup>99m</sup> Tc in the presence of human albumin, ascorbic acid, FeCl <sub>2</sub> , and SnCl <sub>2</sub> .	Primarily used for lung imaging. Also used for imaging of coronary, urogenital, liver, gastrointestinal, lymphatic, and peripheral circulation.	The biological clearance half-life from the lungs of 14 to 15 hours.
<sup>111</sup> In Albumin	Incubate <sup>111</sup> In with human albumin in phosphate at pH 3; adjust pH to 11 and heat.	Primarily used for lung imaging. Also used for imaging of coronary, urogenital, liver, gastrointestinal, lymphatic, and peripheral circulation.	The biological clearance half-life from the lungs of 14 to 15 hours.
<sup>113m</sup> In Albumin	Incubate <sup>113m</sup> In with human albumin in phosphate at pH 3; adjust pH to 11 and heat.	Primarily used for lung imaging. Also used for imaging of coronary, urogenital, liver, gastrointestinal, lymphatic, and peripheral circulation.	The biological clearance half-life from the lungs of 14 to 15 hours.
<sup>203</sup> Pb Albumin	Incubate ionic <sup>203</sup> Pb with human albumin at pH 10 with heat.	Primarily used for lung imaging. Also used for imaging of coronary, urogenital, liver, gastrointestinal, lymphatic, and peripheral circulation.	The biological clearance half-life from the lungs of 14 to 15 hours.
<sup>51</sup> Cr Albumin	Incubate <sup>51</sup> CrCl <sub>3</sub> with human albumin	Detection and quantitation of gastrointestinal protein loss and placental localization	Cr (III) has strong affinity for plasma proteins without affecting (binding to) red blood cells.
<sup>125</sup> I Albumin	Mild iodination of human albumin at 10 EC in slightly alkaline medium	Diagnostic aid in determining total blood and plasma volumes	Longer shelf life than <sup>131</sup> I; emits no beta radiation (all gamma emissions); lower doses needed to obtain greater resolution compared to <sup>131</sup> I
<sup>131</sup> I Albumin	Mild iodination of human albumin at 10 EC in slightly alkaline medium	Diagnostic aid in determining total blood and plasma volumes, circulation times, or cardiac output.	May cause sensitization.
<sup>131</sup> I Albumin, aggregated	Mild iodination of human albumin at 10 EC in slightly alkaline medium	Diagnostic study of the lungs, especially the diagnosis of pulmonary embolisms.	Aggregates block a small percentage (<0.5%) of the fine capillaries. Disintegrating aggregates are cleared by phagocytic Kupffer cells in the liver. Thyroid uptake may be blocked by prior administration of Lugol's solution.
<sup>197</sup> Hg Chlormerodrin	Reflux allylurea with <sup>197</sup> Hg mercuric acetate in methanol; add aqueous sodium chloride	Diagnostic aid in scanning the brain for lesions. Also used for scanning kidneys for anatomical and functional abnormalities.	Rapidly cleared by the kidneys. Provides smaller radiation dose compared to <sup>131</sup> I albumin. A high tumor:background ratio is obtained within 4 hours, allowing quicker scans with greater resolution.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
<sup>203</sup> Hg Chlormerodrin	Reflux allylurea with <sup>203</sup> Hg mercuric acetate in methanol; add aqueous sodium chloride	Diagnostic aid in scanning the brain for lesions. Also used for scanning kidneys for anatomical and functional abnormalities.	Rapidly cleared by the kidneys. Provides smaller radiation dose compared to <sup>131</sup> I albumin. A high tumor:background ratio is obtained within 4 hours, allowing quicker scans with greater resolution.
<sup>32</sup> P Chromic phosphate	React Na <sub>2</sub> H <sup>32</sup> PO <sub>4</sub> with chromic nitrate in a saline-carboxymethylcellulose vehicle.	A neoplastic suppressant that provides palliative treatment of pleural and peritoneal effusions.	Emits virtually no gamma radiation; delivers 10-fold greater radiation dose per millicurie compared to <sup>198</sup> Au. Because it remains <i>in situ</i> after injection, it may be injected directly into a malignancy.
<sup>60</sup> Co	Neutron bombardment of <sup>59</sup> Co	Replaced radium in various therapeutic areas.	The gamma radiation matches that of radium very closely.
<sup>192</sup> Ir	Neutron bombardment of <sup>191</sup> Ir	Replaced radium in various therapeutic areas. May be enclosed in nylon mesh for interstitial use.	Provides softer (i.e., less penetrating) radiation compared to radium.
<sup>57</sup> Co Cyanocobalamin	Vitamin B <sub>12</sub> in which a portion of the molecule contains <sup>57</sup> Co.	Diagnostic aid in studying the absorption and deposition of vitamin B <sub>12</sub> , especially the diagnosis of pernicious anemia.	
<sup>60</sup> Co Cyanocobalamin	Vitamin B <sub>12</sub> in which a portion of the molecule contains <sup>60</sup> Co.	Diagnostic aid in studying the absorption and deposition of vitamin B <sub>12</sub> , especially the diagnosis of pernicious anemia.	Although the half-life is 5.24 years, <sup>60</sup> Co cyanocobalamin may decompose in storage; thus, frequent radiochemical analysis may be required.
Exametazime	<sup>99m</sup> TcO is incorporated into the exametazime molecule.	An adjunct in detecting regional cerebral perfusion in stroke; leukocyte labeling.	Is rapidly cleared from the blood. Maximum brain uptake (3.5-7.0%) is reached within 1 minute of injection. Must be used within 30 minutes of reconstitution due to conversion of lipophilic complex to second lipophilic complex that will not cross the blood-brain barrier.
<sup>113</sup> In Ferric hydroxide	<sup>113</sup> In is stirred with FeCl <sub>3</sub> while titrated with 0.5 N NaOH to a pH of 11 to 12. While stirring, a 20% gelatin is added to attain a pH of 7.6 to 8.5 while heating in a boiling waterbath; preparation is then autoclaved.	A diagnostic aid in lung imaging.	Particles are in the 20 to 50 μm range.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
<sup>59</sup> Fe Ferrous Citrate	<sup>59</sup> Fe complexed with citrate; neutron bombardment of <sup>58</sup> Fe.	A diagnostic aid in studying the kinetics of iron metabolism.	It may be administered directly into the bloodstream where it reacts with the metal-binding globulin.
<sup>99m</sup> Tc Ferrous hydroxide	Add <sup>99m</sup> Tc to a vial containing ferrous sulfate; the hydroxide is precipitated with 0.1N NaOH at a pH of 7.5 to 10.7. Gelatin is added to stabilize the particles; final pH should be 7.1 to 8.3.	A diagnostic aid in pulmonary scintigraphy.	Most of the particles are in the 11 to 13 μm range; virtually all particles fall in the 3 to 50 μm range;.
<sup>125</sup> I Fibrinogen	<sup>125</sup> I in the form of I <sub>2</sub> , ICl or I <sup>-</sup> is combined with fibrinogen and is oxidized by chloramine-T, electrolytically or enzymatically. Unreacted iodine is removed by the addition of sodium thiosulfate.	A diagnostic aid in the localization of deep vein thrombosis. Other applications include detection of renal transplant rejection, tumors, and the study of fibrinogen turnover.	Accumulates in clots, the radiation is easily detected at the external surface of the affected limb.
<sup>67</sup> Ga Gallium citrate	<sup>67</sup> Ga is produced by proton irradiation of <sup>67</sup> Zn-enriched ZnO <sub>2</sub>	Used in the diagnosis of lesions of the lung, breast, maxillary sinuses and liver. A positive <sup>67</sup> Ga uptake is potentially indicative of malignancies such as lymphomas, bronchogenic carcinoma, and Hodgkin's disease. Also useful for placental localization and diagnosis of pancreatitis and disk disc-space infection.	Concentrates in tumors of soft tissues and bone. The half-life of the isotope is 78 hours; the biological half-life of the citrate compound is 53 days.
<sup>111</sup> In Indium chlorides	A cadmium target is bombarded with deuterons. The <sup>111</sup> In is then etched from the target with HCl, carrier Fe <sup>3+</sup> is added, and the mixture is precipitated with NH <sub>4</sub> OH. The precipitate is dissolved in HCl and the ferric iron is removed by extraction with isopropyl ether.	<sup>111</sup> In has been used as a tag for a variety of compounds such as transferrin, EDTA and DTPA (used in cisternography), bleomycin (used for tumor localization), platelets (detection of coronary thrombi), lymphocytes (monitoring cardiac antirejection therapy), and leukocytes (diagnosis of upper-abdominal infections).	Indium normally exists in aqueous solution as a trivalent cation. In aqueous solution InCl exists as a mixture of hydrated chlorides.
<sup>111</sup> In Oxyquinoline		Used to label various blood components such as neutrophils, platelets and lymphocytes; cardiac imaging (labeled platelets); localization of infectious and inflammatory processes (labeled leukocytes).	

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
$^{113m}\text{In}$ Indium Chloride	$^{113m}\text{In}$ is formed by the radioactive transformation of $^{113}\text{Sn}$ . $^{113m}\text{In}$ is separated from $^{113}\text{Sn}$ using sterile, pyrogen-free dilute HCl.	Used in blood-pool studies, including visualization of aneurysms, and placental scintigraphy; also used for bone, liver, lung, brain, and renal imaging.	Indium normally exists in aqueous solution as a trivalent cation. In aqueous solution $\text{InCl}$ exists as a mixture of hydrated chlorides. Urinary excretion is low, resulting in low urinary bladder activity.
$^{113m}\text{In}$ Indium hydroxide	$^{113m}\text{In}$ Indium chloride is adjusted to a pH of 4 or more, whereupon the indium is converted to the insoluble hydroxide. The particle size and stability are controlled by heating and the addition of a stabilizer (gelatin, mannitol, etc.).	Used in liver, spleen and bone marrow scintigraphy.	
$^{125}\text{I}$ or $^{131}\text{I}$ Insulin	Prepared by mild iodination with high-specific-activity radioactive iodine followed by purification via dialysis, ion-exchange or other process.	Used for <i>in vitro</i> assay of circulating insulin; study of <i>in vivo</i> insulin kinetics	Longer shelf life than $^{131}\text{I}$ ; emits no beta radiation (all gamma emissions); lower doses needed for correct resolution compared to $^{131}\text{I}$
$^{123}\text{I}$ or $^{131}\text{I}$ Na Iodohippurate	Iodobenzyl chloride is condensed with glycine with the aid of a dehydrochlorinating agent. The resulting <i>o</i> -iodohippuric acid is reacted with NaOH.	Used in the detection of renal malfunction.	Excreted almost exclusively by the kidneys.
$^{81m}\text{Kr}$ gas	A transformation product of $^{81}\text{Rb}$ , which is produced by alpha bombardment of $^{79}\text{Br}$ .	Used for lung function, ventilation, and perfusion. Also used in radiocardiology.	
$^{123}\text{I}$ Iofetamine Hydrochloride		Used in assessing regional cerebral blood flow.	Crosses the intact blood-brain barrier. Concentrates in metabolically active brain cells. Binding by relatively nonspecific high-capacity binding sites results in brain retention.
$^{125}\text{I}$ or $^{131}\text{I}$ Liothyronine	Synthetic liothyronine is exchanged with $^{131}\text{I}$ . The mixture is then purified by column or strip paper chromatography.	Used for <i>in vitro</i> evaluation of thyroid function.	$^{125}\text{I}$ binds to thyroxine-binding proteins. Due to the high specific activity, this compound may not be taken internally as radiation damage can occur easily. This may be prevented in part by the use of propylene glycol (50%) as a solvent. The materials should be refrigerated or frozen and should be used within 2 weeks.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
<sup>125</sup> I or <sup>131</sup> I Levothyroxine	Obtained by synthesis, with the I-tag in the 3'-position.	Used to study the endogenous metabolism of endogenous thyroxine; to measure thyroxine-binding protein capacity.	Binds to thyroxine-binding proteins.
<sup>111</sup> In Pentetate Indium Disodium	Cyclotron-produced indium chlorides mixed with pentetic acid at low pH (#3.5) to form an indium-DTPA chelate. A trisodium salt of the complex is formed by increasing the pH to 7.0–7.5.	Used as a diagnostic aid for studies of cardiac output, glomerular filtration evaluation; used for cisternography and renal scintigraphy.	Shelf-life is limited by the half-life of <sup>111</sup> In (67.5 hours).
<sup>113m</sup> In Pentetate Indium Trisodium	Pentetic acid containing some ferric ion and HCl is mixed with <sup>113m</sup> In. The resulting chelate is stabilized by increasing the pH to 7.0–7.5, resulting in the formation of a trisodium salt.	Used as a diagnostic aid for studies of glomerular filtration; also used for brain scanning and kidney imaging, and cisternography of spinal fluid circulation.	
<sup>169</sup> Yb Pentetate Ytterbium Trisodium	Buffered, lyophilized pentetic acid is mixed with <sup>169</sup> Yb.	Used as a diagnostic aid for brain scanning and kidney imaging, and cisternographic diagnosis of CSF rhinorrhea.	May be administered orally or intravenously.
<sup>42</sup> K Potassium Chloride	By neutron bombardment of natural potassium.	Used for tumor localization and studies of renal blood flow measuring total exchangeable potassium.	Suitable for intravenous administration.
<sup>43</sup> K Potassium Chloride	By alpha bombardment of a natural argon target.	Used as a diagnostic aid for heart imaging.	
<sup>131</sup> I Rose Bengal Sodium	Prepared by thermal condensation of tetrachlorophthalic anhydride with 2,4-diiodoresorcinol. The resulting phthalein is reacted with NaOH, and the purified product is labeled by isotope exchange	A diagnostic aid for liver function; especially useful for differential diagnosis of hepatobiliary disease.	Accumulates in the polygonal cells of the liver and is excreted via the biliary system. If liver function is impaired, it is excreted via the kidneys.
<sup>75</sup> Se Seleno- methionine	Extracted from yeast grown on sulfur-free medium to which trace amounts of radiolabeled sodium selenite have been added. Selenomethionine is separated from the yeast proteins.	Used for scintigraphy of the pancreas and parathyroid glands; also used to visualize the parotid and prostate glands.	Incorporated into newly-formed proteins. Blood levels decline to a minimum value at 20 to 45 minutes after IV injection; blood levels then rise to about 3/4 that seen at 2 minutes postinjection.
<sup>22</sup> Na Sodium Chloride	By deuteron bombardment of <sup>24</sup> Mg	Used for determination of circulation times, sodium space, and total exchangeable sodium.	The usual required tracer dose is well within tolerated levels. Emits positrons which are easily detected by coincidence counting.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
<sup>51</sup> Cr Sodium Chromate	By neutron bombardment of enriched <sup>50</sup> Cr.	Used as a biological tracer to measure red-cell volume, red-cell survival time, and whole-blood volume. Also used to detect blood cell loss due to hemolytic anemia or GI bleeding.	Requires 15–60 minutes to diffuse into red cells; binds to globin molecules. Has no deleterious effect on erythrocytes.
<sup>18</sup> F Sodium Fluoride	By neutron bombardment of enriched <sup>6</sup> Li in the form of lithium carbonate. Contamination with <sup>3</sup> H must be removed prior to use.	Useful for bone imaging, especially areas of altered osteogenic activity.	
<sup>123</sup> I Sodium Iodide	By proton bombardment of enriched <sup>124</sup> Te or by deuteron bombardment of enriched <sup>122</sup> Te or by transformation of <sup>123</sup> Xe.	For diagnostic procedures in thyroid function studies; for organ imaging including the thyroid, liver, lung, and brain.	Short half-life (13.2 hours) and radiation characteristics result in a smaller radiation dose compared to other iodine isotopes.
<sup>125</sup> I Sodium Iodide	By neutron bombardment of xenon gas.	For diagnostic procedures in thyroid function studies; for organ imaging including the thyroid, liver, and brain; treatment of deep-seated non-resectable tumors.	For organ imaging, dose to patient may be decreased with better delineation of organ and clearer resolution than <sup>131</sup> I.
<sup>131</sup> I Sodium Iodide	By neutron bombardment of enriched <sup>131</sup> Te or as a byproduct of uranium fission.	For diagnostic procedures in thyroid function studies; a neoplastic suppressant.	
<sup>99m</sup> Tc Sodium Pertechnetate	Produced by the elution of sodium pertechnetate through a generator containing <sup>99</sup> Mo which decays to <sup>99m</sup> Tc.	Used in the detection and localization of cranial lesions, thyroid and salivary gland imaging, placenta localization, and blood-pool imaging.	<sup>99m</sup> Tc has an ideal half-life which is long enough for diagnostic procedures but is short enough to minimize radiation doses to the patient. Also, it lacks a beta radiation component. Pertechnetate is readily absorbed by the thyroid; this can be reduced by preinfusion of potassium perchlorate.
<sup>32</sup> P Sodium Phosphate	By neutron bombardment of elemental sulfur in an atomic reactor. <sup>32</sup> P is then separated by leaching with NaOH.	A neoplastic and polycythemic suppressant; a diagnostic aid for the localization of certain ocular tumors.	
<sup>85</sup> Sr Strontium	By neutron bombardment of a strontium salt enriched in <sup>85</sup> Sr.	A diagnostic aid for scanning bones and bony structures to detect and define lesions and to study bone growth and abnormal formations.	Has a long half-life (64 days), resulting in high bone doses.
<sup>99m</sup> Tc Albumin	Albumin is tagged with a reduced form of the pertechnetate. The pertechnetate may be reduced by one of several methods.	Diagnostic aid in determining total blood and plasma volumes, circulation times, or cardiac output.	See earlier comment on <sup>99m</sup> Tc.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
$^{99m}\text{Tc}$ Albumin, Aggregated	Denatured human albumin is tagged with a reduced form of the pertechnetate. The pertechnetate may be reduced by one of several methods.	Diagnostic aid in study of the lungs. Primary use is for diagnosing pulmonary embolism. Also useful for static blood-pool imaging, angiography, dynamic function tests and visualization of placental tissues.	See earlier comment on $^{99m}\text{Tc}$ . $^{99m}\text{Tc}$ is preferred over $^{131}\text{I}$ as the radioactive tag because of the smaller delivered dose.
$^{99m}\text{Tc}$ Etidronate or $^{99m}\text{Tc}$ Oxidronate	Acetic acid is treated with $\text{PCl}_3$ ; the disodium salt is formed when a solution of etidronic acid is adjusted to a pH of 8.5. Stannous chloride and sometimes a stabilizer such as sodium ascorbate are added. Freshly eluted $^{99m}\text{Tc}$ is added.	Useful for bone imaging.	See earlier comment on $^{99m}\text{Tc}$ . This compound is superior to $^{18}\text{F}$ bone scans and to roentgen studies and is frequently more sensitive in detecting metastases to the bone.
$^{99m}\text{Tc}$ Iminodiacetic Acid (IDA)	Usually provided in kit form, the compound is reconstituted and tagged by adding sterile $^{99m}\text{Tc}$ sodium pertechnetate.	Useful for hepatobiliary imaging.	See earlier comment on $^{99m}\text{Tc}$ .
$^{99m}\text{Tc}$ Ferpentate	Usually in kit form, the compound is made by adding a solution of $^{99m}\text{Tc}$ sodium pertechnetate; the pH is adjusted with sodium hydroxide and a solution of pentetic acid is added. The chelate is formed by gentle mixing.	Useful for kidney imaging.	See earlier comment on $^{99m}\text{Tc}$ .
$^{99m}\text{Tc}$ Pentetate	Prepared by adding sterile $^{99m}\text{Tc}$ pertechnetate saline solution to an aliquot of buffered stock solution of DTPA containing stannous chloride as a reducing agent. Instant DTPA $^{99m}\text{Tc}$ kits are available.	Useful for brain and kidney visualization, for vascular dynamic studies for measurement of glomerular filtration and for lung ventilation studies.	See earlier comment on $^{99m}\text{Tc}$ . Does not concentrate in any organ. DTPA is uniformly distributed throughout the extracellular space and is rapidly cleared by the kidneys without retention.
$^{99m}\text{Tc}$ Pyrophosphate	Sodium pyrophosphate, mixed with stannous tin, are combined with a solution of $^{99m}\text{Tc}$ sodium pertechnetate. Kits are available commercially.	Used as a skeletal imaging agent; used to demonstrate areas of altered osteogenesis; also used as a cardiac imaging agent, as an adjunct in the diagnosis of myocardial infarction.	See earlier comment on $^{99m}\text{Tc}$ . The pyrophosphate compound has been found to concentrate in muscle tissue, especially contused muscle tissue. myocardium.
$^{99m}\text{Tc}$ Sestamibi	Usually in kit form, the compound is reconstituted and tagged with sterile $^{99m}\text{Tc}$ sodium pertechnetate.	Used as a myocardial perfusion agent in the evaluation of ischemic heart disease and distinguishing and locating abnormal myocardium.	Accumulates in viable myocardial tissue.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
<sup>99m</sup> Tc Succimer	Usually in kit form, the compound is reconstituted and tagged with sterile <sup>99m</sup> Tc sodium pertechnetate.	Used in renal cortical imaging.	Use within 30 minutes of formulation.
<sup>99m</sup> Tc Sulfur Colloid	A colloidal suspension of sulfur labeled with <sup>99m</sup> Tc.	Used as a diagnostic aid for liver, spleen, and bone marrow scanning. Also used in detection of intrapulmonary and lower GI bleeding, as well as visualization of the lungs by inhalation of the colloid.	See earlier comment on <sup>99m</sup> Tc. Colloids are phagocytized by the liver. The plasma clearance is rapid (approximately 2.5 min). At least 80% of dose accumulates in the liver.
<sup>99m</sup> Tc Tebroxime		Used for myocardial perfusion imaging for distinguishing normal versus abnormal myocardium.	Use within 6 hours of reconstitution.
<sup>99m</sup> Tc Gluceptate	Freshly eluted <sup>99m</sup> Tc sodium pertechnetate is added to sodium glucoheptonate in combination with stannous chloride.	Useful as a renal imaging agent; possibly useful for localization of brain, lung, and gallbladder lesions.	See earlier comment on <sup>99m</sup> Tc. Optimal results are obtained 1–2 hours after administration.
<sup>99m</sup> Tc Sodium Methylene Diphosphonate	Sodium methylene diphosphonate, available in kit form, is mixed reconstituted with <sup>99m</sup> Tc sodium pertechnetate.	Useful for skeletal imaging.	See earlier comment on <sup>99m</sup> Tc. When administered by IV, compound concentrates in areas of altered osteogenesis.
<sup>99m</sup> Tc Mertiatide	Usually provided in kit form, the compound is reconstituted and tagged by adding sterile <sup>99m</sup> Tc mertiatide.	Useful as a renal imaging agent; provides information on renal function, split function, renal angiograms and renogram curves for whole kidney & renal cortex.	Is reversibly bound to serum protein and is excreted rapidly by kidneys and cleared by the blood.
<sup>99m</sup> Tc Sodium Phosphates	Polyphosphate polymer, available in kit form mixed with stannous chloride, is mixed with <sup>99m</sup> Tc pertechnetate.	Useful for bone and renal imaging.	See earlier comment on <sup>99m</sup> Tc.
<sup>99m</sup> Tc Sodium Phytate	Sodium phytate, available in kit form mixed with stannous chloride, is reconstituted with <sup>99m</sup> Tc pertechnetate.	Useful for liver and spleen imaging	See earlier comment on <sup>99m</sup> Tc. Cleared rapidly from the blood by the reticuloendothelial system. Over 80% of compound localizes in the liver and spleen within 30 minutes of an iv injection. The addition of ionic calcium to the <sup>99m</sup> Tc stannous phytate mixture enhances splenic uptake.
<sup>99m</sup> Tc Tetracycline	Tetracycline, available in kit form, is reconstituted with stannous chloride and <sup>99m</sup> Tc pertechnetate.	For imaging kidneys and gall bladder; myocardial imaging is possible with larger doses.	See earlier comment on <sup>99m</sup> Tc. The compound localizes in the gall bladder.

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Table 6-4. Some Radiopharmaceuticals Used in Medicine (continued)**

Radionuclide	Preparation	Use	Properties
<sup>201</sup> Tl Thallium Chloride	Thallium target material is bombarded with protons to produce <sup>201</sup> Pb. The unused thallium material is removed by ion exchange, and the remaining <sup>201</sup> Pb subsequently decays to <sup>201</sup> Tl.	Used for myocardial perfusion imaging for the localization of myocardial ischemia and infarction; used as an adjunct to angiography. Also useful for thyroid imaging, particularly the detection of goiter and thyroid carcinoma.	Thallium mimics potassium ions and is taken up by the cells of the heart; decreased cell vitality is indicated by decreased thallium uptake. Rapidly disappears from the blood.
<sup>15</sup> O Water	Prepared by labeling with a cyclotron-generated radionuclide.	Used in blood-flow imaging using positron emission tomography scanning.	
<sup>127</sup> Xe Xenon gas	Produced by proton bombardment of cesium-133 with <sup>127</sup> Xe.	As a gas, used for lung imaging to detect alveolar block-age; also used for mapping cerebral blood flow. Dissolved in saline used as a tracer for measurement of regional blood flow.	The biological half-life of the gas is approximately 15 minutes.
<sup>133</sup> Xe Xenon	A product of nuclear fission; also formed by neutron activation of <sup>132</sup> Xe.	As a gas, used for lung imaging to detect alveolar blockage; also used for mapping cerebral blood flow. Dissolved in saline used as a tracer for measurement of regional blood flow.	The biological half-life of the gas is approximately 15 minutes.

Source: Remington 1985; Remington and Gennaro 1995

**6.6 EXPOSURE FROM CONSUMER PRODUCTS**

Consumer products contribute an estimated 3% to the average population radiation dose. Several consumer products, used both within the home and in many public areas, emit minuscule amounts of radiation. Among these are ionization-type smoke detectors, television sets, and liquid propane gas (LPG) appliances. The first smoke detectors contained radium (approximately 20  $\mu\text{Ci}$  [0.7 MBq]), but now contain americium-241 (<sup>241</sup>Am), which is more economical and produces much less radiation dose. While present-day detectors contain 0.5–1.0  $\mu\text{Ci}$  (0.02–0.04 MBq) of <sup>241</sup>Am, the original units contained approximately 80  $\mu\text{Ci}$  (3 MBq). In the 1980s, annual sales of smoke detectors approached 12 million, representing approximately 8.5 Ci (300 GBq) of <sup>241</sup>Am. Smoke detectors contain a small ionization chamber in which the air between two electrodes is ionized by the source radionuclide. This ionization allows the flow of current across the gap between the electrodes. When the flow is stopped by smoke particles, the interruption in current flow is interpreted by the detector to indicate the presence of smoke. Television sets accelerate electrons that bombard the screen; in the process, low-energy x rays are emitted. The total annual dose associated with watching a color television has been estimated to be 2–3 mrad per year (0.02–0.03 mGy/yr). Radon is found in LPG, which may be used in water heaters,

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

stoves, and fireplaces; it has been estimated that exposure to radon in homes using natural gas results in an average annual dose of approximately 5 mrem (0.05 mSv) in the United States.

Among consumer products of the past are items that contained radium, such as medicines, tonics, luminous paints, and ceramic glazes. After its discovery in the early part of the 20th century, radium was used for many years in the treatment of rheumatism (arthritis) and mental disorders; oral solutions contained  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  at concentrations up to 2  $\mu\text{Ci}/60\text{ mL}$ , while ampules for intravenous administration contained 5–100  $\mu\text{g}$   $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . Radium was also used to produce luminescent paints that were applied to wristwatches, clocks, static eliminators, fire alarms, electron tubes, and military and educational products. During the peak years of production, approximately 3 million radium-laden timepieces were sold annually in the United States. The radium content of a man's wristwatch ranged from 0.01 to 0.36  $\mu\text{Ci}$  (370–13,000 Bq), resulting in potential gonadal doses of 0.5–6 mrem/year (0.005–0.06 mSv/yr). Radium has been replaced with  $^3\text{H}$  and promethium-147 ( $^{147}\text{Pm}$ ), and watch cases are sufficiently thick to absorb the beta emissions from these radionuclides. Uranium has been used as a coloring agent for ceramic glazes, resulting in doses to the hands of up to 20 mrad/hour (0.02 mGy/hr). The dose from ceramics produced since 1944 is thought to be five-fold less than that from earlier pieces. For more than 40 years,  $^{224}\text{Ra}$  has been used in Europe to treat the symptoms of tuberculosis and ankylosing spondylitis. Although its use in children was curtailed in the 1950s,  $^{224}\text{Ra}$  has been used for treating the pain associated with ankylosing spondylitis. In two studies of patients treated with  $^{224}\text{Ra}$ , average calculated skeletal doses ranged from 0.65–4.2 Gy (65–420 rad) (Eisenbud 1987; Harley 1996; Harvard Medical School 1996; NCRP 1993).

## 6.7 EXPOSURE FROM OTHER SOURCES

Other sources contribute less than 1% to the average population radiation dose. This radiation exposure may result from several anthropogenic sources, including the radioactive debris still remaining from atmospheric and underground detonation of nuclear weapons, electrical energy production, radiopharmaceuticals, and radionuclide production and use (Shapiro 1990; UNSCEAR 1993).

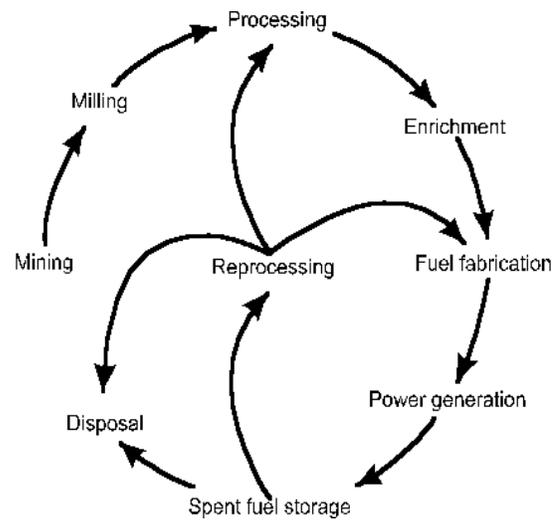
## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**6.7.1 Exposure from the Nuclear Fuel Cycle**

The nuclear fuel cycle contributes around 0.1% to the average population radiation dose and refers to the mining, milling and enrichment of uranium; fabrication of fuel elements; the production of electricity; and the recycling, transportation, and waste storage/disposal of radioactive materials used in nuclear weapons or reactor-grade nuclear fuel. The steps involved in the uranium fuel cycle are depicted in Figure 6-2 and described in the Toxicological Profile for Uranium (ATSDR 1999b).

The primary radionuclide components of nuclear weapons and reactors include  $^{239}\text{Pu}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^3\text{H}$ . Radionuclides associated with uranium mining and milling

include  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and their natural decay chain radionuclides, such as  $^{226}\text{Ra}$ ,  $^{234}\text{Th}$ ,  $^{234\text{m}}\text{Pa}$ ,  $^{230}\text{Th}$ , and  $^{222}\text{Rn}$ , while those associated with power production and subsequent waste disposal include (but are not limited to)  $^{60}\text{Co}$ ,  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{129}\text{I}$ ,  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$ . Noble gas radionuclides of Kr, Xe, and Zr are only associated with operational reactor releases. The various steps within this cycle provide multiple opportunities for the exposure of humans to these materials.



**Figure 6-2. Schematic of the Nuclear Fuel Cycle**

**Mining and milling.** Uranium ore typically contains uranium at concentrations ranging from a tenth of a percent to a few percent; thus, millions of tons of ore are mined and processed annually to meet the needs of nuclear power plants for uranium fuel. Radon is the predominant radionuclide released from uranium mines. Radon containing air is discharged from mines at a rate of approximately  $0.5\text{--}20\ \mu\text{Ci}/\text{min}/1,000\ \text{ft}^3$ ; these are point releases whose concentrations dilute quickly with distance from the release shaft and, thus, pose no additional health risk to the general public. Incomplete extraction of uranium during milling results in uranium concentrations in mill tailings of  $0.001\text{--}0.01\%$ . The presence of radon precursors ( $^{226}\text{Ra}$  and  $^{230}\text{Th}$ ) in mill tailings presents a potential long-term source for atmospheric radon. The rate of radon emanation varies with meteorological factors such as barometric pressure and humidity. The rate of soil and mill tailings migration depends primarily on wind and water erosion of the site (Eisenbud 1987; UNSCEAR 1993).

## 6. SOURCES OF POPULATION EXPOSURE TO IONIZING RADIATION

**Enrichment and fuel fabrication.** In both nuclear weapons and nuclear fuel production, after being mined and milled, uranium must be converted to uranium hexafluoride gas, which is then enriched and converted to uranium oxide or metal. If enrichment is carried to about 90%, the uranium may be used to make nuclear weapons or to fuel naval warships; alternatively, the uranium may be enriched by only a small percentage for use in civilian nuclear energy facilities. Metallic uranium is capable of reacting with both air and water exothermically; because of this reactivity, the more stable uranium oxide is the most commonly used fuel in reactors. While this form is more stable, it has poor thermal conductivity, necessitating the use of small-diameter fuel rods. The fuel is in the form of high melting point ceramic pellets, about 0.5 inches in diameter and 1 inch long, in which  $\text{UO}_2$ -enriched to 3–4%  $^{235}\text{U}$  is dispersed. These pellets are stacked end to end in zirconium alloy or stainless steel tubes about 12 feet long (called cladding) and then sealed to retain the fission products that are produced during operation. These fuel filled tubes are then assembled in groups of 8 x 8 to 17 x 17 arrays into fuel rod assemblies. About 500 of these assemblies make up the core of a nuclear power reactor. For a frame of reference, a single pellet contains the energy equivalent of about one ton of coal or 3 barrels of oil. Emissions from fabrication facilities usually consist of the long-lived isotopes  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ , and the short-lived nuclides  $^{234}\text{Th}$  and  $^{234\text{m}}\text{Pa}$ ; however, the relative value of the refined and enriched uranium and the high level of accountability for uranium stock preclude any long-term or widespread loss of material. The major route of exposure from this source is inhalation (Eisenbud 1987; UNSCEAR 1993). More information about the toxicological properties of uranium can be found in the *Toxicological Profile for Uranium* (ATSDR 1999b).

**Power generation.** Power production from nuclear plants has increased steadily since the industry's birth in the 1950s. During the years between 1970 and 1989, the number of nuclear reactors worldwide increased from 77 to 426, and total nuclear power generation increased from 9 to 212 gigawatts per year. In 1996, nuclear power plants produced 17% and 19.4% of the world's and U. S. electrical energy, respectively (DOE 1997c). The annual worldwide production of uranium from 1979 to 1989 ranged from 19,000 to 44,000 tons; from 1985 to 1990, the annual production was approximately 50,000 tons. USNRC regulations require that all the component parts of the nuclear fuel cycle be designed and operated to limit the annual dose to a member of the public from the total nuclear fuel cycle to a maximum of 25 mrem (0.25 mSv). Various regulations identified in Chapter 7 of this profile are designed to limit human exposure to radiation and radioactive materials. Guidance documents, such as the ANSI air sampling standard (ANSI 1999), are available to aid in establishing monitoring programs for assessing discharges from nuclear facilities. During the energy production phase, radioactive contamination of the coolant occurs through small defects in the protective cladding surrounding the fuel

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pellets, through fission of “tramp” uranium contamination on the outside surface of the fuel rods and through neutron activation of contaminants in the cooling medium.

In general, the levels of radionuclide emissions from reactors are not typically detectable, except at points close to effluent discharges; because of this, estimates of radionuclide discharge levels must be modeled. Based on such models, the total collective dose due to reactor discharges through 1989 was estimated to be 370,000 man•rem (3,700 man•Sv) over a 45-year period. This may be compared to the collective dose to the U.S. public from the natural radioactive potassium within everyone’s body, which is about 5,000,000 man•rem (50,000 man•Sv) each year. A 1981 U.S. Nuclear Regulatory Commission (USNRC) study of the doses received by 98 million people living within 80 km of 48 nuclear facilities concluded, on the basis of a zero threshold model, that 0.02 excess fatal cancers per year, or 1 every 50 years, could be attributed to exposures from nuclear facilities (USNRC 1981). A study of the cancer rates in populations surrounding 62 U.S. nuclear facilities, performed by the National Cancer Institute (NCI) in 1990, found no evidence of a relationship between proximity to a nuclear facility and the occurrence of cancer (Eisenbud 1987; NCI 1990; UNSCEAR 1993).

**Weapons production.** There is little public information regarding the amount of radioactive materials produced for use as weapons. The atmospheric content of krypton-85 ( $^{85}\text{Kr}$ ), a by-product of plutonium extraction, has been used to estimate the plutonium stockpiles in both the United States and Russia. After adjusting for production and release of  $^{85}\text{Kr}$  from nuclear reactors, it is estimated from the atmospheric content of  $^{85}\text{Kr}$  that plutonium stockpiles in both the United States and Russia is about 100 tons each. United Nations estimates from 1981 and 1990 state that nuclear arsenals are comprised of 40,000 weapons with a combined explosive power of 13,000 Mt. Tritium, which has a half-life of 12.32 years, must be continually produced to replace aging stockpiles. It is estimated that an annual production of 3 kg is sufficient to replace that lost by transformation in the United States. By inference, this would indicate a total U.S. stockpile of 55 kg and a world stockpile of about 110 kg (UNSCEAR 1993).

The dose from nuclear weapons research, development, and production is less than 1% of the dose from atmospheric testing. Variations in local exposure from former weapons plants have been reported. In the United States, the Hanford nuclear weapons facility has released a significant amount of radioactive material into the atmosphere and the Columbia River from its plutonium production and reprocessing plants. The majority of the radioactive material ( $^{131}\text{I}$ ) was released between 1944 and 1946 (18 PBq; 486 kCi), although additional releases are known to have occurred from 1947 to 1956 (2 PBq; 54kCi). A

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recent study reconstructed the radiation doses to individuals who were exposed to the  $^{131}\text{I}$  released from the Hanford site, medically determined their thyroid health, and found that the Hanford releases had not caused thyroid problems. The thyroid radiation doses to the 3,193 participants averaged 0.186 Gy (18.6 rad) and ranged from 0–2.842 Gy (0–284.2 rad). Health impacts to the thyroid which have been related to radiation exposure in other studies include thyroid cancer, benign thyroid nodules, hypothyroidism, and autoimmune thyroiditis. The study assessed these conditions and concluded that the occurrence of these diseases in the population were not related to the Hanford releases' radiation dose to the thyroid (CDC 1999).

The Chelyabinsk-40 center, located near Kyshtym in the Soviet Union, was the first nuclear weapons processing facility in Russia. A uranium-graphite-moderated reactor and a fuel reprocessing plant were opened in 1948. Due to poor waste handling and the storage of radioactive wastes in the open, significant liquid releases (100 PBq; 2.7MCi) to the Techa River occurred from 1949 to 1956, with the majority of the releases (95%) occurring from March 1950 to November 1951. The main nuclides released included  $^{89}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ , and ruthenium and rare-earth nuclides. The population along the Techa River was exposed to both external and internal doses of radiation; a total of 20 settlements (7,500 people) were eventually evacuated. The average doses to persons living in the village of Metlino, 7 km downstream from the plant, were estimated to be as much as 140 rad (1.4 Gy) (UNSCEAR 1993).

**Fuel reprocessing.** Fuel reprocessing allows the recovery of uranium and plutonium from the irradiated fuel pellets. Less than 10% of the nuclear fuel is consumed in a spent fuel rod. The radionuclides most commonly associated with reprocessing waste are:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ ,  $^{129}\text{I}$ ,  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and transuranium nuclides. At present, reprocessing is carried out in only a few countries, and only a small portion of the total fuel inventory is being reprocessed (4% from 1985 to 1990). The remainder is retrievably stored (UNSCEAR 1993).

The reprocessing of nuclear fuel has been performed almost exclusively at government-owned facilities designed to meet military needs. Only 1 operable reprocessing facility exists in the U.S. As with nuclear reactors, the facility and the equipment used have been designed with numerous safeguards to prevent criticality and to ensure containment of radioactive material in the event of a non-nuclear explosion or system failure. The estimated collective dose due to reprocessing to date is estimated to be 460,000 man•rem (4,600 man•Sv); the main radionuclide constituents (>90%) of these releases have been  $^{137}\text{Cs}$  and  $^{106}\text{Ru}$ . Releases of gaseous  $^{85}\text{Kr}$  and  $^3\text{H}$  from reprocessing facilities have also been reported. The main pathways of exposure are consumption of locally caught fish and shellfish, external (whole-body) irradiation from intertidal areas, and external (dermal) irradiation of fishermen handling pots and nets.

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Annual individual doses were estimated for critical populations living near three foreign reprocessing plants (Sellafield, England; Cap de la Hage, France; and Tokai-Mura, Japan) for which records of radioactive effluent exist. For the critical population living near Sellafield, annual individual doses from ingestion were estimated to be approximately 350 mrem (3.5 mSv) during the early 1980s and declined to approximately 20 mrem (0.2 mSv) by 1986. The estimated doses in the same group due to external irradiation were estimated to be about 100 mrem (1 mSv) in the early 1980s and 30 mrem (0.3 mSv) by 1986. In contrast, annual individual doses for critical populations living near the Cap de la Hage and Tokai-Mura reprocessing plants were approximately 25 mrem (0.25 mSv) and 0.1 mrem (1  $\mu$ Sv) (Eisenbud 1987; UNSCEAR 1993). A nuclear criticality accident occurred at the Tokai-Mura, Japan uranium reprocessing plant on September 30, 1999 with acute exposures to workers and general population (UPI 1999).

**Waste disposal.** Solid wastes derived from reactor operations and from the handling, processing, and disposal of spent fuel are classified as low-, intermediate-, or high-level wastes. While low- and intermediate-level wastes had been packaged and placed into shallow burial sites, high-level waste disposal strategies have only recently been implemented. Some low-level wastes were packaged and disposed at sea from 1946 to 1982. Currently, some high level waste is being vitrified (mixed with hot liquid glass), solidified inside double-walled stainless steel containers, and prepared for long-term retrievable storage. Exposure to uncontained buried wastes is thought to occur through groundwater migration from leakage at the burial site. The major radionuclide found in reactor waste is  $^{14}\text{C}$  (UNSCEAR 1993).

### 6.7.2 Japanese Atomic Bomb

#### Exposure

A large human cohort (86,572 people who survived the detonation of two atomic bombs in Japan in 1945 and whose radiation doses are reasonably well known) are being studied for the effects of external exposure to ionizing radiation. The Japanese survivors are not the largest cohort; some medically exposed populations are larger. However, the Japanese survivors are probably the most important because of the length of follow-up

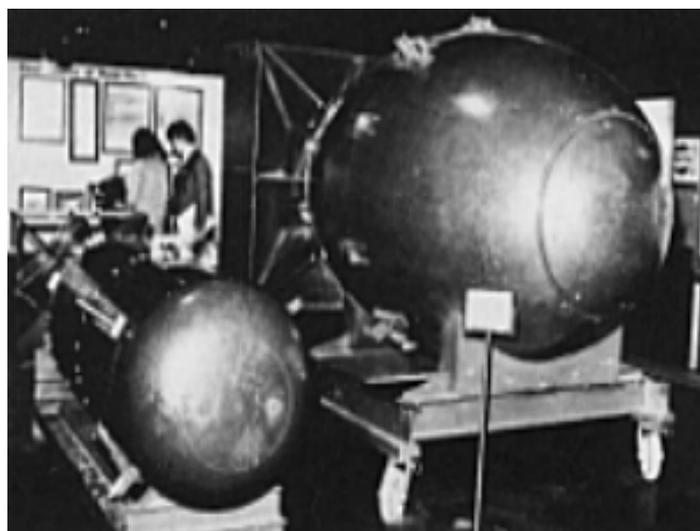


Figure 6-3. Replicas of the “Little Boy” and “Fat Man” Bombs Dropped on Hiroshima and Nagasaki (adapted from A-Bomb WWW Museum, <http://www.csi.ad.jp/ABOMB>).

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and the wide range of doses received. The first atomic bomb was detonated on July 16, 1945, in Alamogordo, New Mexico. The U.S. military, in an effort to bring a swifter end to World War II and to avoid a costly ground invasion of Japan, which could claim more U.S. and Japanese lives, detonated a  $^{235}\text{U}$  atomic bomb, nicknamed “Little Boy,” (see Figure 6-3) over the city of Hiroshima, Japan, on August 6, 1945. Three days later, an atomic bomb using  $^{239}\text{Pu}$ , nicknamed “Fat Man,” was detonated over the city of Nagasaki, Japan.

The uranium used in the “Little Boy” bomb was enriched to  $>80\%$   $^{235}\text{U}$ . (Natural uranium contains 0.7%  $^{235}\text{U}$ , and reactor fuel is enriched to 3–4%  $^{235}\text{U}$ ). The uranium bomb design of Little Boy used a standard explosion trigger, called the “gun” method, because it was originally made using a gun barrel. In this configuration, a sub-critical uranium mass, referred to as the “bullet,” was propelled inside the gun barrel toward a second sub-critical portion of the uranium mass (called the “target”), which was located at the end of the gun barrel. The target contained slightly less than the amount of uranium needed to achieve critical mass (the amount necessary to create a chain reaction). The instant the two subcritical pieces of uranium came together, super-criticality was attained, and an explosion with a force equivalent to 15,000 tons (15 kt) of trinitrotoluene (TNT) occurred (Roesch 1987). In the case of Little Boy, the bullet was a cylindrical stack of nine  $^{235}\text{U}$  wafers about 10 cm wide and 16 cm long, containing 40% of the bomb’s total  $^{235}\text{U}$  mass (25.6 kg). The target was a hollow cylinder 16 cm long and wide; it weighed 38.4 kg and was composed of two separate rings that were inserted into the bomb separately to prevent reaching critical mass during assembly. The complete Little Boy weapon was 10.5 feet long, 28–29 inches in diameter, and reportedly weighed between 8,900 and 9,700 pounds. The firing mechanism was so simple and was considered so failproof that it was not tested prior to its use over Hiroshima. The gun-type firing mechanism was, however, an unsafe weapon design, in that once the firing mechanism was loaded with high explosive, anything that ignited it would cause a nuclear explosion. Also, a crash or even an accidental drop of the bomb could have driven the bullet into the target, potentially resulting in a nuclear explosion. No other weapon of this design was ever tested, and although several Little Boy units were built, none ever entered the U.S. nuclear arsenal.

The plutonium bomb, Fat Man, was dramatically different from the Little Boy design. The gun-type firing mechanism could not be used to unite two pieces of plutonium fast enough to achieve a nuclear blast; impurities in the plutonium would have caused premature detonation. Fat Man contained a ball of subcritical plutonium (plutonium core), which was surrounded with high explosives. The high explosives were cast into spheres, called lenses, and were wired so they would all fire at the same instant. The instantaneous pressure from all sides compressed the plutonium core in on itself, causing it to reach critical mass and density, and create a nuclear blast. The combat configuration for the Fat Man bomb

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consisted of the implosion device encapsulated in a steel armor egg. Fat Man was 5 feet in diameter, 12 feet in length, and weighed 10,300 pounds (American Airpower Heritage Museum 1996; Sublette 1996). The explosion of Fat Man at Nagasaki was equivalent to 21,000 tons (21 kt) of TNT (Roesch 1987).

In Hiroshima and Nagasaki, a total of 64,000 people within 1 km of the hypocenter (the point on the ground directly below where the bomb exploded in the air) died on the first day. The total numbers of acute deaths from the explosions were 90,000–140,000 in Hiroshima and 60,000–80,000 in Nagasaki. For bombs the size (approximate yield of 15 kilotons) and type of Little Boy, the energy released within the first minute after detonation was in the forms of thermal radiation (35%), blast wave (50%), and ionizing radiation (15%); most casualties (including fatalities) resulted from the heat and blast. Two-thirds of those who died during the first day were burned. People close enough to suffer from radiation illness were also well within the lethal zones from blast and heat; thus, the proportion of survivors experiencing radiation illness (30%) was much smaller than the expected proportion based solely on exposure to radiation. People within 1–2 km of the hypocenter who initially survived the blast and received several hundred rad (several grays) of radiation, suffered the ill effects of acute radiation syndrome. It is estimated that all persons whose bodies received a dose of 600 rad (6 Gy) and half of those whose radiation doses were 450 rad (4.5 Gy) died shortly thereafter, as a direct result of radiation exposure. Of those who survived the immediate radiation illness effects, a portion would suffer from the latent effects of radiation-induced cancer (the excess cancer death rate among the exposed population is on the order of 5% greater than that of an unexposed population). It is estimated that approximately 500 survivors have died of radiation induced cancers since 1945. Given the estimated altitudes at which Little Boy and Fat Man detonated (1,900±50 ft. and 1,650±33 ft., respectively), little or no soil was carried up into the fireball, so the fallout produced came mainly from the detonation itself. Of that, very little radioactive material was deposited on the ground in the vicinity of ground zero; the majority was carried high into the atmosphere by heat convection. A small amount of fallout did occur in areas close to the cities due to rainfall that occurred shortly after the explosions; the affected areas were to the west and northwest of Hiroshima and a few miles east of Nagasaki. Fatality rates in the Hiroshima and Nagasaki attacks were 1–2 orders of magnitude greater than rates from conventional bombings because of the nearly instantaneous destruction of buildings that occurred without warning, and because survivors were so incapacitated that they could not escape the rapidly ensuing fire storms. Approximately one-third of all Japanese bombing fatalities occurred in these two cities (Masse 1996; Sublette 1995, 1996; Uranium Information Center 1995; Zajtchak 1989).

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**6.7.3 Exposure from Nuclear Weapons Testing**

Nuclear weapons testing is conducted to assess design efficiency and magnitude of resulting damage. Nuclear explosions world wide were carried out above ground from 1945 to 1980, with the periods of greatest activity occurring from 1952 to 1958, and from 1961 to 1962. A total of 520 tests with an estimated total equivalent energy of 545 megatons (Mt) of TNT was performed, resulting in the release of 220 PBq (6 MCi) of radioactive material. The first U.S. testing of nuclear weapons after World War II was performed in the Marshall Islands in the Pacific Ocean from 1946 to 1948. The Soviet Union conducted its first weapons test in 1948. In the 1950s, as the frequency of weapons testing escalated, so did public concerns over radioactive fallout. In the fall of 1958, the United States, Britain, and Russia, declared a moratorium on weapons testing; however, Russia broke the agreement in 1961, and another rapid escalation in testing ensued. In 1963, the United States, Britain, and Russia, signed the Limited Test Ban Treaty, which prohibited atmospheric testing. Although these three countries have remained faithful to the treaty, other countries such as France, China, India, and Pakistan have since conducted weapons testing (Eisenbud 1987; PBS 1999; UNSCEAR 1993).

The energy from nuclear weapons devices is generated by one or both of the following reactions: (1) the fission of  $^{235}\text{U}$  or  $^{239}\text{Pu}$  in a chain reaction and (2) the fusion of the hydrogen isotopes deuterium and tritium. Fission-type weapons accounted for 217 Mt of the total test yield, while fusion-type weapons accounted for 328 Mt. Many radionuclides are produced by U and Pu fission. Fusion reactions produce helium and result in the neutron activation of the surrounding substance. The most notable neutron activation product is  $^{14}\text{C}$ , which is formed from the neutron bombardment of atmospheric nitrogen. Unused weapons material is also liberated after detonation due to premature loss of critical mass (Radnet 1996; UNSCEAR 1993).

The most important radionuclides associated with nuclear weapons testing exposures are  $^{14}\text{C}$ ;  $^{137}\text{Cs}$ ; zirconium-95 ( $^{95}\text{Zr}$ ); niobium-95 ( $^{95}\text{Nb}$ );  $^{90}\text{Sr}$ ; ruthenium-106 ( $^{106}\text{Ru}$ ); manganese-54 ( $^{54}\text{Mn}$ );  $^{144}\text{Ce}$ ;  $^{131}\text{I}$ ; and  $^3\text{H}$ . This is in addition to the large amounts of  $^{14}\text{C}$  and  $^3\text{H}$  in the biosphere's carbon and hydrogen inventories which are produced by the interaction of cosmic rays with atmospheric gases.

The disposition of ingested strontium-90 ( $^{90}\text{Sr}$ ) has been studied extensively due to its abundance (15 MCi of that introduced into the atmosphere as a result of nuclear weapons testing fell to the earth by January 1970), its long half-life (28 years), and its tendency to localize in bones. Metabolically, Sr follows the pathways of calcium (Ca); however, the body discriminates against Sr in favor of Ca.

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Because of this parallel mechanism, Sr concentrations are often expressed as activity of Sr per gram of Ca. In 1965, bone levels of  $^{90}\text{Sr}$  in 1- to 4-year-old Norwegian children averaged 11.8 pCi/g Ca. In New York City, bone levels of  $^{90}\text{Sr}$  in 1- to 2-year-olds varied from 7 pCi/g Ca in 1965 to 1.6 pCi/g Ca in 1975. In 5- to 19-year-olds, bone levels varied from 3 pCi/g Ca during 1956–1968 to 1.4 pCi/g Ca in 1975 (Shapiro 1990). In an effort to quantify exposure resulting from nuclear testing,  $^{90}\text{Sr}$  deposition has been monitored worldwide at 50 to 200 stations in cooperation with the Environmental Measurements Laboratory (EML), and by a network of 26 stations organized by the United Kingdom Atomic Energy Authority. These data are compiled into a report called Environmental Radiation Data (ERD), which is distributed quarterly by the Office of Radiation and Indoor Air's National Air and Radiation Environmental Laboratory (NAREL). The report contains data from the Environmental Radiation Ambient Monitoring System (ERAMS). ERAMS was established in 1973 by the EPA to provide air, surface and drinking water, and milk samples from which environmental radiation levels are derived. These samples are collected from locations that provide adequate population coverage and function to monitor fallout from nuclear devices and other radioactive contamination from the environment. Samples are subjected to analysis for gross alpha and beta emissions; gamma analyses for fission products; and more specific analysis for uranium plutonium, strontium, iodine, radium, and tritium (EPA 1997).

Zirconium-95 ( $^{95}\text{Zr}$ ) deposition has been monitored as an indicator of exposure to short-lived radionuclides. Monitoring levels of  $^3\text{H}$  and  $^{14}\text{C}$  is more difficult due to the rapid recycling of these elements in a biosphere that contains large cosmogenically produced amounts of these same radionuclides. Interhemispheric transfer is limited due to prevailing trade winds and the scavenging effect of precipitation in the tropics. The best estimate of the average total per person dose, for persons in the northern and southern hemispheres, for all 22 major radionuclides resulting from nuclear testing is 440 and 310 mrem (4.4 and 3.1 mSv), respectively. Worldwide, the average total cumulative dose is 370 mrem (3.7 mSv). However, as noted below, extreme variations in local exposures due to testing have been noted (UNSCEAR 1993).

Approximately 23 billion Ci of  $^{131}\text{I}$  have been introduced into the atmosphere as a result of nuclear weapons testing. In October 1961, the air concentration of  $^{131}\text{I}$  in the United States averaged 3.8 pCi/m<sup>3</sup>. This was estimated to result in an annual dose of 24 mrad to a 1-year-old child. In 1962, the concentration of  $^{131}\text{I}$  in milk in the United States averaged 32 pCi/L. Although about two-thirds of orally administered  $^{131}\text{I}$  is excreted in the urine within the first 24 hours, the remainder concentrates in the thyroid. It has been estimated that an infant receiving milk from cows that grazed on forage contaminated with 1  $\mu\text{Ci }^{131}\text{I}/\text{m}^2$  could receive a dose to the thyroid of 30 rad (Shapiro 1990; UNSCEAR 1993). In addition, studies of pregnant women who died suddenly of non-radiation-induced causes (e.g., car

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accidents) found that fetal tissue concentrations of  $^{131}\text{I}$  were 30% greater than those in maternal tissues (Shapiro 1990). Due to the short 8-day half-life of  $^{131}\text{I}$ , there is no  $^{131}\text{I}$  remaining in the atmosphere or in food from nuclear weapons testing.

Plutonium (Pu) has been introduced into the atmosphere from sources such as weapons testing (>5,000 kg [ $>320$  kCi]) and the vaporization of energy power packs from a Russian satellite (20 kg [0.27 TBq] of  $^{239}\text{Pu}$ ) and U.S. satellites (1 kg [17 kCi] of  $^{238}\text{Pu}$ ) that burned up upon re-entry. Air activity of Pu, monitored in New York, peaked in 1963 at a concentration of 1.7 fCi/m<sup>3</sup>. The cumulative inhalation intake from 1954 to 1975 averaged 43 pCi per person. For comparison, the EPA's annual limit on intake (ALI) for  $^{239}\text{Pu}$  for occupational exposure is 2,000 pCi. Cumulative individual tissue doses (through the year 2000) due to inhalation are predicted to be: lungs, 1.6 mrad; liver, 1.7 mrad; and bone lining cells, 1.5 mrad (Shapiro 1990).

Radioactive debris from nuclear explosions falls into three categories: large particles, which fall out close to the explosion site within hours of the explosion; smaller particles, which penetrate the troposphere, behave like aerosols, and may not fall out for days; and the smallest particles, which penetrate the stratosphere, distribute worldwide, and fall out over many months or years. The greatest portion of fallout from nuclear weapons testing was injected into the stratosphere (78%), while 10% and 12% were injected into the troposphere and in the locality of the test, respectively. As of 1993, the total cumulative worldwide collective dose due to fallout was estimated to be  $7 \times 10^6$  man•Sv. The collective dose will continue to climb, mainly due to long-lived  $^{14}\text{C}$  (Eisenbud 1987; UNSCEAR 1993).

Airborne radioactive materials, both naturally occurring and fallout-derived, usually attach to dust particles; the potential for inhalation of radionuclides bound to particles, and the respective threat this poses to animal and human health, varies considerably. In regions downwind of nuclear weapons test sites or nuclear weapons production facilities, or areas with abnormally high concentrations of naturally occurring radionuclides (e.g., New York state or Denver, Colorado), the potential for inhalation of particulate-bound radionuclides increases. Although most inhaled radioactive particles are eliminated from the lungs by normal clearing mechanisms, some of the particles remain in the lungs for extended periods. Others are carried to lymph nodes by scavenger cells (Eisenbud 1987; Shapiro 1990).

Radiation dose from atmospheric testing is attributed to external exposure to radionuclides on the earth's surface, internal exposure from inhalation of gases or particulate matter, and ingestion of contaminated foods and water. Approximately 80% of the radiation dose from nuclear testing is estimated to be

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delivered through ingestion, with 16% and 4% of the dose delivered through external exposure and inhalation, respectively. Radioactive particles resulting from fallout can contaminate food supplies directly by foliar deposition, or indirectly by entry into the soil and subsequent incorporation into plants, although this delayed incorporation through root uptake accounts for a small portion of that ingested. Surface waters can be contaminated through soil runoff or direct contamination from the atmosphere. The degree of radionuclide incorporation into plants through root uptake varies considerably among radionuclides. For example,  $^{137}\text{Cs}$  and radium bind tightly to clay or organic minerals upon entering the soil and are not amenable to root uptake; thus, foliar deposition is the primary route for oral exposure to  $^{137}\text{Cs}$ . Likewise,  $^{131}\text{I}$  poses little threat through root uptake due to its short half-life (8 days). The concentrations of radionuclides in food vary considerably across food types. The concentration of  $^{226}\text{Ra}$  ranges from 0.15 pCi/kg in cow's milk to 2,000 pCi/kg in Brazil nuts. Concentrations of  $^{137}\text{Cs}$  range from 20 pCi/kg in cow's milk to 5,000 pCi/kg in beef. More important than concentrations in foods is the rate of intake and absorption of a radionuclide. Individual intake varies considerably; for a given radionuclide and locality, intake may vary as much as 500-fold. Likewise, the extent of absorption varies among the various nuclides, from almost completely in the case of  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ , and  $^{131}\text{I}$ , to very poorly (0.003%) in the case of Pu radionuclides. Many alpha emitters, such as  $^{226}\text{Ra}$ , are taken up and retained in the bone, resulting in sustained alpha irradiation of the bone-forming cells and bone surface lining cells (Eisenbud 1987; McClellan 1982; Shapiro 1990).

The primary radionuclides of concern are  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{131}\text{I}$ . Within the first year, 45% of  $^{137}\text{Cs}$  is transferred to the food chain (through milk, grain and meat).  $^{90}\text{Sr}$  enters the food chain primarily through milk and grain products. Exposure to  $^{14}\text{C}$  and  $^3\text{H}$  is through ingestion and inhalation; however, the contribution of  $^{14}\text{C}$  and  $^3\text{H}$  by ingestion is trivial. The dose rate from naturally occurring  $^{14}\text{C}$  is about 1 mrem per year. At its peak effect, the dose rate from  $^{14}\text{C}$  due to weapons testing also was about 1 mrem per year, and is now decreasing. The dose from the tritium due to weapons is considered to be even less. At the peak, the additional  $^3\text{H}$  contributed less than 0.1 mrem per year (UNSCEAR 1993). The radiation dose from  $^{131}\text{I}$  occurs largely during the first 2 months following detonation, due to its short 8-day half-life, and the primary exposure route is via the pasture-cow's milk ingestion pathway.

In addition to exposures from inhalation and ingestion, radiation exposure also occurs externally from particles deposited on the ground. Since the debris spends more time on the ground than in the air, the radiation dose from earthbound particles ranges from 100 to 1,000,000 times that from airborne particles (UNSCEAR 1993).

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**6.7.3.1 Atmospheric Testing**

Atmospheric testing refers to the detonation of nuclear bombs above the earth's surface.

**Nevada Test Site Fallout.** A total of 100 surface or near-surface tests with a total explosive yield of about 1 Mt were performed at the Nevada test site between 1951 and 1962. The population around the site at this time was approximately 180,000 persons. Within this population, thyroid doses in children may have been as high as 100 rad (1 Gy). The collective dose received by this population was approximately 50,000 man•rem (500 man•Sv); 90% of this dose was delivered between 1953 and 1957. The dust from these tests also drifted over the United States, producing bands of exposure to radioactive material. Deposition of fallout varied considerably because of meteorological conditions. For example, the greatest (non-local) fallout levels from one of the Nevada test explosions occurred in New York State, some 2,000 miles away, due to rainfall. The cumulative dose from gamma radiation in New York, approximately 100 mrad (1 mGy), exceeded the doses received by any remote U.S. location for all of 1953 (Eisenbud 1987; UNSCEAR 1993).

**Bikini Atoll Fallout.** Operation Crossroads was a series of nuclear weapons tests that began in the Marshall Islands, a group of atolls in the Pacific, on July 1, 1946. Prior to testing, the inhabitants of the Bikini Island Atoll were evacuated. The second test in this series, designated "Baker," was a 21-kiloton bomb that was detonated underwater. This resulted in contamination of ships staged nearby and the atoll itself. Local soil contamination prevented the return of the Bikini native population until 1969. Although the island was still contaminated in 1969, it was thought that dietary restrictions and the importation of foods would allow safe habitation. However, body burdens of plutonium began to increase in the natives, resulting in their re-evacuation in 1978.

During Operation Castle, another series of nuclear tests, the second test, "Bravo," resulted in significant fallout and contamination of humans. Abrupt changes in wind direction after the 15-Mt detonation on March 1, 1954, resulted in the inadvertent exposure of residents of the Rongelap and Utirik islands, which lie 210 and 570 km to the east of Bikini, as well as exposure of a group of 23 Japanese fishermen whose boat was caught in the fallout approximately 80 miles downwind. Since the device was mounted on a barge situated in shallow water, a considerable amount of coral was incorporated into the fireball. The fishermen reported that the fallout particles resembled snow and that deposits of fallout on the boat were of sufficient depth to allow one to see footprints. Because they were unaware of the circumstances, the fishermen took no precautionary measures to minimize exposure; they remained on the contaminated boat

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until returning to port some 13 days later. Within 1–2 days after exposure to the fallout, the fishermen began to experience itching and burning sensations on exposed skin. By the third day, skin lesions and epilation began to develop; the skin lesions became ulcerous in about 70% of the fishermen. Lesions were less severe in those who had worn protective clothing such as hats (ACHRE 1995; Eisenbud 1987). A more detailed description is available (Simon and Vetter 1997).

Within 78 hours of the explosion, 82 and 159 persons were evacuated from Rongelap and Utirik, respectively. However, as with the fishermen, the island inhabitants took no precautionary measures to minimize exposure to radioactive fallout. Within 1–2 days after exposure to fallout, itching and burning sensations on exposed skin were experienced by the natives of Rongelap, but not those of Utirik. Skin lesions and epilation occurred within 21 days of exposure, becoming ulcerous in about 25% of the Rongelaps; lesions were less severe in those who had worn protective clothing or bathed during the period prior to evacuation. The island of Utirik was not heavily contaminated, and its residents were allowed to return within a few months; however, the Rongelap residents were not allowed to return to their island until 1957, and they were monitored annually by U.S. medical teams thereafter. Despite the monitoring, fears among the island residents that exposure-related health problems were occurring prompted a second evacuation, initiated by the residents, in 1985. External doses, ranging from 10 to 190 rad (0.1 to 1.9 Sv), were mostly from short-lived radionuclides. Mean thyroid doses to adults, 9-year-olds, and 1-year-olds, were 1,300, 2,200, and 5,200 rad (13, 22, and 52 Gy), respectively. Maximum thyroid doses to these groups were 4,200, 8,200, and 20,000 rad (42, 82, and 200 Gy), respectively (ACHRE 1995; Eisenbud 1987; NAS 1994; UNSCEAR 1993).

Average gamma dose rates 3 feet above ground level on Rongelap island, estimated from a survey performed in July 1956, were 0.2–0.5 mR/hr (mean of 0.4 mR/hr) (DOE 1994b). Environmental samples collected in 1964 showed  $^{239}\text{Pu}$  concentrations of 11 pCi/g (0.4 Bq/g) in a soil sample collected at a depth of 0.5–1.0 inch. A more extensive survey that included 14 of the atoll islands was performed in April and May of 1967. External radiation, as well as the radioactive content of food, vegetation, and soil, was measured. On the islands closest to the detonations, the major contributor to the external gamma radiation field was  $^{60}\text{Co}$ , which was associated with neutron activation of scrap metal; the major contributor to the external gamma radiation field on distant islands was  $^{137}\text{Cs}$ . Additional samples were collected during the U.S. cleanup operations in 1969.  $^{239}\text{Pu}$  concentrations on Bikini Island ranged from 1.3 to 190 pCi/g (0.1–7 Bq/g).  $^{239}\text{Pu}$  concentrations on Eneu Island ranged from 0.5 to <3 pCi/g (0.02–<0.1 Bq/g) (DOE 1970). Measurements of gamma radiation exposure performed in June 1975 showed highly variable exposure rates on Bikini island (10–20  $\mu\text{R/hr}$  at the shore versus 30–100  $\mu\text{R/hr}$  in

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the interior), while exposure rates on Eneu island were relatively constant ( $<10 \mu\text{R/hr}$ ) over the entire island. Thirty-year cumulative doses were estimated to be 0.057 and 0.027 Sv (5.7 and 2.7 rem) for those living on Bikini Island (interior portions) and Eneu Island, respectively (USERD 1975). Water samples collected from Eneu Island in 1975 showed  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  at concentrations that would lead to a combined 30-year whole-body and skeletal dose of 25 mrem (0.25 mSv). Sampling of cistern water on Bikini Island during the same period revealed  $^{90}\text{Sr}$  concentrations, which would lead to a 30-year skeletal dose of 9.1 mrem (0.09 mSv), and  $^{137}\text{Cs}$  at concentrations that would lead to a 30-year whole-body dose of 1.9 mrem (0.02 mSv) (DOE 1975). Whole-body counting of Bikini Island residents in 1974, 1977, and 1978 showed that the major contributor to whole body doses was  $^{137}\text{Cs}$ . The average body burden for  $^{137}\text{Cs}$  increased 10-fold between 1974 and 1977 and by 72% between 1977 and 1978. Nine persons had body burdens exceeding the federal standards for non-occupational dose in that year (0.5 rem/year [0.005 Sv/yr]); the highest body burdens were approximately twice the permissible levels (DOE 1978).

**Semipalatinsk Test Site Fallout (Russia).** Approximately 10,000 people living near the Semipalatinsk test site in the Kazakh region of Russia were exposed to radioactive materials from atmospheric testing between the years of 1949 and 1962. Underground testing, which typically retains the radioactive material underground, was conducted between 1964 and 1989. The collective doses to this population from external and internal radiation were estimated to be 260,000 and 200,000 man•rem (2,600 and 2,000 man•Sv), respectively (UNSCEAR 1993).

**Australian Test Site Fallout (United Kingdom).** The United Kingdom performed a total of 12 nuclear tests at 3 sites in Australia with total explosive yields at each site of 100, 16, and 60 kilotons, respectively. The collective dose delivered to the Australian population was estimated to be 70,000 man•rem (700 man•Sv). In addition, several hundred smaller experiments were performed, resulting in the contamination of hundreds of square kilometers with a total of 24 kg of  $^{239}\text{Pu}$ . Potential annual exposures to individuals in these areas, assuming continuous habitation, is estimated to range up to several rem (several hundredths of a Sv) (UNSCEAR 1993).

**Lop Nor Test Site Fallout (People's Republic of China).** China has performed more than 40 nuclear weapons tests. Approximately 23 tests were atmospheric; the last atmospheric test was performed on October 16, 1980 and the fallout was measured worldwide. The remaining tests have been performed underground; the most recent underground test occurred on August 17, 1995. All Chinese nuclear testing occurs at the Lop Nor site, located in the Xinjian region in northwest China. China has not allowed independent assessments of the ecological or health impacts of its testing program; however, increased

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mortality rates due to fallout of radioactive materials have been reported by a political advocacy group in neighboring eastern Turkestan (Eastern Turkestan Information Bulletin 1996). Both the data and the claims are unsubstantiated.

### 6.7.3.2 Underground Testing

Underground testing refers to the detonation of nuclear bombs below the earth's surface. About 1,400 underground nuclear tests have been performed worldwide, with a total explosive yield of 90 Mt. The frequency of underground testing increased dramatically after the 1963 signing of the Limited Test Ban Treaty, which banned atmospheric testing. The nature of underground tests typically causes all the radioactive material produced to be retained underground. However, some radioactive material can be released if the blast penetrates the surface or if inadvertent leaks occur due to ground structure damage or the gradual diffusion of gases. Of the 500 underground tests performed at the Nevada test site, only 32 led to off-site contamination. The total activity of  $^{131}\text{I}$  inadvertently released was about 5 PBq (135 kCi), which is about five orders of magnitude lower than that released during atmospheric testing. Based on calculations of theoretical yields, it is estimated that the total release of noble gases from underground testing resulted in a population dose of 500 man•rem (5 man•Sv). Of the noble gases,  $^{133}\text{Xe}$  is the predominant radionuclide. It is estimated that the total dose from  $^3\text{H}$  resulting from underground testing is 0.1 man•rem (0.001 man•Sv) (UNSCEAR 1993).

In addition to military-sponsored nuclear explosions, a series of about 100 test detonations was carried out during the 1960s for the purpose of developing peaceful applications for nuclear explosives, such as building flood prevention reservoirs and interoceanic canals similar to the Panama canal (designated as Project Plowshare). As the benefits were far outweighed by the issues of contamination, the project was subsequently terminated. Of these tests, six were performed at the Nevada test site. The estimated collective dose delivered to the surrounding population (180,000 persons) from one of these tests (Sedan; 104 kt explosion) was estimated to be 300 man•rem (3 man•Sv). As a result of the Schooner cratering experiment carried out in the United States in 1968, tungsten-181 ( $^{181}\text{Tl}$ ) generated from the neutron shield was detected as far away as Europe. The estimated collective dose from this explosion to the population living in the 40°–50° latitude band of the northern hemisphere was estimated to be 2,000 man•rem (20 man•Sv) (UNSCEAR 1993).

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**6.7.4 Occupational Exposure**

Occupational exposure to radiation occurs when workers handle radioactive materials or are exposed to radiation sources (e.g., x rays and radioactive sources). The history of occupational exposure to radioactivity is as old as its use. In the period between the discovery of x rays and the early 1930s, more than 100 radiologists died of skin cancer, anemia, and leukemia, largely because they knew very little about the hazards of radiation and how to protect themselves adequately from it. The frequencies of anemia and noncancerous skin damage were also elevated. The concept of a “tolerance dose” was developed early, based initially upon the levels of exposure that resulted in erythema. Originally, limits of 5 R/month or 0.2 R/day were established, and these limits were successively lowered as the body of knowledge concerning radiation health effects grew. The current limits are 5 rem/year (0.05 Sv/year) total effective dose equivalent; 50 rem/year (0.5 Sv/year) for the sum of deep-dose equivalent (from external radiation) and committed dose equivalent (from internal radionuclides) to any individual organ or tissue other than the lens of the eye; 15 rem/year (0.15 Sv/year) shallow dose equivalent to the skin or to any extremity. While most exposure is external, internal exposure occurs in several occupations, such as radium dial painters, powerplant workers, uranium miners, radiopharmaceutical manufacturers, and nuclear medicine support staff.

In the early 1900s, it was discovered that radium, when mixed with zinc sulfide causes the zinc sulfide to glow. This discovery spurred the development of radioluminescent paints, which consists of a mixture of finely powdered radium salt and zinc sulfide crystals in an appropriate volatile vehicle. This paint was used in the manufacture of dial faces, wristwatches, static eliminators, emergency exit signs, electron tubes, and educational products. In 1924, bone damage that looked like phosphorus poisoning was observed in radium dial painters employed at a northern New Jersey plant, and later it was determined to be bone cancer caused by radium. It was determined that the young women were inadvertently ingesting radium due to the practice of lip-pointing the brush tips when painting fine numerals. A group of 24 dial painters ingested approximately 900–1,300  $\mu\text{Ci}$  (33–48 MBq) radium during the course of their careers, which resulted in the formation of bone cancers (see Chapter 3 of this toxicological profile) (Eisenbud 1987; Shapiro 1990; UNSCEAR 1993).

Exposure to airborne uranium ore dust occurs in uranium miners and millers, while exposure to airborne elemental uranium or uranium salts occurs in uranium processors. Uranium ore contains other radionuclides including  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$ ,  $^{220}\text{Rn}$ ,  $^{218}\text{Po}$ ,  $^{214}\text{Po}$ , and  $^{210}\text{Po}$ . Radon diffuses from the rock into the mine air, where the radon progeny become attached to particles of dust or moisture and are inhaled into the

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lungs. In the 1800s, silver and uranium miners in Europe were dying of a mysterious malady; the illness was diagnosed as intrathoracic malignancy (lung cancer) in 1879. At that time, it was estimated that the life expectancy of these miners was 20 years after entering the occupation. Death rates from lung cancer in these miners were much higher than expected; as early as 1942, the deaths were attributed to radon exposure. It has been estimated that as much as 40% of all lung cancers in miners may be due to exposure to radon and its progeny (Archer et al. 1973a; Gottlieb and Husen 1982; Lubin et al. 1969, 1995; Samet et al. 1984, 1986). Although radon and its transformation products have been implicated as causative agents in miners with lung cancer, it is difficult to isolate the cancer risk that may be specific to the miners' exposure because they were concurrently exposed to other suspected or known carcinogens such as tobacco smoke, silica and other dusts, and diesel engine exhaust fumes (ACHRE 1995; Auerbach et al. 1978; Band et al. 1980; Lundin et al. 1969; Saccomanno et al. 1971, 1976, 1986; Whittemore and McMillan 1983).

A study of 16 male Navajo uranium miners who developed lung cancer between February 1965 and May 1979 found that the mean cumulative radon exposure was 1,140 working level months (WLM) (Gottlieb and Husen 1982). The working level is a measure of airborne concentration of radon progeny. The WLM is a measure of total exposure. It is the product of the concentration, in WL's and the exposure time, in months (1 working month = 170 hrs). One WLM corresponds to an alpha dose to the tracheobronchial epithelium of approximately 1 rad (0.01 Gy). An excess of lung cancer deaths was also found in uranium miners who had worked underground for at least 1 year in the Grants mineral belt area of New Mexico. Mean exposures in these studies ranged from 2.6 to 42 WLM from 1954 to 1966 and from 0.3 to 21.8 WLM from 1967 to 1982 (Acquavella et al. 1985; Samet et al. 1986). A National Institutes of Health study (NIH 1994) summarized cumulative WLM for several mining cohorts and Colorado miners had the highest average WLM of 807 (follow-up period 1950–1987). Exposed New Mexico miners had an average WLM estimate of 110 and non- U.S. miners ranged from an average WLM of 7 to 370. It should be noted that, in several of these studies, exposure to dust and cigarette smoke was also found to be related in varying degrees to the incidence of cancer.

With the discovery of fission and the development of particle accelerators (Cockcroft-Walton, Van de Graaff generator, cyclotron), numerous new radionuclides and new elements could be readily produced. The number of users and frequency of radionuclide use are both steadily increasing.

Other professions in which workers receive elevated radiation doses include: commercial airline personnel (pilots and flight attendants), military pilots, astronauts, industrial and nuclear power plant workers, radiographers, and dental and medical personnel. A person flying cross-country receives about

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5 mrem (0.05 mSv) per flight due to the increased levels of cosmic radiation associated with the increase in elevation; it has been estimated that pilots and flight attendants receive an annual dose that is approximately 160 mrem (1.6 mSv) higher than that of the average population. Astronauts are exposed to intense radiation emanating from solar flares, the earth's radiation belts, and ambient cosmic radiation. The average radiation doses for crews of the Apollo missions (5–12 days/mission) were 0.16–1.14 rad (0.0016–0.0114 Gy); the average doses for the Skylab missions, which lasted 20–90 days, were 1.6–7.7 rad (0.016–0.77 Gy). These high doses of radiation require attention if people begin living in the environments of outer space (space stations, interplanetary travel, etc.). The processing and blending of LPG tends to enhance radon concentrations, and the long-lived radon daughters ( $^{210}\text{Pb}$  and  $^{210}\text{Po}$ ) tend to accumulate inside LPG processing machinery, resulting in a possible risk of exposure to maintenance workers. A nuclear power plant worker averages 300 mrem (3 mSv) additional dose per year, resulting in doses about 80% higher than the average population (DOE 1996; Eisenbud 1987).

### 6.8 ADEQUACY OF THE DATABASE

The database is considered to be adequate for use as the basis for radiation safety standards.

### 6.9 CONCLUSIONS

The issue of radiation exposure is a matter of interest to the general public; however, radiation exposure is inevitable as it is a natural part of the environment. Indeed, radioactive materials have always existed around and even within us. While the risk of exposure to radiation from man-made sources exists, with the exception of locally high exposures, the average individual dose received from man-made radiation is small compared to that received from natural sources. When assessing the risks associated with a radiation exposure, one must weigh the potential benefits (e.g., gain in quality of life related to medical diagnoses and treatments) against the potential detriments (acute radiation sickness, cancer risk) associated with the exposure. Conversely, in situations presenting minimal risks of exposure to radiation and radioactive materials, one may also compare the potential risks associated with the use of alternatives. For example, in the case of nuclear power versus power from fossil fuels, one may want to weigh the risk of exposure to coal dust, radioactive materials, combustion products, and waste materials associated with coal power versus the risk of radiation exposure from nuclear power production and waste disposal. The regulations concerning radiation exposure limitations are based upon the studies and recommendations of numerous scientific organizations to ensure the health of occupational workers and the public.