6. POTENTIAL FOR HUMAN EXPOSURE

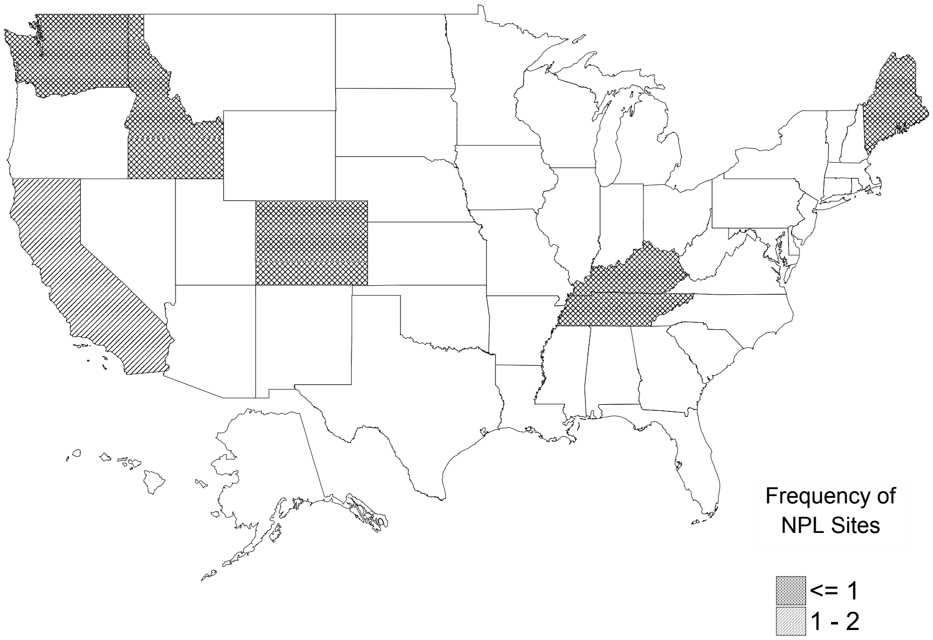
6.1 OVERVIEW

Americium has been identified in at least 8 of the 1,636 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat 2004). However, the number of sites evaluated for americium is not known. The frequency of these sites can be seen in Figure 6-1. Of these sites, all are located within the United States.

Americium is a human-made element. In the environment, ^{241}Am is present as a result of basically three sources: fallout from nuclear explosions, releases from nuclear reactors and reprocessing plants, and production and disposal of smoke detectors by producers and consumers. The majority of the americium present in the environment was the result of atmospheric nuclear weapons testing during the 1950s and 1960s (EPA 2004c). Additionally, there are accidental releases such as the Chernobyl accident, the burn-up of a satellite powered by the aerospace reactor SNAP 9A, and the crash of a B-52 aircraft carrying nuclear weapons in Thule, Greenland. Today, ^{241}Am along with ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239}Pu, and ^{240}Pu, are the isotopes that produce the majority of radiation associated with nuclear fallout (DOE 1997a). Low levels of ^{241}Am may be found in environmental media worldwide from atmospheric nuclear weapons tests; these levels are referred to as fallout background levels. In nuclear reactors, ^{241}Am has been detected in primary coolant water, stack aerosols, and waste water (Rosner et al. 1978). Because ^{241}Am is produced from the decay of ^{241}Pu (half-life=14.4 years), releases of ^{241}Pu will result in an ingrowth of ^{241}Am; clearly, releases of ^{241}Pu and ^{241}Am must be considered collectively. The maximum quantity of ^{241}Am following an accidental release of ^{241}Pu occurs in 70–80 years following the production of the ^{241}Pu (EPA 1976). Consequently, the impact of ^{241}Am from atmospheric nuclear weapons testing will reach its peak in about the year 2035 and then, based on this model, levels will decline according to its combined rate of formation and decay. It should be noted that when the ^{241}Am level reaches its peak, there will still be ^{241}Pu remaining. ^{241}Am is used commercially as an ionization source in smoke detectors and it may be released into the environment from its production, use, and disposal. ^{241}Am is the only americium isotope that is used commercially.

Americium released to the atmosphere will be associated with particulate matter and will be deposited on land or surface water by dry or wet deposition (Essien et al. 1985). Dry deposition results from

Figure 6-1. Frequency of NPL Sites with Americium Contamination



Derived from HazDat 2004

gravitational settling and impaction on surfaces. Wet deposition returns americium to earth in precipitation. Radionuclides resulting from atmospheric weapons tests are often injected into the lower stratosphere, while other atmospheric releases are into the troposphere. The residence time of particles in the atmosphere will depend on their aerodynamic size, as well as the altitude, latitude, season, and hemisphere because of atmospheric circulatory patterns. For example, residence half-times of particles in the lower stratosphere range from 3 to 12 months in polar regions and from 8 to 24 months in equatorial regions, with removal being highest in the spring and lowest in the fall. Half-times for particles in the troposphere are about 1 month. Larger particles will remain in the atmosphere for shorter periods of time and are deposited closer to their emission source, while smaller particles remain in the air for longer periods of time and are deposited farther from their source. In the absence of actual measurements, the deposition of ^{241}Am in a region can be estimated by using the relationship between air concentration and deposition quantities for other isotopes that are attached to particulate matter in air. Using this methodology, the annual deposition of ^{241}Am in New York was estimated to have reached a maximum of 0.010 mCi/km^2 (0.37 MBq/km^2) in 1964 (Bennett 1979). The cumulative ^{241}Am deposition in the year 2000 is estimated to be 0.058 mCi/km^2 (2.1 MBq/km^2). If the ingrowth from ^{241}Pu decay is included, the cumulative ^{241}Am deposited by the year 2000 would be 0.80 mCi/km^2 (30 MBq/km^2). Americium may also be injected into the atmosphere from water surfaces by the action of bubbles scavenging the water column and wave action (McKay et al. 1994; Walker et al. 1986). Similarly, ^{241}Am-contaminated soil may be released into air by wind action.

Americium released to water in effluent, runoff, or atmospheric deposition adsorbs to particulate matter and is rapidly depleted from the water column and deposited in surface sediment (Murray and Avogadro 1979). Adsorption is very high with distribution coefficients between the particulate-associated phase and the dissolved phase in sediment and water of the order of 10^5–10^6 (Davis et al. 1997; NRC 1981; Pattenden and McKay 1994). Aquatic organisms can bioaccumulate americium via intake from the water or ingestion of sediment or organisms lower on the food scale. In crustacean zooplankton, crustaceans, and molluscs, americium is accumulated from water by passive absorption onto exoskeletons (Fisher et al. 1983). Ingested in suspended diatoms or other food, americium largely passes through the gut and is eliminated. Biogenic debris from these animals, such as fecal pellets and discarded molts, facilitates vertical transport of ^{241}Am into the depths and sediment.

If land deposition occurs, americium will sorb to the soil, leach into the subsurface soil, and redistribute by the action of wind, water, or biotic transport. Small mammals inhabiting areas containing radioactive contamination or radioactive waste sites may become contaminated with americium by consuming contaminated soil or plants and disturb ^{241}Am-contaminated soil through their burrowing and excavating activities. In addition, small mammals may be consumed by animals higher in the food chain such as hawks and coyotes, which would add to the dispersal of americium from disposal areas. Since fallout ^{241}Am is accompanied by its precursor, ^{241}Pu, which also leaches into the ground, the amount of ^{241}Am at a depth is a function of the leaching of both the americium and plutonium as well as the decay of these isotopes. Fallout ^{241}Am is predominantly retained to the upper few centimeters of soil where it is associated with organic matter and bound to manganese and iron oxides (Bennett 1976; Bunzl et al. 1995; Vyas and Mistry 1980). However, transuranic radionuclides, including americium, have been found to migrate in groundwater, and have traveled underground from 80 to over 3,000 meters at several national laboratories. Colloidal materials can be mobile in groundwater systems for great distances and are capable of binding and transporting radionuclide contaminants, including americium, in subsurface systems (McCarthy et al. 1998a, 1998b; Penrose et al. 1990).

Americium may be taken up from the soil by plants and translocated to other parts of the plants. ^{241}Am-contaminated dust may also be deposited on the above-ground parts of the plant. Uptake of ^{241}Am from soil is low (Bennett 1979; EPA 1979; Nisbet and Shaw 1994; Romney et al. 1981; Schultz et al. 1976; Zach 1985). Uptake from soil is a function of the chemical species, soil chemistry, plant species, and environmental conditions. Concentration ratios of ^{241}Am in plants to that in soil range over about 2 orders of magnitude depending on crop and soil type. Uptake is higher under acidic conditions and lower in soil with high fractions of organic matter that play an important role in complexing americium, making it less available for uptake. Translocation of transuranic elements taken up through the roots into seeds and fruit is generally very low (Bennett 1979; Schreckhise and Cline 1980). Concentration ratios in seed, grain, and fruit range from 10^{-6} to 10^{-4}, about a factor of 10 lower than in foliage. Animals can accumulate americium by eating contaminated plants as well as by ingesting soil or other animals and by breathing air. Biological uptake in aquatic organisms may depend on factors such as temperature, time after intake, season of the year, and water quality. The bioconcentration of ^{241}Am in fish is low, especially to the edible parts of the fish (DOE 1996). In a study performed in a nuclear waste pond where the levels of ^{241}Am were about 3 orders of magnitude above background levels, concentrations in fish filet were rarely >10 times that of controls (Emery et al. 1981). In marine organisms consumed by humans, uptake is generally highest in mussels where the target organs are mainly the digestive gland, gill, and exoskeleton (Chassard-Bouchaud 1996; Fisher et al. 1996; Hamilton and Clifton 1980). Transfer coefficients have been reported for many of these processes. Available evidence suggests that americium is not biomagnified up the food chain to humans (Bulman 1978; Jaakkola et al. 1977). More recent reports on the biomagnification of americium have not been located.

Exposures of the general population to ^{241}Am via air, water, soil, and food are generally very low; these 'background' levels are a result of fallout from past atmospheric nuclear weapons tests. Since 1973, ^{241}Am air concentrations have been $<10^{-18}$ Ci/m^3 (0.037 μBq/m^3) and are continuing to decline (Bennett 1979). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000) reported that deposition and air concentrations of radionuclides from fallout dropped rapidly once atmospheric testing ceased in 1980. Inhalation exposures for even long-lived transuranic radionuclides became insignificant after 1985. Levels around nuclear power plants are indistinguishable from fallout background (EPRI 1981). ^{241}Am levels in surface seawater of the North Sea and North Atlantic Ocean stayed around 10 Bq/m^3 (270 pCi/m^3) between 1976 and 1988 (Pattenden and McKay 1994), but may be considerably higher near discharges from nuclear facilities. ^{241}Am in soil resulting from cumulative deposition and ingrowth in the New York region should reach a maximum of 0.88 mCi/km^2 (33 MBq/km^2) in the year 2035. In the FDA Total Diet Studies in 1983–1986 and 1987–1992, the concentrations of radionuclides were sufficiently low as to require no specific action or simple monitoring.

The general population may be exposed to elevated levels of ^{241}Am from nuclear accidents or from residing in areas in the proximity of hazardous waste sites where this radionuclide is present. Exposure is generally through the inhalation and ingestion of dust from these sites. Workers involved in producing ionization smoke detectors or other devices containing ^{241}Am, workers at nuclear reactors or Department of Energy (DOE) facilities, and workers who use americium-containing devices (neutron backscatter sources for checking roof leaks and road undermining, and well logging equipment) may also be exposed to higher levels of ^{241}Am. In the environment and most occupational situations, americium is usually encountered in combination with plutonium and curium; exposure to americium alone is unlikely. Elevated levels of ^{241}Am have been identified in at least 8 of the 1,636 current or former EPA NPL hazardous waste sites (HazDat 2004). However, the number of sites evaluated for americium is not known. The distribution of these sites within the United States is shown in Figure 6-1.

6.2 RELEASES TO THE ENVIRONMENT

Throughout this chapter, the units used to express concentration or intake of ^{241}Am are the same units reported by the authors, which may be followed by converted units in parenthesis. Concentrations of ^{241}Am are generally expressed in terms of activity, either in the conventional unit, the curie (Ci) or the SI unit, the becquerel (Bq), where 1 Ci=3.7×10^{10} Bq=0.037 TBq or 1 Bq=2.7×10^{-11} Ci=27 pCi. Activities may be converted into mass units using the specific activity, 3.43 Ci/g. It should be remembered that since ^{241}Pu (half-life=14.4 years) decays into ^{241}Am, over time, inventories of ^{241}Am will be augmented by releases of ^{241}Pu. The ratio of americium to plutonium isotopes is often used to characterize the nature of a radionuclide source.

6.2.1 Air

^{241}Am has been detected in air at 3 of the 1,636 current or former NPL hazardous waste sites where it was detected in some environmental media (HazDat 2004).

^{241}Am is released into the atmosphere primarily by nuclear reactor operations, nuclear accidents, and nuclear weapons testing, but previous releases from plutonium production reactors have ended since they are no longer operational. The ^{241}Am released is augmented by the decay of ^{241}Pu released during these events. The amount of ^{241}Am released is often estimated by the amount deposited on earth. In 1982, UNSCEAR estimated that the integrated deposition density of ^{241}Am in the north temperate zone of the earth was 25 Bq/m^2 (0.68 nCi/m^2) (Suchanek et al. 1996). Since then, the effective deposition of ^{241}Am has grown by 1 Bq/m^2 (0.03 nCi/m^2) per year because of the radioactive decay of 730 Bq/m^2 (20 nCi/m^2) of deposited ^{241}Pu (to approximately 45 Bq/m^2 [1.2 nCi/m^2]).

In atmospheric nuclear weapons tests, radionuclides are often injected into the stratosphere. The amount of ^{241}Am present in the atmosphere from nuclear weapons testing depends on the amount of unfissioned ^{241}Am and ^{241}Pu, as well as lower isotopes of plutonium neutron activated to ^{241}Pu during detonation and injected into the atmosphere, and the transport and residence time of both ^{241}Am and its precursor in the various atmospheric compartments (e.g., stratosphere and troposphere). For a particular weapon's test, this will depend on such factors as the amount of plutonium and americium in the weapon, detonation yield, altitude and latitude of the detonation, and season. High-yield U.S. tests were characterized by very high ratios of ^{241}Pu/^{239,240}Pu, and these would produce relatively large quantities of ^{241}Am (Roos et al. 1994). At this time, the legacy of past atmosphere nuclear weapons tests is a fairly uniform low level radiation background. The peak global activity of ^{241}Am resulting from nuclear weapons testing between 1951 and 1978 and receiving global distribution is estimated to be 130 kCi (4,800 TBq), taking into account the simultaneous production of ^{241}Am from ^{241}Pu and its decay (Bennett 1979). At the same time, it should be realized that the detonation of uranium bombs can produce some americium.

The reactor accident at Chernobyl in April 1986 released radionuclides into the atmosphere, mostly between April 26 and May 6. Estimates of quantities released are based on observations of deposition within 30 km of the reactor. Releases in this area were predominantly highly irradiated fuel particles. It is estimated that the discharge of ^{241}Pu through May 6, 1986 was 5,200 TBq (140 kCi), which amounted to 3% of the reactor content of this radionuclide (Askbrant et al. 1996; Pattenden and McKay 1994). The material was released mainly in the lower troposphere.

Subsequent to the primary events releasing radionuclides into the atmosphere, radionuclides can enter the atmosphere from resuspension of soil particles and forest fires (Lujaniene et al. 1999). Resuspension studies performed with ^{241}Am indicate that the activity to mass ratio is greatest for particles <45 μm in diameter (Cooper et al. 1994). Therefore, the fine, resuspended soil released into the air would have a higher concentration of ^{241}Am than the remaining surface soil. This enhancement factor for resuspended soil in the inhalable fraction (<7 μm) ranged from 5.4 to 32.5 at six sites at Taranaki, a weapons testing range at Maralinga, South Australia (Cooper et al. 1994). Another potential source of release is from windblown soil from LLW disposal sites. While low-level radioactive waste is required to be buried in a manner protective of near-term release, biotic transport by plants and small mammals may transport americium to the soil surface where it is accessible to wind. In order to predict the impact of biotic transport, a model was applied to six composite waste streams containing isotopic concentrations for the western United States prepared by the USNRC to which current and past LLW disposal conditions applied. The containers and wastes were assumed to decompose with a 35-year half-life and active biotic transport was assumed to occur for 100 years following site closure and before institutional control of the site was lost. After 100 years, the activity at the soil surface (assumed to be uniformly mixed in the upper 0.5 m of the entire burial ground) was estimated to be 8.7×10^3 and 2.8×10^5 pCi/m^2 (0.32 and 10 kBq/m^2) for ^{243}Am and ^{241}Am, respectively (Kennedy et al. 1985).

^{241}Am is released to the atmosphere at DOE facilities. Air sampling data were used to estimate effluent release from the Savannah River Site (SRS) from the plant's start-up in 1954 through 1989 (DOE 1991a). From this monitoring, it was estimated that 5.49 mCi (200 MBq) of ^{241}Am was released to the atmosphere between 1977 and 1989. The SRS was a major U.S. nuclear production facility and included five nuclear reactors, a fuel and target fabrication plant, a naval fuel materials facility, two chemical separation plants, a heavy water production plant, and a laboratory. Between 1977 and 1996, the total reported release from the SRS of ^{241}Am to the atmosphere was 0.0061 Ci (230 MBq). Prior to 1977, no specific anaylsis for ^{241}Am was done (DOE 1998). In 1999, the total reported releases to atmosphere from the SRS were

$3.86x10^{-5}$ and $4.28x10^{-6}$ Ci (1.43 and 0.158 MBq) for ^{241}Am and ^{243}Am, respectively (DOE 1999b). ^{241}Am has been identified in air at the Hanford Site, Washington; Idaho National Engineering and Environmental Laboratory (INEEL), Idaho; and Los Alamos National Laboratory, New Mexico. A maximum concentration of 13 MBq/L (350 μCi/L) was measured in 1991 in air at a point source (stack) at Hanford (HazDat 2004). In 1994, atmospheric discharges of ^{241}Am from four areas at the Hanford Site ranged from 0.067 to 66 μCi (2.5–2,400 kBq) (PNL 2000).

6.2.2 Water

^{241}Am has been detected in surface water samples at 2 and in groundwater at 1 of 1,636 NPL hazardous waste sites where americium has been identified in some environmental component (HazDat 2004).

Americium is released into surface water primarily from plutonium production reactors, nuclear fuel reprocessing facilities, or nuclear accidents. It may also be released from radioactive waste storage facilities. Since ^{241}Pu decays into ^{241}Am, ^{241}Am is also released as a result of ^{241}Pu releases. Water sampling data were used to estimate effluent releases from the SRS from the plant's startup in 1954 through 1989 (DOE 1991a). From this monitoring, it was estimated that 290 mCi (11 GBq) of ^{241}Am was released into seepage basins between 1977 and 1989; however, no ^{241}Am was released directly into streams. In 1999, $1.34x10^{-5}$Ci (0.496 MBq) of ^{241}Am was released from the SRS into surface waters (DOE 1999b).

Radionuclides have been released since 1952 at Sellafield, United Kingdom on the Irish Sea by the British Nuclear Fuels Limited (BNFL) reprocessing plant and, before that, by two plutonium-producing nuclear reactors. In 1988, the estimated environmental inventory of ^{241}Am from all previous operations totaled 859 TBq (23.2 kCi), consisting of 537 TBq (14.5 kCi) actually released plus 322 TBq (8.7 kCi) produced from the decay of ^{241}Pu that was also released. Annual discharges of ^{241}Am from Sellafield ranged from 4.5 to 120 TBq/year (120–3,200 Ci/year) between 1964 and 1989, while those for ^{241}Pu ranged from 1 to 2,800 TBq/year (27–76,000 Ci/year) between 1952 and 1989; maximum annual discharges occurred in 1973 for ^{241}Pu and 1974 for ^{241}Am (Pattenden and McKay 1994). The annual liquid effluent discharges from a typical U.K. Magnox Station contained 34.3–121 Bq/L (0.926–3.27 nCi/L) of ^{241}Am for the years 1991–1995 (Ware et al. 1998). These discharges are from the final monitoring and delay tank. (Magnox, an alloy of magnesium oxide and aluminum, has been used as a protective covering or cladding material for fuel elements in British nuclear reactors. The name

"Magnox" is widely used to refer to the early gas cooled because it provides a simple way to uniquely identify the group.)

^{241}Am was identified in surface water at Oak Ridge, Tennessee and Paducah Gaseous Diffusion Plant, Kentucky, as well as in groundwater as an unspecified form at the Hanford, Washington and Rocky Flats, Colorado NPL sites (HazDat 2004). The distribution of NPL sites is shown in Figure 6-1.

6.2.3 Soil

^{241}Am was detected in soil and sediment at 5 and 2 of the 1,636 NPL hazardous waste sites, respectively, where it was detected in some environmental media (HazDat 2004).

As a manufactured element, americium is not naturally present in the environment. Contamination of the soil can occur either from deposition of americium or precursor plutonium originally discharged into the atmosphere, or from waste products discharged directly into or on the ground. Except for the reentry into the atmosphere of the SNAP 9A satellite and the French atmospheric tests in the Mururoa Atoll between 1966 and 1974 that occurred in the southern hemisphere, the main events resulting in the deposition of artificial radionuclides occurred in the northern hemisphere. As a consequence, there is a progressive decline in the levels of artificial radioactivity from northern to southern latitudes. Recent studies of the multi-isotopic radioactive content in ice layers in Antarctica have determined that 0.21 Bq/m^2 (5.7 pCi/m^2) of ^{241}Am was deposited in fallout between 1969 and 1993 (Baeza et al. 1999). Therefore, the mean annual fallout in Antarctica is about 8.3 mBq/m^2 (0.22 pCi/m^2).

The USNRC has projected the activities and volumes of low level radioactive waste from all sources buried at commercial nuclear waste burial sites to the year 2000 using information from the INEEL waste retrieval project and assuming that the waste disposal practices then used would continue into the future. The estimated 20-year decayed ^{241}Am and ^{243}Am concentrations were 380 and 230 pCi/m^3 (14 and 8.5 Bq/m^3), respectively (Kennedy et al. 1985).

The largest releases of ^{241}Am to soil in the United States occur at DOE facilities. The ^{241}Am inventory in surface soil at the Nevada Test Site (NTS) estimated as part of the DOE's Radionuclide Inventory and Distribution Program between 1981 and 1986 was 150 Ci (5,600 GBq) (DOE 1991b). The highest concentration of ^{241}Am was found in the Yucca Flats area. The United States began testing nuclear weapons at the NTS in January 1951. Since then, the NTS has become the primary site for testing nuclear weapons and studying the effect of these weapons on structures and military equipment; >600 nuclear explosions have taken place at this site. At Rocky Flats, ^{241}Am contamination in soil was largely formed from ^{241}Pu in the storage tanks or in the ground (Litaor et al. 1996). Waste oil stored in the tanks was from processing weapons-grade plutonium, which contained 0.58% ^{241}Pu and <10^{-4}% ^{241}Am. Between 1978 and 1981, 48 and 179 kCi (1,800 and 6,600 TBq) of ^{241}Am and ^{241}Pu, respectively, were disposed in the Subsurface Disposal Area of INEEL (Arthur and Janke 1986). No estimates are available of the quantities of these isotopes disposed of between 1952 and 1971 at the Radioactive Waste Disposal Complex of INEEL. ^{241}Am is present at plutonium facilities and at plutonium reclamation facilities where ^{241}Am is routinely separated from plutonium. The Hanford plant was the site of a 1976 column explosion, which resulted in extensive contamination and worker exposure (PNL 2000).

6.3 ENVIRONMENTAL FATE

6.3.1 Transport and Partitioning

Atmospheric Transport. In the atmosphere, americium is associated with particulate matter and the transport of americium in air will therefore be governed by that of its host particles (Bennett 1979). Radionuclides resulting from atmospheric weapons tests are often injected into the lower stratosphere, while other atmospheric releases are into the troposphere. The residence time of particles in the atmosphere will depend on the aerodynamic particle size, altitude, latitude, season, and hemisphere because of atmospheric circulatory patterns. For example, residence half-lives of particles in the lower stratosphere range from 3 to 12 months in polar regions and from 8 to 24 months in equatorial regions, with removal being highest in the spring and lowest in the fall. Half-lives for particles in the troposphere are about 1 month. Larger particles will remain in the atmosphere for short periods of time and are deposited closer to their emission source, while small particles remain in the air for longer periods of time and are deposited further from their source. There are three deposition mechanisms: rain-out caused by droplet formation on aerosols within clouds, wash-out by falling precipitation, and dry deposition of particles onto the ground. Rain-out is the most important deposition process for americium, wash-out occurs relatively slowly and is dependent on the solubility of the pollutant in rainwater, and dry deposition makes an important contribution in areas of low rainfall. Therefore, deposition of ^{241}Am-containing particulate matter will depend on location, season, and meteorological conditions. In the absence of actual measurements, the deposition of ^{241}Am in a region can be estimated by using the relationship between air concentration and deposition quantities for other isotopes that are attached to particulate matter in air. In this way, using the average annual air concentration to annual deposition rate for strontium 90 (^{90}Sr), the deposition rate of ^{241}Am in the New York area was estimated to reach a maximum of 10 μCi/km^2 (37 kBq/km^2) in 1964 and decline thereafter to 0.06 μCi/km^2 (2.2 kBq/km^2) by 1981 (Bennett 1979). This would be expected to result in cumulative ^{241}Am deposit of 58 μCi/km^2 (2.1 MBq/km^2) in the New York region in 2000, assuming no additional atmospheric nuclear testing after 1976. If the ingrowth from ^{241}Pu decay is included, the cumulative ^{241}Am deposit in the year 2000 would be 800 μCi/km^2 (30 MBq/km^2).

Sea-air Transport. Sea-air transport may occur as a result of bubble scavenging in the water column coupled with ejection into the air when bubbles burst on the water surface. Laboratory and field studies of this process showed respective ^{241}Am enrichments in aerosols of 92–580 and 20–40 over that in the bulk seawater (Belot et al. 1982; McKay et al. 1994; Walker et al. 1986). In extensive field studies, Mackay et al. (1994) found clear evidence of localized temporal and spatial variability in the enrichment of americium in sea spray over that in adjacent surf-zone seawater. While the enrichment is typically between 50 and 80, it ranges from 4 to 400. The aerosols produced by the sea spray are deposited nearby. Coastal strips in Cumbria, England have elevated levels of ^{241}Am from sea spray (Pattenden and McKay 1994; Walker et al. 1986). The water in this region of the Irish Sea receives effluent from the BNFL Sellafeld reprocessing plant. Studies of coastal soils in Northern Scotland are also believed to obtain their ^{241}Am contamination from the sea, albeit by a different mechanism. The activity level decreases with distance from the sea and is indistinguishable from background levels more than a few hundred meters inland. The actinide levels in these soils are in excess of those due to past nuclear explosions. The influence of sea spray was ruled out because of the much lower actinide concentrations in the Scottish water compared with that in the Irish Sea. In Northern Scotland, a stable foam or spume is formed along the coast that collects in rocky inlets. It is thought that this spume, which is rich in particulate matter and actinides, builds up in inlets, both overflowing onto the surrounding land and blowing inland on onshore wind (Pattenden and McKay 1994).

Leaching in Soil. After being deposited on soil, ^{241}Am may then leach into the soil. Since fallout of ^{241}Am is accompanied by its precursor, ^{241}Pu, which also leaches into the ground, in addition to being mobilized by colloidal action, the amount of ^{241}Am at a depth is a function of the leaching of both americium and plutonium, the colloidal movement of plutonium, and the decay of these isotopes. A depth profile of fallout americium was performed on samples at a representative site in North Eastham, Massachusetts in 1972 (see Table 6-1). Sixty-two percent of the activity resided in the upper 2 cm of soil and an additional 21% in the 2–4 cm depth (Bennett 1976). A comparison of modeling and experimental

Table 6-1. Concentrations of ^{241}Am in Soil and Sediment

Site/sample type (year)	Depth, cm	Number of samples	Concentration	Units[a]	Type	Reference
Bikini Atoll (not specified)[b], U.S. Nuclear Test Site from 1946 to 1958, including BRAVO explosion on 3/1/54						Robison et al. 1997b
Interior of island	0–5	157	0.26 (0.30)	Bq/g d.w.	Median (mean)	
	5–10	151	0.19 (0.27)	Bq/g d.w.	Median (mean)	
	10–15	127	0.081 (0.18)	Bq/g d.w.	Median (mean)	
	15–25	80	0.026 (0.11)	Bq/g d.w.	Median (mean)	
	25–40	59	0.012 (0.051)	Bq/g d.w.	Median (mean)	
	40–60	23	0.017 (0.035)	Bq/g d.w.	Median (mean)	
	0–40	53	0.11 (0.14)	Bq/g d.w.	Median (mean)	
Village area	0–5	63	0.11 (0.22)	Bq/g d.w.	Median (mean)	
	5–10	62	0.13 (0.20)	Bq/g d.w.	Median (mean)	
	10–15	63	0.12 (0.19)	Bq/g d.w.	Median (mean)	
	15–25	59	0.064 (0.15)	Bq/g d.w.	Median (mean)	
	25–40	52	0.059 (0.13)	Bq/g d.w.	Median (mean)	
	40–60	20	0.012 (0.11)	Bq/g d.w.	Median (mean)	
	0–40	51	0.13 (0.17)	Bq/g d.w.	Median (mean)	
Northern Marshall Islands Radiological Survey (NMIRS)[c], site of U.S. atmospheric weapons testing from 1946 to 1958, including BRAVO explosion on 3/1/54						Robison et al. 1997a
16 Atolls/islands	0–5	182	0.49–569	Bq/g d.w.	Range of means	
	5–10	182	0.26–435	Bq/g d.w.	Range of means	

Table 6-1. Concentrations of ^{241}Am in Soil and Sediment

Site/sample type (year)	Depth, cm	Number of samples	Concentration	Units[a]	Type	Reference
	10–15	182	0.47–207	Bq/g d.w.	Range of means	
	15–25	182	0.15–48	Bq/g d.w.	Range of means	
	25–40	182	0.038–21	Bq/g d.w.	Range of means	
Nuclear Weapons Testing Range, Maralinga, South Australia, site of 12 hydronuclear experiments at Taranaki in 1960, 1961, and 1963 (Vixen B Trials) in which 22.2 kg of plutonium was dispersed, having been ejected 2,500 feet vertically into the atmosphere.						Cooper et al. 1994
Taranaki, 5 sites	0–1	1	0.227–4.40	Bq/g d.w.	Range	
	1–2	1	0.026–0.799	Bq/g d.w.	Range	
	2–3	1	0.005–0.099	Bq/g d.w.	Range	
	3–4	1	0.009–0.022	Bq/g d.w.	Range	
	4–5	1	0.004–0.019	Bq/g d.w.	Range	
Lake Michigan, site 7 km from Grandhaven, Michigan, water depth 67 m (cores and sediment trap at 60 m)						Alberts et al. 1989
Core	0–1	NS	651±118	μBq/g d.w.	Mean±SD	
	1–2	NS	1,147±111	μBq/g d.w.	Mean±SD	
	2–3	NS	1,427±181	μBq/g d.w.	Mean±SD	
	3–4	NS	1,110±148	μBq/g d.w.	Mean±SD	
	4–5	NS	363±67	μBq/g d.w.	Mean±SD	
Suspended sediment, 60 m		NS	555±107	μBq/g d.w.	Mean±SD	
Cruise of RV Scotia, Orkney Islands, and northeast Scottish coast						Pattenden and McKay 1994
Bottom sediment	Surface	NS	0.44–21	mBq/g d.w.	Range	

Table 6-1. Concentrations of ^{241}Am in Soil and Sediment

Site/sample type (year)	Depth, cm	Number of samples	Concentration	Units[a]	Type	Reference
Savannah River Estuary sediment[d]						DOE 1980
Tidal freshwater	0–5	NS	426±150	µBq/g d.w.	Mean±SD	
	5–15	NS	140±70	µBq/g d.w.	Mean±SD	
	15–30	NS	200±70	µBq/g d.w.	Mean±SD	
	30–50	NS	74±30	µBq/g d.w.	Mean±SD	
	50–70	NS	8.5±8.5	µBq/g d.w.	Mean±SD	
Mouth of estuary	0–5	NS	411±63	µBq/g d.w.	Mean±SD	
	5–15	NS	110±85	µBq/g d.w.	Mean±SD	
	15–25	NS	67±20	µBq/g d.w.	Mean±SD	
Soil in northeastern United States from 1970 to 1974, undisturbed soil unless otherwise indicated[e]						Bennett 1979
Brookhaven, New York	0–5	1	16[f]	mBq/km^2		
North Eastham, Massachusetts	0–30	1	17[f]	mBq/km^2		
	0–30	2	18[f]	mBq/km^2	Mean	
North Eastham, Massachusetts-cultivated soil	0–25	5	19[f]	mBq/km^2	Mean	
North Eastham, Massachusetts, background fallout[e]						Bennett 1979
	0–2	NS	8.1±2	mBq/km^2	Mean±SD	
	2–4	NS	2.7±0.3	mBq/km^2	Mean±SD	
	4–6	NS	1.1±0.2	mBq/km^2	Mean±SD	
	6–8	NS	0.59±0.1	mBq/km^2	Mean±SD	
	8–10	NS	0.1±0.07	mBq/km^2	Mean±SD	
	10–12	NS	0.1±0.07	mBq/km^2	Mean±SD	

Table 6-1. Concentrations of ^{241}Am in Soil and Sediment

Site/sample type (year)	Depth, cm	Number of samples	Concentration	Units[a]	Type	Reference
	12–14	NS	0.1±0.04	mBq/km^2	Mean±SD	
	0–30	NS	19±2	mBq/km^2	Mean±SD	

[a] conversion: 1 Bq=2.7x10^{-11} Ci=27 pCi
[b] Continuous monitoring program 1975–1993; decay corrected to 1999
[c] Decay corrected to 1996
[d] Concentration units converted from fCi/g
[e] Concentration units converted from mCi/km^2
[f] Decay corrected to sampling date

d.w. = dry weight; NMIRS = Northern Marshall Islands Radiological Survey; NS = not specified; SD = standard deviation results indicates that americium is slightly less mobile than plutonium in this soil. Leaching studies of surface-deposited ^{241}Am in three Indian soils of widely differing characteristics, namely laterite, medium black, and alluvial, were conducted in soil columns using simulated rain corresponding to the mean annual precipitation (Vyas and Mistry 1980). It was found that 98% of the americium was retained in upper 2 cm of soil; amending the soil with organic matter only slightly reduced its mobility. The greatest retention was obtained with the medium black and laterite soils, where >99% of the ^{241}Am was found in the upper 1 cm. In 1990, Bunzl et al. (1994) investigated the residence time, and consequently the migration rates, of fallout ^{241}Am in a typical undisturbed grassland soil (Alfisol) in Germany. Since ^{241}Am is present from its own deposition and also due to ingrowth from ^{241}Pu, the residence time of plutonium also had to be determined and a compartment model had to be employed. The residence half-time in a particular zone and mean migration rates of americium at various soil depths were (depth, residence half-time, mean migration rate): 0–2 cm, 8 years, 0.3 cm/year; 2–5 cm, 8 years, 0.4 cm/year; 5–10 cm, 10 years, 0.50 cm/year; 10–15 cm, 10 years, 0.5 cm/year; 15–20 cm, 4 years, 1 cm/year; and 20–30 cm, 9 years, 1 cm/year. The migration rates were similar to, but somewhat greater than, those of plutonium. After a period of 30 years, the ^{241}Am concentration on the surface layer was significantly diminished as a result of the migration. Stability constants for Am(III) indicate strong complexation with soil organic matter, and this could explain its increased mobility with depth. In another study, Bunzl et al. (1995) investigated the association of ^{241}Am from global fallout with various soil components in six successive layers of undisturbed grassland in Germany from 0 to 30 cm using a sequential extraction procedure. In this way, the fractions that were readily exchangeable, bound to iron and manganese oxides, bound to organic matter, and bound to mineral (residual fraction) were identified. The association of a chemical with soil components controls their availability for plant uptake and transport. The americium was mainly deposited on the soil surface 30–40 years ago as a result of atmospheric nuclear weapon tests. The total deposition (i.e., sum over all layers) of ^{241}Am was 18±2 Bq/m^2 (490±50 pCi/m^2). On a Bq/kg basis, the data show that the highest activity was contained in the 2–5 cm layer, slightly less was present in the 5–10 cm layer, and activity declined markedly below 10 cm. On a Bq/m^2-cm thickness basis, the maximum activity was in the 5–10 cm layer. The attachment of americium to soil component varied considerably with soil layer. The largest amount of ^{241}Am was attached to organic matter, 18–74%, depending on the soil layer. A substantial fraction, 12–64%, was bound to oxides. In the 5–10 cm layer, the ^{241}Am in the readily exchangeable fraction was a minimum, 1.5%, while that attached to soil minerals was a maximum, 68%. In contrast, very little ^{241}Am (<0.5%) was bound to mineral in the surface (0–2 cm) layer and the deepest soil layers (15–20 and 20–30 cm). Compared with plutonium, much more americium is contained in the exchangeable fraction, a result that has been ascribed to americium's greater hydrolyzability. Americium has also been shown to be largely associated with the high molecular weight organic factions of dissolved organic matter in the soil solution of two grassland soils, a soddy podzolic soil and a peat soil, in the vicinity of the nuclear reactor at Chernobyl. The distribution coefficients for ^{241}Am in these soils were (soil type [depth], K_d in mL/g): soddy podzolic—sod layer (0–2 cm), 1,220; soddy podzolic—mineral layer (2–5 cm), 810; peat (0–2 cm), 2,760; and peat (2–5 cm), 4,550 (Bunzl et al. 1998). While it was similarly shown that the concentration of ^{241}Am was 2–3 times higher in organic matter than in whole sediment from Lake Michigan, organic matter was a very minor constituent of the sediment (<0.5%), so organic matter was associated with a smaller percentage of americium despite its higher concentration (Alberts et al. 1989). The bulk of the ^{241}Am in Lake Michigan was found in the hydrous oxides fraction of both the sediment core samples and the suspended particulate matter.

While laboratory studies predicted very little movement of actinides in subsurface environments, these radionuclides have been found to migrate in groundwater, and have traveled underground from 80 to over 3,000 meters at Los Alamos National Laboratory (LANL), New Mexico and Oak Ridge National Laboratories (ORNL), Tennessee. Colloidal materials, which can be mobile in underground systems over large distances, can bind and transport radionuclide contaminants in subsurface systems (McCarthy et al. 1998a, 1998b; Penrose et al. 1990).

Sorption to Sediment and Particles in Water. When discharged into water, americium rapidly sorbs onto particulate matter in the water column, settles, and adsorbs to bottom sediment (Murray and Avogadro 1979). The distribution coefficient, K_d, of americium between the particulate-associated phase and the dissolved phase is $2.0\text{x}10^6$ (Pattenden and McKay 1994). The sediment-water distribution coefficient, K_d, was also $2.0\text{x}10^6$ for intertidal sediment from the Ravenglass estuary near Sellafield, United Kingdom, in agreement with other investigators (Davis et al. 1997). Equilibrium was established in 30 minutes. Adsorption is greatest for fine-grained particles. A large fraction of the heavier sediment particles will settle out quickly and remain close to the source. Finer particles (and soluble species) will remain suspended for longer periods and be dispersed by currents and tides. The distribution in sediment will be site specific, being largely determined by hydrology and the nature of the underlying sediment. Being a particle-reactive nuclide, like plutonium, americium's distribution in sediment after release will mirror that of plutonium, although its loss from the water column appears to be more rapid. The distribution of americium released from the BNFL reprocessing plant at Sellafield, United Kingdom near the Irish Sea changed very little between 1978 and 1988 (McCartney et al. 1994). Americium in the effluent was entirely as Am(III). In their study of actinide activity in waters entering the northern North Sea, Murray et al. (1978) noted that the activity level of ^{241}Am relative to ^{238,239,240}Pu near the northwest coast of

Scotland was very low, 0.06. Assuming that the contamination was derived from discharges into the Irish Sea 2–2.5 years earlier and 300–500 km to the south where Am-to-Pu alpha activity ratios varied from 0.9 to 2.56, there was a decrease of about 1.7 in the ratio from the point of discharge in spite of the increase in ^{241}Am due to the decay of ^{241}Pu. Similar results have been observed during the transport of actinides discharged from the nuclear fuel reprocessing plant at La Hague, France and in the case of fallout into Mediterranean waters (Murray et al. 1979). These observations have been ascribed to americium's greater adsorption to suspended particulate matter, which then settle out; americium consequently disappears more rapidly from surface water than plutonium. Support for this conclusion was found in another study where the percentage activities associated with suspended materials were 36±8 and 80±7 for plutonium and americium, respectively, <10 km from the point of discharge. Therefore, twice as much americium was associated with suspended matter than plutonium. At a distance of 100 km from the point of discharge, this fraction was still >50%. Core profiles of sediment in the northeast Irish field reveal major subsurface peaks of Sellafield-derived ^{241}Am in both the solid phase and interstitial water (Malcolm et al. 1990b). These peaks were not related to the distribution of organic carbon or iron or magnesium oxides in the sediment or composition of the interstitial water, but rather resembled the discharge of ^{241}Am and plutonium. Studies of radionuclide concentrations in sediment cores from the Ravenglass salt marsh in West Cumbria, United Kingdom, suggest that the concentration profile of ^{241}Am in sediment bear a qualitative resemblance to the Sellafield discharge patterns and to other cores from the eastern Irish Sea coastline. A maximum concentration of 29,300 Bq/kg (0.792 μCi/kg) dry weight was found in the sediment core at 8.9 cm. This maximum concentration for ^{241}Am is in the correct section of the sediment core to correlate with discharges in 1974 based on a sedimentation rate of 4 mm/year (Morris et al. 2000).

Davis et al. (1997) investigated how various environmental factors influenced K_d. The presence of CO_2 gas had a marked effect, increasing the amount of americium leached from sediment by a factor of 40. The increase is attributable to the presence of stable americium-carbonate complexes in seawater and, to a lesser extent, the accompanying decrease in pH to 5.5 as a result of purging with CO_2. Experiments on the effect of pH on K_d suggested that desorption of americium from sediment at low salinity is due to low pH rather than to a drop in ionic strength. Therefore, the effect of low salinity and low pH in an estuary at low tide and when fresh water is backed up by the incoming tide is the remobilization of ^{241}Am. Resorption to sediment entering the estuary from rivers will result in the redistribution of activity.

The disposal of large quantities of radionuclides in Arctic seas has raised concern about the behavior of these chemicals in these waters. Fisher et al. (1999) found that the mean (range) K_d of americium in Kara

Sea sediment was $1x10^6$ ($1x10^5$–$3x10^6$) mL/g, which is in accord with that for temperate coastal sediment, $2x10^6$ ($1x10^5$–$2x10^7$) mL/g. The mean K_d values for americium in two areas in the Kara Sea, an Arctic bay where radionuclides have been dumped, were $4.3x10^4$ and $35x10^4$ for ambient water column samples and $57x10^4$ and $16x10^4$ for bottom sediment samples (Carroll and Harms 1999; Carroll et al. 1999). The mean sediment water distribution function recommended by the International Atomic Energy Agency (IAEA) is $2x10^6$ (range $2x10^5$–$2x10^7$). Therefore, use of the IAEA value in risk assessment modeling would underestimate the americium concentration in seawater. Carroll et al. (1999) stressed that for particle-reactive elements like americium, it is important to use site-specific information for K_d.

Extensive adsorption determinations of ^{241}Am were obtained in freshwater, estuarine, and marine sediment-water systems (NRC 1981). The K_d values for sorption range from 10^4 to 10^6 mL/g, accounting for upward of 95% removal of americium to the solid phase. There were no apparent trends in K_d with salinity. There was an increase in K_d with increasing pH that is typical of hydrolyzable metal ions. The K_d declined with increasing sediment concentration. The mean sediment-water distribution coefficients obtained were (sample type, K_d with standard deviation [SD]$x10^{-4}$): Lake Michigan—three sites, 54.8 (37.3); Clinch River, Tennessee, 73.3 (36.4); Hudson River estuary—three sites, 6.76 (2.43)–29.6 (8.8); Lake Washington, Washington, 18.0 (4.3); Lake Nitinat, Washington—anoxic marine, 25.4 (3.1); Washington Continental Shelf, 10.3 (1.8); Sinclair Inlet, Puget Sound, Washington, 45.4 (12.0); Saanich Inlet, British Columbia—anoxic fjord, 37.1 (8.4); and Skagit Bay, Puget Sound, Washington—estuary—two sites, 26.8 (7.4) and 38.0 (10.6). Sediment-water distribution coefficients for ^{241}Am in various environments obtained from the literature include (site, K_d in mL/g): Lake Michigan, $>50x10^4$; Hanford, U-Pond, $1.9x10^4$ and $8.7x10^4$; and Rocky Flats, Pond A, $0.65x10^4$ (NRC 1981). Laboratory measurements yielded K_d values of 5,000 and 400 in Dutch subsoil and river sand, respectively (NRC 1981).

Despite its high absorptivity, americium has migrated into groundwater and traveled underground at several national laboratories; the mobile forms were colloids and anionic complexes (McCarthy et al. 1998a, 1998b; Penrose et al. 1990; Toran 1994). Artinger et al. (1998) found that increasing concentrations of humic substances reduced americium adsorption on sandy aquifer material, confirming the involvement of colloid-sorbed americium in its subsurface transport. The leachability of radionuclides in waste was assessed in soil column experiments in Carjo loam soil (B horizon) collected in the vicinity of the low level radioactive burial site at Los Alamos. In the case of ^{241}Am, 90% was retained in the silica sand layer placed on top of the soil column, 5% was in the upper 5 cm of the column, and the remainder was distributed at greater depths. In batch experiments, 40 and 49% of soluble ^{241}Am in waste, 0.3% of the total ^{241}Am present, sorbed to Puyo sandy loam and Carjo loam, respectively (Fowler et al. 1981). While americium should strongly adsorb to silicates and mineral oxides of the highly weathered shale saprolite in the unlined disposal trenches used for transuranic waste (TRU) at Oak Ridge National Laboratory, rapid migration in groundwater with little retardation over distances of 80 meters has been observed (McCarthy et al. 1998a, 1998b). This occurred when seasonal fluctuations in the water table permitted groundwater to contact TRU in the trenches. As a result of laboratory studies, the mobilization and transport of americium has been demonstrated to result from complexation of the actinide by natural organic matter. Storm events aid the mobilization by providing the physical connection between the TRU and groundwater and increasing the amount of organic matter in the soil solution and groundwater.

In the absence of natural organic matter, the adsorption coefficient on the site-specific saprolite was 8,900. A similar situation was documented in Los Alamos where nuclear wastes infiltrated a small aquifer. While the movement of ^{241}Am was predicted to be limited to a few meters, the actinide was detectable in monitoring wells as far as 3,390 m downgradient from the discharge (Penrose et al. 1990). Further investigation indicated that the mobile americium was tightly bound to colloidal material between 25 and 450 nm in dimension. The distributions of ^{241}Am activity with colloid particle size (<450 nm) in the well water were (particle size, percent): 25–450 nm, 28%; 5–25 nm, 0.75%; 2–5 nm, 26%; and <2 nm (filtrate), 43%. The unbound americium fraction appeared to be a stable anionic complex of unknown composition. At other sites at Los Alamos, contaminated plumes traveled 30 m in <30 years and 40 m in <20 years (Nyhan et al. 1985; Toran 1994). Migration appeared to be related to the water content of the tuff (a rock composed of the finer kinds of volcanic detritus usually fused together by heat) bed underlying the disposal site, and not simply to fractures in the rock (Nyhan et al. 1985). Investigation of the possible mobility of ^{241}Am in soils around Rocky Flats contaminated by leaking drums under simulated conditions of extreme rain events found that 90% of ^{241}Am activity resided in the upper 12 cm of soil, below which it rapidly decreased; ^{241}Am activity in interstitial water never exceeded 0.4 Bq/L (11 pCi/L) at the deepest sampling layers even in a 100-year rain event (Litaor et al. 1996). However, evidence was found, in line with other investigators, that actinides may move to greater depths in macropores possibly formed by decaying root channels or other biological processes.

The behavior of colloidal americium was studied in an oligotrophic soft water lake used as the source of cooling water for a nuclear power plant in Wales (Orlandini et al. 1990). ^{241}Am concentrations in the dissolved fraction (≤450 nm) were 5.8–18.2 fCi/L (210–673 μBq/L). Most of the americium was distributed in two size ranges, filterable particles (≥450 nm) that contained ~37% of the activity and medium-sized colloids (5–100 nm) that contained ~54% of the activity; only a small fraction (~7%) of americium was 'dissolved' or in true solution, size <1.5 nm (3,000 molecular weight [MW]), where the americium would occur as free ions. The K_d of americium to filterable particles (≥450 nm), the minimum particle size that would settle by gravity, was found to be 470,000; the K_d of americium to colloidal particles was estimated to be about a factor of 20 higher. Lower K_d values obtained for ^{241}Am in dialysis experiments, which restrict the size of particles, compared to those obtained in shaking sorption experiments is a further indication that chemical species affects sorption (NRC 1981).

Distribution of ^{241}Am in a dialysis system containing sediment, phytoplankton, and detrital matter established that a substantial amount of americium accumulated in all three phases both in fresh and marine waters (NRC 1981). The adsorption process was not reversible, and the longer the americium was adsorbed, the more difficult the chemical was to desorb. Appreciable amounts of americium have been shown to adsorb to bacterial cells such as those found in the Waste Isolation Pilot Plant in New Mexico (Francis et al. 1998). There is a potential that americium attached to biocolloids may facilitate its transport from the waste site.

Biotic Transport. Biotic transport can be defined as the actions of plants and animals that result in the transport of a radioactive material or other substance from a waste site to locations where it can enter pathways that may result in exposure to humans. Small mammals inhabiting areas containing radioactive contamination or radioactive waste sites may become contaminated with americium by consuming contaminated soil or plants and may disturb americium-contaminated soil through their burrowing and excavating activities. These animals may therefore affect the distribution of americium within the waste site or transport americium to previously uncontaminated areas. In addition, small mammals may be consumed by animals higher in the food chain, such as hawks and coyotes, which would add to the dispersal of americium from disposal areas. However, results of studies of numerous species of wildlife that inhabit the waste disposal area at INEEL indicate that wildlife do not contribute significantly to the spread of radionuclides to the surrounding environment (Arthur and Janke 1986). What may be more significant is the contribution of biotic intrusion on the soil covering the waste areas and the effect on moisture infiltration rates. A study was conducted of the dispersal of radioactive isotopes by deer mice at the Subsurface Disposal Area (SDA) at INEEL in southwest Idaho (Arthur et al. 1987). These animals, which make up 69% of the small mammal biomass at the SDA, contained significantly higher levels of ^{241}Am than in control areas; the isotope accumulated primarily in the pelt and lung tissue, but also the gastrointestinal tract. Of the 22.8 μCi (0.844 MBq) radionuclide inventory estimated to be contained in deer mice inhabiting the SDA during a 1-year period, about 8.4 μCi (0.31 MBq) was estimated to have been transported off site, of which about 1% was the transuranic radionuclides ^{238}Pu, $^{239+240}$Pu, and ^{241}Am. These transuranic radionuclides also make up about 1% of the 28.8 μCi (1.07 MBq) inventory present in deer mice feces. It was estimated that during 1978–1979, deer mice excavated 12,450 kg of contaminated soil to the surface, which could be further dispersed by wind. Evidence of burrowing activity by earthworms and ants were also found at Rocky Flats, Colorado and other sites (Litaor et al. 1994). Models have been developed to estimate the amount of radioactivity transported to the surface as a result of burrowing activity by animals and plant uptake at a generic LLW disposal site in an arid region (Kennedy et al. 1985). It was found that for ^{243}Am, plants were about 200 times more effective at transporting waste to the surface than burrowing animals. These results indicate that long-term biotic transport may affect the integrity of land burial, redistributing radionuclides such that they become a source of human exposure. Biotic transport will be affected by the oxygenation of soil, as waterlogged and anaerobic soils are less likely to be disturbed by earthworms and other burrowing animals (Livens and Hursthouse 1993).

Uptake by Plants. The uptake of chemicals by plants and their ingestion and bioaccumulation by animals is one mechanism by which a chemical present in soil, air, and water enters into the food chain of humans. Plants may become contaminated with americium as a result of uptake via the root and deposition on the upper part of the plant. Direct deposition occurs during the deposition period, but surface contamination may also occur from resuspension of or direct contact with contaminated soil. Surface deposits may be reduced by weathering or by washing or processing, thereby reducing the amount of americium consumed by humans. Some studies indicate that foliar uptake and subsequent translocation, which is affected by rainfall and size of the deposited particles, may rival the soil-root pathway in magnitude (Cataldo et al. 1980). Translocation of transuranic elements taken up through the roots into seeds and fruit is generally very low (Bennett 1979). Uptake in seeds is significantly lower than for other above ground plant parts, and legumes accumulate about 10 times more than grasses (Schreckhise and Cline 1980).

Plant uptake from soil depends on factors such as the form and availability of the chemical, soil chemistry, plant species, and environmental conditions. Various studies indicate that ^{241}Am uptake by plants is on the order of 10^{-2} to 10^{-6}, about an order of magnitude greater than ^{239}Pu under the same conditions (Bennett 1979; EPA 1979; Romney et al. 1981; Schultz et al. 1976; Zach 1985). Uptake is somewhat greater under acidic conditions and greater to the leaves than to the grain or fruit. Plant uptake experiments with potatoes, peas, and corn were performed using ‘fallout background’ soil in North Eastham, Massachusetts. The ^{241}Am in the plant was compared with the average activity in the upper

30 cm of dried soil, 0.0033 Bq/g (89 fCi/g). The concentration ratios (CRs) (activity per unit wet weight of plant/activity per unit dry weight of soil), also referred to as the transfer coefficient, were 1.7×10^{-3} for husked corn plus cob, 1.0×10^{-3} for peeled potatoes, and $<1.6 \times 10^{-3}$ for shelled peas (Bennett 1979). Flours that were milled from crops of wheat, rye, barley, and oats grown on German soil where radionuclides resulted exclusively from global fallout contained #5.2, 11, 31, and 70 μBq/kg (1.9 fCi/kg) of ^{241}Am, respectively (Bunzl and Kracke 1987). The respective soil to plant transfer factors were $\#0.86 \times 10^{-4}$, 1.5×10^{4}, 4.7×10^{-4}, and 37×10^{-4}. The concentration of ^{241}Am in the bran was a factor of 10–38 higher than in the flour for wheat, rye, and oats. Romney et al. (1981) studied ^{241}Am uptake in crops of peas, soybeans, tomatoes, and wheat grown in seven soils representative of major food production areas of the United States. The CRs in the foliage ranged from 10^{-5} to 10^{-2} and the CRs in the seed, grain, and fruit parts ranged from 10^{-6} to 10^{-4}, about a factor of 10 lower than in the foliage. Uptake varied among the difference soils by a factor of about 100. However, Schulz et al. (1976) obtained less of a variation in uptake of ^{241}Am by wheat in three California soils. The CRs were 6.3×10^{-6}, 16.4×10^{-6}, and 6.1×10^{-6} in Aiken (a slightly acidic forest soil), Yolo (an agricultural soil), and Panoche (a calcareous soil) soil, respectively. The CRs for $^{239, 240}$Pu were approximately the same. Nisbet and Shaw (1994) obtained CRs ranging from 0.04×10^{-4} to 4×10^{-4} for ^{241}Am for carrots, cabbage, and barley grown in artificially-contaminated peat, loam, and sand soils; the CRs were in the order carrot > cabbage > barley. The CRs therefore ranged over 2 orders of magnitude with crop and soil type. Uptake was lowest in peat soil where the organic matter plays an important role in complexing the ions, making them less available for uptake. The CR remained unchanged for plants grown ≥3 years after contamination of the soil. The soil-plant transfer appears to be related to the concentration of ^{241}Am in the soil solution, rather than the bulk soil. While Livens et al. (1994) obtained plant-soil CRs for ^{241}Am in the range of 10^{-5}–10^{-6}, the plant-soil solution CR obtained was much higher, 210–640 L/kg (Bennett 1979). The concentration ratio for ^{241}Am in composite vegatation samples (grasses and herbaceous species) collected in Lady Wood, a coniferous wood located 500 meters northeast of BNFL Sellafield, Cumbria, United Kingdom, ranged from 0.02 to 0.32 (Copplestone et al. 1999). Amendment of soil with lime and organic matter decreases uptake (Adriano 1979; Hoyt and Adriano 1979). This is thought to reduce the solubility or increase americium binding, thereby reducing its availability.

The uptake of radionuclides into plants from soil at contaminated sites at DOE facilities and elsewhere has been the impetus for several studies. Such concerns led to experiments conducted in May 1996 at the Los Alamos National Laboratory (LANL), New Mexico using stream channel sediment from Los Alamos Canyon (LAC). This stream received various amounts of untreated radioactive waste in the 1940s (Fresquez et al. 1998b). Pinto beans, sweet corn, and zucchini were grown in this contaminated soil as well as in soil from seven regional background locations (RBG) between 32 and 96 km around the LANL. The radionuclide content of this latter soil is reflective of global fallout. The ^{241}Am levels in LAC and RBG soils were 0.708 and 0.005 pCi/g dry weight (0.0262 and 0.0002 Bq/g), respectively. The mean ^{241}Am concentrations (dry weight basis) in edible tissue of crops grown on LAC soil and RBG soil were (crop, LAC level [pCi/g (μBq/g)], RBG level [pCi/g (μBq/g)]): beans, 21.5x10^{-5} (7.96), 7.0x10^{-5} (2.6); corn, 27.8x10^{-5} (10.3), 4.1x10^{-5} (1.5); and zucchini, 182.5x10^{-5} (67.5), 27.1x10^{-5} (10.0). The CRs (activity of ^{241}Am in dry plant tissue to that in dry soil) in the edible LAC crop tissues were (crop, concentration ratio): LAC beans, 3.04x10^{-4}; LAC corn, 3.93x10^{-4}; and LAC zucchini, 25.78x10^{-4}. These ratios are much lower than those in composite nonedible crop tissue, which ranged from 48x10^{-4} to 740x10^{-4}. The mean concentration (dry weight) of ^{241}Am found for pinon pine nuts collected around LANL was 9.8x10^{-5} pCi/g (3.6 μBq/g) with a mean concentration ratio to soil of 0.0193 (dry basis) (Fresquez et al. 2000). A garden plot was established on a portion of a drained contaminated pond at the Savannah River Site (SRS) having a mean ^{241}Am concentration of 1.60±0.59 Bq/kg (43.2±15.9 pCi/kg) in soil (0–15 cm). Geometric mean concentration ratios of ^{241}Am (Bq/kg dry plant/Bq/kg dry soil) in the various crops were: corn kernel, 0.0011; corn husk, 0.0016; bush beans, 0.0072; turnip green, 0.0528; and turnip tuber, 0.0053 (Whicker et al. 1999). Uptake for americium was similar to that of curium, but was much higher than for plutonium, thorium, and uranium. Based on the conditions of the study, it was thought that root uptake was the primary mechanism of plant contamination. The concentration ratios for americium are about an order of magnitude higher than a similar study in Tennessee. Since it has been demonstrated that liming decreased uptake, the high uptakes in the SRS study have been ascribed to the acidity of the soil (pH 3.8-4.5 vs. 7–7.5). SRS served as a primary source of reactor-produced nuclear materials, primarily plutonium and tritium, for nearly 4 decades. The CR for ^{241}Am in barley grown on contaminated soil at the Nevada Test Site was of the order of 10^{-4} for vegetative growth and 25–100 times lower for the grain (Schulz et al. 1976). Similar CRs (3x10^{-4}–7x10^{-4}) were obtained for green and root vegetables on land reclaimed from the sea that was contaminated with Sellafield discharges (Green and Wilkins 1995); an earlier study with potatoes, obtained CRs about an order of magnitude less (Popplewell et al. 1984).

Bioconcentration in Aquatic Organisms. Transuranic elements are generally associated with the gut or external surfaces of aquatic organisms. Bioconcentration factors (BCFs) for ^{241}Am measured at a pond at Savannah River, Georgia for fish and other plants and animals are presented in Table 6-2. In a study performed at a nuclear waste pond at Hanford, the maximum concentration of actinides, including ^{241}Am, that would accumulate in the whole fish and fish fillet were measured. In this waste pond, the sediment concentration of ^{241}Am was about 150 pCi/g (5.5 Bq/g), approximately 3 orders of magnitude above background levels (Emery et al. 1981). Both the bluegill and largemouth bass were studied. The

Table 6-2. Bioconcentration Factors for ^{241}Am

	Bioconcentration factor[a]			
Species	Value	Mean	Number of samples	Site
Large-mouth bass, muscle	2,500		1	SRS
Fish muscle		50		Literature value[b]
Fish muscle		25		USNRC[c]
Bullhead catfish, bone	4,200		1	SRS
Macroinvertebrates, larvae	78,000–240,000		2	SRS
Macrophyte, rooted vascular	1,400–21,000		3	SRS
Macrophyte, floating vascular	75,000		1	SRS
Turtle, muscle	5,600		1	SRS
American coot, muscle	650		1	SRS
Bahia grass	0.03–0.12	0.067	6	SRS
	2.3×10^{-7}–0.005			Literature value[b]
		2.5×10^{-4}		USNRC[c]
Pine tree leaves	0.0052–0.021	0.013	2	SRS
	0.0001–0.1			Literature value[b]

[a] The bioconcentration factor is the concentration in organism dry weight/concentration in medium (i.e., water, soil as appropriate).
[b] Values obtained from various literature sources cited in DOE 1996
[c] USNRC values cited in DOE 1996

USNRC = U.S. Nuclear Regulatory Commission; SRS = Savannah River Site

Source: DOE 1996

concentration of ^{241}Am in the water was about 0.2 fCi/mL (7 μBq/mL). The results from the Hanford study indicate that both short- and long-term uptake of americium were low; that uptake was similar for short-term (5 days) and long-term (430 days), experiments; and that direct sediment-to-fish transfer was the primary route for americium uptake. In both species of fish, there were only a few cases where fillet concentrations were >10 times those in controls. The maximum concentration of ^{241}Am obtained in bass and bluegill were 0.030 and 0.028 pCi/g (1.1 and 1.0 mBq/mL) dry weight in fillet and 0.067 and 2.0 pCi/g dry weight (2.5 and 74 mBq/mL) in whole fish (Emery et al. 1981).

In marine organisms, the target organs and tissues of americium bioaccumulation are mainly the digestive gland, gill, and exoskeleton (Chassard-Bouchaud 1996). Uptake in mussels appears to be primarily from seawater rather than ingested sediment (Hamilton and Clifton 1980). Americium taken up by mussels from water was primarily associated with the shell, while that obtained from food was bound to soft tissue (Fisher et al. 1996). Absorption efficiencies from ingested food in mussels were also critical in determining bioaccumulation. Clifton et al. (1983) studied the depuration profiles in mussel populations chronically exposed to radioactive waste discharges from the BNFL reprocessing plant at Sellafield, Cumbria, into the northeast Irish Sea. Apart from clearance from the digestive tract (half-life of 0.9 hours), the half-time of ^{241}Am is 303 days. Essentially all of the ^{241}Am taken up by a euphausiid, a shrimp-like zooplankton, was by passive adsorption onto exoskeletons, so that negligible americium was retained by the animal after molting (Fisher et al. 1983). Wet weight concentration factors were of the order of 100 after a week of exposure, with the BCFs varying linearly with the surface area-to-weight ratio of the organism. Americium was also taken up by feeding on suspensions of diatoms, but there was negligible assimilation and most americium passed through the gut and was defecated.

Uptake and retention of americium by benthic organisms is quite variable (Fowler and Carvalho 1985). In general, filter feeders such as tunicates can clear particles containing ^{241}Am from seawater by filtration through the granchial basket and accumulate small amounts in internal tissue. Mucous feeding guilds of suspension-feeders like crinoids do not efficiently trap americium; uptake appears to occur by complexation or adsorption of dissolved americium to the body wall. Echinoderms and some large crustaceans assimilated americium with their ingested prey, although large differences in half-times and assimilation efficiencies in different groups suggest different feeding-digestion physiologies. Uptake in benthic marine isopods, is to a great extent, by surface adsorption to the exoskeleton and to a lesser extent through the gut, digestive gland, muscle, and haemolymph (Carvalho and Fowler 1985). The half-time in the long-lived compartment was 261 days. Elimination from the internal tissue was more rapid than from the exoskeleton. The fraction assimilated into tissue from food was <5%. The transfer of ^{241}Am from marine sediments contaminated by nuclear tests to benthic polychaetes was <1% (dry weight) (Hamilton et al. 1991).

The biological uptake in aquatic organisms may depend on factors such as temperature, time after intake, season of the year, and water quality. In the brittle star (*Ophiothrix fragilis*), a species that dominates Arctic benthic communities, the initial uptake was 3 times greater at 12 EC than at 2 EC, but depuration was not temperature dependent (Hutchins et al. 1996a). After 14 days, the BCF was 48 at 2 EC and 53 at 12 EC. In the sea star (*Asteruas forbesi*), another Arctic organism, temperature greatly enhanced the retention of americium ingested in food (Hutchins et al. 1996b). The biological half-time of americium ingested in food was 31 days at 12 EC and essentially infinite at 2 EC. Experiments performed on the freshwater snail (*Lymnaea stagnalis*) found higher ^{241}Am uptakes from mesotrophic pond water at acid pH 5.0 than at pH 8.0 (Thiels et al. 1984). In summer, the BCF in the freshwater snail was more than twice as high in acidic media as in alkaline media. In addition, uptake was a function of season, being greater in August than in December. This has been ascribed to changes in metabolic rate with changing photoperiods. Uptake of ^{241}Am by two freshwater benthic invertebrates, *Hyallella* and *Tubifex,* from water was two orders of magnitude higher than from detritus or sand, suggesting that soluble ^{241}Am is the principal route of uptake and that desorption from particulate matter must occur before the substance is available for uptake (Sibley and Stohr 1990). No uptake was observed from labeled National Bureau of Standards (NBS) standard sediment.

Mahon (1982) measured BCFs of 1,576 and 459 in algae and plankton, respectively. Horikoshi et al. (1981) determined BCFs 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.

Biomagnification in the Food Chain. The available evidence suggests that the transport of americium up the food chain to man is slight (Bulman 1978). Americium in the food chain, lichen-reindeer-man, was studied by determining the concentrations ^{241}Am in lichen and reindeer liver in Finnish Lapland (Jaakkola et al. 1977). During 1963–1974, ^{241}Am in lichen ranged from 1 to 40 pCi/kg (0.037-1.48 Bq/kg) dry weight. Levels in six reindeer livers in 1974–1976 contained 0.3–5.4 pCi/kg (0.01-0.20 Bq/kg) fresh weight.

6.3.2 Transformation and Degradation

Americium isotopes are transformed by radioactive decay. However, the half-lives of the principal americium isotopes, ^{241}Am and ^{243}Am, are very long, 432.2 and 7,370 years, respectively (Baum et al. 2002), and there is only a small amount of transformation over a human lifetime. ^{241}Am is formed by the decay of ^{241}Pu (half-life of 14.4 years) (Baum et al. 2002) and this can lead to a significant transformation of that isotope to ^{241}Am in humans, especially for ^{241}Pu that is fixed in the bone.

6.3.2.1 Air

There is limited information available regarding the abiotic transformation of americium in the atmosphere. Oxidation is the most common reaction that occurs in the atmosphere. This would not be expected for americium compounds, which are generally present in the +3 oxidation state under environmental conditions.

6.3.2.2 Water

The principal abiotic processes affecting americium in water are precipitation and complex formation. In natural waters, americium solubility is limited by the formation of hydroxyl-carbonate ($AmOHCO_3$) precipitates. Solubility is unaffected by redox condition. Increased solubility at higher temperatures may be relevant in the environment of radionuclide repositories. In environmental waters, americium occurs in the +3 oxidation state; oxidation-reduction reactions are not significant (Toran 1994).

6.3.2.3 Sediment and Soil

Americium will occur in soil in the trivalent state. The transformations that may occur would involve complexation with inorganic and organic ligands (see Section 6.3.1) and precipitation reactions with anions and other substances present in the soil solution. The ^{241}Am occurring as an ingrowth progeny of ^{241}Pu and trapped in a plutonium matrix will exhibit solubility and biokinetic characteristics of plutonium, rather than americium.

6.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

In 1973, the EPA established The Environmental Radiation Monitoring System (ERAMS), a nationwide network 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 2000b). Sampling locations for ERAMS are located near primary population centers to provide optimal population coverage. ERAMS does not analyze samples for americium, but does measure concentrations of $^{238,239/240}$Pu in air, from which it may be possible to infer levels of ^{241} Pu and ^{241} Am. The most recent ERAMS report containing plutonium data is that covering January–December 1998 composites (EPA 2000b).

6.4.1 Air

The annual average ^{241}Am concentration in surface air in the mid-latitudes resulting from nuclear testing between 1951 and 1978 was estimated using the atmospheric transport model and the amount of ^{241}Am that would have been produced as a result of the testing. The results show a rapid increase in ^{241}Am surface air concentrations, which reached a maximum of 23 aCi/m^3 (1 aCi=10^{-18} Ci) (0.85 µBq/m^3) in 1963–1964. Since 1973, levels have been <1 aCi/m^3 (0.037 µBq/m^3) and are continuing to decline, assuming no additional atmospheric nuclear testing (Bennett 1979). Measurement of these low 1973 levels would have required large sample volumes and exceedingly long sampling periods at common sampling rates. Integral concentration over the entire fallout period is estimated to be 0.12 fCi-year/m^3 (4.4 µBq-year/m^3). Confirmation for these concentration estimates comes from two sources: comparison with plutonium fallout and cumulative deposits of ^{241}Am activity in soil, which is a reflection of integral air concentrations. UNSCEAR (2000) reported that deposition and air concentrations of radionuclides from fallout dropped rapidly once atmospheric testing ceased in 1980. Inhalation exposures for even long-lived transuranic radionuclides became insignificant after 1985.

The Electric Power Research Institute (EPRI 1981) conducted a survey of transuranic radionuclides in the terrestrial environs of nuclear power plants in the United States in 1978–1979. The plants included two pressurized water reactors (PWRs) and two boiling water reactors (BWRs) that were of modern design and had been in operation at least 3 years. The ^{241}Am air concentrations around all of the power plants were extremely low and indistinguishable from fallout background.

Selected radionuclide levels in air were measured at Bragin, 55 km north of Chernobyl for 18 days from April 28, 1986, 2 days after the start of releases from the Chernobyl accident, until May 15. The mean activity concentration of ^{241}Am during this period was 40 μBq/m^3 (1.1 fCi/m^3) and the mean total radionuclide activity was 20 Bq/m^3 (0.54 nCi/m^3) (Knatko et al. 1993). Debris from the Chernobyl accident arrived in Denmark 1.5 days after the explosion. Air samples collected at Roskilde, Denmark on April 27–28, contained a mean air concentration of ^{241}Am of 5.2 μBq/m^3 (0.14 fCi/m^3). In May 1986, the mean concentration was 11 μBq/m^3 (0.30 fCi/m^3) (Aarkrog 1988). Whereas debris from nuclear weapons testing is injected into the stratosphere, debris from Chernobyl was injected into the troposphere. As the mean residence time in the troposphere is 20–40 days, it would appear that the fallout would have decreased to very low levels by the end of 1986. However, from the levels of other radioactive elements, this was not the case. Sequential extraction studies were performed on aerosols collected in Lithuania after dust storms in September 1992 carried radioactive aerosols to the region from contaminated areas of the Ukraine and Belarus. The fraction distribution of ^{241}Am in the aerosol samples was approximately (fraction, percent): organically-bound, 18%; oxide-bound, 10%; acid-soluble, 36%; and residual, 32% (Lujaniene et al. 1999). Little americium was found in the more readily extractable ‘exchangeable and water soluble’ and ‘specifically adsorbed’ fractions.

The accident at the Three Mile Island nuclear reactor in March 1979 resulted in contamination of the containment and auxiliary buildings. An aerosol sample from the auxiliary building obtained by filtering about 1.4x10^9 cm^3 of air through a fiberglass filter for 8 days contained an estimated total transuranic actinide radioactivity of 13 Bq (350 pCi), for an average concentration of 0.0093 Bq/m^3 (0.25 pCi/m^3) (assuming no oversampling in the confined space) of which ^{241}Am was the major contributor (Kanapilly et al. 1983).

Air monitoring of ^{241}Am was started at Hanford in 1993 to estimate regional background concentrations of the radionuclide before large-scale remediation work commenced (PNL 2000). Maximum and mean onsite levels of ^{241}Am were 0.50 and 0.46 aCi/m^3 (0.019 and 0.017 μBq/m^3) in 1994 compared with 0.90 and 0.41 aCi/m^3 (0.033 and 0.015 μBq/m^3) in 1993. Concentrations were not measurable at perimeter sites or at distant community-operated surveillance stations in 1994 (PNL 1995).

Annual average ^{241}Am air concentrations of 2.9x10^{-5}, 1.4x10^{-5}, and 0.55x10^{-5} pCi/m^3 (1.1, 0.52, and 0.20 μBq/m^3) at the 100-K Area, and 2.5x10^{-5}, 1.6x10^{-5}, and 0.32x10^{-5} pCi/m^3 (0.93, 0.59, and 0.12 μBq/m^3) at the 200-East Area of the Hanford site were measured in 1999, 2000, and 2001, respectively. No data were reported for ^{241}Am levels measured at distant community sites. Average gross alpha concentrations in air on the Hanford Site in 1999, 2000, and 2001 were comparable to levels measured at distance stations. These data indicate that the onsite levels were likely to be due to natural sources and worldwide radioactive fallout (PNNL 2000, 2001, 2002). ^{241}Am was detected in 15 quarterly air samples measured at distant sites, boundary sites, and the INEEL site in 2002. Concentrations ranged from 1.6×10^{-12} to 8.4×10^{-12} pCi/mL of air (5.9×10^{-8}–31×10^{-8} μBq/mL). ^{241}Am was not detected in the other 16 samples. Concentrations were within historical measurements and were below regulatory standards (DOE 2003b).

In an attempt to obtain background ^{241}Am concentrations around the Waste Isolation Pilot Plant (WIPP) in southeastern New Mexico, a plant developed for demonstrating the safe disposal of transuranic waste, high volume samples were obtained continuously over a 14-day period between February 23 and May 6, 1996 1 km from the facility. Samplers concurrently obtained total suspended solids (TSP) and particles >10 μm (PM_{10}) at a 5-m level elevation and PM_{10} at a 2-m level. The activities of ^{241}Am were 8±2, 6±2, and 11±1 nBq/m^3 (0.2±0.05, 0.16±0.054, and 0.31±0.03 aCi/m^3) in the PM_{10}-2 m, PM_{10}-5 m, and TSP-5 m samples, respectively (Lee et al. 1998). Approximately 50% of the ^{241}Am activity concentration was in the PM_{10} fraction. Using values for the ^{239,240}Pu concentrations measured at the site and those obtained in the early 1980s and estimated mean residence times for Pu in the troposphere and stratosphere, it is likely that the observed ^{241}Am levels reflect resuspension of dust contamination at the WIPP site and not background ^{241}Am levels. Present background levels result from past atmospheric nuclear testing and would be fairly uniform throughout the northern hemisphere.

6.4.2 Water

^{241}Am may enter surface water from nuclear power plants sited on the shores. A region of possible concentration is the Great Lakes where 35 nuclear facilities are located in the lakes basin. A sample of water from Lake Ontario collected in 1985 and filtered through a 0.45-μm Millipore filter contained 0.3 mBq ^{241}Am/L (8 fCi/L) (Platford and Joshi 1986). A few water samples from the Savannah River, which received runoff and discharges from the DOE Savannah River Plant, contained 0.05 fCi ^{241}Am/L (2 μBq/L), about the same levels as other American rivers (DOE 1980).

In some surface waters that have been contaminated by waste discharge, the concentrations of americium may be higher than background levels. For example, at times, measurable levels of ^{241}Am have been detected in the creek where waste water is discharged from Argonne National Laboratory's treatment plant. However, this creek is not used for drinking water and no ^{241}Am has been detected in the Des Plains River into which it empties (ANL 1999). While ^{241}Am is associated with waste from the Hanford site operations, concentrations in groundwater were considered low (no value was given) and were not discussed in the PNNL (2002) report. The 1995–1996 levels of ^{241}Am in rivers and lakes in the exclusion zone of the Chernobyl Nuclear Power Plant ranged from 20 to 200 μBq/L (0.5–5 fCi/L) with 70% of the radioactivity being contained in the particulate phase (Matsunaga et al. 1998). The particles appear to have originated from the erosion of surface soil in the area.

The concentration of ^{241}Am in filtered seawater is very variable. Some ^{241}Am levels (filtered through a 0.22 μm filter) are (site, date, and concentration): Arctic Ocean, 7/81, 1.77 μBq/L (0.048 fCi/L); English Chanel, 4/85, 4.96 μBq/L (0.13 fCi/L); and Eastern Irish Sea, 4/84, 4,840 μBq/L (131 fCi/L) (Lovett et al. 1990). ^{241}Am concentrations in filtered seawater in the North Channel of the Irish Sea were about 200 and 120 mBq/m^3 (5.4 and 3.2 pCi/m^3) in 1979 and 1986, respectively (Pattenden and McKay 1994). Because of the higher sediment load, near shore water generally has a larger fraction of americium bound to particles, and therefore, seawater from the northern coast of Scotland (which is impacted by the United Kingdom Atomic Energy Agency [UKAEA] Technology Dounreay Establishment, a nuclear research and development facility at Dounreay, Scotland) shows a large variation in near-shore concentrations (see Table 6-3). Levels were much higher in the immediate vicinity of Dounreay than elsewhere along the coast. The 1984 level was unusually high because the monthly discharge from the plant before the sampling was 40 times the mean monthly level. Time series measurements on seaweeds and winkles show that the americium levels peaked in the early 1980s and then declined by 1989, when levels were the lowest they had been in 12 years. In comparison, offshore ^{241}Am levels in surface seawater of the North Sea and North Atlantic Ocean has remained relatively constant, at around 10 Bq/m^3 (270 pCi/m^3), between 1976 and 1988 (Pattenden and McKay 1994). Nearshore levels of ^{241}Am were much greater than offshore concentrations, and the activity in the dissolved and particle-associated fractions were broadly similar.

Mururoa and Fangataufa Atolls were used from 1975 to 1996 for underground testing of nuclear weapons. Of the nine sites at Mururoa sampled for underground water, two, Aristee and Ceto, had measurable ^{241}Am concentrations, 0.064 and 0.104 mBq/L (1.7 and 2.8 fCi/L), respectively. The remaining seven sites at Mururoa and the two sites at Fangautaufa had ^{241}Am had concentrations below the detection limit (<0.002–<0.008 mBq/L [<0.05–<0.2 fCi/L]). At Aristee and Ceto, the ^{241}Am concentrations in the particle fraction of groundwater samples were 0.037 and 0.114 mBq/L (1.0 and 3.08 fCi/L) of filtered

Table 6-3. Concentrations of ^{241}Am in Water

Site/sample type	Year	Number of samples	Concentration (mBq/m^3)	Concentration (pCi/m^3)	Type
Dounreay, Scotland (north coast) near U.K. AEA Technology Dounreay Establishment R&D (nuclear reactor, reprocessing)					
Filtered seawater (0.45 μm)	1984	13	6,058±9,912	164±268	Mean±SD
	1985	16	36±18	0.97±0.49	Mean±SD
	1986	14	47±62	1.3±1.7	Mean±SD
	1987	30	15±17	0.41±0.46	Mean±SD
Unfiltered seawater	1986	14	110±147	3.0±4.0	Mean±SD
	1987	30	35±65	0.95±1.8	Mean±SD
Scotland mainland and some island coasts (near shore)					
Filtered seawater (13 sites)	1982	NR	1.9–140	0.051–3.78	Range
Cruise of RV Scotia, Orkney Islands, and northeast Scottish coast					
Near surface unfiltered seawater	1982	NR	6–270	0.2–7.3	Range
30–40 m unfiltered seawater		NR	220–650	5.9–17.6	Range

NR = not reported; R&D = Research and Development; SD = standard deviation; U.K. AEA = United Kingdom Atomic Energy Agency

Source: Pattenden and McKay 1994

water, respectively, and at the remaining seven sites at Mururoa, the ^{241}Am concentrations were below the detection limit (<0.003–<0.001 mBq/L [0.08–0.03 fCi/L]. At the two Fangatufa Atoll sites, ^{241}Am concentrations were 0.016 and 0.006 mBq/L (0.43 and 0.16 fCi/L) of filtered water (Mulsow et al. 1999).

The concentration of ^{241}Am in New York City tap water in 1974 was estimated to be 0.03 fCi/L (1 μBq/L) based on the average ^{241}Am/^{239,240}Pu ratio of 0.10 obtained in 1975–1976 and the 1974 ^{239,240}Pu concentration (Bennett 1979).

6.4.3 Sediment and Soil

Soil may become contaminated with ^{241}Am from the deposition of the element released to the atmosphere from nuclear weapons' testing and nuclear reactors, or secondarily, from the resuspension of contaminated soil. ^{241}Am levels may then increase due to ingrowth from the decay of ^{241}Pu (half-life 14.4 years). Soil surveillance provides baseline information with which unplanned releases can be compared, and long-term contamination trends can be determined. It also provides information about direct exposure to humans (inhalation of dust, ingestion of dirt) as well as potential sources of exposure (contamination of groundwater, uptake into plants and animals). The concentrations of ^{241}Am in soil samples from the northeastern United States from 1970 to 1974 are shown in Table 6-1. The areal activity at the sampling date ranged from 0.37 to 0.49 mCi/km^2 (14–18 MBq/km^2), which compares well with that calculated from estimated values obtained from releases from nuclear weapons testing combined with an atmospheric transport model (Bennett 1979). ^{241}Am in soil resulting from cumulative deposition in the New York region was estimated as being fairly constant since about 1969 at about 0.060 mCi/km^2 (2 MBq/km^2), whereas ^{241}Pu peaked at about 20.1 mCi/km^2 (744 MBq/km^2) in 1965. The decay of ^{241}Pu adds to the cumulative deposits of ^{241}Am in soil, bringing the total ^{241}Am to about 0.8 mCi/km^2 (30 MBq/km^2). The ^{241}Am inventory should reach a maximum of 0.88 mCi/km^2 (33 MBq/km^2) in 2035. The radioactive decay of ^{241}Am will then exceed its ingrowth, and the cumulative deposits of ^{241}Am will then slowly be depleted.

Nuclear Power Plants. EPRI (1981) conducted a survey of transuranic radionuclides in the terrestrial environs of nuclear power plants in 1978–1979. The plants included two PWRs and two BWRs that were of modern design and had been in operation at least 3 years. The ^{241}Am levels in soil samples collected around all of the power plants were indistinguishable from fallout background.

A survey of Big Trees Park was conducted in 1998 after soil samples taken in the park in 1993 and 1995 revealed higher-than-expected concentrations of plutonium (LLNL 1999). Big Trees Park is a public park in the city of Livermore, California located about one-half mile from Lawrence Livermore National Laboratory (LLNL). The pattern of ^{241}Am distribution was the same as that for plutonium with levels significantly higher inside ornamental tree wells than outside the wells or at depths. Activity levels were well below EPA's residential preliminary remediation goal of 2.5 pCi/g (93 mBq/g), which may have necessitated cleanup. The source of the contamination was most likely sewage sludge from LLNL that was applied to the ornamental trees in the park.

Great Britain. In 1993, the Ministry of Agriculture, Fisheries, and Food (MAFF) in the United Kingdom systematically surveyed the concentrations of ^{241}Am in soil and grass samples near 18 nuclear sites in England and Wales (Sanchez et al. 1996). ^{241}Am was detected in most soil samples; the range was 0.03–0.40 Bq/kg (0.8–11 pCi/kg) soil; three samples were below the detection limit. Sanchez et al. (1998) measured the ^{241}Am activity in root mat samples from tide washed pastures in 17 estuaries spanning the eastern seaboard of the Irish Sea from Solway in northwest England to David's Head in southern Wales. Fourteen of the pastures had detectable levels of ^{241}Am with median levels of 64–5,570 Bq/kg (1.7–150 nCi/kg) dry weight and individual samples of 14–10,230 Bq/kg (0.38–276 nCi/kg) dry weight. ^{241}Am accumulations in soils (to 10–15 cm depth) near Greeny Geo, Scotland ranged from 2,000 to 11,400 Bq/m^2 (54–308 nCi/m^2) (Pattenden and McKay 1994). This coastal area is contaminated by foam or spume that forms in seawater in rocky inlets and blows over the land. The seawater in the area receives discharges from the BNFL Sellafield facility. Sediment from two Harbors on the Isle of Mann, which is impacted by the reprocessing plant at Sellafield in the United Kingdom, contained ^{241}Am ranging from <2.0 to 8.1 Bq/kg (<50–220 pCi/kg) dry weight during 1990–1993 (McKenna and Longworth 1995). Nearshore sediment core samples taken at several sites off the northern coast of Scotland (see Table 6-1) had fairly constant ^{241}Am levels with depth showing that mixing had occurred to a depth of 74 cm (Pattenden and McKay 1994). ^{241}Am concentrations found in a sand dune ecosystem near the British Nuclear Fuels reprocessing plant at Sellafield, Cumbria United Kingdom, were 183–250 Bq/kg (4.95–6.76 nCi/kg) (Copplestone et al. 2001).

Los Alamos National Laboratory. Soil surveillance is conducted at national laboratories to monitor the levels of radionuclides and ascertain their possible migration. Data from such a program at the LANL in 1974–1996 was evaluated to ascertain whether there were changes in radionuclide concentrations during that period and whether changes could be ascribed to air emissions and fugitive dust (Fresquez et al. 1998a). Surface samples (0–5 cm) were obtained on-site and from perimeter and background sites.

Americium levels at LANL sites were significantly higher than at background sites. The mean 1974–1996 concentrations of ^{241}Am in soil at LANL and background sites were 0.011±0.006 and 0.006±0.002 pCi/g (0.41±0.22 and 0.22±0.07 mBq/g) dry weight, respectively. Sediment 2.1 km downstream of a National Pollutant Discharge Elimination System (NPDES) outfall in Mortandad Canyon, Los Alamos County, New Mexico in 1994 contained high levels, 8.93 pCi/g (330 Bq/g) dry weight, of ^{241}Am. However, the ^{241}Am concentrations in sediment upstream of the outfall and 4.0 km downstream below a sediment trap were below upper limits of regional background levels, 0.023 pCi/g (0.85 Bq/g) dry weight. (Bennett et al. 1996).

Brookhaven National Laboratory. Sediment samples were analyzed from the Peconic River system on Long Island, NY, downstream of Brookhaven National Laboratory (BNL). Near the sewage treatment plant closest to BNL, respective mean concentrations (dry weight) of ^{241}Am from three locations at four depth intervals (0.00-0.06, 0.06-0.15, 0.15-0.24, and 0.24-0.37 m) were 31, 24, 5.6, and 1.7 Bq/kg (0.83, 0.64, 0.15, and 0.045 nCi/kg). At one location at the BNL property boundary mean concentrations of ^{241}Am were 18, 2.8, 0.29, and 0.28 Bq/kg (0.49, 0.076, 0.0078, and 0.0076 pCi/kg), respectively, for the same depth intervals. Sediment samples from a control river, Connetquot River, were 1.4 and 0.74 Bq/kg (0.038 and 0.020 nCi/kg) (0.00-0.06 m) and 0.30 and 0.081 Bq/kg (0.0081 and 0.00022 nCi/kg) (0.06-0.15 m) in two locations (Rapiejko et al. 2001).

Rocky Flats. Rocky Flats (RF), near Golden, Colorado, was contaminated with ^{241}Am as a result of past waste storage practices, which resulted in drums leaking contaminated oil (Litaor et al. 1994). Sampling was performed in four regions of expected actinide activity (26 pits) east of RF. Activity decreased with increasing distance east of the former storage site and was predominantly confined to the upper 12 inches of soil. The mean ^{241}Am activities in the upper 3 inches of soil in the four regions were 1,653, 47.4, 2.9, and 3.7 Bq/kg (44.6, 1.28, 0.078, and 0.100 nCi/kg). In the region with the highest activity, the mean concentrations were 1,653, 1,063, 758, 595, 122, and 2.6 Bq/kg (44.6, 28.7, 20.5, 16.1, 3.3, and 0.070 nCi/kg) at depths of 0–3, 3–6, 6–9, 9–12, 12–18, and 36–48 inches, respectively, indicating significant retention near the surface (Litaor et al. 1994). Soil sampled at 118 plots around RF contained ^{241}Am ranging from 0.18 to 9,990 Bq/kg (0.0049–270 nCi/kg) with a mean and SD of 321 and 1,143 Bq/kg (8.67 and 30.9 nCi/kg), respectively (Litaor 1995). The distribution pattern reflects wind dispersion consistent with the prevailing winds at RF.

Idaho National Engineering and Environmental Laboratory. ^{241}Am contamination occurred outside the Subsurface Disposal Area (SDA) to a distance of 2,500 meters at the INEEL. Maximum concentrations of ^{241}Am, 2,048 nCi/m^2 (75.8 kBq/m^2) in the 0–4 cm surface layer, near the perimeter of the SDA, were thought to be due to flooding and to localized drainage of water, while low concentrations away from the SDA perimeter area resulted from wind transport (Markham et al. 1978). With the exception of samples collected from the Waste Experimental Reduction Facility, soil samples collected in 2002 from within the INEEL ranged from not detected to 0.889 pCi/ g (dry weight) and were within the background range for the INEEL and surrounding areas. These levels were attributable to past fallout. Soil samples collected from the perimeter and off-site around INEEL ranged from approximately 0.001 to 0.015 pCi/g (dry weight). There were no significant differences in ^{241}Am concentrations between boundary and distant samples, indicating that INEEL inputs did not contribute significantly to these concentrations (DOE 2003b).

Superfund National Priorities List (NPL) Sites. ^{241}Am has been detected in soil at 5 of the 1,636 NPL hazardous waste sites where americium has been identified in some environmental compartment (HazDat 2004); it was found in sediment at two of these sites. The distribution of Superfund NPL sites is shown in Figure 6-1.

Chernobyl. ^{241}Am, in five samples of surface soil collected 1.5–15 km from the Chernobyl accident in July 1987, was predominantly in the 0.005–0.25 mm size fraction of soil, which comprised 65% of the mass (Berezhnoi et al. 1991).

Bikini Atoll. Bikini Atoll was the site of 23 nuclear detonations between 1946 and 1958. Surface soil collected at 20 sites on Bikini Atoll in 1972 contained 1.2–45 pCi/g (0.044–1.7 Bq/g) of ^{241}Am (Nevissi et al. 1976). This level will increase for several decades as a result of ingrowth from ^{241}Pu before it begins to decrease, unless weathering causes the americium and plutonium to spread away from the surface and cause the concentration to decrease.

More recent mean surface soil (0–5 cm) ^{241}Am concentrations were 0.30 and 0.22 Bq/g (8 and 5.9 pCi/g) for the interior of the island and the village area, respectively (Robison et al. 1997b). This indicates either that soil samples were collected at too great a depth (adding uncontaminated soil from deeper layers) or that weathering of surface media carried contamination into deeper layers (results in decreased concentrations for the samples). Concentrations of ^{241}Am in sediment from the surface of the lagoon floor ranged from 0.010 to >12 Bq/g from (0.27 to >320 pCi/g) with 1.1 and 0.60 Bg/g (30 and 16 pCi/g) being the average concentrations associated with fine and coarse sediments, respectively. The highest ^{241}Am concentrations were found in surface sediments (0–2 cm) in the northwestern area of the lagoon, which was adjacent to the locations of the largest explosions. The ^{241}Am concentrations were reported to decrease from north to south and from east to west (Robison and Noshkin 1999).

Alaska. The National Oceanic and Atmospheric Administration (NOAA) conducted a study in the Beaufort Sea in northwestern Alaska during the summer of 1993 to ascertain the levels of radionuclide activity in the U.S. Arctic (DOE 1997). ^{241}Am was present in sediment at detectable levels at 8 of the 13 sites sampled, ranging from 0.06 to 0.12 Bq/kg (1.6–3.2 pCi/kg). From the average ratio of ^{241}Am:$^{239+240}$Pu, it was concluded that the americium originated from global fallout.

The Great Lakes. In 1976, sediment at several sites was sampled in Lakes Erie and Ontario. ^{241}Am was detected at only one site in Lake Erie, a site influenced by the Nuclear Fuels Reprocessing plant at West Valley, New York (U.S. Energy Research and Development Administration 1977). Levels of ^{241}Am increased with depth at concentrations of 3.4, 7.9, 11.1, and 11.8 Bq/kg (92, 210, 300, and 319 pCi/kg) dry weight at 0–2, 2–4, 4-6, and 6–8 cm, respectively.

Arctic. ^{241}Am concentrations were measured in the surface marine sediments collected in the Spitsbergen-Bear Island region, located between the Arctic Ocean, Barents, Greenland, and Norwegian Seas, an area important to Norwegian fisheries. Highest concentrations (dry weight) were found in the Spitsbergen area, ranging from 0.37 to 0.85 Bq/kg (10–23 pCi/kg). Lower levels were found in the deeper parts of the Norwegian Sea, ranging from 0.30 to 0.007 Bq/kg (8.1–0.2 pCi/kg) (Heldal et al. 2002).

6.4.4 Other Environmental Media

Concentrations of ^{241}Am have been determined in various foods (Table 6-4) and biota (Table 6-5), some of which may be eaten by humans. Levels in food items and animals include those from diet studies as well as from areas contaminated by nuclear weapons testing and fuel reprocessing. The highest levels were in clams, crabs, and lobsters (up to 0.46 Bq/kg [12 pCi/kg]). In studies at DOE facilities and in the vicinity of the BNFL reprocessing plant at Sellafield and the Ravenglass Estuary, where pastures are subject to tidal inundations with seawater receiving discharges from the plant, results clearly show the effect on animals from feeding on contaminated pasture land (Arthur and Janke 1986; Bradford and Curtis 1984; DOE 1979c; Janke and Arthur 1985).

Table 6-4. Average Concentrations of ^{241}Am in Food

	Concentration[a]		
Local Food	mBq/kg	fCi/kg	Reference
Bikini Atoll[b], U.S. Nuclear Test Site from 1946 to 1958, including BRAVO explosion on 3/1/54			Robison et al. 1997b
Reef fish	6.5	180	
Tuna	1.3	35	
Mahi mahi	1.3	35	
Marine crabs	26	700	
Lobster	26	700	
Clams	460	12,000	
Coconut crabs	28	760	
Land crabs	28	760	
Octopus	6.5	180	
Turtle	6.5	180	
Chicken, muscle	6.0	160	
Pork, muscle	6.0	160	
Eggs, chicken	6.0	160	
Pandanus fruit (n=69)	3.8	100	
Breadfruit (n=41)	1.2	32	
Coconut juice, drinking (n=747)	8.5	230	
Coconut meat, drinking (n=812)	3.6	97	
Copra meat (n=188)	1.1	30	
Papaya (n=93)	0.36	9.7	
Squash (n=53)	3.0	81	
Pumpkin	3.0	81	
Banana (n=39)	0.36	9.7	
Citrus	0.36	9.7	
Rain water	0.0037	0.10	
Well water	0.044	1.2	

Table 6-4. Average Concentrations of ^{241}Am in Food

	Concentration[a]		
Local Food	mBq/kg	fCi/kg	Reference
Northern Marshall Islands Radiological Survey (NMIRS)[c], fallout from U.S. atmospheric weapons testing from 1946 to 1958, including BRAVO explosion on 3/1/54			Robison et al. 1997a
Reef fish, muscle; 11 atolls/islands	<0.23–1.4	<6.2–38	
Pelagic fish, muscle; 9 atolls/islands	<0.14–0.27	<3.8–7.3	
Clams, soft parts; 10 atolls/islands	<1.4–46	<38–1,200	
Pork; 6 atolls/islands	<0.11–2.8	<3.0–76	
Chicken; 4 atolls/islands	1.2–4.1	32–110	
Coconut crab; 2 atolls/islands	4.3–28	120–760	
Drinking coconut meat; 5 atolls/islands	<1.3	<35	
Copra meat; 10 atolls/islands	<0.42–7.4	<11–200	
Pandanus; 12 atolls/islands	<0.23–8.5	<6.2–230	
Breadfruit; 7 atolls/islands	0.26–8.2	7.0–220	
Total Diet Study—New York, 1974[d]			Bennett 1979
Shellfish	0.48±0.15	13±4	
Whole grain products	0.074±0.044	2.0±1.2	
Bakery products	0.063±0.02	1.7±0.6	
Fresh vegetables	0.048±0.015	1.3±0.4	
Fresh fruit	0.041±0.015	1.1±0.4	
Root vegetables	0.030±0.007	0.80±0.19	
Dry beans	0.022±0.012	0.60±0.34	
Poultry	0.019±0.011	0.52±0.31	
Potatoes (peeled)	0.013±0.002	0.36±0.06	
Flour	0.013±0.007	0.36±0.18	
Fruit juice	0.011±0.006	0.29±0.16	

Table 6-4. Average Concentrations of ^{241}Am in Food

	Concentration[a]		
Local Food	mBq/kg	fCi/kg	Reference
Meat	0.010±0.006	0.28±0.17	
Rice	0.010±0.012	0.27±0.34	
Milk	0.0093±0.0004	0.25±0.01	
Fresh fish	0.0089±0.0030	0.24±0.08	
Eggs	0.0059±0.002	0.16±0.06	
Macaroni	0.0059±0.009	0.16±0.25	
Canned fruit	0.0026±0.0033	0.07±0.09	
Near Sellafield, England, 1994			Fulker et al. 1998
Honey	0.0007	0.02	
Blackberry	0.043	1.2	
Pheasant	0.029	0.78	
Mushrooms	0.0065	0.18	

[a] Concentrations on a wet or fresh weight basis. Concentrations originally expressed in mBq/kg unless otherwise indicated.
[b] Terrestrial food is from a continuous monitoring program in 1975–1993; marine food is from a 1988 report.
[c] Decay correction to 1996.
[d] Concentrations originally expressed as pCi/kg.

Table 6-5. Concentrations of ^{241}Am in Fauna

Species	Sample	Location	Concentration	Units[a]	Type	Reference
Fish						
Farallon Islands Nuclear Waste Disposal Site (FINWDS) and Point Arena (reference site), 1986–1987						Suchanek et al. 1996
Dover sole	Flesh	FINWDS (n=3)	1,630±2,360	mBq/kg w.w.	Mean±SD	
		Point Arena (n=6)	126±88			
Sablefish	Flesh	FINWDS (n=1)	2,850	mBq/kg w.w.	Mean±SD	
		Point Arena (n=5)	1,000±1,400			
Thornyheads	Flesh	FINWDS (n=2)	186±35	mBq/kg w.w.	Mean±SD	
		Point Arena (n=3)	299±157			
Vicinity of Los Alamos National Laboratory, 1997, Bottom-feeding fish (n=10/site)						Fresquez et al. 1999
Fish (2 sites)	Muscle+bone	Reservoirs	1.7 (19.8)	10^{-5} pCi/g d.w.	Mean (SD)	
Fish (4 sites)	Muscle+bone	Rio Grande	84.1 (54.6)	10^{-5} pCi/g d.w.	Mean (SD)	
Fish (5 sites)	Viscera	Rio Grande	31.2–198.0	10^{-5} pCi/g d.w.	Range of means	
Fish from Irish ports possibly impacted by Sellafield, 1988–1997						Ryan et al. 1999
Plaice	Edible portion		0.4–144	mBq/kg w.w.	Range of means	
Whiting	Edible portion		0.42–2.8	mBq/kg w.w.	Range of means	
Cod	Edible portion		0.62–26	mBq/kg w.w.	Range of means	
Crustacea						
Fish ports possibly impacted by Sellafield, 1988–1997						Ryan et al. 1999
Prawns	Edible portion		12–138	mBq/kg w.w.	Range of means	
North Eastern Irish Sea, Sellafield coastal area, U.K., commercially caught crabs and European lobsters, 1997						Swift and Nicholson 2001
crab (n=34)	Edible portion		1.7(0.8)	Bq/kg w.w.	Mean(SD)	
lobster (n=35)	Edible portion		8.3(11.3)	Bq/kg w.w.	Mean(SD)	

Table 6-5. Concentrations of ^{241}Am in Fauna

Species	Sample	Location	Concentration	Units[a]	Type	Reference
Bivalves						
Farallon Islands Nuclear Waste Dump Site (FINWDS) and Point Arena (reference site), 1986–1987						Suchanek et al. 1996
Mussels	Flesh	FINWDS (n=1)	6	mBq/kg w.w.	Mean	
Mussels	Flesh	Point Arena (n=2)	126±88	Bq/kg d.w.	Mean±SD	
U.S. Mussels Watch, 1976–1978 (East Coast: Maine-Virginia; East/Gulf Coast: Virginia-Texas; West Coast: Washington-California)						EPA 1984
Mussels and oysters	Flesh	East coast	0.43–0.65 (0.03–1.7)	Bq/kg d.w.	Range of means (range)	
Mussels and oysters	Flesh	East/Gulf coasts	0.32–0.60 (0.03–1.95)	Bq/kg d.w.	Range of means (range)	
Mussels and oysters	Flesh	West coast	0.49 (0.14–18.5)	Bq/kg d.w.	Range of means (range)	
Carlinford, Ireland (port city), impact from Sellafield Reprocessing Plant, 1991–1997						Ryan et al. 1999
Mussels	Edible portion		<11–109 (69)	mBq/kg w.w.	Range of means (median)	
Oysters	Edible portion		14–27	mBq/kg w.w.	Range of means	
Irish Sea, site impacted by Windscale Reprocessing Plant and reference sites, 1977						EPA 1984
Mussels	Flesh	Isle of Man	550 (55)	Bq/kg d.w.	Mean (SD)	
Mussels	Flesh	Ravenglass estuary	63.9x10^{3} (6,390)	Bq/kg d.w.	Mean (SD)	
Mussels (2 sites)	Flesh	Ebro estuary, Spain	0.52–0.88	Bq/kg d.w.	Range of means	
Sea mammals						
North Rona and Isle of May, Scotland, Possible impact from Sellafield and Chernobyl, 1987						Anderson et al. 1990
Grey seals (n=8)	Milk		<0.2–<0.3	Bq/L	Range	
Grey seals (n=2)	Liver		<0.3	Bq/kg f.w.	Range	
Grey seals (n=3)	Muscle		<0.3	Bq/kg f.w.	Range	

Table 6-5. Concentrations of ^{241}Am in Fauna

Species	Sample	Location	Concentration	Units[a]	Type	Reference
Mammals						
Halla, Finland, reindeer graze on lichen impacted by Chernobyl fallout						Paatero and Jaakkola 1998
Reindeer	Meat		0.014 (0.003)	Bq/kg d.w.	Mean (SD)	
Reindeer	Liver		0.013 (0.004)	Bq/kg d.w.	Mean (SD)	
Reindeer	Kidney		0.0043 (0.0010)	Bq/kg d.w.	Mean (SD)	
Material Disposal Area G, Los Alamos National Laboratory, New Mexico, mule deer and Rocky Mountain elk, 1991–1998						Ferenbaugh et al 2002
Deer (n=11)	Muscle		0.28 (0.22)	mBq/kg w.w.	Mean (SD)	
Elk (n=21)	Muscle		0.47 (0.34)	mBq/kg w.w.	Mean (SD)	
Nevada Test Site: Area 13 (outer compound) and near Area 13, but outside the area, 1973–1976						DOE 1979
Cattle (11 animals)	Muscle	Area 13	0.020–0.17	pCi/kg w.w.	Range	
Cattle (4 animals)	Muscle	Near Areas 13	<0.05–<0.11	pCi/kg w.w.	Range	
Fox (adult)	Muscle	Area 13	25.0	pCi/kg w.w.		
Fox (adult)	Muscle	Area 13	17.0	pCi/kg w.w.		
Coyote	Muscle	Area 13	4.7	pCi/kg w.w.		
Vicinity of Sellafield and the Ravenglass Estuary, England, 1977–1982						Bradford and Curtis 1984
Cattle	Liver	Sellafield (n=1)	0.23	Bq/kg f.w.		
		Ravenglass (n=3)	0.037–0.48		Range	
		Control (n=2)	0.003, 0.005			
Cattle	Meat	Ravenglass (n=2)	0.002, 0.012	Bq/kg f.w.		
		Control (n=1)	0.008			

Table 6-5. Concentrations of ^{241}Am in Fauna

Species	Sample	Location	Concentration	Units[a]	Type	Reference
Cattle	Lung	Ravenglass (n=3) Control (n=2)	0.05–0.085 0.003, 0.002	Bq/kg f.w.	Range	
Cattle	Bone	Ravenglass (n=3) Control (n=2)	0.25–0.29 0.013, 0.018	Bq/kg f.w.	Range	
Sheep	Meat	Ravenglass (n=3) Control (n=1)	0.040–0.064 0.0038	Bq/kg f.w.	Range	
Sheep	Liver	Ravenglass (n=3) Control (n=2)	2.6–6.6 0.006–0.0012	Bq/kg f.w.	Range	
Sheep	Lung	Ravenglass (n=3) Control (n=1)	0.018–0.24 0.017	Bq/kg f.w.	Range	
Sheep	Bone	Ravenglass (n=3) Control (n=1)	0.79–1.7 0.006	Bq/kg f.w.	Range	
Idaho National Engineering Laboratory–Subsurface Disposal Area (SDA) and control area, 1978–1979						Arthur and Janke 1986; Janke and Arthur 1985
Coyote	Feces	SDA (n=24) Control (n=12)	0.4±0.9 0.003±0.002	pCi/kg w.w.	Mean±SD	
Cottontail rabbit	Carcass	SDA (n=10) Control (n=5)	0.010±0.009 0.001±0.001	pCi/kg w.w.	Mean±SD	
Cottontail rabbit	Hide	SDA (n=10) Control (n=5)	0.029±0.046 0.002±0.001	pCi/kg w.w.	Mean±SD	
Deer mice	Carcass	SDA (n=21) Control (n=5)	0.010±0.02 0.003±0.002	pCi/kg w.w.	Mean±SD	

Table 6-5. Concentrations of ^{241}Am in Fauna

Species	Sample	Location	Concentration	Units[a]	Type	Reference
Deer mice	Hide	SDA (n=21) Control (n=5)	0.27±0.83 0.039±0.050	pCi/kg w.w.	Mean±SD	
<u>Invertebrates</u>						
Idaho National Engineering Laboratory, Subsurface Disposal Area (SDA) and control area, 1978–1979						Arthur and Janke 1986
Invertebrates	Composite	SDA (n=3) Control (n=1)	0.022±0.020 <0.004	pCi/kg w.w.	Mean±SD	

[a] conversion: 1 Ci=3.7×10^{10} Bq=0.037 TBq or 1 Bq=2.7×10^{-11} Ci=27 pCi

d.w. = dry weight; FINWDS = Farallon Islands Nuclear Waste Dump Site; f.w. = fat weight basis; n = number of samples; SD = standard deviation; SDA = subsurface disposal area; w.w. = wet weight

As part of the U.S. Mussel Watch program, monthly samples of mussels were collected from Bodega Head, California and Narragansett Bay, Rhode Island in 1977 and 1978 (Goldberg 1986). The mean concentration of ^{241}Am in samples ranged from 4.2 to 11.6 Bq/kg (0.11–0.31 nCi/kg) dry weight at Bodega Head and from 0.2 to 0.7 Bq/kg (0.005–0.02 nCi/kg) dry weight at Narragansett Bay. No seasonal pattern was evident. The ratio of ^{241}Am to $^{239+240}$Pu ranged from 1.3 to 5.3 on the west coast site and from 0.2 to 1.0 on the east coast. A surveillance program of U.S. coastal waters involving somewhat over 100 sites was another component of the Mussel Watch program. The flesh of both oysters and mussels, collected from the three major coastlines of the United States in 1976, 1977, and 1978, was analyzed (EPA 1984). The results are included in Table 6-5. The levels of ^{241}Am were higher on the west coast, which was ascribed to seasonal upwelling of the ^{241}Pu- and ^{241}Am-rich mid-depth north Pacific Ocean waters (affected by nuclear weapons test fallout), rather than from releases from the low-level nuclear waste site off the Farallon Islands. Another factor is the association of ^{241}Am with fine particles and the greater exposure of west coast bivalves to open ocean fine particles than east coast bivalves. Paradoxically, americium does not show the same seasonal fluctuations associated with upwelling as plutonium; this may be due to americium's greater adsorptivity and associated removal from the water column, or its slower uptake in bivalves. The NOAA conducted a study in the Beaufort Sea in northwestern Alaska during the summer of 1993 to ascertain the levels of radionuclide activity in the U.S. Arctic (DOE 1997a). As part of this study, ^{241}Am was determined in ashed samples of a variety of biota, including fish, invertebrates, crustaceans, whales, seal, and ducks, collected mostly in the coastal waters off Barrows, Alaska and analyzed by high-resolution gamma spectroscopy. ^{241}Am was not detectable in any of the samples (detection limits ≤0.6–≤80 mBq/kg [<20–<2,000 fCi/kg]).

Fish and mussels were sampled near the Farallon Islands Nuclear Waste Dump Site (FINWDS), approximately 30 miles west of San Francisco and a reference site (see Table 6-5) (Suchanek et al. 1996). While many of the drums have been breached or are nearing their expected lifetime, no statistical difference was found between the levels of ^{241}Am in the FINWDS fish and those from the reference site. A simple explanation is the fact that the fish species tested are quite mobile. However, ^{241}Am levels found in fish samples, 1.0 and 1.5 Bq/kg (27 and 41 pCi/kg), were higher than comparable fish muscle reported from relatively contaminated sites such as the Pacific Testing Grounds in Micronesia and in the Irish Sea near the Windscale Nuclear Plant at Sellafield, none of which exceeded 0.74 Bq/kg (20 pCi/kg). ^{241}Am concentrations were determined in fish and shellfish collected in 1990 from fish markets in the vicinity of the Vandellós I nuclear power plant (NPP) on the Spanish Mediterranean coast. The Vandellós I NPP was decommissioned after 1989. Mean ^{241}Am concentrations (dry weight) for large prawn, mollusc, red mullet, and hake collected near the Vandellós I NPP for were 62, 25, 54, and

6.0 mBq/kg (1.7, 0.68, 1.5, and 0.16 pCi/kg), respectively. At other locations along the Catalan coast, mean ^{241}Am concentrations were 5.2, 10, 3.0, and 5.2 mBq/kg (0.14, 0.27, 0.081, and 0.14 pCi/kg), respectively, for the same species (Sanchez-Cabeza and Molero 2000). As part of the Environmental Surveillance Program (at LANL), bottom-feeding fish were collected from reservoirs upstream and downstream from LANL in September 1997, as well as from five sites on the Rio Grande River starting at one site about 1 mile upstream of any stream crossing LANL land and analyzed for ^{241}Am and other radionuclides (Fresquez et al. 1999). The concentration of ^{241}Am in muscle + bone in fish collected in the reservoirs was significantly lower than that collected in the river (see Table 6-5). Mussel samples collected from two sites in the Irish Sea where the Windscale nuclear reprocessing plant is located show the influence of low levels of radionuclides from the nuclear fuel cycle (Table 6-5). The ^{241}Am found in the mussels were orders of magnitude higher than those on the U.S. coast. The Isle of Mann site is in the middle of the Irish Sea about 50 km from Windscale and the Ravenglass estuary site is 16 km to the south of the plant. The ^{241}Am input into the Irish Sea from fallout is dwarfed by the contribution by the Sellafield reprocessing facility in England. Observations from surveys of ^{241}Am in edible parts of fish and shellfish sampled from commercial landings at major Irish shipping ports between 1988 and 1997 (Table 6-5) indicate that activity levels in fish landed at northeast Irish ports have remained low, generally a fraction of a mBq/kg wet weight, while those in mussel, prawns, and oysters are much higher and have fluctuated in activity (up to 180 mBq/kg [4.9 pCi/kg] wet weight in prawns) over time (Ryan et al. 1999). Concentrations of ^{241}Am in the edible portions of molluscs (*Adamussium colbecki*) and fish (*Trematomus bernacchii*) collected during the years 1987–1996 from Terra Nova Bay (Ross Sea) in Antarctic were reported to be 0.0087 and 0.0006 Bq/kg (0.24 and 0.02 pCi/kg), dry weight, respectively (Marzano et al. 2000).

Enewetak and Bikini Atolls were used for nuclear weapons testing by the United States between 1946 and 1958. Log mean normal concentrations (wet weight) of ^{241}Am found in fish muscle and invertebrate flesh collected since 1976 and decay corrected from collection date to October 1998 were determined. At Enewetak Atoll, concentrations of ^{241}Am in reef fish, pelagic fish, mollusks, and crustaceans were 0.0050, 0.0024, 0.0010, and <0.1 Bq/kg (0.14, 0.064, 0.027, and <2.7 pCi/kg), respectively. At Bikini Atoll, ^{241}Am concentrations in reef fish, pelagic fish, mollusks, and crustaceans were 0.0072, 0.0018, 0.32, and <0.1 Bq/kg (0.19, 0.049, 8.6, and <2.7 pCi/kg), respectively. ^{241}Am concentrations were higher in the surface sediments at Bikini Atoll, ranging from 10 to >12,000 Bq/kg (0.27–>320 nCi/kg) as compared to Enewetak Atoll, ranging from <1 to 1,300 Bq/kg (0.027–35 nCi/kg). The authors noted that it is difficult to conclude that there is a difference in the mean concentrations of radionuclides in the muscles of fish from Bikini Atoll as compared to Enewetak Atoll, and that this supports earlier studies that found that at

Bikini and Enewetak Atolls, the transuranics in some fish may be unrelated to the environmental concentrations (Robison and Noshkin 1999).

Fish samples were analyzed from the Peconic River system on Long Island, New York, downstream of BNL. Near the sewage treatment plant closest to BNL, mean concentrations of ^{241}Am were 0.070 and 0.019 Bq/kg (1.9 and 0.51 pCi/kg) in bullhead catfish and chain pickerel (whole fish), respectively. A mean ^{241}Am concentration in bullhead catfish from a control river, Connetquot River, was <0.02 Bq/kg (<0.54 pCi/kg). In ponds farther downstream of BNL and the sewage treatment plant ^{241}Am concentrations were <0.03 Bq/kg (<0.8 pCi/kg) for all fish and <0.07 Bq/kg (<2 pCi/kg) for mussel flesh (Rapiejko et al. 2001).

Fifteen cattle grazing for 1,064 days on a site at the Nellis Bombing and Gunnery Range, Nevada, where a nuclear device was explosively destroyed 16 years previously, had ^{241}Am levels ranging from 0.74 to 590 mBq/kg (0.02–16 pCi/kg) wet weight, 4.4 mBq/kg (0.10 pCi/kg) geometric mean in muscle (Gilbert et al. 1989). Levels in liver and kidney were much higher, 72 and 11 mBq/kg (1.9 and 0.30 pCi/kg), geometric mean, respectively.

In 1993, the MAFF, United Kingdom systematically surveyed the concentrations of ^{241}Am in grass samples near 18 nuclear sites in England and Wales (Sanchez et al. 1996). ^{241}Am levels in vegetation samples were <0.10 Bq/kg (<2.7 pCi/kg). Sanchez et al. (1998) measured the ^{241}Am activity in vegetation from tide-washed pastures in 17 estuaries spanning the eastern seaboard of the Irish Sea from Solway in northwest England to David's Head in southern Wales. In 11 of the pastures, the median level of ^{241}Am in vegetation ranged from 14 to 575 Bq/kg (0.38–15.5 nCi/kg) dry weight; the others were <1 Bq/kg (<30 pCi/kg) dry weight. ^{241}Am levels in individual samples in the 11 pastures were 13–1,260 Bq/kg (0.35–34 nCi/kg) dry weight.

Brauning et al. (1986) reported radioactivity levels in municipal sludge. Sludge from Stickney, Illinois that was used as an organic fertilizer contained 3.3 and 1.5 fCi/g (14 and 575 Bq/kg) dry weight of ^{241}Am in 1974 and 1975, respectively. Potential sources in this case were thought to be from background concentrations and fallout from nuclear weapons testing. In Texas, ^{241}Am was present in sludge samples in 1983 and 1986 from a facility that received sanitary waste from the Gulf Nuclear, Inc. facility. Potential sources of radionuclides in the sludge were industrial wastes. Two incidents of contaminated sludge from companies producing foils for smoke detectors occurred in New York State. One of the receiving facilities that incinerated its sludge had 180–750 pCi/g (6.7–28 Bq/g) of ^{241}Am in dried incinerator ash between April 17, and May 7, 1984, after which it dropped to 13 pCi/g (0.48 Bq/g), corresponding to a sludge concentration of 5.2 pCi/g (0.19 Bq/g). During the latter half of 1984 and 1985, the ^{241}Am concentrations in sludge were 3.5 and 2.5 pCi/g (130 and 93 mBq/g), respectively. In the second incident, ^{241}Am in sludge was as high as 120 pCi/g (4,400 mBq/g) in 1984, but dropped to an average of 24.8 pCi/g (920 mBq/g) in the latter half of 1985. This facility placed its sludge in landfills (Brauning et al. 1986).

6.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

Americium in the environment may be transferred to humans by inhalation and ingestion of food and, in the case of children, ingestion of soil; in general, intake of americium should be small. Much of the ^{241}Am so taken is found in the lungs, skeleton, and liver. In addition to the ^{241}Am taken up, additional ^{241}Am is produced by the decay of ^{241}Pu that has been inhaled or ingested and absorbed. No data on the body burden of ^{241}Am in the general population were found. However, the body burden from fallout was estimated from the concentration of ^{241}Am in air and estimates of intake by ingestion. The modeling study indicated that most of the ^{241}Am body burden, 89% in 1978, was due to ingrowth from ^{241}Pu. The model also estimated that the contribution of ingestion to the ^{241}Am body burden was <1% of that due to inhalation. The total skeletal burden for 1970–1974 in New York was estimated from ashed skeletal samples to be 0.27 pCi (10 mBq) (Bennett 1979). UNSCEAR (2000) reported that inhalation exposures for even long-lived transuranic radionuclides became insignificant after 1985.

Ash from a 1974 ^{90}Sr total diet study in New York consisting of representational foods from 19 separate categories was analyzed for ^{241}Am to ascertain the dietary intake of the radionuclide from fallout (Bennett 1979). It was estimated that over 95% of the ^{241}Am transfer to food was from cumulative deposits in soil, rather than from direct deposition. The highest concentrations of ^{241}Am in food occurred in shellfish, followed by grain products and fresh fruits and vegetables. The lowest concentrations were in meat, milk, eggs, fish, and processed foods. The same patterns of concentrations were obtained for ^{239,240}Pu. The activity levels of ^{241}Am in foods from New York from a total diet study, and in areas near nuclear weapons test sites and a fuel reprocessing facility are shown in Table 6-4. Using dietary consumption estimates and the activity levels in the food items and tap water, the ^{241}Am ingestion intake for a New Yorker was 0.40±0.05 pCi/year (15±2 mBq/year) in 1974. The concentration of ^{241}Am in tap water was estimated at 30±3 aCi/kg (1±0.1 μBq/kg) with 1,200 kg of water being consumed in a year. The food samples obtained in 1974 occurred at a time when direct deposition from fallout was very low and the concentration of ^{241}Am in food was largely a result of uptake from soil derived from cumulative deposits. Direct deposition from fallout was greatest in 1963–1964. Additionally, direct deposition is a much less significant means of plants acquiring activity than root uptake. The ^{241}Am intake in diet from fallout will continue to increase due to ingrowth from ^{241}Pu until the cumulative deposit reaches its projected maximum of 0.88 mCi/km^2 (33 MBq/km^2) in the year 2035, at which time, the dietary intake of ^{241}Am will have reached a maximum of 0.7 pCi/year (26 mBq/year) (Bennett 1979).

As part of its Radionuclides in Food Program, the FDA determined concentrations of radionuclides in their Total Diet Study of food originating from the vicinity of nuclear reactors including raw vegetables, food crops (primarily fruits), fish, and milk. While not specifically analyzed for, ^{241}Am concentrations of radionuclides in both of these surveys in 1983–1986 and 1987–1992 were sufficiently low as to require no specific action or simple monitoring. Imported foods were extensively monitored starting in May 1986 for contamination resulting from the Chernobyl accident. While ^{241}Am was identified in samples of imported food that probably originated from Chernobyl, the vast majority of samples were below FDA's level of concern with only one oregano and three cheese shipments being detained (Cunningham et al. 1989, 1994). FDA has since published guidelines for radionuclide concentrations, including isotopes of americium and plutonium, in food (FDA 1998).

A study in the Ravenglass estuary, where tidal action inundates pasture land with water that receives discharges from the BNF reprocessing plan in Sellafield, concluded that consumption of a diet consisting of locally produced food would result in dose levels far below recommended limits (Bradford and Curtis 1984). Sanchez et al. (1998) measured the ^{241}Am activity in vegetation from tide washed pastures in 17 estuaries spanning the eastern seaboard of the Irish Sea from Solway in northwest England to David's Head in southern Wales. From the mean and median ^{241}Am activity in grass and appropriately reduced transfer factors, they predicted the activity in animal products, namely milk, beef, beef liver, lamb, and lamb liver. From this, they estimated the committed effective dose for a 1-year-old child, a 10-year-old child, and an adult from eating these foods. Assuming that the land above the high water mark was to be reclaimed for growing vegetables, the authors calculated estimates of dose derived from consumption of potatoes, which were determined to be the critical vegetable group in terms of radionuclide contamination. The results are shown in Table 6-6, in which the relative doses for the various foods are beef liver>potatoes>lamb liver>beef>milk>lamb.

Using transfer factors derived for uptake into plants and animals, Friberg and Vesanen (1999) have listed critical activity levels for combined α-emitting isotopes of Pu, Am, and Cm and other nuclides deposited

Table 6-6. Estimated Effective ^{241}Am Dose from Food Raised on Land Impacted by Tidal Estuaries along the Irish Sea

Food item	Transfer factors		^{241}Am concentration[a]		Committed effective dose (μSv) from ^{241}Am[b]		
	Mean	Median	Mean	Median	Adult	10-year old	1-year old
Milk	1.0×10^{-7}	1.0×10^{-7}	6.3×10^{-4}	2.5×10^{-4}	0.01	0.02	0.03
Beef	1.0×10^{-5}	1.0×10^{-5}	0.06	0.02	0.2	0.2	0.07
Beef liver	2.0×10^{-3}	2.0×10^{-3}	12.61	4.94	14.6	8.3	4.9
Lamb	4.0×10^{-5}	4.0×10^{-5}	0.03	0.01	0.05	0.03	0.01
Lamb liver	3.0×10^{-3}	3.0×10^{-3}	2.18	0.86	2.5	1.4	0.8
Potatoes					3.3	3.1	1.2

[a] Food concentrations were estimated from measured ^{241}Am levels in grass (485 Bq/kg dry weight mean, 190 Bq/kg dry weight median), by applying grass-to-food transfer factors. Units are Bq/L for milk and Bq/kg fresh weight for other food items.

[b] The average consumption rates for the animal products were obtained from Byrom et al. (1995) and the dose coefficients were taken from SEPA (2000) (conversion: 1 Sv=100 rem).

Source: Sanchez et al. 1998

on soil and pasture vegetation from a nuclear accident required to exceed the action level for foodstuffs recommended by the IAEA. Critical activity levels for the α-emitting isotopes of Pu, Am, and Cm were determined to be 8 kBq/kg (0.2 μCi/kg) for soil top-layer and 3.5 kBq/kg (0.095 μCi/kg) for pasture vegetation. The lowest detection limit required for the α-emitting isotopes of Pu, Am, and Cm was determined to be 2 kBq/kg (0.05 μCi/kg). Detection limits for the α-emitting isotopes of Pu, Am, and Cm achieved with thorough radioanalytical procedures have been reported as low as 0.07 Bg/kg (2 pCi/kg) in soil (Friberg and Vesanen 1999).

The consumption of fish exposed to actinide contamination is a potential source of ^{241}Am exposure. This potential source of exposure has been evaluated by measuring the concentration of ^{241}Am in bass and bluegill living in a nuclear waste pond on the Hanford site where the levels of ^{241}Am in sediments are approximately 3 orders of magnitude above background levels. Assuming that actinide behavior in the body is similar to that defined by the International Commission on Radiation Protection (ICRP) for Standard Man, the maximum levels occurring in fish fillets after 1 year in the waste pond (see Table 6-5) would not be sufficient to produce a significant radiation dose to a human eating 1 pound of fish fillets every day for 70 years (Emery et al. 1981). Similarly, the consumption of fish from the Farallon Islands Nuclear Waste Dump Site off of San Francisco is expected to add ~1.5% to the typical annual dose to the whole body effective dose equivalent value to pooled radionuclide concentrations committed to age 70 (Suchanek et al. 1996). This increase is derived largely from the ingestion of ^{241}Am.

Lichens are very efficient collectors of deposited radionuclides. Since caribou and reindeer depend on lichens for food during the winter season, the lichen-reindeer/caribou-human pathway can lead to elevated exposures to indigenous Inuit and Sami populations who eat large quantities of meat and organs from caribou and reindeer. Reindeer tissue samples were analyzed for transuranium elements in Finland following the Chernobyl accident in 1986. Paatero and Jaakkola (1998) established that americium was transferred from lichen to reindeer via the gastrointestinal tract and not inhalation, and obtained a gastrointestinal-tract absorption coefficient of 0.00075 for ^{241}Am. This gastrointestinal-tract absorption coefficient is similar to that estimated for humans (0.0006–0.0008, see Section 3.3.1.2). Concentrations of ^{241}Am in reindeer resulting from Chernobyl fallout appear in Table 6-5. Global fallout of plutonium, which correlates with that of americium, was considerably higher than Chernobyl fallout, ca. 70 Bq/m^2 (2 pCi/m^2) compared to 1.5 Bq/m^2 (0.04 pCi/m^2) (Paatero and Jaakkola 1998). Based on ^{241}Am concentrations in reindeer in Finnish Lapland, estimated ^{241}Am intake is 2 pCi/year (74 mBq/year) for men and 1 pCi/year (37 mBq/year) for women (Jaakkola et al. 1977).

A modeling study for estimating exposure to man from dumping of radioactive waste in the Arctic Seas concluded that ingestion of molluscs was the dominant pathway for ^{241}Am, and that ^{241}Am along with ^{239}Pu would dominate the collective dose at longer time scales (Nielsen et al. 1997). Another modeling study dealing with the effect of long-term release of ^{241}Am into an estuarine coastal sea ecosystem found that the greatest intake by man is from eating marine food items such as seaweed, molluscs, crustaceans, and fish (Murray and Avogadro 1979). Another important route of exposure is the inhalation of contaminated airborne coastal sediment. Ingestion of sediment by children and the contamination of crops by airborne sediment were estimated to be of minor importance. Radiation from contaminated mud flats and direct exposure from water were considered to be of little consequence. A survey of ^{241}Am and plutonium in fish and shellfish in Ireland between 1988 and 1997 estimated that the mean doses from ^{241}Am for the typical and heavy Irish consumer were 0.024 and 0.104 μSv/year (2.4 and 10.4 μrem/year), respectively (Ryan et al. 1999). Americium contributes somewhat over a quarter of the combined plutonium-americium dose. In 1988, this combined dose was roughly 10% of that which the typical Irish seafood consumer obtained from ^{137}Cs; in 1997, this percentage had increased to 48%.

Both the general population and workers may be exposed to ^{241}Am from the production, distribution, and use of ionization chamber smoke detectors. The USNRC investigated exposure relating to this device. Their 1979 report concluded that the sum of doses to the population resulting from production, distribution, use, and disposal of 14 million smoke detectors containing 3 μCi (100 kBq) of ^{241}Am each (over 3 times that presently used) that were distributed in 1978 would result in a collective total body dose of 1,100 person-rem (11 person-Sv)—much lower than that which could potentially result in one cancer death (USNRC 1979). The risks reported in the 1979 report may be overestimates for current times, since radiation risk coefficients have changed and the amount of americium used per unit has been reduced. More recent information also indicates that exposure to ^{241}Am in a home smoke detector is negligible (<0.01 mrem/year [$<1\text{x}10^{-7}$ Sv/year]). Smoke detectors today typically contain approximately 1 μCi (37 kBq) of ^{241}Am (EPA 2004b). Exposure to ^{241}Am could result from improper disposal of smoke detectors; for example, if a detector was broken and then disposed of in a municipal landfill or incinerated. The ^{241}Am from the broken detector could be consumed by a child, but uptake would be negligible based on human study results (Rundo et al. 1977).

Occupational exposure to ^{241}Am may occur to workers manufacturing the ^{241}Am-containing foil used in ionization-type smoke detectors or assembling the device. Two manufacturers of the ^{241}Am foils were surveyed in 1977 (USNRC 1979). One reported an external population dose of 0.8 person-rem per curie (0.2 person-pSv/Bq) of ^{241}Am processed for the 150 workers involved and the 16.6 Ci (0.614 TBq) of

^{241}Am they processed. The maximum dose for an individual was approximately 0.8 rem (8 mSv). The second manufacturer reported 0.2 person-rem per curie (0.05 person-pSv/Bq) of ^{241}Am processed. It should be noted that manufacturers of ionization chamber smoke detectors must be licensed by the USNRC or an agreement state, and that certain work practices are required to minimize exposure. Workers wear shoe covers, lab coats, and gloves, and wipe tests are performed weekly. During the manufacture of the smoke detector units, one manufacturer reported the yearly external doses among badged workers in 1978: 40% had no measurable dose, 40% had <100 mrem (<1 mSv), 15% had doses between 250 and 500 mrem (2.5 and 5.0 mSv), 4% had doses between 500 and 750 mrem (5.0 and 7.5 mSv), and one individual had a dose between 750 and 1,000 mrem (7.5 and 10 mSv). The total occupational dose in the ionization chamber smoke detector industry in 1978 was estimated to range from 15 to 48 person-rem. The external doses to people involved in the distribution and retail sale of smoke detectors would be much less. The hand dose to installers is estimated to be 0.6 μrem (6 nSv) per unit installed. An individual having one detector in the hallway and one in the bedroom would have an annual dose of 9.3 μrem (93 nSv).

Workers at plutonium reprocessing facilities, nuclear reactors, or transuranium and low level waste storage facilities, or those engaged in the production or processing of ^{243}Am or ^{241}Am may be occupationally exposed to americium. In addition, workers at sites where nuclear testing was conducted may also be exposed to americium. Workers in nuclear power stations may be exposed to airborne radionuclides. The USNRC publishes annual reports on occupational radiation exposure at USNRC-licensed facilities. Monthly cumulative 24-hour air filter samples were obtained from five different working areas of a BWR in India during normal operation and during refueling in order to measure the concentrations of transuranic radionuclides, including americium, in air. The average air ^{241}Am concentrations during normal operation were (work area, concentration in mBq/m^3 [fCi/m^3]): reactor building—103 feet elevation, 0.95 (26); reactor building—143 feet elevation, 0.22 (5.9); reactor building—200 feet elevation, 0.25 (6.8); turbine building—133 feet elevation, 0.07 (1.9); and radwaste building—2.50 (68) (Hedge and Bhat 1983). During refueling, the respective concentrations were: 1.64, 1.33, 0.78, 0.19, and 6.34 mBq/m^3 (44.3, 35.9, 21, 5.1, and 171 fCi/m^3). The derived air concentration recommended by the ICRP for occupational exposure is 80.0 mBq/m^3 (2,200 fCi/m^3) (Hedge and Bhat 1983). In 1997, the French radiation protection office conducted monitoring (24-hour urine analysis/whole body activity measurements) of workers in the non-nuclear energy field (i.e., nuclear medicine, research laboratories, and non-nuclear industries) to ascertain the occupational intake of radionuclides (De Vathaire et al. 1998). ^{241}Am was not detected in samples from any of the 37 workers who worked with the radionuclide.

The U.S. Transuranium and Uranium Registries (USTUR) include The National Human Radiobiology Tissue Repository, which contains postmortem tissue from persons (primarily workers) with documented intakes of uranium or transuranic elements, such as americium. The USTUR publishes annual reports detailing cases of accidental exposure and tissue levels of the different radioactive isotopes. These reports and other information are available over the internet at http://www.tricity.wsu.edu/ustur/. Exposure is generally by inhalation, but entry into the body may also occur through wounds. Americium is largely retained in the lung, skeleton, and liver, but after long term exposure, significant amounts of ^{241}Am are found in the muscle, respiratory tract, and other soft tissue (Filipy et al. 1994; McInroy et al. 1989). In 25 USTUR cases involving the registry of individuals who typically had ≥20 years of employment in facilities where ^{241}Am was processed and who were likely to have had chronic, low-level inhalation exposure to the actinide, the ^{241}Am organ content of the liver and skeleton ranged from 0.01 to 554 Bq (0.26 Bq, median) (0.3–15,000 pCi, 7.0 pCi, median) and from 0.04 to 6,623 Bq (0.99 Bq, median) (1–179,000 pCi, 27 pCi, median), respectively (Kathren et al. 1988). Seventy-five percent of the combined activity in the liver and skeleton resided in the skeleton (see Section 3.4). Measurements of ^{241}Am were performed on the skulls of 66 residents of the Caithness area of Scotland near the Dounreay Nuclear Establishment between October 1990 and December 1991. No ^{241}Am was detected in any of these subjects (Ham et al. 2003).

6.6 EXPOSURES OF CHILDREN

This section focuses on exposures from conception to maturity at 18 years in humans. Differences from adults in susceptibility to hazardous substances are discussed in 3.7 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, 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, put things in their mouths, sometimes eat inappropriate things (such as dirt or paint chips), and spend more time outdoors. Children also are closer to the ground, and they do not use the judgment of adults to avoid hazards (NRC 1993).

Specific information on the exposure of children to americium is not available. As for adults in the general population, small exposures to ^{241}Am occur from normal ingestion of food and drinking water and inhaling air. While the potential for americium exposure is greater for children who consume foods grown in areas with elevated levels of americium in the soil, the uptake of americium into plants and animals is very low. Children may be exposed to a substance brought into the home on parents' work clothes, skin, hair, tools, or other objects from the workplace; however, with one exception, no instances of americium exposure in children by this route were found in the literature. In this case, a man who did some experiments using an americium source brought home a secondary source and stored it in a dresser drawer (Kelly and Dagle 1974). No information was located on americium levels in amniotic fluid, meconium, cord blood, neonatal blood, or breast milk, or other potential measurements relating to fetal or childhood body burdens.

^{241}Am may also find its way into municipal landfills because of improper disposal of smoke detectors or rubble of buildings containing smoke detectors. Smoke detectors may be broken, exposing the ^{241}Am ionization source. Young children might get access to the ^{241}Am and ingest it, although a human study indicates that uptake from ingesting the americium source from a smoke detector is insignificant (Rundo et al. 1977). At hazardous waste sites, americium that is found in excess of fallout background levels is most likely to be in soil and presents a special hazard for young children. Hand-to-mouth activity and inadvertent or intentional (pica) eating of contaminated dirt will result in oral exposure to americium.

6.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Occupational exposures and general population exposures to potentially high levels of ^{241}Am and other radionuclides might occur to people occupying land like the southern Solway and Dee marshes, areas tidally inundated by water contaminated by the BNFL nuclear fuel reprocessing plant at Sellafield, United Kingdom plant in England (Rose et al. 1996). Modeling studies of human populations exposed to ^{241}Am in such coastal environments show that the highest levels of ^{241}Am would occur in sediment and marine organisms, especially molluscs and seaweed (Murray and Avogadro 1979). These human populations include farm workers and turf cutters and recreational users such as birdwatchers, hikers, fishermen, and bird shooters, as well as those who consume contaminated marsh produce, molluscs, and seaweed. Exposure routes would be ingestion and inhalation of dust. Even so, the dose rates that would be received by these pathways are very low (Murray and Avogadro 1979).

Populations with potentially high exposure to ^{241}Am from commercial devices such as ionization chamber smoke detectors might include production workers and firemen attending to fires in production facilities, or warehouses where a large number of sources or devices are housed.

6.8 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 americium is available. Where adequate information is not available, ATSDR, in conjunction with the National Toxicology Program (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 americium.

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 proposed.

6.8.1 Identification of Data Needs

Physical, Chemical, and Radiological Properties. Data on the physical and chemical properties of americium and americium compounds are available in the literature (Cotton and Wilkinson 1980; Lide 1994, 1998; Nenot and Stather 1980; Seaborg 1991). Data are also available on the radiological properties of americium isotopes (Baum et al. 2002; DOE 2003a; ICRP 1983; Lide 1998).

Production, Import/Export, Use, Release, and Disposal. Data regarding the past and present production and import/export volumes for ^{241}Am and ^{243}Am are not available. The limited commercial use of ^{241}Am provides a good estimate of probable production volumes. The uses of americium are known (Budavari 1996; Seaborg 1991; UIC 1997; USNRC 1979). However, information on the production, import/export volumes, and the amounts of ^{241}Am used in various devices would be useful for estimating exposure. Estimates of the amounts of ^{241}Am released as a result of nuclear tests and accidents are available (Bennett 1979; Suchanek et al. 1996). The disposal of americium is governed by the

USNRC regulations (10 CFR 61). The Manifest Information Management System (MIMS), maintained by the INEEL, contains information on low-level radioactive waste shipments received at commercial low-level radioactive waste disposal facilities and the WIPP transuranic waste facility (INEEL 2000). The DOE stores most of its spent fuel at three primary locations: the Hanford site, Washington, the INEEL, Idaho, and the Savannah River Site, South Carolina. The DOE National Spent Fuel Program maintains a database of spent nuclear fuel stored in each of the three locations (DOE 1999a). Information concerning the americium content of civilian nuclear power plants and naval nuclear propulsion plant spent fuel now in storage and awaiting disposition would be useful for estimating the amount of americium stored in the country.

Environmental Fate. The environmental fate of americium has been extensively studied in relation to its introduction into the Irish Sea from the BNFL nuclear fuel reprocessing plant at Sellafield, United Kingdom (Belot et al. 1982; Bennett 1976; Bunzl et al. 1994, 1995; Malcolm et al. 1990b; McCartney et al. 1994; McKay et al. 1994; Murray et al. 1978, 1979; Pattenden and McKay 1994; Walker et al. 1986).

Other studies have investigated transport in soil and at DOE waste disposal sites (Fowler et al. 1981; McCarthy et al. 1998a, 1998b; Penrose et al. 1990). As a result of these studies, the environmental fate of americium is reasonably well understood. However, DOE has begun using the RESRAD Biota computer program, and it would be useful to validate uncertainties in associated input parameter values used in the model.

Bioavailability from Environmental Media. The absorption and distribution of americium as a result of inhalation and ingestion exposures have been discussed in Sections 3.3.1 and 3.3.2. EPA lists identical uptake factors for inhaled and ingested americium (as well as all of the other transuranics other than plutonium) regardless of compound solubility, indicating that the knowledge base for americium is not sufficiently developed to quantify the differences that are recognized for most other elements, and additional studies would be helpful.

Food Chain Bioaccumulation. Information about the levels of americium in aquatic and terrestrial organisms and its bioaccumulation in these organisms is available (DOE 1996; Fresquez et al. 1999; Suchanek et al. 1996). Data are also available on the uptake of americium in plants (Bennett 1979; Cataldo et al. 1980; EPA 1979; Romney et al. 1981; Schreckhise and Cline 1980; Schulz et al. 1976; Zach

1985) and levels in food (Bennett 1979; Cunningham et al. 1989, 1994; Robison et al. 1997a, 1997b). These data indicate that americium does not biomagnify in the food chain (Bennett 1979; Bulman 1978).

Exposure Levels in Environmental Media. Monitoring data for the levels of americium in contaminated media at hazardous waste sites would be useful. These data can then be used to estimate the intake and body burden of americium and assess the potential risk of adverse health effects in populations living in the vicinity of these sites.

Information is available on the levels of ^{241}Am in soil and sediment in areas affected only by global fallout, at DOE installations and other nuclear facilities, and at sites of nuclear explosions and accidents (Alberts et al. 1989; Bennett 1979; Cooper et al. 1994; DOE 1980; Pattenden and McKay 1994; Robison et al. 1997a, 1997b; Sanchez et al. 1996). ^{241}Am levels in soil around nuclear power plants in the United States were indistinguishable from fallout background (EPRI 1981).

For the Radionuclides in Food Program, the FDA determined concentrations of radionuclide activity in their Total Diet Study. Levels of ^{241}Am in both the 1983–1986 and 1987–1992 studies were sufficiently low as to require no specific action or simple monitoring (Cunningham et al. 1989, 1994).
The FDA (1998) has published derived intervention levels for ^{241}Am in food at which action should be considered. Dietary values are 2.0 Bq/kg for 3-month-old children, increasing to 17 Bq/kg for 1-year-old children, and then decreasing to 8.8 Bq/kg for adults. Since 1973, background fallout levels of ^{241}Am in air have been <1 aCi/m^3 (1 aCi=10^{-18} Ci) (0.037 μBq/m^3) and are continuing to decline (Bennett 1979). Levels in tap water are sufficiently low that levels must be estimated (Bennett 1979).

UNSCEAR (2000) reported that deposition and air concentrations of radionuclides from fallout dropped rapidly once atmospheric testing ceased in 1980. Inhalation exposures for even long-lived transuranic radionuclides became insignificant after 1985.

Exposure Levels in Humans. Although some data on the levels of americium in human tissues exposed to natural background levels (food, water, and air) are available, few measurements have been made on the americium content in human tissues. The principal source of information about occupationally exposed individuals is the USTUR database, established to document levels and distribution of uranium and transuranium isotopes in human tissues for occupationally exposed workers (USTUR 1999). 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). The DOE has developed the Comprehensive Epidemiologic Data Resource (CEDR) Program to provide public access to health and exposure data concerning DOE installations. In addition, studies relating to populations residing near DOE installations, and other studies of radiation exposures and health effects, such as atomic bomb survivors, are included in CEDR (CEDR 2000). Additional research is needed to reassess the magnitude of any health risks associated with exposure to americium from smoke detector damage under realistic scenarios.

Exposures of Children. Children will be exposed to americium in the same manner as adults in the general population (i.e., ingestion of food and water and inhalation of air). Americium is potentially found at hazardous waste sites at elevated levels. Since children may have oral exposure to soil through hand-to-mouth activity, bioavailability studies of americium in soil via the oral route may be useful to assess the risk of this type of exposure.

Child health data needs relating to susceptibility are discussed in 3.13.2 Identification of Data Needs: Children's Susceptibility.

Exposure Registries. Americium is not currently one of the chemicals for which a subregistry has been established in the National Exposure Registry. Americium will be considered in the future when chemical selection is made for subregistries to be established. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to the exposure to americium.

Other exposure registries are available for americium. The USTUR established a database to document levels and distribution of uranium and transuranium isotopes in human tissues for occupationally exposed workers who donate their bodies to science (USTUR 1999). The DOE has developed the CEDR Program to provide public access to health and exposure data concerning DOE installations. In addition, studies relating to populations residing near DOE installations, as well as other studies of radiation exposures and health effects, such as atomic bomb survivors, are included in CEDR (CEDR 2000).

6.8.2 Ongoing Studies

The Federal Research in Progress database lists ongoing studies about environmental effects of americium (FEDRIP 2004). The one current study listed in the FEDRIP database was R.D. Lloyd of the University of Utah at Salt Lake City, studying the consequences of exposure to a bone-seeking radionuclide, ^{239}Pu. These studies will provide new knowledge on the biological behavior of plutonium and other bone-seeking radionuclides, including americium, and improve the capability to predict the risks of exposure. This research is supported by the National Cancer Institute. The International Science and Technology Center (ISTC), headquartered in Moscow, which began operations on March 3, 1994, also is active in research concerning transuranic elements.