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Subsurface Behavior of Plutonium and Americium at Non-Hanford Sites and Relevance to Hanford

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RG Riley

February 2008



Pacific Northwest
NATIONAL LABORATORY

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**Subsurface Behavior of Plutonium
and Americium at Non-Hanford
Sites and Relevance to Hanford**

Letter Report

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1.0 Introduction

A common perception is that Pu is not very mobile under typical subsurface conditions. This perception is based on the fact that Pu is generally highly insoluble (e.g., Knopp et al. 1999; Neck and Kim 2001; Haschke et al. 2002) and sorbs strongly to a variety of minerals, soils, and sediments under typical subsurface conditions (e.g., Keeney-Kennicutt and Morse 1985; Sanchez et al. 1985; Duff et al. 1999; Cantrell et al. 2003). Acidic conditions or high concentrations of chelating agents can increase solubility and reduce adsorption significantly. In some instances, where Pu has been found to be unexpectedly mobile, colloidal transport has been invoked as a likely process (Penrose et al. 1990; Kaplan et al. 1994a; Kersting et al. 1999; Novikov et al. 2006). From the perspective of regulators and other public organizations responsible for the health and safety of the public and environment, unexpectedly high migration of Pu in the past suggests possible unexpected exposure in the future. From the perspective of the responsible parties, this can mean significant financial impact to address required remediation and possible litigation. In such high-profile cases where contaminant migration has been high relative to expectations, it is particularly imperative to have a well-founded understanding of the specific processes responsible for this behavior so that appropriate corrective measures can be taken.

In this document, the transport behavior of Pu is reviewed at several sites within the DOE complex as well as at one U.S. commercial site and one site in Russia. This review focuses primarily on sites at which Pu contamination has occurred and migration of Pu has been observed. Although the primary focus of this review is on Pu, data for Am also are included when available. As with Pu, Am tends to be relatively immobile as a result of its low solubility and high affinity for mineral surfaces under typical environmental conditions. Particular attention is paid to what is currently known regarding the physical and chemical processes related to the mobility behavior of Pu and Am at these sites and to the possible analogies or implications with regard to Pu and Am migration at Hanford. This report is organized by facility at which Pu-containing waste has been disposed and for which site-specific information regarding Pu subsurface behavior has been reported previously.

2.0 Los Alamos National Laboratory

Los Alamos National Laboratory (LANL) was built in the early 1940s and served as the center for design and construction of atomic weapons in the United States. The site is in northern New Mexico and, like Hanford, has an arid climate. The first atomic weapons were built at LANL during World War II, and the laboratory subsequently was used to improve weapons design, build new weapons, and develop nuclear technologies. These activities resulted in environmental contamination at a number of locations throughout the site.

The LANL complex is on the Pajarito Plateau, which slopes eastward from the Jemez Mountains to the Rio Grande Valley (Marty et al. 1997). Erosion has cut the plateau into a series of east-west trending mesas and canyons. At LANL, these canyons were used to dispose of solid and liquid wastes. Treated liquid wastes from the LANL Central Waste Treatment Plant containing traces of Pu and Am were released into Mortandad Canyon beginning in 1963. Surface-water flow in the canyon is ephemeral and drains to the east. Treated effluent discharges produce perennial flow in the middle of the canyon. The water generally evaporates or percolates into the canyon fill within 2 km of the waste discharge point.

Spring snowmelt and periodic storms can produce flow 3.4 km or farther from the outfall. The shallow alluvium in the canyon is composed of lenses of sandy to silty clay materials formed by weathering of volcanic rocks (Bandelier tuff) and contains a small elevated aquifer with a storage capacity of $(20-30) \times 10^3 \text{ m}^3$ (Penrose et al. 1990). The alluvium in the canyon forms a thin ribbon that thickens down-canyon from 0 m in the west to 30 m thick in the east.

Based on adsorption behavior alone, liquid waste containing Pu and Am would be expected to move overland less than a few meters within the soils and sediments below the point of discharge. Contrary to this expectation, traces of Pu have migrated as far as 3.4 km from the waste outfall within the canyon. As a result, this site has received considerable attention (e.g., Purtymun et al. 1977; Penrose et al. 1990; Marty et al. 1997; Smith and Amonette 2006). Several early studies concluded that surface-deposited Pu and Am moved downslope primarily through erosion and redeposition of the contaminated soils and sediments (Abrahams et al. 1962; Dahlman et al. 1980; ESG 1985). In a later study, Penrose et al. (1990) found that all of the Pu and part of the Am in groundwater samples were associated with colloidal material in the particle size range of 25 to 450 nm. They suggested that the distant migration of Pu and Am observed in the canyon was the result of colloidal transport (Penrose et al. 1990). Penrose's work has been cited frequently as an example of the significant potential of colloidal transport in contaminant migration (Ryan and Gschwend 1990; Choppin 1992; Gaffney and Marley 1992; Puls and Powell 1992; Gounaris et al. 1993; Litton and Olson 1993; Marley et al. 1993; Nuttal and Kale 1993; Hering and Kraemer 1994; Kaplan et al. 1994a; Kretzschmar et al. 1994; Litaor et al. 1996; Novikov et al. 2006).

In a more recent study, Marty et al. (1997) reexamined available groundwater isotopic data with respect to Pu migration at the Mortandad Canyon site. It was found that ^{239}Pu appeared in one downgradient well before it appeared in wells closer to the point of discharge and that the ^{239}Pu arrived prior to tritium, also present as a contaminant in the groundwater. These results suggest that if Pu were moving through the aquifer as soluble Pu, it would have had to move twice as fast as groundwater. It was determined also that ^{239}Pu arrived in the surface alluvium and groundwater simultaneously and that the isotopic composition of Pu in the alluvium and groundwater were identical. These results demonstrated that Pu did not move down-canyon through groundwater and that the potential for Pu migration by colloidal transport at Mortandad Canyon is likely overstated. It would appear that erosion and redeposition of Pu contaminated soils and sediments is the source of colloidal bound Pu found in the groundwater of Mortandad Canyon.

3.0 Nevada Test Site

The Nevada Test Site (NTS) is located in an arid region about 105 km northwest of Las Vegas. The site was the location of 828 underground nuclear tests conducted by the United States between 1956 and 1992 (DOE 1994). The NTS is in the southern part of the Great Basin, the northern-most subprovince of the Basin and Range physiographic province. The area is geologically complex, encompassing at least six Tertiary-age calderas, many relatively young basin-and-range-style normal faults, and thrust faults and intrusive bodies of Mesozoic age, all superimposed on a basement complex of highly deformed Proterozoic and Paleozoic sedimentary and metasedimentary rocks (Slate et al. 1999).

The NTS is situated within the Death Valley region groundwater flow system, one of the major hydrologic subdivisions of the southern Great Basin (Laczniaik et al. 1996). The depth to groundwater in

the area varies from the surface and near-surface at limited discharge areas in Oasis Valley to more than 610 m below ground surface in the northwestern NTS (Drellack and Prothro 1997). Perched groundwater occurs locally throughout the NTS area, mainly in the volcanic rocks. The lower carbonate aquifer consists of a very thick, widespread section of Paleozoic carbonate rocks and is considered to be the primary regional aquifer in the NTS area, while the Proterozoic and lowermost Paleozoic siliciclastic rock form the regional hydrologic basement (Winograd and Thordarson 1975).

During a nuclear explosion, the energy release is significant and leads to instantaneous mechanical disruption and shock melting of the rock adjacent to the explosion. The extreme temperatures (~1M°C) and pressure (~1M bar) creates a shock wave that propagates outward radially away from ground zero. A cavity is created that expands outward until the internal pressure and momentum of the shock wave are equalized by the confining strength of the surrounding rock. More than 700 metric tons of rock are fused per kiloton of nuclear yield. The melted rock coalesces at the bottom of the standing cavity until, within minutes to hours, the cavity pressure is relieved and the cavity collapses. Rubble invades the still molten glass and the former standing cavity.

Borehole characterization studies have been conducted to investigate the behavior of radionuclides adjacent to the edge of a nuclear explosion cavity, where groundwater can be eliminated as a possible transport process (the nuclear test was conducted above the static water level). Results of these studies indicated that radionuclides were detectable at a maximum of a few hundred meters from the original detonation point (Nimz and Thompson 1992; Smith et al 1996). These radionuclides, including Pu would have been transported at the time of the original detonation by gas movement through fractures or fracture injection of vaporized material.

Based on results of serial filtration of groundwater samples collected downgradient of two underground nuclear tests, it was concluded that more than 99% of the Pu in the samples was adsorbed onto colloidal particles. It was concluded from the isotopic signature of the Pu that the origin of the Pu was the Benham nuclear test site, a distance of 1.3 km from the sample well (Kersting et al. 1999). Colloid-facilitated transport was suggested to be a major factor in the transport of Pu at this site and potentially other sites (Kersting et al. 1999).

Although the work of Kersting and colleagues demonstrated that colloids are present and that Pu is associated with these colloids, it did not prove that colloid-facilitated transport was the process resulting in Pu migration to a distance of 1.3 km from the blast zone (Honeyman 1999). Although the reports of Nimz and Thompson (1992) and Smith et al. (1996) would seem to support the colloidal transport scenario, the issue remains unresolved.

4.0 Rocky Flats Environmental Technology Site

From 1952 to 1989, the Rocky Flats Nuclear Weapons Plant made components for the U.S. nuclear arsenal using various radioactive and hazardous materials. At the end of the nuclear weapons production mission, the area became known as the Rocky Flats Environmental Technology Site (RFETS). Pu and Am are significant contaminants at the site. The most important source of Pu and Am contamination at the site came from leaking drums stored at the 903 Pad (Clark et al. 2006). Eventually, increased concentrations of Pu were measured at various surface-water monitoring locations. These results were in

contrast to expectations of low mobility as a result of the low solubility of Pu and high affinity for natural mineral surfaces. Because of this discrepancy, scientific studies were undertaken to determine how Pu and Am, among other radionuclides, behave in the air, surface water, groundwater, and soil at the RFETS.

Water at Rocky Flats and the surrounding area is distributed among surface water, shallow groundwater, and deep groundwater (Kaiser-Hill 2002). A series of detention ponds has been constructed along creeks to manage plant waste and surface water runoff. Shallow groundwater refers to water within the alluvium and weathered bedrock and is found to a depth of 30 m. Precipitation infiltrates through the vadose zone to recharge the shallow groundwater, which recharges the stream channels at certain times of the year. Beneath the alluvium is highly impermeable bedrock that inhibits vertical flow. As a result, shallow groundwater flows laterally and either discharges into streams or emerges as hillside springs and seeps. Deep regional groundwater occurs at about 200 to 300 m below ground surface. Because of the intervening bedrock, the regional groundwater aquifer is hydrologically isolated from the surface water, groundwater, and contaminants (Hurr 1976).

Based upon existing knowledge of plutonium chemistry, it was expected that Pu solubility would be limited by the formation of amorphous $\text{Pu}(\text{OH})_4(\text{am})$. In addition, transport of Pu at Rocky Flats would occur primarily through migration of fine organic and mineral particles to which Pu(IV) was adsorbed (Clark et al. 2006). Earlier work by Harnish et al. (1994) determined that 65% of the Pu collected from groundwater was associated with particulate and large colloidal size fractions. Harnish et al. (1994) suggested that these colloidal phases could facilitate transport of actinides in groundwater. However, no evidence beyond this was presented to indicate that colloidal transport of Pu through the shallow aquifer was a significant transport flowpath.

Later synchrotron-based x-ray absorption studies determined that the particulate Pu occurs as the relatively insoluble $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (Conradson et al. 2004). Studies conducted by Santschi et al. (2002) demonstrated that Pu and Am were transported from contaminated soils to streams as particulate (greater than $\sim 0.45 \mu\text{m}$) and colloidal ($\sim 0.002 \mu\text{m}$ to $0.45 \mu\text{m}$) phases. In addition, the evidence of Santschi et al. (2002) suggested that the colloidal fraction of Pu occurred as PuO_2 imbedded in or adhered to organic matter containing some iron.

With the knowledge that essentially all the Pu occurs as particulate or colloidal phases, computer modeling was conducted to simulate Pu and Am transport at the site. Approximately 90% of the radioactive inventory was in the uppermost 12 cm of the soil (Litaor et al. 1996). Concentrations in soils and sediments ranged up to a few nanocuries per gram, and concentrations in streams and ponds ranged up to several picocuries per liter. The wind is an important factor in the dispersal of soil and actinides at RFETS. The winds at RFETS blow predominantly from the northwest to the southeast, periodically becoming strong and gusty, exceeding 160 km/h. Air monitoring and geostatistical modeling of the actinide loads indicated that air transport was a dominant actinide migration pathway before and during cleanup.

Additional state-of-the-art process-oriented computer models were used to simulate hillside erosion processes and estimate the spatial and temporal distributions of soil erosion and sediment deposition in stream channels and impoundments. The Water Erosion Prediction Projection (WEPP) model was selected for this work because it can account for enrichment of sediments transported as fine particles. The WEPP model was routed into another model, the U.S. Army Hydrologic Engineering Center sediment-transport model, HEC-6T, which can accommodate up to 100 tributaries flowing into a main

channel. Results of this modeling indicate that up to 99% of Pu in the surface water at RFETS comes from hillside erosion, and colloidal transport is therefore insignificant.

5.0 Idaho National Engineering and Environmental Laboratory

The Idaho National Engineering and Environmental Laboratory (INEEL), originally established in 1949 as the National Reactor Testing Station, is a U.S. Department of Energy-managed reservation that historically has been devoted to energy research and related activities. The INEEL is located in southeastern Idaho and occupies 2,305 km² in the northeastern region of the Snake River Plain. The Snake River Plain Aquifer underlies the Radioactive Waste Management Complex (RWMC) at an approximate depth of 177 m. The RWMC, located in the southwestern quadrant of the INEEL, encompasses a total 7.16 x 10⁵ m² and is divided into three separate areas by function: the Subsurface Disposal Area (SDA), the Transuranic Storage Area, and the administration and operations area (Holdren et al. 2002; EG&G 1989).

The SDA is a radioactive waste landfill. Contaminants in the landfill include hazardous chemicals, remote-handled fission and activation products, and transuranic radionuclides. The most extensively characterized waste site within the SDA is Pit 9. The pit was operational between November 1967 and June 1969, measures approximately 4000 m² in size, and was excavated to basalt located at an average depth of 5.3 m below ground surface and backfilled with approximately 1.1 m of soil before disposal operations commenced. The volume of backfill material was approximately 4.5 x 10⁶ L with an estimated pore volume of 1.4 x 10⁶ L, assuming a porosity of 0.3. Backfill soil was placed over the waste after each disposal and a final soil layer averaging about 1.8 ft thick covered the entire pit when the pit was closed (Holdren et al. 2002).

During an 8-month period in 1968, waste shipments from the Rocky Flats Nuclear Weapons Plant (RFP) in Colorado were transported from the RFP to INEEL and buried in Pit 9. The RFP waste, a major source of waste disposed to the pit, consisted of 208 L steel drums of sludge contaminated with a mixture of transuranic (TRU) elements, metals, and organic solvents. Mean failure rate on the drums was estimated at 22.6 years (Becker 1997). Among the contaminants disposed in Pit 9 were Pu (29 kg), Am (0.94 kg), Texaco Regal Oil (2.22 x 10⁴), carbon tetrachloride (1.39 x 10⁴), organic acids (4.87 x 10³), and ethylenediaminetetraacetic acid (EDTA, 4.87 x 10³). The total volume of liquid wastes disposed to the pit (organic and inorganic co-contaminant waste) was estimated at 2.8 x 10⁵ L (EG&G 1989; Holdren et al. 2002).

During periods of intense precipitation or flooding, ponding occurred at the SDA, allowing the potential for infiltration and possible contact with buried waste. A diversion drainage system was first constructed around the perimeter of the SDA in 1962. The dikes established after the 1962 flood again overflowed during the 1969 flood. Further improvements were made following this flood. As a result of the floods, some water may have entered Pit 9. The diversion drainage system was expected to have prevented any subsequent floodwater from overflowing the trenches, and it is assumed no further potential impacts on release and migration occurred. Subsequent to backfilling, no subsidence has occurred at Pit 9. The INEEL region is classified as arid to semi-arid because of the low average rainfall of 22.1 cm/yr. Net drainage or recharge measured at the SDA ranged from a high of 49.4 cm/yr to less than 0.3 cm/yr (EG&G 1989; Holdren et al. 2002).

The regional subsurface consists mostly of layered basalt flows with a few comparatively thin layers of sedimentary deposits. Layers of sediment, consisting of mixtures of sand, silt, and clay, referred to as interbeds, tend to retard infiltration to the aquifer. Three major interbeds (A-B [9.1 m], B-C [33.5 m], and C-D [73.2 m]) are present in the interval between the surface and the aquifer. The 9.1 m interbed consists of silty clay. The 33.5 m interbed consists of an organic-rich soil horizon intermixed with a calcium carbonate crust. The 73.2 m interbed consists of an intermixture of clay, silt, sand, and gravels (Rightmire and Lewis 1987).

To better understand the potential for migration of contaminants below Pit 9, hydrologic and geochemical characterization measurements have been made on samples from the soil-sediment/basalt interval (Holdren et al. 2002). Table 5.1 summarizes the concentrations of Pu and Am measured in sediments underlying the SDA. Concentrations were very low and comparable to background levels found in surficial sediments.

Table 5.1. Plutonium and Americium Concentrations in the Vadose Zone^(a)

Depth Interval (m)	^{239/240} Pu (pCi/g)	²⁴¹ Am (pCi/g)
0 to 10.7	0.026 to 1.13	0.012 to 9.6
10.7 to 42.7	0.009 to 0.74	0.006 to 0.908
42.7 to 76.2	0.013 to 0.14	0.016 to 0.033
(a) Background concentrations of ²³⁹ Pu in surficial sediments ranged from 0.18 to 0.93 pCi/g. A background level for ²⁴¹ Am was 0.008 pCi/g.		

The SDA soil was determined to have a high clay content (approximately 36%) and a high silt content (approximately 56%). As a result, the soil has very low vertical hydraulic conductivity (approximately 4.7×10^{-6} cm/sec), which would retard the downward migration of liquids from the buried waste (Barraclough et al. 1976). In addition, the clay may retard the migration of radionuclides and chemicals through adsorption. The clay content, mineralogy, and cation exchange capacity of the soils at the SDA imply that moderate retardation of migrating cations is possible. Sediments may have been compacted due to significant overburden pressures (Holdren et al. 2002). Under unsaturated conditions with limited water, flow will move preferentially through small openings in sediment or basalt, bypassing large openings. Flow through the basalt layers is reduced as a result of open fractures being filled with clay particles carried by infiltrating water.

Both batch and column experiments have been performed to determine K_{ds} for Pu and Am in SDA vadose zone sediments (Holdren et al. 2002). Plutonium (total) and Am showed an enhanced mobility fraction of about 2% with K_{ds} in the range of about 0.02 to 0.25 for crushed basalt. Although no direct observations were made, this mobile fraction has been attributed to colloidal transport (Miner et al. 1982; Newman et al. 1996; Fjeld et al. 2000). The remaining 98% of the Pu and Am had a retardation of about 60. Interbed sediment K_{ds} were greater than 48 for Pu and Am. Oxidation state determinations on Pu in batch experiments showed that when the solid-to-solution ratio in experiments more closely resembled natural conditions in the vadose zone, Pu(V) and Pu(VI) were reduced to Pu(IV). The Pu(IV) K_d was >250 and 8.8 to 20 in the presence of EDTA. Historically, assessment modeling has used conservative values of K_d (i.e., 5100 for Pu and 450 for Am).

It has been previously estimated that 4.9% (55.5 kg) of the plutonium (as PuO₂) disposed at the SDA was colloid-size in nature. Under flooding and snowmelt conditions, it has been hypothesized that this colloidal fraction would pass through fractured basalt and sorb to interbed sediments underlying the SDA. Once sorbed, the plutonium would not be remobilized in a colloid form. Rather, remobilization would occur through dissolution and transport in the dissolved phase (Batcheller and Redden 2004). There have been no direct measurements of PuO₂ colloids along subsurface flow paths below the SDA. Plutonium colloids were not clearly evident in water samples collected from below the INEEL (Cleveland and Rees 1982).

The potential for transport of contaminants to the subsurface underlying Pit 9 has been primarily by natural infiltration under unsaturated conditions. A possible exception was during the time of the past flooding event and periodic precipitation events that might have resulted in enhanced infiltration of water to the pit. Concentrations of Pu and Am measured in the vadose zone underlying the SDA suggest little migration of these contaminants from any of the waste sites within the SDA, including Pit 9. Contaminant levels detected are comparable to background levels of contamination measured in surface soils. It has been suggested that some contamination may have been introduced artificially during past drilling operations.

The absence of significant Pu and Am in the near-subsurface sediments suggests that the 1.1 m layer of backfill soil placed above the basalt at the bottom of Pit 9 prior to waste disposal and other backfill soil surrounding the waste drums have served as a significant barrier to migration. This barrier persists despite the fact that large quantities of organic acids and EDTA are present in the waste that could complex with Pu and Am and potentially enhance their mobility. A laboratory column experiment showed Pu migration in SDA sediment to be enhanced by a factor of ten or more in the presence of EDTA (K_d in the range of 8.8 to 20) but still significantly less than highly mobile species (Holdren et al. 2002). Decades of slow release of waste from the drums to adjoining backfill soil have not exceeded the capacity of the soil pore space to accommodate the liquid waste volumes disposed to the pit (i.e., liquid waste disposed amounts to approximately 20% of the pore volume capacity of the backfill material). Slow release, low recharge rates, and low hydraulic conductivity, coupled with a high sorption capacity of the backfill soil, could explain the absence of Pu and Am migration. Small amounts of a fast-release fraction of Pu and Am in SDA sediment column studies have been identified and suggest some potential for subsurface colloidal transport of these contaminants. The possibility of such historical occurrence in the vadose zone under Pit 9 appears low when considering the very low concentrations of Pu and Am measured in the SDA subsurface in past years.

6.0 Savannah River National Laboratory

The Savannah River Site is located in the Upper Coastal Plain east of the Appalachian Mountains of western South Carolina near Aiken. The site was constructed in the early 1950s for production of nuclear material, mainly ³H and ²³⁹Pu for defense purposes but also ²³⁸Pu and various transplutonium radionuclides for medical, industrial, and scientific applications (Dai et al. 2002). The facilities that produced these radionuclides have been shut down or placed on standby status, and the main mission at the site has been directed toward cleanup of chemical and radioactive contamination that resulted from the radionuclide production operations.

Beginning in late 1954, the plutonium and uranium extraction (PUREX) process was used in the F Area to recover ^{239}Pu , ^{237}Np , and ^{238}U from irradiated ^{238}U targets. The seepage basins in this area received waste effluent from the separation facilities, including waste from the nitric acid recovery unit and the evaporators that concentrated the dilute uranium nitrate solutions. Discharges to the F-Area seepage basins were stopped in 1988, and low-permeability clay closure caps were placed over the basins in 1990.

Aqueous wastes from a nuclear materials processing facility on the Savannah River Site were released over a 33-year period to unlined seepage basins (Kaplan et al. 1994a). The wastes released to the seepage basins contained high concentrations of acids (pH 2.9), Na (790 mg/L), tritium (27 $\mu\text{Ci/L}$), and trace amounts of numerous actinides and metals. A number of radionuclides, including Pu and Am and metals, have been reported to be moving rapidly through the aquifer underlying the seepage basins (Looney et al. 1987, 1988). The surface aquifer is located in the Barnwell formation, which has a sandy-clay and clayey-sand texture. Kaplan et al. (1994a) investigated the association of actinide elements with groundwater colloids, a potential cause for the apparent enhanced transport. Kaplan and colleagues found that the ratio of filterable to nonfilterable activity increased for Pu and Th in groundwater as the distance from the seepage basins increased. For Am, Cm, and Ra, this ratio decreased as the distance from the seepage basins increased. At the most distant sampling point (0.55 km), all the Pu and Th and none of the Am and Cm were in the filterable fraction. Based on these results, Kaplan et al. (1994a) concluded that Pu had a much greater potential than Am or Cm for moving through the study site in association with mobile colloids.

In a later study, Dai et al. (2002) used cross-flow ultrafiltration to determine that less than 4% of the Pu was associated with colloids in groundwater collected from the same site. Dai et al. (2002) attributed this major discrepancy in results to sampling artifacts that likely occurred in the Kaplan et al. (1994a) study. In the Kaplan et al. (1994a) study, the ultrafiltration work took up to 3 days and likely resulted in exposure to ambient air, resulting in the formation of colloidal iron during sample processing (Hasselov et al. 2001; Dai et al. 2002). Other differences in equipment and protocols also may have contributed to the contradictory results found in the two studies. Dai et al. (2002) also applied isotopic analysis techniques in their study. From the results of the Dai et al. work, it was determined that Pu migration from the seepage basins was minor and that Pu found downstream of the seepage basins was the result of preferential migration of transplutonium isotopes (mainly ^{244}Cm), which decays to yield ^{240}Pu .

7.0 Maxey Flats, Kentucky

Maxey Flats, an eroded plateau in the knobs region of northeastern Kentucky (Dayal et al. 1986a), contains a commercial shallow land burial site that was used for disposal of low-level radioactive waste. The plateau rises approximately 90 to 120 m above the surrounding alluvium-filled valley. The climate is humid continental, with an annual mean precipitation of 110 cm and sharp contrasts between winter and summer months.

At the site, burial trenches were excavated in the Nancy Member of the Borden Formation to a typical depth of 7.5 m. The site began operations in 1963 and was closed in 1977. The upper 0.3 to 7.5 m of the site are composed of weathered shale. Approximately 80% of the rocks beneath the site are shale; the remainder is sandstone. The bedrock has an extremely low hydraulic conductivity and, as a result,

essentially all groundwater flow occurs through fractures and joints. Zehner (1983) estimated the maximum potential discharge from bedrock in the drainage below the site to be 0.25 cm/yr (~5% of the mean annual base flow). The remaining ~95% of the groundwater discharge occurs in the colluvium and alluvium that cover the valley slopes and bottom.

Most of the wastes buried at the site are believed to be large-volume, low-level radioactive solid wastes such as paper trash, decontaminated materials, solidified cleanup solutions, packing materials, broken glassware, plastics, protective clothing, radioactive carcasses of experimental animals, and contaminated equipment. Approximately 104,000 m³ of low-level radioactive wastes were buried at the site (Meyer 1976). Higher-activity wastes such as sealed sources, reactor resins, filters, and irradiated reactor parts, also were buried at the site. It is estimated that 80 kg of ²³⁹Pu and significant quantities of ²³⁸Pu were disposed at the site (Clark 1973).

Unsegregated, poorly packaged, and unstabilized wastes resulted in cap subsidence and significant infiltration of water into the trenches. Infiltration of water has resulted in migration of contaminants away from the trenches. A humid climate, combined with rapid infiltration and a layer of bedrock of low permeability underlying the trenches, eventually resulted in saturated conditions within the trenches (the so-called bathtub effect). This bathtub effect allowed for prolonged contact of water with the wastes and reactions to occur (e.g., microbial degradation), leading to the formation of leachates highly enriched in contaminants. Microbial degradation resulted in anoxic conditions and leachates that contained elevated concentrations of Na⁺, K⁺, Fe, Mn, Cl⁻, dissolved organic and inorganic carbon, ammonia, several organic compounds, and a number of radionuclides, including ³H, ²⁴¹Am, ⁶⁰Co, ¹³⁴Cs, ¹³⁷Cs, ⁹⁰Sr, ²³⁸Pu, and ^{239,240}Pu. The organic compounds present were a mixture of those of waste origin and those produced from anaerobic degradation of waste.

Of the organic compounds present, EDTA was of particular interest. EDTA is a strong chelating agent that forms very stable complexes with many actinide elements and transition metals, resulting in increased solubility and reduced adsorption. It is believed that the EDTA complexed with Pu and other radionuclides, resulting in elevated concentrations of radionuclides in the leachates (Cleveland and Rees 1981; Means and Alexander 1981; Polzer et al. 1982; Dayal et al. 1986a, 1986b). Dissolved concentrations of ²³⁸Pu in the range of 600–16,100 pCi/L had been measured in trench leachates collected from the site (Cleveland and Rees 1981; Means and Alexander 1981). It is hypothesized that complexed Pu and other radionuclides migrated away from the trenches contaminating the surrounding aquifer. This hypothesis was based on trace levels of both ²³⁸Pu and ²³⁹Pu that were detected in groundwater hundreds of meters from the original solid-waste disposal trenches.

8.0 Mayak Production Association, Russia

The Mayak Production Association was commissioned in 1948 in the northern part of the Chelyabinsk region of Russia for production of nuclear weapons material (Myasoedov and Drozko 1998; Christensen et al. 1997). The plant had five special nuclear reactors for production of ²³⁹Pu and a facility for separation of weapons-grade Pu. Very large quantities of radioactive wastes (76,000,000 m³) resulted from the Pu production processes. Production of weapons-grade Pu was terminated in 1987. The activities of the plant led to substantial radioactive contamination to both the immediate environment and large areas several tens of kilometers distant. In addition, considerable quantities of radioactive wastes

remain stored at the Mayak site. Major factors that have led to the environmental contamination include discharge of large quantities of liquid radioactive waste into the Techa River of the Ob River system from 1949 through 1956; an explosion in 1957 of a tank containing high-level liquid radioactive waste, leading to contamination of an area of approximately 20,000 km²; and aeolian resuspension of radioactivity from the shores of Lake Karachay in 1967, which was in use as a storage pond for liquid radioactive waste.

Since 1951, liquid intermediate-level waste was discharged to Lake Karachay, a lake with no outlet. The total activity discharged to the lake was estimated to be 120 x 10⁶ Ci. Current discharges are approximately 270 x 10³ Ci/yr. Most of the activity is currently due to ⁹⁰Sr, ¹⁰⁶Ru, and ¹³⁷Cs. Because the lake has no outlet, soluble radionuclides and chemicals infiltrate the sediment bed and act as a source of groundwater contamination.

Novikov et al. (2006) studied the transport of Pu in the far-field at the Mayak Production Association. Weakly alkaline NaNO₃ brine solutions contaminated with ⁹⁰Sr, ¹³⁷Cs, ²⁴¹Am, and ²³⁹Pu were discharged into Lake Karachay (Myasoedov and Drozko 1998; Christensen et al. 1997). The lake is connected to an aquifer composed of fractured metavolcanic rocks. Plutonium concentrations were determined to be approximately 27,000 pCi/L at the source (Lake Karachay) and 4.3 pCi/L in groundwater 4 km from the source. The Pu measured in groundwater was found to be 70 to 90 mole percent sorbed onto colloids. Novikov et al. (2006) asserted that this confirmed that colloids were responsible for long-distance transport of Pu. To the contrary, these results suggested evidence but not proof of colloid-facilitated transport of Pu in the groundwater.

An alternative explanation for the Pu migration in groundwater is complexation with oxalate. Concentrations of oxalate in the waste effluents ranged from 0.9 to 14 g/L. If oxalate were diluted in a similar fashion to nitrate following waste discharge, it is estimated that oxalate concentrations in groundwater 4 km from the source would be as high as 0.003 molar. Such high oxalate concentrations would be more than sufficient to complex Pu levels present in groundwater. Ternary complexes [Pu(IV)-oxalate-OH] are likely to be the most important Pu-oxalate complexes to form at neutral to alkaline pH values. Unfortunately, stability constants for Pu(IV)-oxalate-OH complexes are not available (Hummel et al. 2007).

9.0 Summary and Implications for Hanford

Seven sites at which Pu release to the environment has raised significant environmental concerns have been reviewed. A summary of the most significant contaminant release events, site features and processes, and far-field impacts relevant to Pu migration at the seven sites is presented in Table 9.1. At four of these sites (LANL, NTS, SRS, and Mayak), colloidal transport through groundwater has been implicated as the primary mechanism responsible for unanticipated migration of Pu. For two of these sites (LANL and SRS), later studies demonstrated that colloidal transport was not the process responsible for the Pu migration at the site. Erosion and redeposition of Pu contaminated soils and sediments was the mechanism of Pu transport at LANL. Transport and decay of Cm was determined to be the source of Pu at the SRS site review. Only circumstantial evidence was presented to support the case for colloidal transport of Pu through groundwater (the presence of Pu on colloids was demonstrated) at the NTS and Mayak sites. In these two cases, alternative explanations are possible for the observed migration behavior of Pu. Colloidal transport through groundwater also was suggested as a possible process for Pu transport

through groundwater at Rocky Flats (Harnish et al. 1994). Later work indicates that this pathway is not likely to be significant at Rocky Flats, but rather wind driven re-suspension and hillside erosion were dominant transport mechanisms for Pu (Santschi et al. 2002; Clark et al. 2006). Pu migration at Maxey Flats is due primarily to increased solubility and reduced adsorption of Pu as a result of complexation with EDTA. Significant migration of Pu through the vadose zone to groundwater at INEEL appears to not have occurred.

The sources, processes, and pathways of migration of Pu and Am (when available) at the seven sites reviewed in this study have little in common with the 200-PW-1/3/6 Operable Unit site at Hanford, where Pu and Am had migrated to unanticipated depths in the subsurface. This deep migration is due primarily to the unique features of the organic wastes disposed at 200-PW-1/3/6 Operable Units that do not occur at any of the other sites considered in this review. The potential for colloidal transport is sometimes a source of uncertainty with respect to future Pu migration at Hanford. Our review suggests that colloid-facilitated transport has generally been overstated in site assessments described in this study. This position also is supported by Hanford-specific studies demonstrating that colloid facilitated transport of highly sorptive contaminants in groundwater is minimal (Flury et al. 2002; Cherrey et al. 2003; Zhuang et al. 2003, 2004; Marshal et al. 2004; Dai et al. 2005; Chen and Flury 2005; Chen et al. 2005; Czigany et al. 2005). Colloid-facilitated transport of highly sorptive contaminants in the vadose zone would be expected to be even less than in saturated groundwater, due to the much higher ratio of surface area to water volume and thin water-film thicknesses, which would be conducive to filtration of particles from solution.

Table 9.1. Contaminant Release Events, Site Features and Processes, and Far-Field Impacts Relevant to Pu Migration at the Reviewed Sites

Waste Site	Contaminant Release Event	Features and Processes	Far-Field Impact
Los Alamos National Laboratory– Mortandad Canyon	Waste discharge to canyon	Waste releases of low volume; ephemeral surface water flow; erosion, redeposition, and infiltration influenced by episodic rainfall and snowmelt events ^(a)	Trace Pu detected in groundwater 3.4 km from the point of waste discharge
Nevada Test Site – Benham Nuclear Test	Nuclear device detonation	Near-field (few hundred meters)-gas or vaporized contaminant movement through fractured subsurface material; Far-field (1.3 km)-transport process unknown ^(b)	Trace Pu detected in groundwater 1.3 km from test
Rocky Flats Environmental Technology Site – 903 Pad	Waste leakage from drums	Surface transport via wind-driven resuspension/ deposition of particulates; hillside erosion with particulate and colloidal transport in surface water; infiltration influenced by episodic rainfall and snowmelt events ^(c)	Surface-water contamination; trace contamination of shallow groundwater aquifer
Idaho National Engineering and Environmental Laboratory – Pit 9	Waste leakage from drums	Liquid waste volume disposed did not exceed pore volume capacity of backfill material; infiltration influenced by episodic rainfall and snowmelt events ^(d)	No significant Pu or Am migration in subsurface
Savannah River Site – F-Area Seepage Basins	Waste discharge to seepage basins	Infiltration influenced by episodic rainfall events; acidic waste resulted in increased radionuclide mobility ^(e)	Trace Pu detected 0.55 km down gradient of basins (from transport and decay of Cm)
Maxeys Flats, Kentucky – Low-Level Waste Burial Ground	Leachate release from burial ground trenches	Saturated wastes (bathtub effect); chemical/biological reactions of waste with infiltrating water; radionuclide complexation with EDTA ^(f)	Trace Pu detected hundreds of meters away from burial grounds
Mayak Production Association, Russia – Lake Karachay	Waste discharge to lake	Lake interconnected with aquifer; process leading to far-field Pu migration hypothesized to be complexation with oxalate ^(g)	Trace Pu detected in groundwater 4 km from source
<p>(a) Colloidal transport of Pu in groundwater unlikely. (b) The issue of colloidal transport of Pu in groundwater remains unresolved. (c) No evidence of colloidal transport of Pu in surface or groundwater. (d) Potential of colloidal transport is limited by entrapment in interbed sediments. (e) Significant colloidal transport of Pu appears unlikely. (f) Colloidal transport of Pu not identified as an important process in groundwater transport. (g) Evidence but no proof of colloid-facilitated transport of Pu in groundwater.</p>			

10.0 References

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