URANIUM 257

5. POTENTIAL FOR HUMAN EXPOSURE

5.1 OVERVIEW

Elevated levels of uranium have been identified in at least 54 of the 1,517 current or former EPA National Priorities List (NPL) hazardous waste sites (HazDat 1999). However, the number of sites evaluated for uranium is not known. The distribution of these sites within the United States is shown in Figure 5-1.

Uranium is a naturally occurring radioactive element that is present in nearly all rocks and soils; it has an average concentration in U.S. soils of about 2 pCi/g (3 ppm) (du Preez 1989; NCRP 1984a). Some parts of the United States, particularly the western portion, exhibit higher than average uranium levels due to natural geological formations. Most uranium ores contain between 0.05 and 0.2% uranium, up to 1,000 times the levels normally found in soil (Uranium Institute 1996).

Uranium can undergo oxidation-reduction reactions in the environment or microbial reactions to form complexes with organic matter (Premuzie et al. 1995). The only mechanism for decreasing the radioactivity of uranium is radioactive decay. Since all three of the naturally occurring uranium isotopes have very long half-lives (²³⁴U, 2.4x10⁵ years; ²³⁵U, 7.0x10⁸ years; and ²³⁸U, 4.5x10⁹ years), the rate at which the radioactivity diminishes is very slow (NCRP 1984a). Therefore, the activity of uranium remains essentially unchanged over periods of thousands of years.

Uranium may be redistributed in the environment by both anthropogenic and natural processes. The three primary industrial processes that cause this redistribution are operations associated with the nuclear fuel cycle that include the mining, milling, and processing of uranium ores or uranium end products; the production of phosphate fertilizers for which the phosphorus is extracted from phosphate rocks containing uranium; and the improper disposal of uranium mine tailings (Cottrell et al. 1981; Hart et al. 1986; NCRP 1984a; Yang and Edwards 1984). Essentially no uranium is released from nuclear power plants because of the fuel assembly design and the chemical and physical nature of the uranium oxide fuel. Examples of uranium redistribution by natural processes include activities and processes that move soil and rock, such as resuspension of soils containing uranium through wind and water erosion, volcanic eruptions (Kuroda et al. 1984), operation of coal-burning power plants (coal containing significant quantities of uranium), and construction of roads and dams.

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Figure 5-1. Frequency of NPL Sites with Uranium (U-238) Contamination

Uranium becomes airborne due to direct releases into the air from these processes. Deposition of atmospheric uranium may occur by wet (rain, sleet, or snow) or dry (gravitation or wind turbulence) processes. The rate of uranium deposition is dependent upon such factors as particle size, particle density, particle concentration, wind turbulence, and chemical form. Data are lacking on residence times of particulate uranium in the atmosphere, although UNSCEAR (1988) assumed that it behaves like atmospheric dust, for which meteorological models exist.

Uranium deposited by wet or dry precipitation will be deposited on land or in surface waters. If land deposition occurs, the uranium can be reincorporated into soil, resuspended in the atmosphere (typically factors are around 10⁻⁶), washed from the land into surface water, incorporated into groundwater, or deposited on or adsorbed onto plant roots (little or none enters the plant through leaves or roots). Conditions that increase the rate of formation of soluble complexes and decrease the rate of sorption of labile uranium in soil and sediment enhance the mobility of uranium. Significant reactions of uranium in soil are formation of complexes with anions and ligands (e.g., CO₃-2, OH-1) or humic acid, and reduction of U⁺⁶ to U⁺⁴. Other factors that control the mobility of uranium in soil are the oxidation-reduction potential, the pH, and the sorbing characteristics of the sediments and soils (Allard et al. 1979, 1982; Brunskill and Wilkinson 1987; Herczeg et al. 1988; Premuzie et al. 1995).

Uranium in surface water can disperse over large distances to ponds, rivers, and oceans. The transport and dispersion of uranium in surface water and groundwater are affected by adsorption and desorption of uranium on aquatic sediments. As with soil, factors that control mobility of uranium in water include oxidation-reduction potential, pH, and sorbing characteristics of sediments and the suspended solids in the water (Brunskill and Wilkinson 1987; Swanson 1985). In one study of a stream with low concentrations of inorganics, low pH, and high concentrations of dissolved organic matter, the concentration of uranium in sediments and suspended solids was several orders of magnitude higher than in the surrounding water because the uranium was adsorbed onto the surface of the sediments and suspended particles (Brunskill and Wilkinson 1987).

The levels of uranium in aquatic organisms decline with each successive trophic level because of very low assimilation efficiencies in higher trophic animals. Bioconcentration factors measured in fish were low (Mahon 1982; Poston 1982; Waite et al. 1988) and were thought to arise from the extraction of uranium from the water or simply from the accumulation of uranium on gill surfaces (Ahsanullah and Williams

1989). In plants, uptake of uranium may be restricted to the root system and may actually represent adsorption to the outer root membrane rather than incorporation into the interior of the root system (Sheppard et al. 1983). Most of this uranium may be removed by washing the vegetable surfaces; cutting away the outer membrane will essentially result in complete removal. No significant translocation of uranium from soil to the aboveground parts of plants has been observed (Van Netten and Morley 1983).

The EPA has established a nationwide network called the Environmental Radiation Ambient Monitoring System (ERAMS) for obtaining data concerning radionuclides, including natural uranium isotopes, in environmental media. Sampling locations for ERAMS were selected to provide optimal population coverage (i.e., located near population centers). Airborne uranium concentrations and precipitation levels of uranium were quite low, in the attocurie/m³ (10⁻³ nanoBg/m³) and 0.006–0.098 picocurie/L (0.0002–0.004 Bg/L/m³) ranges, respectively (EPA 1994). However, both air samples and water samples taken near facilities producing uranium ore or processing uranium were found to be higher, in the pCi/L range (Eadie et al. 1979; Lapham et al. 1989; Laul 1994; NCRP 1984a; Tracy and Meyerhof 1987). The ERAMS reports document ²³⁴U to ²³⁸U concentration ratios in drinking water which deviate from the equilibrium value of unity found in undisturbed crustal rock. Theories proposed to account for this natural phenomenon involve water contact with soil and permeable rock containing uranium. The ²³⁸U atoms transform through ²³⁴Th to ²³⁴U, and any process which removes either of these radionuclides from the solid changes the ²³⁴U to ²³⁸U ratio. ²³⁸U atoms at the solid-liquid interface which emit decay alpha particles inward may experience a kinetic energy recoil sufficient to either tear the ²³⁴Th progeny from the solid or fracture the surficial solid layer making the ²³⁴Th more accessible for the enhanced dissolution that thorium typically experiences relative to uranium in mineral matrices. Either process can enhance the relative ²³⁴U content of the liquid. Should that liquid stabilize in another location and evaporate, the localized solids could show an enhanced ²³⁴U ratio.

A large drinking water study was performed in which data from the National Uranium Resource Evaluation (NURE) program plus data prepared for the EPA (Drury 1981) were compiled for a total of over 90,000 water samples. Domestic water supplies were represented by 28,000 samples and averaged 1.7 pCi/L ($2.5 \mu g/L$) uranium, with a population weighted mean value for finished waters, based on 100 measurements, of 0.8 pCi/L ($1.2 \mu g/L$). Other studies show the population weighted average concentration of uranium in U.S. community drinking water to range from 0.3 to 2.0 pCi/L ($1.4 \mu g/L$)

(Ohanian 1989), while concentrations of uranium from selected drinking water supplies analyzed by EPA laboratories were generally <1 pCi/L $(1.5 \mu g/L)$ (EPA 1985j).

The uranium content of food has been studied extensively; human daily intake has been estimated to range from 0.6 to 1.0 (0.9–1.5 µg/day) pCi/day of natural uranium. Uranium is adsorbed onto the roots of plants; thus unwashed potatoes, radishes, and other root vegetables which retain their surface membrane are a primary source of uranium in the diet. Based on consumption rates, potatoes constitute the highest dietary intake of uranium (EPA 1985). One study showed that the concentration of uranium in plant roots was proportional to the uranium concentration in the soil (NCRP 1984a), while a second study did not support a linear relationship (Mortvedt 1994).

Estimates of uranium intake for drinking water vary widely, but the mean is approximately 0.8 pCi/L uranium. Drinking water intake is in the range of 0.6–1.0 pCi/day (0.9– $1.5 \text{ }\mu\text{g/day}$). Uranium intake from food and water sources is approximately equal (EPA 1991b). Compared with food and drinking water, intake of uranium by the inhalation route is small, with values reported at 0.007 pCi/day ($0.010 \text{ }\mu\text{g/day}$) (Cothern 1987) and at 0.0007 pCi/day ($0.0010 \text{ }\mu\text{g/day}$) (UNSCEAR 1988).

Higher rates of uranium intake have been reported for some populations. The potential for uranium intake is greater for individuals who consume foods grown in areas with elevated concentrations of uranium in the soil, and for individuals with elevated concentrations of uranium in their drinking water (EPA 1985; NCRP 1984a). Workers engaged in the extraction and processing of uranium are occupationally exposed to uranium. Industries where uranium exposure are known to have occurred are uranium mining and milling, uranium conversion and enrichment, uranium fuel fabrication, and uranium weapons production (BEIR IV 1988; Miller 1977; NCRP 1984a; West et al. 1979). Other groups with the potential for exposure due to technologically enhanced natural background radioactivity include populations involved in producing and using phosphate fertilizers, and individuals living and working near fossil fuel plants (Jaworowski and Grzybowska 1977; NCRP 1984a; Tadmor 1986; Weissman et al. 1983).

5.2 RELEASES TO THE ENVIRONMENT

Throughout this chapter, the units used to express concentration or intake of uranium are the same units reported by the authors, and are sometimes followed by converted units using the conversion factor of 0.68 pCi/µg, or other multiplier based on the information provided about the isotopic mixture. In some cases, values are expressed in mass units while in other cases values are expressed as activities. In the case of natural uranium with a fixed abundance of the three isotopes, conversion from one unit to the other is possible using the conversion factor of 0.68 pCi/g (EPA 1991b). Likewise, 1 ppm is equivalent to 1 µg/g and, therefore, to 0.68 pCi/g. Other conversion values have been used, such as 0.72 pCi/µg (EPA 1985b) and 0.67 pCi/µg (NCRP 1984a). These different values are largely accounted for by the periodic refinement of values for uranium isotopic half-lives and relative percentages in crustal rock.

5.2.1 Air

Uranium is introduced into the atmosphere primarily by resuspension of soil, but also by the intentional or accidental release of uranium from mining and milling activities, by uranium processing facilities, or by burning coal.

Natural processes that involve the weathering of crustal rock and soil can change the crustal ratio of uranium isotopes. In some cases, human activities have also altered the normal crustal distribution of naturally occurring radioactive materials, resulting in what has been termed technologically enhanced naturally-occurring radioactive material (TNORM) (NCRP 1984a). No new radioactivity is produced by the enhancement, but uranium, its isotopes, and its progeny are redistributed in such a way that real exposure or the potential for human exposure may increase. A major localized source of enhanced natural uranium can result from mining and milling operations (Table 5-1).

Uranium ore with concentrations of uranium up to 1,000 times the average concentration normally found in soil (NCRP 1984a) is removed from its natural location during open-pit, *in-situ* leach, or underground mining operations. The primary sources of airborne releases from these sources are from the actual mining, from ore crushing and grinding, from high-temperature processes such as calcining or sintering, and from yellowcake drying and packaging at the mill. Ore stockpiles can also be a source of airborne emissions (NCRP 1993).

Production in the front end of the nuclear fuel cycle (uranium mining and milling) has undergone a significant reduction since its peak in the early 1980s (ABMS 1994; EPA 1985a). Currently, mining and milling activities represent a source of minimal uranium release. In the United States in 1994, uranium was mined using *in situ* leaching methods and uranium was recovered as a by-product of the processing of phosphate ore (DOE 1995). In 1995, for the third straight year, no conventional uranium mills, which recover uranium from ores mined from the ground, were in operation in the United States (DOE 1996).

As part of the nuclear fuel cycle, uranium conversion (Stelling 1980), uranium enrichment (MMES 1985), and fuel fabrication facilities also release small amounts of uranium to the atmosphere (Table 5-1). Uranium is converted to uranium hexafluoride (UF₆) prior to enrichment. There is the potential for release of some UF₆ to the atmosphere during the conversion process or from storage of the depleted UF₆. Upon release to the atmosphere, gaseous UF₆ is rapidly hydrolyzed to UO₂F₂ (particulate) and hydrofluoric acid (gas) (Bostick et al. 1985). Uranium is enriched in the United States by the gaseous diffusion process, which produces appropriate enrichments for use in both commercial and government-operated facilities. A large amount of depleted uranium is the by-product of the enrichment process; the depleted uranium is kept in storage for further potential use (DOE 1994) or used in commercial or military applications. In one study, concentrations of uranium in the air near a uranium refinery (crude U refined to highly pure UO₃) were found to be 200 times higher than background concentrations, which are typically low (Tracy and Meyerhof 1987).

In addition to gaseous diffusion operations, the DOE is involved in other activities that release uranium to the atmosphere. These include activities at two national laboratories, sites undergoing remedial actions, weapons facilities, facilities classified as waste management operations (active facilities), or environmental restoration sites (inactive and surplus facilities). DOE facilities are present in many states with operations ranging from small laboratories to large, highly complex processing plants (Figure 5-2).

Another method by which uranium may be introduced into the atmosphere is the natural process of erosion and wind activity. In geographic areas that contain higher levels of uranium in the rocks and soil, such as the western United States, additional natural uranium is introduced into the air. Wind erosion of tailings at uranium mining and milling activities will also result in the resuspension of uranium and uranium progeny (e.g., radium-226 and radon-222) (Bigu et al. 1984; Hans et al. 1979). Approximately 5–10% of the uranium initially present ends up in the mill tailing (Uranium Institute 1996).



Figure 5-2. Major DOE Offices, Facilities, and Laboratories

Operations Offices (O)

Oakland CA Idaho Falls IH Chicago IL Las Vegas NV Albuquerque NM Savannah SC Oak Ridge TN Richland WA

Laboratories/National

Laboratories (*)

Lawrence Berkeley Natl Lab, Berkeley CA Lawrence Livermore Natl Lab, Berkeley CA Stanford Linear Accelerator Menio Park CA Natl Renewable Energy Lab, Golden CO Idaho National Energy & Env Lab IH Fermi Natl Accelerator Lab. Batavia IL Argonne Natl Lab, Argonne IL Ames Laboratory, Ames IA

MIT Bates Lab, Cambridge MA Princeton Plasma Physics Lab NJ Inhalation Toxicology Res Inst., Albuquerque NM Los Alamos Nat Lab, Los Alamos NM

Sandia Nat Labs, Albuquerque NM Livermore CA

Brookhaven Nat Lab, Brookhaven NY Oak Ridge Nat Lab, Oak Ridge TN T Jefferson Natl Accelerator, Newport News VA

Pacific Northwest Nat Lab, Richland WA

Special Purpose Offices (❖)

Energy Technology Eng Center, LA, CA Naval Petroleum Reserves 1&2, Kern Co. CA Naval Oil Shale Reserves 1&3, Rifle CO

Yucca Mountain Project NV

Nat Petroleum Technology Office, Tulsa OK

Federal Energy Technology Centers, Pittsburgh PA, Morgantown WV Oak Ridge Inst for Science &

Education TN

Naval Oil Shale Reserves 2, Vernal UT Naval Petroleum Reserve 3, Casper WY

Facilities (+)

Grand Junction CO Rocky Flats CO Pinellas FL Idaho Falls IH Kansas City Plant Kansas City MO Las Vegas NV Waste Isolation Pilot Plant. Carlsbad NM Fernald Env Management Project, Cincinnati, OH

Miamisburg Environmental Managemnt Project, OH

Savannah River SC

Oak Ridge Reservation, Oak Ridge TN Pantex Plant, Amarillo TX

Hanford WA

Field Offices (#)

Rocky Flats CO Golden CO Ohio (4 sites in OH, 1 site

in New York)

Source: DOE 1999b

Volcanic eruption is another natural phenomenon that may increase the concentration of natural uranium in the air. After the eruption of Mount St. Helens, increased levels of ²³⁸U were observed in rainwater at Fayetteville, Arkansas, due to precipitation of ²³⁸U from the atmosphere (Essien et al. 1985; Kuroda et al. 1984). Other studies indicated that long-lived natural radionuclide (²³²Th, ²²⁶Ra, and ⁴⁰K) content in the ash was comparable to that of crustal material (Fruchter et al. 1980).

Uranium releases occur as a result of phosphate mining for production of phosphorous, which is used in phosphoric acid and phosphate fertilizers (NCRP 1984a). Phosphate rock from Florida, Texas, and southeastern Idaho contains as much as 120 ppm (80 pCi/g) uranium, a concentration sufficiently high to be considered as a commercial source of uranium (NCRP 1975).

Coal also contains variable amounts of uranium and other elements such as sulfur. The amount discharged to the atmosphere depends on the concentration in the coal, the amount burned, the method of combustion, the plant design, and the efficiency of fly ash recovery. Approximately 90% of the coal mass is consumed during combustion. Retained uranium concentrates in the nonvolatile fraction or ash. The uranium concentration of the fly ash, which has been quantified by several investigators (Jaworowski and Grzybowska 1977; Tadmor 1986; Weissman et al. 1983), has been found to be higher than in the original coal (NCRP 1984a), indicating that less than 90% of the uranium is released to the atmosphere. A 550 MWe plant with a coal input of 1.5 million tons/year with a uranium content of approximately 3 tons may release 0.06–0.2 Ci (90–300 kg) of ²³⁸U and ²³⁴U per year (NCRP 1984a), indicating that modern coal power plants release more like 10% of the uranium. A nuclear power plant by comparison releases essentially no uranium.

Raw shale oil is also known to contain uranium, and retorting operations may result in the release of uranium to the environment. Studies indicate that shale oil processing operations may increase atmospheric levels of ²³⁸U and ²³⁴U by a maximum of 0.04 fCi/m³ over background levels of uranium (Gogolak 1985).

Various regulations identified in Chapter 7 are designed to limit human exposure to uranium. 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.

5.2.2 Water

The redistribution of uranium and uranium progeny to both surface water and groundwater occurs primarily from the natural erosion of rock and soil; some redistribution also comes from the mining, milling, and, to a lesser extent, conversion portions of the nuclear fuel cycle (Table 5-1). Contamination of surface water and groundwater by effluents from uranium mining, milling, and production operations has been documented (Brandvold et al. 1981; Eadie and Kaufmann 1977; Hart et al. 1986; Swanson 1985; Yang and Edwards 1984).

Uranium is discharged to surface water and/or groundwater during mining operations. If an open-pit or underground mine extends below the water table, groundwater must be removed to permit mining operations to continue. This is usually accomplished by pumping and discharging excess water into the ground or nearby bodies of water. Since mine water is generally concentrated with uranium, its introduction into surface water bodies may produce measurable increases in uranium levels.

Waste waters from open-pit mines are typically one to two orders of magnitude greater in volume and radioactivity content than waters from shaft or underground mines (AEC 1974). A typical open-pit mine may discharge a million gallons of water daily, giving releases of approximately 0.5 mCi per day or 200 mCi per year, these releases consist of less uranium than other elements like radium and radon (AEC 1974).

Liquid releases from uranium mills have been implicated in contamination of surface water and nearby wells and groundwater (Table 5-1). Contamination of groundwater and surface water can also occur by water erosion of tailings piles (Goode and Wilder 1987; Veska and Eaton 1991; Waite et al. 1988). Since extraction of uranium ore during the milling process averages 90–95% recovery, the primary contaminants from uranium tailings disposal sites are uranium progeny (e.g., radium-226).

Generation of liquid waste from the uranium conversion process (see Table 5-1) is generally small and is handled by placing liquid effluent in lined ponds with sealed bottoms. The pond effluent is chemically neutralized to precipitate out uranium and uranium progeny in pond sludge. Water in the ponds is permitted to evaporate and sludge is disposed of as waste under controlled conditions (AEC 1974).

Liquid discharges containing uranium resulting from uranium enrichment and fuel fabrication are generally quite small (see Table 5-1).

In addition to processes of the nuclear fuel cycle, release of uranium has been detected in surface water adjacent to a radioactive waste disposal site in Massachusetts (Cottrell et al. 1981). ²³⁸U measurements indicated that surface water located adjacent to the waste disposal site had concentrations of up to 155 pCi/L. Additionally, groundwater measurements of ²³⁸U and ²³⁵U at the disposal site were 4,400 pCi/L and 2,400 pCi/L, respectively. These values were elevated compared to values obtained in a study performed for the EPA (Drury 1981). For the EPA study, a total of 35,000 surface water samples from across the United States were analyzed; the average total uranium concentration was 1.1 pCi/L (range 0.01–582 pCi/L). Of these, 28,000 were considered samples of domestic water supplies. In this same study, 55,000 groundwater samples had a total mean uranium concentration of 3.2 pCi/L (range 0.01–635 pCi/L).

²³⁸U has been detected in surface water samples at 10 of 51 hazardous waste sites and in groundwater samples at 23 of 51 hazardous waste sites where uranium has been identified in some environmental component (HazDat 1998). Examples of uranium values in groundwater and surface water include the Uravan site (uranium levels ranged from 1,500 to 16,000 pCi/L) and the tailings pond of the United Nuclear site (uranium concentrations were as high as 3,900 pCi/L). A break in the tailings pond dam in 1979 sent 93 million gallons of tailings liquid into the Rio Puerco (EPA 1988b). The distribution of Superfund NPL sites is shown in Figure 5-1.

5.2.3 Soil

Uranium is a naturally occurring radionuclide that is present in nearly all rocks and soils (soils being derived from erosion of the rocks). The average concentration in U.S. soils is about 2 pCi/g (3 ppm); however, much higher levels are found in areas such as the Colorado Plateau and lands supporting current and previous phosphate mining in Florida, Texas, and South Carolina. Lower concentrations of uranium are found in basic rocks (e.g., basalt, 0.02–0.03 pCi/g), while acidic rocks contain higher uranium concentrations (e.g., sedimentary . 1.0 pCi/g) (NCRP 1984a; Stokinger 1981). The uranium present in the rocks and soil as a natural constituent represents natural background levels.

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Table 5-1. Normalized Uranium Effluent Discharges from Uranium Mining, Milling, Conversion, Enrichment, and Fuel Fabrication

Uranium-238ª	Curies per GWy(e) ^b		
Atmospheric releases			
Mining			
Milling	1.8x10 ⁻²		
Mill tailings	1.9x10⁻⁵		
Conversion	2.0x10 ⁻³		
Enrichment	9.9x10⁻⁴		
Fabrication	2.0x10⁻⁵		
Liquid releases			
Conversion	2.2x10 ⁻²		
Enrichment	9.9x10 ⁻³		
Fabrication	9.9x10 ⁻³		

Source: UN 1982

 $^{^{}a}$ In equilibrium with progeny through U-234 b GWy (e) = gigawatts (10 9 watts) of electricity generated for 1 year

Contamination of the soil can occur either from deposition of uranium originally discharged into the atmosphere, or from waste products discharged directly into or on the ground (e.g., water containing uranium from either underground or open-pit mines). Examples of industrial activities that may result in soil deposition include uranium mining and milling, uranium processing, phosphate mining, heavy metal mining, coal use, and inappropriate waste disposal.

In the process of mining uranium, when the depth of the mine is below the water table (either an open-pit or underground mine), the resulting water is pumped from the mine and often discharged directly to the ground or into surface water. For uranium milling, uranium in the ore is extracted (currently 90–95%, originally >50%) so that wastes from uranium milling contain only low levels of uranium; however, the levels of uranium progeny (e.g., radium) remain essentially unchanged. Uncontrolled erosion of these wastes from open tailings piles not protected from the weather occurred at a Shiprock, New Mexico, uranium mill site, resulting in contamination of the surrounding area (Hans et al. 1979). Uncontrolled erosion also occurred in storage areas such as the St. Louis Airport Storage Site in Missouri (Seelley and Kelmers 1985). Increased levels of uranium, radium, radon, and other decay products of uranium have also been measured around these sites, particularly in the soil. A number of controlled disposal locations on government-owned mill sites exist, but the ones identified involved uncontrolled disposal.

At various facilities that process uranium for defense programs, uranium is released to the atmosphere under controlled conditions, resulting in deposition on the soil and surface waters. Monitoring data from the area surrounding the Fernald Environmental Management Project (formerly the Fernald Feed Materials Production Center) showed that soil contained uranium released from the facility (Stevenson and Hardy 1993).

The uranium content of phosphate rock, a source of phosphorus for fertilizers and phosphoric acid for the chemical industry, ranges from several pCi/g to 130 pCi/g (several \sim 200 µg/g) (Boothe 1977; UNSCEAR 1977, 1982). During milling, much of the uranium content becomes concentrated in slag by-products (Melville et al. 1981). The slag by-products are often used for bedrock in the paving of roads, thus transferring the uranium-rich slag to the soil (Melville et al. 1981; Williams and Berven 1986). Because of the large amounts of phosphate fertilizer produced annually (12–15 million tons), trace amounts of uranium progeny remaining in the fertilizer result in the distribution of about 120 Ci (180 metric tons) per year over U.S. agricultural lands (Kathren 1984).

Combustion of coal is a significant source of enhanced natural radioactivity (especially combustion of coal from the western United States, which contains significantly more uranium than coal from the eastern United States). When coal is burned, some of the radioactivity is released directly to the atmosphere, but a significant fraction is retained in the bottom ash. Enhanced concentrations of uranium have been found on the ground around coal-fired power plants (UNSCEAR 1982).

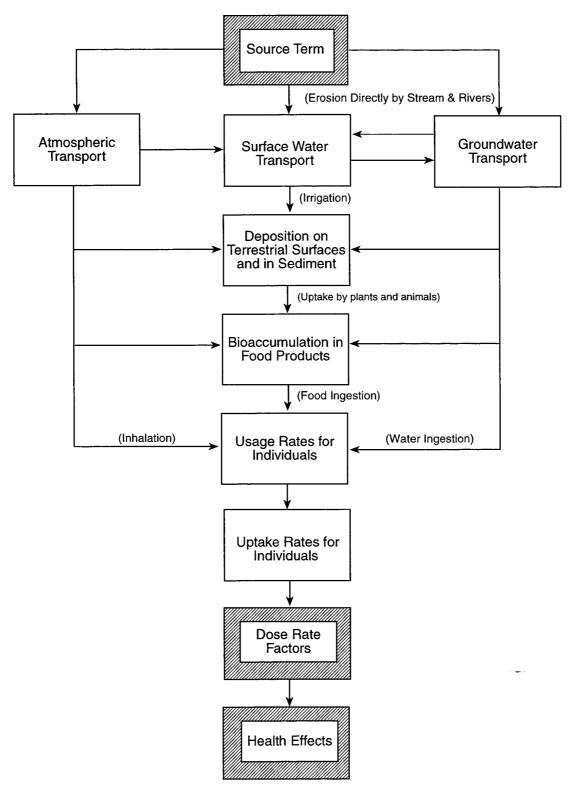
Unauthorized landfill disposal of uranium processing wastes (e.g., Shpack Landfill in Norton, Massachusetts, and the Middlesex Municipal Landfill in Middlesex, New Jersey) has resulted in soil contamination (Bechtel National 1984; Cottrell et al. 1981). Also, elevated uranium concentrations have been measured in soil samples collected at 30 of 51 hazardous waste sites and in sediment samples at 16 of 51 hazardous waste sites (HazDat 1998). The HazDat data includes both Superfund and NPL sites. Elevated concentrations of uranium have been detected in soil, in surface water, in groundwater, or in all three of these environmental media from these sites. In several cases, the uranium concentrations in soils were significantly elevated. For example, uranium concentrations from the Shpack/ALI site were found to be 16,460 pCi/g (24,000 µg/g). At the United States Radium Corporation site (New Jersey), uranium concentrations ranged from 90 to 12,000 pCi/g (130–18,000 µg/g); for the Monticello site (Utah), uranium levels were reported to range from 1 to 24,000 pCi/g (1.5–36,000 µg/g) (HazDat 1998).

5.3 ENVIRONMENTAL FATE

5.3.1 Transport and Partitioning

The components of an ecosystem can be divided into several major compartments (Figure 5-3) (NCRP 1984b). None of the environmental compartments exist as separate entities; they have functional connections or interchanges between them. Figure 5-3 also shows the transport pathways between the released uranium and the environmental compartments as well as the mechanisms that lead to intakes by the population. Initial uranium deposition in a compartment, as well as exchanges between compartments (mobility), are dependent upon numerous factors such as chemical and physical form of the uranium, environmental media, organic material present, oxidation-reduction potential, nature of sorbing materials, and size and composition of sorbing particles.

Figure 5-3. Environmental Pathways for Potential Human Health Effects from Uranium



Source: NCRP 1984b

Natural processes of wind and water erosion, dissolution, precipitation, and volcanic action acting on natural uranium in rock and soil redistribute far more uranium in the total environment than the industries in the nuclear fuel cycle. However, those industries may release large quantities of uranium in specific locations, mainly in the form of solids placed on tailings piles, followed by liquids released to tailings ponds and then airborne releases, both directly from the facilities and by wind erosion of the tailings piles. Although solid releases represent the largest quantity of uranium redistribution, they remain on the facility grounds and are normally inaccessible to the public. It is the airborne (direct and wind erosion on tailings piles) and liquid releases (tailings pond runoff and water erosion of tailings) which most likely represent the important pathways for public exposure (i.e., inhalation and ingestion) if pathways can be completed.

While entrained in the air, particulate uranium represents an inhalation source for humans, the extent of which is dependent upon concentration and particle size. For particulate uranium to be an inhalation hazard to humans, the particulates must be in the size range of 1–10 µm (Bigu and Duport 1992; ICRP 1979). In some cases, the solid tailings have been removed from the site for use as fill or construction material, which can lead to external radiation exposures primarily from the uranium progeny.

Deposition of the atmospheric uranium can occur by dry deposition or wet deposition (Essien et al. 1985). Dry deposition results from gravitational settling and impaction on surfaces exposed to turbulent atmospheric flow. The rate of dry deposition is dependent upon particle size distribution, chemical form, particle density, and degree of air turbulence. Few experimental data on the particle size and residence time of uranium and uranium compounds present in ambient atmospheres are available; however, uranium particles are expected to behave like other particles for which data are available, which show that smaller uranium particles (<5 µm) travel longer distances than larger particles because of their longer residence time in the atmosphere due to their low settling velocity.

The chemical form of the uranium affects the atmospheric residence time. One uranium compound for which there are data regarding residence time and particle size is uranium hexafluoride, a soluble compound, which will hydrolyze in the atmosphere to particulate UO₂F₂•nH₂O and hydrogen fluoride gas (Bostick et al. 1985). In the case of UO₂F₂, although the particles were small (<2.5 µm), its atmospheric residence time was estimated to be only 35 minutes as a result of rapid hydration and agglomeration to larger particles that have faster settling velocities (Bostick et al. 1985).

In wet deposition of airborne contaminants, the uranium is washed from the atmosphere by rain, sleet, snow or other forms of moisture. The rate of wet deposition depends upon particle size and solubility (chemical form).

Uranium thus deposited (dry or wet) will usually reside on land or be deposited on surface waters. If land deposition occurs, the uranium can incorporate into the soil or adhere to plant surfaces, be resuspended in the atmosphere as a result of wind action, or be washed from the land into surface water and groundwater. Resuspension factors are typically quite low (10⁻⁶) and protective against significant exposures, but this may not apply to windy and arid areas. Resuspension into the air can be an inhalation source even after the plume or source has disappeared.

In addition to the migration of dissolved or suspended uranium due to the movement of water in the environment, the transport and dispersion of uranium in surface water and groundwater are affected by adsorption and desorption of the uranium on surface water sediments. On the other hand, migration of uranium in soil and subsoil and uptake in vegetation are usually quite local involving distances from several centimeters to several meters.

In most waters sediments act as a sink for uranium and the uranium concentrations in sediments and suspended solids are several orders of magnitude higher than in surrounding water (Brunskill and Wilkinson 1987; Swanson 1985). Factors that control the mobility of uranium from sediment to the water phase are the oxidation-reduction potential, the pH, the characteristics of complexing agents or ligands, and the nature of sorbing materials in the water. Inorganic or organic ligands that can form soluble complexes with uranium will result in mobilization of the uranium in water. However, the stability of such complexes is dependent on the pH. For example, uranium is likely to be in solution as a carbonate complex in oxygenated water with high alkalinity (Herczeg et al. 1988); however, in acidic waters (pH <6 containing low concentrations of inorganic ions and high concentrations of dissolved organic matter), the uranium is in solution as the soluble organic complex (Brunskill and Wilkinson 1987).

The oxidation-reduction potential of water is important in controlling the mobility of uranium. In anoxic waters where the aquatic environment is reductive, U(VI) will be reduced to U(IV) (e.g., changed from a soluble compound to an insoluble one). The U(IV) will be deposited into the sediment due to the insolubility of the resulting U(IV) salts (Allard et al. 1979; Herczeg et al. 1988). Mobilization and

deposition of uranium as defined by the oxidation-reduction potential of the water has been observed by several investigators (Barnes and Cochran 1993; Shaw et al. 1994). Uranium can also be removed from solution by physical adsorption processes, such as adsorption onto oxides of iron or manganese that occur as coatings on the particles of soil and sediment (Ames et al. 1982).

The mobility of uranium in soil and its vertical transport (leaching) to groundwater depend on properties of the soil such as pH, oxidation-reduction potential, concentration of complexing anions, porosity of the soil, soil particle size, and sorption properties, as well as the amount of water available (Allard et al. 1982; Bibler and Marson 1992). Retention of uranium by the soil may be due to adsorption, chemisorption, ion exchange, or a combination of mechanisms (Allard et al. 1982). Any soil property that alters the sorption mechanism will also alter the mobility of uranium in the soil. The sorption of uranium in most soils is such that it may not leach readily from soil surface to groundwater, particularly in soils containing clay and iron oxide (Sheppard et al. 1987), although other geological materials such as silica, shale, and granite have poor sorption characteristics (Bibler and Marson 1992; Erdal et al. 1979; Silva et al. 1979; Tichnor 1994).

Sorption in most soils attains a maximum when the neutral hydroxy complex of uranium is at a maximum. However, at pH 6 and above, and in the presence of high carbonate or hydroxide concentrations, uranium may form anionic complexes such as $[UO_2(OH)_4]^{-2}$. The mobility of anionic uranium complexes in soil is dependent upon the nature of the soil. For example, the decrease in sorption in soil with little anion-exchange capacity may result in increased mobility; however, increased sorption in soil with high anion-exchange may result in decreased mobility (Allard et al. 1982; Ames et al. 1982; Brookins et al. 1993; Ho and Doern 1985; Hsi and Langmuir 1985; Tichnor 1994).

Other factors also affect the mobility of uranium in soil. A field study performed near an active carbonate leach uranium mill showed that uranium in an alkali matrix can migrate to the groundwater (Dreesen et al. 1982). Uranium mobility may also be increased due to the formation of soluble complexes with chelating agents produced by microorganisms in the soil (Premuzie et al. 1995).

Uranium may be transported to vegetation by air or by water. It can be deposited on the plants themselves by direct deposition or resuspension, or it can adhere to the outer membrane of the plant's root system with potential limited absorption. Similarly, uranium deposited on aquatic plants or water may be adsorbed or taken up from the water. The plants, aquatic or terrestrial, may be eaten directly by humans or consumed

by land or aquatic animals, which provide food for humans. The uptake or bioconcentration of uranium by plants or animals is the mechanism by which uranium in soil, air, and water enters into the food chain of humans.

Numerous factors influence the bioaccumulation of uranium, such as the chemical and physical form of the uranium; the season of the year and other climatic factors such as temperature, age of the organism, specific tissue or organs involved; and the specific characteristics of the local ecosystem, such as total suspended and dissolved solids. Bioconcentration factors for uranium have been measured by several investigators in various aquatic organisms. Mahon (1982) measured bioconcentration factors of 1,576 and 459 in algae and plankton, respectively. Horikoshi et al. (1981) determined bioconcentration factors in several species of bacteria that ranged from 2,794 to 354,000. However, bioconcentration by the bacteria represented adsorption onto the cell surfaces of the bacteria rather than true biological uptake.

Low bioconcentration factors for uranium were observed in fish. The highest bioconcentration factors observed in fillet of rainbow trout (*Salmo gairdneri*), white and finescale suckers (*Castastomus catactomus*), and lake whitefish (*C. clupeaformis*) did not exceed a value of 38 (Mahon 1982; Poston 1982; Swanson 1983, 1985). Ahsanullah and Williams (1989) concluded that the primary source of uranium for crab (*Pachygrapsus laevimanus*) and zebra winkle (*Austrocochlea constricta*) was from water since both fed and starved animals took up uranium at the same rate.

Uranium is transported poorly from soils to plants (Dreesen et al. 1982; Moffett and Tellier 1977). As with aquatic organisms, the uptake of uranium by plants is dependent on the nature of the soils (soil texture and organic content), the pH, and the concentration of uranium in the soil. Greater plant uptake is expected to occur in soils that contain higher levels of available uranium (i.e., less sorption of uranium to soil particles or formation of soluble uranium complexes). Swiss chard grown in sandy soils contained 80 times the levels of uranium found in Swiss chard grown in peat soil (Sheppard et al. 1983). The uptake of uranium by native plants, expressed as plant/soil concentration ratio (CR), grown near a mining and milling complex was 0.8 compared to a CR of 0.09 for plants grown in soil with background levels of uranium (Ibrahim and Wicker 1988). The effect of soil and plant type on CR values has been reviewed by Mortvedt (1994).

Reported CR values for plant/soil interaction vary widely (range, 0.0025–0.81) (Garten 1978; Ibrahim and Wicker 1988; Mortvedt 1994). Although some studies indicate that CR values in plants do not vary linearly with the concentration of uranium in the soil (Mortvedt 1994), other reported studies show a linear relationship between plant content and soil content of uranium (NCRP 1984a). It has been postulated that uranium uptake by plants may be limited to the outer membrane of the root system and may not occur on the interior of the root at all (Van Netten and Morley 1983; Sheppard et al. 1983). However, other investigators have reported the transfer of uranium from soil to the stems and leaves of plants in which the CR decreased in the following order: fruit < leaf < root (Morishima et al. 1977). Because of the higher root sorption of uranium, it has been postulated that consumption of radishes and other root vegetables grown in uranium-containing soils may be a source of human exposure (Van Netten and Morley 1983). Thorough cleansing of the plant exterior, especially if performed in conjunction with removal of the outer membrane, may remove most or all of the uranium.

5.3.2 Transformation and Degradation

5.3.2.1 Air

The presence of uranium and uranium compounds in the atmosphere results from activities associated with uranium mining, milling, processing, and use. There is limited information available regarding the abiotic transformation and degradation of uranium and uranium compounds, except for uranium hexafluoride. Uranium hexafluoride immediately hydrolyzes on contact with moisture in the air to form uranyl fluoride (UO_2F_2) and hydrofluoric acid (HF). Uranyl fluoride is hygroscopic and will absorb moisture from the air, resulting in an increased settling velocity associated with the larger particle size. The half-life of a release of airborne UF_6 is about 35 minutes (Bostick et al. 1985). Uranyl fluoride is a stable oxohalide compound of uranium which is soluble in water, a factor that will increase its mobility in the environment once deposition from the air has occurred.

5.3.2.2 Water

The principal abiotic processes that transform uranium in water are formation of complexes and oxidation-reduction reactions that have been described in Section 5.3.1. In seawater at pH 8.2, it was shown that U(IV) exists as 100% neutral hydroxo complexes, and UO_2^{+2} and U(VI) exist as 100% carbonato

complexes. In freshwater at pH 6, U(IV) was shown to exist as 100% hydroxo complexes, and UO_2^{+2} existed as 12% hydrated complexes, 18% hydroxo complexes, 8% fluoro complexes, and 60% carbonato complexes. In freshwater at pH 9, U(IV) exists as 100% hydroxo complexes, but UO_2^{+2} exists as 100% carbonato complexes (Boniforti 1987).

Oxidation-reduction conditions are important in the geologic transport and deposition of uranium. Oxidized forms of uranium (U[VI]) are relatively soluble and can be leached from the rocks to migrate in the environment. When strong reducing conditions are encountered (e.g., presence of carbonaceous materials or H₂S), precipitation of the soluble uranium will occur.

5.3.2.3 Sediment and Soil

The primary abiotic and biological processes that transform uranium in soil are oxidation-reduction reactions that convert U(VI) (soluble) to U(IV) (insoluble). Reduction of U(VI) to U(IV) can occur as a result of microbial action under anaerobic soil or sediment conditions, thereby reducing the mobility of uranium in its matrix (Barnes and Cochran 1993; Francis et al. 1989). Further abiotic and biological processes that can transform uranium in the environment are the reactions that form complexes with inorganic and organic ligands (see Section 5.3.1).

Certain microorganisms (e.g., *Thiobacillus ferrooxidans*) can facilitate the oxidation of Fe^{+2} to Fe^{+3} . The Fe^{+3} ion, in turn, can convert insoluble uranium dioxide to soluble UO_2^{+2} ions by the following reaction:

$$2Fe^{+3} + UO_2 6 UO_2^{+2} + 2Fe^{+2}$$

This reaction enhances the mobility of uranium in soil from mining and milling wastes (Barnes and Cochran 1993; de Siloniz et al. 1991; Scharer and Ibbotson 1982).

Uranium may be removed from the pore water of sediments under sulfate reduction conditions; microbes may control this process indirectly (Barnes and Cochran 1993).

5.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

In 1973, the EPA established a nationwide network, called ERAMS, for obtaining data in environmental samples. ERAMS consists of a network of sampling stations that provide air, surface and drinking water, and milk samples that the EPA uses to obtain environmental concentrations of radioactive material. The objective of this system is to identify trends in the accumulation of long-lived radionuclides in the environment (EPA 1994). Sampling locations for ERAMS are located near primary population centers to provide optimal population coverage.

The ratio of ²³⁴U to ²³⁸U would be expected to be unity as long as the uranium stays locked inside undisturbed crustal rock in secular equilibrium with its progeny, but measurements show that the ratio is typically different than unity (EPA 1994). This disequilibrium occurs when the rock is disturbed by chemical or physical changes involving water. In the environment, a portion of the ²³⁴U separates from the ²³⁸U by what is theorized to be a physical process (alpha recoil ejection of the ²³²Th decay product from surfaces of soil particles) or a combination of physical and chemical processes (a ²³⁸U transformation at the soil particle surface fractures the surface allowing access for water to dissolve the more soluble ²³⁴Th product) (NCRP 1984a). These processes can change the uranium isotope ratios in air, soil, and water.

5.4.1 Air

For airborne particles collected for the ERAMS program, ²³⁴U, ²³⁵U, and ²³⁸U analyses are performed on semiannually composited air filters collected from continuously operating airborne particulate samplers. Following chemical separation, the uranium is quantified by α-spectroscopy.

Table 5-2 shows the results of monitoring for uranium in airborne particles for the January to June 1993 composites as published in Report 74 (EPA 1994). Results from April through June 1984 are included as well (EPA 1986). The locations of air samples with the highest total uranium concentrations were Las Vegas, Nevada; El Paso, Texas; Ross, Ohio; Lynchburg, Virginia; and Phoenix, Arizona (listed in descending concentrations of airborne total uranium). In all cases, atmospheric levels of total uranium were low, in the attocurie/m³ range. The airborne data show ²³⁴U to ²³⁸U ratios that range from 1.0 to 7.4, many of which are significantly different from the one-to-one ratio found in crustal rock.

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Table 5-2. Uranium in Airborne Particles (Composites)

			aCi/m	1 ³		
	Ap	ril-June 198	4	Janua	ıry-June 199	3
Location	²³⁴ U	²³⁵ U	²³⁸ U	²³⁴ U	²³⁵ U	²³⁸ U
AK, Anchorage				11.9	0.7	9.1
AK, Juneau				12.7	0.7	10.9
AL, Mongomery	12.4ª	1.3ª	11.1 ^a	24.6	0.4	19.3
AR, Little Rock				34.2	2.0	26.7
AZ, Phoenix				51.4	1.8	41.4
CA, Berkeley	26.6ª	2.0 ^a	19.2ª	9.2	1.1	6.9
CA, Los Angeles	43.8ª	2.2ª	33.1ª	29.8	1.8	20.4
CO, Denver	49.7ª	1.0ª	43.0ª	43.2	2.0	40.1
CT, Hartford	15.8ª	0.3ª	11.7 ^a	17.8	0.7	15.6
DE, Wilmington	14.0ª	0.7ª	11.3ª	17.8	1.5	15.2
FL, Miami	17.7ª	0.7ª	18.0ª	29.3	2.5	22.1
HI, Honolulu	11.7ª	0.2a	6.2ª	6.9	0.3	5.6
IA, Iowa City	23.5ª	2.2ª	20.3ª	17.7	0.8	13.3
ID, Boise	35.7ª	4.3ª	27.3ª	24.2	1.8	21.9
IL, Chicago	36.9ª	0.2ª	30.1ª	25.1	2.2	22.6
KS, Topeka	3.9ª	0.2ª	2.7ª	31.6	1.4	29.6
LA, New Orleans	5.5	V. _		17.2	1.1	17.7
ME, Augusta	17.5ª	0.6ª	11.5ª	40.4	1.3	40.1
MI, Lansing	10.9ª	0.9ª	10.4ª	9.7	0.4	9.6
NC, Charlotte	21.8ª	0.6ª	19.7ª	31.7	1.4	25.1
ND, Bismarck	49.1ª	0.4ª	32.7ª	21.6	0.9	18.5
NH, Concord	11.5ª	0.9ª	9.9ª	15.5	0.7	14.6
NJ, Trenton	15.7ª	0.0ª	12.3ª	9.3	0.5	9.4
NM, Santa Fe	22.1ª	0.6ª	18.3ª	28.5	1.2	24.1
NV, Las Vegas	82.2ª	0.9ª	49. 1 °	161 ^b	5.7	98.1
NY, Niagara Falls	37.9ª	1.8ª	49.5°	42.5	4.5	40.2
NY, Syracuse	2.5ª	0.1ª	2.0°	13.0	0.7	10.8
OH, Columbus	45.1ª	2.0ª	40.3ª	17.1	0.5	15.3
OH, Ross	70.1	2.0	+0.0	56.5	3.5	55.7
PA, Harrisburg	22.7ª	1.4 ^a	12.7ª	11.8	0.7	11.4
SD, Pierre	20.3	1.1	17.0	13.7	0.7	12.1
TN, Knoxville	33.2	1.2	17.7	19.9	1.2	17.3
TX, Austin	39.3	1.5	33.2	11.6	0.4	9.8
TX, El Paso	132.7	3.6	105.0	77.2	1.7	65.4
· · · · · · · · · · · · · · · · · · ·	318.6	6.3	16.5	89.3	2.8	12.1
VA, Lynchburg	32.9	1.5	26.6	09.3 13.5	2.8 0.7	10.8
VA, Virginia Beach	35.8	1.0	20.0 31.8	25.3	1.4	21.0
WA, Spokane	აე.ი	1.0	٥١.٥	25.5	1.4	21.0

aCi = attocurie, 10⁻¹⁸ curie

Source: EPA 1986d, 1994a

^a Data from July-September 1984 composites ^b Measured uranium is enriched in U^{234} , as reflected in ration of U^{234} : U^{238} = (82.2/49.1) and (161/98.1) = 1.65 > 1

Uranium in airborne dust appears to result from resuspension of soil and, consequently, airborne dust has the same uranium concentration as the soil particles that produce it. Airborne dust near uranium mining or milling operations would be expected to contain higher than background levels of total uranium and have an isotope ratio the same as crustal rock as long as the surface material from which it originated had not experienced significant weathering by moisture. Some examples of airborne uranium levels near mining and milling operations when the industry was actively producing uranium ore are included below for comparison with EPA values in Table 5-2. The annual average concentration of uranium in ambient air taken near the Jackpile Open Pit mine (New Mexico) was 2.4 fCi/m³ (Eadie et al. 1979), and the concentration of uranium in air measured near a Canadian refinery ranged between 1.3 and 134 fCi/m³ (2–200 ng/m³) with a geometric mean of 13 fCi/m³ (20 ng/m³) (Tracy and Meyerhof 1987). Air samples taken near a uranium mill tailings pile showed a uranium concentration of 1 pCi/m³ (NCRP 1984a). Near the Paducah Gaseous Diffusion Plant in Kentucky, where uranium enrichment is performed, the maximum total air alpha activity in 1979 at one location was 0.7 pCi/m³ (UCC 1980).

5.4.2 Water

Until the early 1980s, uranium in drinking water was not often measured except when contamination was suspected. Welford and Baird (1967) found a concentration of 0.02 pCi/L in New York City tap water. UNSCEAR (1977) reported that tap water usually contains less the 0.03 pCi/L.

A large study was performed in which data from the NURE program plus data prepared for the EPA (Drury 1981) were compiled. Over 90,000 water samples were analyzed for uranium. The total data included approximately 35,000 surface water samples that averaged 1.1 pCi/L and approximately 55,000 groundwater samples that averaged 3.2 pCi/L (NCRP 1984a). The population mean was 0.8 pCi/L, which was higher than the 0.03 pCi/L reported by UNSCEAR (1977). Ohanian (1989) reported a population-weighted average concentration of uranium in U.S. community drinking water ranging from 0.3 to 2.0 pCi/L. Another study showed that the average uranium concentrations in drinking water exceeded 2 pCi/L in South Dakota, Nevada, New Mexico, California, Wyoming, Texas, Arizona, and Oklahoma. States in which the average drinking water uranium levels exceeded 1 pCi/L are shown in Figure 5-4 (Cothern and Lappenbusch 1983; EPA 1985j). In another study based on NURE data, the mean uranium concentration in samples of more than 28,000 domestic water supplies was 1.73 pCi/L, with a median concentration range of 0.1–0.2 pCi/L (Cothern and Lappenbusch 1983). The level of uranium

Figure 5-4. Average Uranium Concentrations in Drinking Water for States Where Concentration Exceeds 1pCi/L



Source: NCRP 1984b

in 2,228 water supplies was 10 pCi/L or more, while in 979 water supplies, the uranium concentrations were 20 pCi/L or greater. Most of these water supplies were in small towns and served less than a few thousand people (Cothern and Lappenbusch 1983;EPA 1985j).

The EPA ERAMS program measures the uranium content of precipitation. Precipitation samples are only collected during the months of March through May since these spring rain months usually contain the year's highest concentrations of uranium. The data for ²³⁴U, ²³⁵U, and ²³⁸U for 1993 are presented in Table 5-3. The precipitation samples with the highest total uranium concentrations were obtained from Berkeley, California; Niagara Falls, New York; Santa Fe, New Mexico; Wilmington, Delaware; Salt Lake City, Utah; Jacksonville, Florida; and Denver, Colorado (listed in descending concentration of total uranium). In all cases, the uranium concentrations were low, confirming that the atmospheric content of airborne uranium is small.

In some surface waters that have been contaminated by waste discharge and in groundwaters from natural uranium-bearing aquifers, the concentrations of uranium may be higher than the average natural background levels for that area. For example, higher levels of uranium have been observed in water from Ambrosia Lake in New Mexico (uranium milling and mining) (Lapham et al. 1989), the agricultural draining and evaporation pond water of the San Joaquin Valley in California (Bradford et al. 1990), and groundwater from Rocky Flats, Colorado (Laul 1994). The concentration of uranium in creek waters that lead to the Ohio River near the Paducah Gaseous Diffusion Plant in Kentucky ranged from <0.7 to 470 pCi/L (1–700 μg/L) (UCC 1980). Mono Lake, a natural alkaline, saline lake in California, contained 185 pCi/L ²³⁸U and 222 pCi/L ²³⁴U during the period 1978–80 (Simpson et al. 1982). Analysis of water from the Colorado River and its tributaries during 1985 and 1986 showed that the levels of total uranium ranged from 3.4 to 60 pCi/L (Stewart et al. 1988). In the United States, the highest concentrations of uranium found in surface water and groundwater used as a source of drinking water were 582 and 653 pCi/L, respectively (Drury 1981).

Discharge of dewatering effluents from underground uranium mines and runoff from uranium mine tailings piles have contaminated surface waters and aquifers in New Mexico with elevated levels of gross alpha activity and uranium (NMHED 1989). The concentration of uranium in mine discharge water in New Mexico was 31,500 µg/L (equivalent to 22,680 pCi/L assuming the uranium content is natural uranium)

Table 5-3. Uranium Analyses of Select Precipitation Composite Samples March–May 1993

	234 _U		²³⁵ U		²³⁸ U	
Location	pCi/L	±2 σ	pCi/L	±2 σ	pCi/L	±2 σ
AK, Anchorage	0.073	0.005	ND	ND	0.047	0.020
AK, Juneau	0.086	0.024	0.008	0.012	0.026	0.020
AL, Montgomery	0.061	0.018	0.005	0.005	0.028	0.012
AR, Little Rock	0.083	0.021	0.001	0.004	0.032	0.013
AZ, Phoenix	0.007	0.006	0.004	0.004	0.009	0.007
CA, Berkeley	0.097	0.023	0.002	0.004	0.043	0.015
CO, Denver	0.085	0.022	0.008	0.007	0.028	0.014
CT, Hartford	0.022	0.010	0.005	0.005	0.007	0.006
DE, Wilmington	0.080	0.027	0.004	0.006	0.047	0.021
FL, Jacksonville	0.098	0.026	0.007	0.007	0.023	0.013
FL, Miami	0.063	0.023	0.010	0.009	0.032	0.015
HI, Honolulu	0.047	0.016	0.006	0.006	0.033	0.014
ID, Boise	0.054	0.018	0.004	0.005	0.038	0.015
ID, Idaho Falls	0.079	0.022	0.005	0.009	0.014	0.013
IL, Chicago	0.056	0.018	0.007	0.007	0.043	0.016
LA, New Orleans	0.053	0.016	0.004	0.004	0.036	0.014
ME, Augusta	0.071	0.021	0.003	0.004	0.034	0.015
MI, Lansing	0.076	0.022	0.002	0.005	0.037	0.015
MN, Minneapolis	0.057	0.018	0.002	0.004	0.046	0.016
MO, Jefferson City	0.063	0.017	0.002	0.003	0.032	0.012
MS, Jackson	0.064	0.020	0.006	0.006	0.030	0.014
NC, Charlotte	0.071	0.023	0.001	0.004	0.029	0.014
NC, Wilmington	0.049	0.022	0.001	0.004	0 . 032	0.016
ND, Bismarck	0.065	0.019	0.003	0.005	0.033	0.014
NH, Concord	0.058	0.018	0.004	0.005	0.014	0.010
NJ, Trenton	0.063	0.020	0.003	0.004	0.016	0.010
NM, Santa Fe	0.086	0.021	0.003	0.004	0.042	0.014
NV, Las Vegas	0.079	0.030	0.015	0.013	0.029	0.018

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Table 5-3. Uranium Analyses of Select Precipitation Composite Samples March–May 1993 (continued)

	224		235		2381 1	
Location	234U		²³⁵ U		238U	
	pCi/L	±2σ	pCi/L	±2 σ	pCi/L	±2σ
NY, Albany	0.068	0.020	0.007	0.006	0.033	0.014
NY, Niagara Falls	0.083	0.024	0.011	0.009	0.038	0.016
NY, Yaphank	0.010	0.007	0.002	0.003	0.006	0.006
OH, Painesville	0.066	0.019	0.005	0.006	0.027	0.012
OH, Toledo	0.092	0.023	0.003	0.005	0.023	0.012
OR, Portland	0.038	0.014	ND	ND	0.022	0.011
PA, Harrisburg	0.070	0.023	0.002	0.010	0.056	0.024
SC, Barnwell	0.042	0.016	0.003	0.004	0.029	0.014
SC, Columbia	0.061	0.019	0.004	0.005	0.036	0.015
TN, Knoxville	0.063	0.019	0.003	0.004	0.034	0.014
TN, Nashville	0.055	0.019	0.009	0.007	0.027	0.013
TX, Austin	0.067	0.018	0.002	0.003	0.024	0.011
UT, Salt Lake City	0.085	0.026	0.006	0.006	0.037	0.017
VA, Lynchburg	0.068	0.024	0.007	0.009	0.042	0.019
WA, Olympia	0.077	0.024	ND	ND	0.031	0.015
WI, Madison	0.008	0.006	ND	ND	0.006	0.007

 σ = Counting error; ND = Not detectable

Source: EPA 1994a

(EPA 1985j). Groundwater from an aquifer adjacent to a uranium mill tailings pile in Falls City, Texas, was also found to have concentrations of uranium above natural background levels (DOE 1994).

The concentrations of ²³⁴U and ²³⁸U in groundwater from Cambrian-Ordovician sandstone aquifers in Illinois range from <0.1 to 8.0 pCi/L (Gilkeson and Cowart 1987). The ratio of the activity of ²³⁴U to ²³⁸U ranged from 2.0 to >40. The lowest ratios were found in unconfined aquifers in primary recharge zones while ratios >20 were found in the confined zones aquifer. It was suggested that glacial recharge in unconfined zones might be responsible for the high ²³⁴U to ²³⁸U ratios (Gilkeson and Cowart 1987). Fifty-five groundwater samples from the Lockatong and Passaic Formation in the Newark Basin in New Jersey were analyzed during 1985–1987. These samples were found to contain 0.1–40 pCi/L total uranium, with a median value of 2.1 pCi/L (Szabo and Zepecza 1987). Uranium concentrations measured in 7 samples of groundwater from the Raymond Basin in California ranged from 5.3 to 43.7 pCi/L (Wiegand et al. 1987).

Water in a private well in Maine, thought to be of geologic origin, was reported to contain as much as 403 µg/L uranium (approximately 270 pCi/L) (Lowry et al. 1987). Elevated levels of uranium measured in waters from private wells in northern and northeastern Nebraska were thought to be due to the upward migration of uranium from bedrock and heavy use of phosphate fertilizers. Uranium values up to 110 pCi/L were measured (NEDH 1989). The concentrations of uranium in U.S. groundwaters were estimated using a conceptual model based on the geochemical and hydrological characteristics of aquifers.

The population-weighted average uranium concentration in groundwaters used as sources of drinking water in all 50 states was found to range from 0.05 to 4.6 pCi/L, with a mean value of 0.55 pCi/L (Longtin 1988). This mean is lower than the population-weighted uranium value for finished waters of 0.8 pCi/L (NCRP 1984a). Some methods which may be suitable for reducing the concentration of uranium in drinking water include lime softening, coagulation/precipitation, and filtering; however, these methods may not efficiently remove the uranium.

5.4.3 Soil

Table 5-4 shows the average concentrations of uranium in several types of rocks and soils (NCRP 1984a). The radioactivity in soils is similar to that in the rocks, usually bedrock, from which it derives. The average soil concentration of ²³⁴U from Table 5-4 is 0.6 pCi/g. Since the activity of ²³⁴U accounts for

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Table 5-4. Uranium In Rocks and Soils

Material	pCi/g ²³⁸ U ^a
Igneous Rocks	0.2–0.3
Basalt (crustal average) Mafic ^b	0.2, 0.3
Salic ^b	1.3, 1.6
Granite (crustal average)	1
Sedimentary Rocks Shale	1
Sandstones	
Clean quartz	<0.3
Dirty quartz	1.0°
Arkose	0.3-0.7 ^c
Beach Sands (unconsolidated)	1
Carbonate Rocks	0.7
Soils	0.6

^aTo obtain the series equilibrium radioactivity for total alpha, beta or approximate gamma emission (excluding bremsstrahlung and X-rays), multiply by 8, 6, or 3, respectively.

^bThe median and mean value are given

Source: NCRP 1975

^cIndicates that the values are not well defined.

approximately one-half of the total activity in natural uranium (see Chapter 3), the value in Table 5-4 may be multiplied by two to obtain the total uranium in soils (approximately 1.2 pCi/g).

There are wide variations from the values presented in the table, particularly in areas where uranium minerals are more concentrated. Concentrations of uranium in Louisiana soils ranged from 2.35 to 3.98 μ g/g (1.6–2.7 pCi/g) (Meriwether et al. 1988), while uranium concentrations in phosphate rock in north and central Florida ranged from 4.5 to 83.4 pCi/g (6.8–124 μ g/g) (EPA 1985j).

Soil samples adjacent to Los Alamos, New Mexico, taken during 1974–1977 contained total uranium in the range of $0.1–5.1~\mu g/g$ ($0.067–3.4~\mu Ci/g$), with a mean concentration of $1.6~\mu Ci/g$ ($2.4~\mu g/g$) (Purtymun et al. 1987). The concentrations of uranium in soils adjacent to the Hanford Fuel Fabrication Facility near Richland, Washington, that were collected during 1978–81 ranged from 0.51~to $3.1~\mu Ci/g$ ($0.76–4.6~\mu g/g$), with a median value of $1.2~\mu Ci/g$ ($1.8~\mu g/g$). The control samples for the Hanford Fuel Fabrication Study contained uranium at concentrations of $0.21–0.86~\mu Ci/g$ ($0.32–1.128~\mu g/g$), with a median value of $0.49~\mu Ci/g$ ($0.73~\mu g/g$) (Price and Kinnison 1982). Uranium in the soil within the property boundary of the Paducah Gaseous Diffusion Plant in Kentucky ranged from 3.3~to $4.8~\mu Ci/g$ ($4.9–7.1~\mu g/g$), whereas off-site samples taken as far as 12 miles away contained uranium at levels of $3.8–6.0~\mu Ci/g$ ($6.4–9.0~\mu g/g$) (UCC 1980). Soil monitoring data from the area surrounding the Feed Material Production Center at Fernald, Ohio, showed that the uranium concentrations within an $8-\mu Ci/g$ area were between $3~\mu Ci/g$ and $23~\mu Ci/g$ ($4.5–34~\mu g/g$) compared to an mean of $2.2~\mu Ci/g$ ($3.3~\mu g/g$) for natural background levels (Stevenson and Hardy 1993). Other investigators have detected uranium levels in surface soils at the Fernald site as high as $50~\mu Ci/g$ (4.19).

5.4.4 Other Environmental Media

Concentrations of uranium have been determined in meat and fish (Table 5-5). The uranium content measured in tissues of cattle herds grazing in pastures close to the Rocky Flats Plant in Colorado was slightly higher than in other cattle, reflecting possible contamination from this source (Smith and Black 1975). The average concentrations of uranium in game fish (surface feeders) collected from a reservoir at locations upstream and downstream from the Los Alamos National Laboratory were 2.9 ng/g dry weight (dw) (0.0019 pCi/g) and 4.9 ng/g dw (0.0033 pCi/g), respectively (Fresquez et al. 1994). The corresponding values in nongame, bottom-feeding fish were 7.9 ng/g dw (0.0058 pCi/g) and 17.7 ng/g dw

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Table 5-5. Concentrations of Uranium in Some Foods

	Uranium concentration	
Type of food	(ng/g raw weight)	Reference
Whole grain products	1.45	NCRP 1984a
Potatoes	2.66–2.92; 15–18	NCRP 1984a; EPA 1985j
Carrots	7.7	EPA 1985j
Root vegetables	0.94–1.20	NCRP 1984a
Cabbage	4.7	EPA 1985j
Meat	0.58–1.32; 20	NCRP 1984a; EPA 1985j
Poultry	0.14-0.42	NCRP 1984a
Beef	14	EPA 1985j
Beef liver	26	EPA 1985j
Beef kidney	70	EPA 1985j
Eggs	0.23; 9.6	NCRP 1984a; EPA 1985j
Dairy products	0.08-0.31	NCRP 1984a
Cow milk	4	EPA 1985j
Milk	1–2	EPA 1985j
Fresh fish	0.43–0.85; 11	NCRP 1984a; EPA 1985j
Shellfish	9.5–31.0	NCRP 1984a
Welsh onion	69	EPA 1985j
Flour	0.25-0.68	NCRP 1984a
Wheat bread	19	EPA 1985j
Baked products	1.32–1.5; 12	NCRP 1984a; EPA 1985j
Polished rice	1.43–6.0; 15	NCRP 1984a; EPA 1985j
Macaroni	0.4–0.63	NCRP 1984a
Tea	5	EPA 1985j
Coffee	6	EPA 1985j
Parsley	60	EPA 1985j
Red pepper	5	EPA 1985j
Mustard	0.2	EPA 1985j
Table salt	40 EPA 1985j	
Canned vegetables	0.09–0.18 NCRP 1984a	
Fruit juices	0.04-0.12 NCRP 1984a	
Canned fruits	0.18-0.29	NCRP 1984a
Fresh fruits	0.71–1.29	NCRP 1984a
Dried beans	1.5–3.67	NCRP 1984a
Fresh vegetables	0.52-0.92 NCRP 1984a	

(0.012 pCi/g), respectively. The concentrations of uranium in fish muscle (dw) from a Canadian lake receiving uranium mill effluents were 7–11 times higher than in fish caught in uncontaminated lakes, but this uranium may have only been attached to the gills (Swanson 1985).

The mean uranium concentration in vegetation from Ambrosia Lake, New Mexico, (site of mining and milling activities) was measured at 0.3 pCi/g dw compared to 4 fCi/g dw for vegetation from a control site (Lapham et al. 1989). Although the concentrations of uranium in muscle from exposed cattle were indistinguishable from uranium levels in muscle from control cattle, levels of uranium in liver and kidney tissues were 4 times higher in exposed cattle than in control cattle, and levels of uranium in femur samples were 13 times higher than in controls, indicating that kidney and liver slightly bioconcentrate uranium while muscle does not (Lapham et al. 1989).

5.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

General population exposure to uranium occurs by ingestion of food and drinking water and by inhalation of air. The pathways are shown in Figure 5-3.

Table 5-5 depicts uranium levels in various types of food in the United States. Measurements of normal levels of dietary ²³⁴U and ²³⁸U indicate that foods consumed contain about 0.3–0.5 pCi/day for each uranium isotope (0.6–1.0 pCi/day [0.9–1.5 μg/day] total uranium) (EPA 1985j; Welford and Baird 1967). Based on consumption rates, root crops such as potatoes, parsnips, turnips, and sweet potatoes contribute approximately 38% of total dietary intake of uranium (EPA 1985j).

Ingestion of food grown in the vicinity of a uranium mill may lead to an intake up to 3 pCi/day uranium (Rayno 1983). Other investigators have estimated a dietary intake of 2.86–4.55 mg/day for individuals living near a uranium mine (Yamamoto et al. 1971).

An alternate method for estimating uranium intake is to measure the daily excretion of uranium in urine and feces. Using this method in a study of 12 subjects in Utah, it was estimated that the average dietary intake for the Salt Lake City population was $4.4\pm0.6~\mu g$, an intake that is higher than that reported for New York City, Chicago, and San Francisco residents (1.3–1.4 μg) (Singh et al. 1990).

Intakes of uranium in food may also increase when certain ceramic glazed dishes are used for serving or storing food (Landa and Councell 1992). Leaching occurs on contact with acidic foods or beverages. Experiments show that when a ceramic glazed plate was kept in contact with a 4% acetic acid solution for 24 hours, the concentration of uranium in the leachate was 31.8 mg/L (Landa and Councell 1992).

Uranium glazed commercial ceramic dinnerware is no longer made and sold because it was determined that the uranium is leachable by acidic foods and beverages (Landa and Councell 1992). Experiments show that when a Fiesta tableware plate was kept in contact with 20 mL of 4% acetic acid solution for 24 hours, the quantity of uranium in the leachate was $600 \mu g$ ($400 \mu g$). Other liquids were much less effective at leaching uranium, with water giving a value over three orders of magnitude lower, and other uranium glazed ceramics were much less leachable (Landa and Councell 1992).

Uranium intakes from food in Japanese diets from two control areas ranged from 0.86 to 1.02 μg/day (Yamamoto et al. 1971). A more recent study reported a mean value of 0.71 μg/day for Japanese males from 31 prefectures (Shiraishi et al. 1992). Worldwide intake values for uranium have been reported at an average of 1 pCi/day (1.5 μg/day) (range 0.6–3.2 pCi/day [0.9–4.8 μg/day]) (Linsalata 1994).

Concentrations of uranium from selected drinking water supplies in the United States were analyzed by the EPA laboratories and found to be generally <1 pCi/L (EPA 1985j). Based on data obtained from the NURE program plus data prepared for the EPA (Drury 1981), a population-weighted average of 0.8 pCi/L uranium was determined. In another study, Ohanian (1989) reported population-weighted average concentrations of uranium in U.S. community drinking water ranging from 0.3 to 2.0 pCi/L. Considering an individual water intake of approximately 1.7 L/day, and an average intake of uranium from drinking water of 0.8 pCi/L as reported in the EPA study, the total intake of uranium for an individual from drinking water each day is approximately 1.4 pCi.

Uranium is also taken into the body by the inhalation route. The average daily intake of uranium from inhalation of air has been estimated to range from 0.007 pCi/day (0.010 μ g/day) (Cothern 1987) to 0.0007 pCi/day (0.0010 μ g/day) (UNSCEAR 1988). This value may be somewhat higher for persons living near sources of uranium emission. Glass makers and potters who use uranium-containing enamels may be exposed to small amounts of uranium from handling the powder or from fuming operations in glass making (Rossol 1997). In general, however, exposure to uranium from inhalation is small compared to exposure from food and drinking water.

Measurements of concentrations of uranium have been made in human tissues and body fluids resulting from consumption of food and water and from natural background sources. These are non-occupationally exposed populations. Two longtime residents of Los Alamos, New Mexico (one a smoker and one not) were shown to have uranium tissue concentrations for the skeleton (average $5.8~\mu g/g$ wet weight) and liver (average $0.08~\mu g/kg$) in closer agreement with the Reference Man (Kathren 1997; ICRP 1975) than those reported in New York City residents (Fisenne and Welford 1986). Values of uranium in whole blood measured in New York City residents and Illinois residents averaged $0.14~\mu g/kg$ ($0.09~\mu ci/kg$) and $0.1~\mu g/kg$ ($0.07~\mu ci/kg$), respectively, compared to a mean value worldwide of $0.58~\mu g/kg$ (Fisenne 1988). Mean concentrations of uranium were measured in the organs of persons representing all age groups from different parts of the United States. The uranium values for lungs, liver, kidney, and bone (vertebrae, rib, and skeleton) were $0.5-1.17~\mu g/kg$ ($0.34-0.78~\mu ci/kg$), $0.12-0.33~\mu g/kg$ ($0.08-0.22~\mu ci/kg$), $0.39-1.00~\mu g/kg$ ($0.26-0.67~\mu ci/kg$), and $0.25-1.9~\mu g/kg$ ($0.17-1.3~\mu ci/kg$), respectively (Fisenne et al. 1988; Fisenne and Welford 1986; Singh et al. 1986b). These differences reflect dietary variations.

Workers engaged in the extraction and processing of uranium are occupationally exposed to uranium. Industries where uranium exposures are known to have occurred are uranium mining and milling, uranium conversion and enrichment, uranium fuel fabrication, and nuclear weapons production. Epidemiologic surveys were initiated in the United States as early as 1950 to study the effects of uranium exposure on uranium millers, and similar studies were performed of workers at the Oak Ridge Gaseous Diffusion Plant in Oak Ridge, Tennessee, where uranium conversion and enrichment were performed. Those studies attributed the health decrement to radon progeny and other toxicants and not directly to the uranium (BEIR IV 1988).

5.6 EXPOSURES OF CHILDREN

This section focuses on exposures from conception to maturity at 18 years in humans and briefly considers potential pre-conception exposure to germ cells. Differences from adults in susceptibility to hazardous substances are discussed in Section 2.6, Children's Susceptibility.

Children are not small adults. A child's exposure may differ from an adult's exposure in many ways.

Children drink more fluids, eat more food, and breathe more air per kilogram of body weight, and have a larger skin surface in proportion to their body volume. A child's diet often differs from that of adults. The

developing human's source of nutrition changes with age: from placental nourishment to breast milk or formula to the diet of older children who eat more of certain types of foods than adults. A child's behavior and lifestyle also influence exposure. Children crawl on the floor; they put things in their mouths; they may ingest inappropriate things such as dirt or paint chips; they spend more time outdoors. Children also are closer to the ground, and they do not have the judgement of adults in avoiding hazards (NRC 1993).

Specific information on the exposure of children to uranium is not available. As for adults in the general population, small exposures occur from normal ingestion of food and drinking water and inhaling air. These exposures may be higher in areas with naturally high uranium soil levels or near uranium processing sites and hazardous waste sites containing uranium. A study of uranium content in bone from three age groups (<13, 13–20, 20–25 years old) reported somewhat higher uranium content in the youngest compared to the oldest age group (approximately 1.5–3-fold); however, there were only 2–4 subjects in each group and the results were not statistically significant (Broadway and Strong 1983). No information on uranium levels in amniotic fluid, meconium, cord blood, neonatal blood, or breast milk was located.

At hazardous waste sites, uranium that is found in excess of natural background levels is most likely to be in soil and presents a special hazard for young children. Hand-to-mouth activity and eating contaminated dirt will result in oral exposure to uranium. The hazard in this case depends on the form of uranium present at the waste site. Soluble uranium compounds (e.g., uranyl nitrate) are absorbed by the gastrointestinal tract to a much greater degree than insoluble uranium compounds (e.g., insoluble oxides of uranium), and a large toxicity database in animals supports the higher toxicity of the soluble forms (see Chapter 2). Uranium in soil at non-hazardous waste sites is almost entirely (>99%) in the form of insoluble oxides of uranium which have very low bioavailability.

As for adults, the potential for uranium exposure is greater for children who consume foods grown in areas with elevated concentrations of uranium in the soil and for children with elevated concentrations of uranium in their drinking water (EPA 1985; NCRP 1984a). Other home exposures are unlikely since no household products or products used in crafts, hobbies or cottage industries contain significant amounts of uranium, except in cases where uranium-bearing rocks are used in and around the home for decorative, collection, or construction purposes (ATSDR 1997)

No information is available on whether children differ from adults in their weight-adjusted intake of uranium. The fractional absorption of uranium (as uranyl nitrate and uranyl citrate) by the oral route was higher in neonatal than in adult rats and swine (Sullivan 1980b; Sullivan and Gorham 1982). In a mathematical model developed by the International Commission for Radiological Protection (ICRP) for risk assessment, one of the assumptions is that the fractional absorption of ingested uranium is twice as high in children under the age of 1 year compared to adults.

Uranium exposure to children from parents' work clothes, skin, hair, tools, or other objects from the workplace is possible if the parent uses uranium at work. However, in a comprehensive review of incidents of home contamination by workers (NIOSH 1997), no cases of uranium contamination were described.

As a radionuclide, uranium is potentially genotoxic and thus it is important to know if parental exposure to uranium could affect the developing fetus or germ cells. However, epidemiological studies of workers exposed to uranium show no evidence of genotoxic effects. This is most likely due to the very low specific activity, the low systemic absorption of uranium, and the lack of concentration of uranium in the germ cells. Genotoxic effects to parental germ cells or to a developing fetus are not likely at probable levels of exposure to uranium from the environment or at hazardous waste sites. Some uranium is stored in bone, but it is not known if this uranium is released during pregnancy and lactation, when it could result in exposure to the fetus or infant.

5.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Higher rates of uranium exposure have been reported for some populations. The potential for uranium exposure is greater for individuals who consume foods grown in areas with elevated concentrations of uranium in the soil, and for individuals with elevated concentrations of uranium in their drinking water (EPA 1985; NCRP 1984a). Industries where higher exposures to uranium are known to occur include uranium mining and milling, uranium conversion and enrichment, uranium fuel fabrication, and nuclear weapons production (BEIR IV 1988; Miller 1977; NCRP 1984a; West et al. 1979). Other groups with the potentially higher exposures include persons involved in producing and using phosphate fertilizers and individuals living and working near fossil fuel plants (Jaworowski and Grzybowska 1977; NCRP 1984a; Tadmor 1986; Weissman et al. 1983). Uranium compounds were previously used in dental appliances, and individuals with dental work of this kind have potentially higher exposures (Sairenji et al. 1980).

5.8 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of uranium is available. Where adequate information is not available, ATSDR, in conjunction with NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of uranium.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be prepared.

5.8.1 Identification of Data Needs

Physical and Chemical Properties. Pertinent data on the physical and chemical properties of uranium and uranium compounds are available in the literature.

Production, Import/Export, Use, Release, and Disposal. Data regarding the past and present production (ABMS 1994; EPA 1985a) and import/export volumes (USDOC 1995) for uranium are available. The uses of uranium and uranium compounds are well known (EPA 1985j; Stokinger 1981; USDI 1980). Other than glazed ceramic foodware and decorative items (Landa and Councell 1992) and dental appliances (Sairenji et al. 1980), consumer contact with uranium products is negligible. Since uranium is not covered under SARA, Title III, manufacturers and users are not required to report releases to the EPA. There is a lack of data on the release and disposal of uranium during mining, milling, and chemical processing and its use during fuel cycle operations. The disposal of uranium is governed by the U.S. Nuclear Regulatory Commission (NRC) regulations (10 CFR 61), and releases of uranium to the environment are governed by NRC and EPA regulations (10 CFR 20, Appendix B; 40 CFR 190; 40 CFR 192). Since significant amounts of depleted uranium are used on modern battlefields, it would be useful to have more information on the export of depleted uranium to other nations, the disposal of related wastes in

the United States, and the mass of depleted uranium released to long-distance air transport when projectiles are used against different target types.

Environmental Fate. For solids, there is a need to determine uptake factors into edible portions of plants and not just adherance to the root structure. For the solid-liquid interface, a method is needed to determine method by which ²³⁴U to ²³⁸U ratio deviates from unity such that the EPA ERAMS water sample results indicate disequilibrium. Uranium enters the atmosphere in particulate form from natural sources and from uranium mining, milling, and processing. Dry or wet deposition from the atmosphere to soil and water can occur (Essien et al. 1985). Little experimental data on the particle size and residence time of uranium and uranium compounds present in ambient atmospheres are available. Additional data regarding the measured particle size of uranium compounds in ambient air, settling velocity, and knowledge of the chemical forms and lifetime of the particles in air would be useful. Although recent studies have characterized the oxidation states and chemical forms of some uranium compounds (UO₂ and UO₃) (Dodge and Francis 1994; Wersin et al. 1994), more data identifying the chemical forms of uranium in the environment are needed to better understand the fate and transport of uranium. Since significant amounts of depleted uranium are used on modern battlefields, it would be useful to have more information on the export of depleted uranium to other nations and the disposal of related wastes in the United States, as well as estimates of projectile quantities that aerosolize to a significant extent and associated downwind air contamination levels

Bioavailability from Environmental Media. The absorption and distribution of uranium as a result of inhalation and ingestion exposures have been discussed in Sections 2.3.1 and 2.3.2. However, quantitative data relating to physical/chemical properties such as particle size, chemical form, and degree of absorption with the bioavailability of uranium from inhaled air particles and inhaled and/or ingested soil particles, are lacking. Such studies would be useful in assessing potential public health impact of uranium to people living near a hazardous waste site.

Food Chain Bioaccumulation. Information about uranium bioaccumulation in fish (Mahon 1982; Poston 1982; Swanson 1983; Waite et al. 1988) is available. Also data concerning levels of uranium in various foods (EPA 1985j) are available. These data indicate that uranium does not biomagnify in the food chain (Ahsanullah and Williams 1989; Morishima et al. 1977; Swanson 1983, 1985). Data on the levels of

uranium in food grown in contaminated areas are limited. Additional data are needed on whether the uptake of uranium in fish is restricted to the gills and how much actually distributes to the meat.

Exposure Levels in Environmental Media. Reliable monitoring data for the levels of uranium in contaminated media at hazardous waste sites are needed so that the information obtained on levels of uranium in the environment can be used in combination of the known body burden with uranium to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

The levels of uranium in airborne particles and precipitation have been monitored since 1973 (EPA 1986, 1994). Data from several large studies of uranium in domestic water supplies are available (Cothern and Lappenbusch 1983; Drury 1981), as are data from studies of groundwater and surface water (NCRP 1984a). The primary source of recent information on the occurrence of uranium in drinking water is the National Inorganics and Radionuclides Survey (NIRS) conducted by EPA (EPA 1991). Some monitoring data are available for uranium-contaminated soils and sediments associated with the nuclear fuel cycle. Better information on background levels in the environment and speciation of uranium in soils and sediments would be useful for determining which species lead to actual public exposure.

Exposure Levels in Humans. Although some data on the levels of uranium in human tissues exposed to natural background levels (food, water, and air) are available, few measurements have been made on the uranium content in human tissues. The principal source of information about occupationally exposed individuals is the U.S. Transuranium and Uranium Registries (USTUR) Tissue Program and database, established to document uranium levels and distribution in human tissues for occupationally exposed workers (PNL 1981). Several major database files are available. The Radiochemical file contains information about radiochemical analysis of tissue donations from occupationally exposed individuals. The Health Physics file contains bioassay and other health physics data. These two databases are regularly updated. The Medical file contains abstracted personal, medical, and clinical data; the Pathology file contains autopsy and pathology information; and the Skeletal Estimate file contains estimated actinide concentrations for unanalyzed half skeletons from donors (USTUR 1999).

Exposures of Children. Children will be exposed to uranium in the same manner as adults in the general population (i.e. ingestion of food and water and inhalation of air). A study of uranium content in bone from three age groups (<13, 13–20, 20–25 years old) reported somewhat higher uranium content in the

youngest compared to the oldest age group (approximately 1.5–3 fold); however, there were only 2–4 subjects in each group and the results were not statistically significant (Broadway and Strong 1983). Since the skeletons of children are growing (higher rate of bone formation), it is possible that a higher fraction of circulating uranium will be deposited in bone than in adults. Further information is needed on bone levels of uranium in children to determine if this is the case. Uranium is found in all soil, and at potentially higher levels at some hazardous waste sites. Since children may have oral exposure to soil through hand-to-mouth activity, bioavailability studies of uranium in soil may be useful to assess the risk of this type of exposure. There is some evidence that neonatal animals absorb uranium in the gastrointestinal tract to a greater extent than adults. Experiments to confirm this finding and to determine how long into maturation a difference exists would help refine risk assessment for uranium exposure in children.

Exposure Registries. A voluntary exposure registry, the U.S. Transuranium and Uranium Registry (USTUR) for occupationally exposed individuals, was established at Richland, Washington, in 1968 as the National Plutonium Registry for investigation of the potential hazards for occupational exposure to uranium. In 1971, additional radiochemistry support was provided by Los Alamos National Laboratory. The United States Uranium Registry was created as a separate entity in 1978, and the two registries operated in parallel until 1987, when a single director was given responsibility for both registries. In 1992, the management and operation of the registries was combined at Washington State University under a grant from the U.S. Department of Energy. The primary goals are to develop information on the distribution and dose of uranium and transuranic elements in humans, providing data for verification or development of radiation protection standards, and to determine and evaluate health effects due to exposure to these radioactive elements.

5.8.2 Ongoing Studies

The Federal Research in Progress database lists ongoing studies about environmental effects of uranium (FEDRIP 1999). These are shown in Table 5-6.

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Table 5-6. Ongoing Studies on Environmental Effects of Uranium

Investigat	Affiliation	Study	Sponsor
Lioy P	New Jersey University of Environmental and Occupational Health Sciences Institute, Piscataway NJ	Refinement of exposure/dose models. Comparison of bioavailability of elemental waste laden soils using in vivo and in vitro analytical methodology	DOE
Davidson P	Stanford University Palo Alto CA	Sorption of heavy metals and radio- nuclides on mineral surfaces in the presence of organic co-contaminants	DOE
Gilliland F	Univ of New Mexico Albuquerque,NM	Improved exposure assessment using ²¹⁰ Pb measurements for epidemiological study	NIH, Natl Center For Res Resources
Chaney RL	Beltsville Agr Res Center Beltsville, MD	Phytoremediation technology for cleanup of radionuclide contaminated sites	U.S. Department of Agriculture, Agricultural Research Service
Harsh JB	Washington State U, Agriculture and Soils, Pullman, WA	Physical chemical state and plant availability of uranium in contaminated mine soil	U.S. Departmentt of Agriculture Cooperative State Res Service
Huang JW	Phytotech, Monmouth Junction, NJ	Phytiremediation of uranium contaminated soils	DOD, Army
Gibson DK	Australian Nuclear Science and Technology Organisation, Palmerston, N Territory, Australia	Water pollution by a uranium mine	DOE
Jeffree RA	Australian Nuclear Science and Technology Organisation, Menai, New South Wales, Australia	Biological impacts of uranium mining	DOE
USTUR	Washington State University	Various studies including determining relationships between uranium concentrations of body organs and between organs and body burdens in healthy and non-healthy individuals	DOE
Beasley T	Oregon State U College of Oceanography	Cycling of transuranic and other long-lived radionuclides in Columbia River	USDOE Energy Research
Barton L	U of New Mexico Albuquerque NM	Mechanisms of metal transformation in bacteria	NIH, Natl Inst of Gen Med Science
Faison B	Oak Ridge National Laboratory	Crada: biosorption of uranium.	USDOE Environmental Restoration and Waste Management

DOD = Department of Defense; DOE = Department of Energy; USTUR = United StatesTransuranium Uranium Registry

Source: FEDRIP 1999; USTUR 1999