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ANALYSIS OF POTENTIAL GROUNDWATER CONTAMINATION IN THE
VICINITY OF THE WELDON SPRING RAFFINATE PITS SITE,
WELDON SPRING, MISSOURI

by

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FOREWORD

This report presents the results of one of several waste-management and environmental studies underway or planned for the U.S. Department of Energy's Weldon Spring, Missouri, Raffinate Pits (WSRP) property. As part of a scoping assessment of environmental impacts associated with storage of radioactive materials at the site, this report focuses on the migration of radionuclides contained in materials currently stored in the pits in their existing conditions. The potential benefits associated with engineering improvements that might be undertaken at the site in the future will be described in a later report, following completion of engineering studies currently being conducted by Bechtel National, Inc. (BNI). This report is not meant to be a definitive statement on the adequacy of the raffinate pits for long-term management of radioactive materials; rather it is meant to analyze, in a rather broad sense, the confinement capabilities of these pits.

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OBJECTIVE

Potential contamination of groundwater in the vicinity of the Weldon Spring Raffinate Pits site was analyzed as part of a scoping assessment of environmental impacts associated with the storage of radioactive materials at this site. The objective of this analysis was to provide an estimate of the extent of radionuclide migration associated with existing conditions at the Raffinate Pits site, using readily available environmental data. The analysis presented in this report is limited to quasi steady-state evaluations; potential contamination of groundwater by catastrophic events such as earthquakes is not considered.

DESCRIPTION OF THE RAFFINATE PITS SITE

SITE LOCATION

The Weldon Spring Raffinate Pits site is a U.S. Department of Energy (DOE) surplus facility located in St. Charles County, Missouri, about 23 km (14 mi) southwest of St. Charles and 48 km (30 mi) west of St. Louis (Figure 1). The site is situated near State Route 94, about 3 km (2 mi) southwest of the junction of State Route 94 and U.S. Routes 40 and 61. The site occupies about 21 ha (52 acres) and includes four raffinate pits (Figures 2 and 3) that contain radioactive residues from past uranium and thorium refining and metal production activities at the Weldon Spring Chemical Plant. The site is completely surrounded by land controlled by the U.S. Army--it is adjoined on the west by a large Army Reserve and National Guard training area and on the east by the Weldon Spring Chemical Plant (Natl. Lead Co. Ohio 1977).

DESCRIPTION OF RAFFINATE PITS

The four raffinate pits occupy about one-half of the 21-ha (52-acre) Raffinate Pits site and have a total design volume (waste-holding capacity) of

Figure 1. Location of the Weldon Spring Raffinate Pits Site.

Figure 2. Location of Weldon Spring Raffinate Pits and Weldon Spring Chemical Plant. Source: National Lead Company of Ohio (1977).

Figure 3. Layout of Weldon Spring Raffinate Pits.
Source: Bechtel National, Inc. (1983a).

approximately 500,000 m³ (650,000 yd³). The pits were constructed by excavating into the existing clay formation and using the removed clay for construction of the levees surrounding the pits. Raffinate Pits 1 and 2 were constructed in 1958, side by side, on nearly level terrain; each pit covers an area of about 0.5 ha (1.2 acres) with a depth of about 2.9 m (9.6 ft). The levees of these two pits are approximately 1 m (3 ft) above the surrounding grade. Each of the two pits has a design volume of approximately 14,000 m³ (19,000 yd³) and contain about 13,000 m³ (17,000 yd³) of radioactive residues from past uranium refining and metal production operations at the Weldon Spring Chemical Plant (Natl. Lead Co. Ohio 1977; Bechtel Natl. Inc. 1983a).

Pit 3 was constructed in 1959 with a design volume of approximately 130,000 m³ (170,000 yd³), a surface area of approximately 3.4 ha (8.4 acres), and a depth of about 3.7 m (12 ft). The natural terrain slopes downward toward the west boundary so that the levees around Raffinate Pits 3 and 4, although effectively at the same elevation as those around Pits 1 and 2, are in fact much higher with respect to the original grade. A portion of the levee in the northeast corner of Pit 3 was constructed on existing terrain so that the levee is about 7 m (23 ft) above original grade in that area. Pit 3 contains approximately 99,000 m³ (130,000 yd³) of radioactive residues from past uranium refining and metal production operations at the Weldon Spring Chemical Plant (Natl. Lead Co. Ohio 1977; Weidner and Boback 1982; Bechtel Natl. Inc. 1983a).

Pit 4 was constructed in 1964 with a design volume of about 340,000 m³ (440,000 yd³), a surface area of about 6 ha (15 acres), and a depth of about 5.6 m (18 ft). The east levee of Pit 4 is common to the west levee of Pit 3. The elevations of the tops of the levees of Pits 3 and 4 are the same. The west levee of Pit 4 extends about 11 m (35 ft) above the existing grade at its maximum elevation. Approximately 43,000 m³ (56,000 yd³) of radioactive materials are stored in Pit 4, and the residue fill is irregular. Overflow from Pit 3 is designed to flow into Pit 4 through a connecting pipe 2 m (7 ft) below the top of the common levee (Natl. Lead Co. Ohio 1977; Weidner and Boback 1982; Bechtel Natl. Inc. 1983a).

The radioactive materials in the four pits are predominantly raffinates from refinery operations and magnesium fluoride slurry from uranium recovery processes at the Weldon Spring Chemical Plant. Approximately 70% of the contents of Pits 1, 2, and 3 consists of neutralized raffinates from refinery operations. These residues were pumped as a slurry to the pits where the solids settled out and the supernatant liquor overflowed into the plant process sewer. The solids remaining in the pits were principally acid-insoluble compounds in the feed materials (mainly silica) as well as hydroxides and precipitates formed when the raffinates were neutralized with lime. The remaining 30% of the contents of Pits 1, 2, and 3 consists primarily of washed slag residues from uranium metal production operations. Magnesium fluoride slag was produced as a scrap during the reduction of uranium hexafluoride to metallic uranium with magnesium chips and, as generated, contained approximately 5% uranium. This uranium was leached from the slag with nitric acid, then washed to remove the remaining soluble uranyl nitrate. The washed slag, essentially uranium-free, was pumped as a slurry to the pits, with the solids retained in the pits as a settled sludge. Pit 4 contains uranium residues similar to those in Pits 1, 2, and 3 as well as raffinate solids from the processing of thorium recycle materials. Pit 4 also contains drums and rubble dumped during partial decontamination of the adjacent Weldon Spring Chemical Plant (Natl. Lead Co. Ohio 1977).

The radioactive contaminants in the four raffinate pits belong to the uranium-238 and thorium-232 decay chains (Figures 4 and 5). The materials in these pits resulted from processing refined uranium ore (yellowcake) and recycled uranium and thorium materials. The radionuclides in these two decay chains are in a state of disequilibrium with the parent radionuclides as a result of these processing operations. Preliminary results of a characterization program of the contents of the four raffinate pits are given in Table 1. These concentrations are used in the analyses that follow.

NOTES:

Only the dominant decay mode is shown.
 The times shown are half-lives.
 The symbols α and β indicate alpha and beta decay.
 An asterisk indicates that the isotope is also a gamma emitter.

Figure 4. Uranium-238 Decay Series.
 Source: Argonne National Laboratory (1982).

Figure 5. Thorium-232 Decay Series.
 Source: Modified from
 Momeni (1981).

Table 1. Concentrations of Major Radionuclides in the Materials Currently Being Stored in the Four Raffinate Pits

Radionuclide	Concentration (pCi/g) in Each Pit			
	Pit 1	Pit 2	Pit 3	Pit 4
Uranium-238	710	470	520	620
Uranium-234	810	560	570	610
Thorium-232	100	120	120	120
Thorium-230	24,000	24,000	14,000	1,600
Radium-228	850	200	100	60
Radium-226	430	440	460	11

Source: Adapted from Bechtel National, Inc. (1983c).
The data in this reference include error bars that are not reproduced here.

PHYSICAL CHARACTERISTICS OF RAFFINATE PITS AREA

SITE STRATIGRAPHY AND LITHOLOGY

The Weldon Spring area is covered by several unconsolidated sedimentary units consisting of alluvium, glacial drift, and residuum from weathering rock to a thickness of 5.2 to 15 m (17 to 50 ft). A generalized description of these units is presented in Table 2. It has been estimated that approximately 6 to 12 m (20 to 40 ft) of silty clays underlie the raffinate pits (Bechtel Natl. Inc. 1983b, 1984).^{*} These silty clays thicken across the Raffinate Pits site from the northwest to the southeast. The silty clays overlie an additional 1 m (3 ft) of clay and weathered limestone residuum of the Burlington-Keokuk Formation (Natl. Lead Co. Ohio 1977). This formation is approximately 49 m (160 ft) thick beneath the pit area (Lenhard et al. 1967)[†]. A generalized geologic cross section of this area is given in Figure 6.

^{*}This estimate was made by Bechtel National, Inc. by extrapolating results from borehole investigations outside the actual pits area. No direct measurement of clay thickness beneath the raffinate pits has been conducted to date.

Table 2. Description of Unconsolidated Overburden Units at Weldon Spring

Unit	Description	Thickness	
		meters	feet
Topsoil	Sandy clay; black-brown; organic	0.2 - 1.1	0.5 - 3.5
Modified loess	Clayey silt; mottled gray-yellow-orange; becomes dense and plastic with depth; manganese-stained.	0.8 - 3	2.5 - 10
Clay (Ferrelview Formation)	Clay; mottled gray-dark yellow-orange; plastic; manganese-stained; contains weathered iron nodules.	0 - 3	0 - 10
Clay till	Clay; yellow-brown; plastic; blocky fractures; manganese-stained; contains sand- to pebble-sized quartz, granitic, and chert grains.	0.3 - 11	1 - 37
Basal till	Sandy, clayey silt; yellow-brown; abundant in broken chert nodules loosely bound by matrix.	0.3 - 1.5	1 - 5
Cherty clay	Clay matrix with abundant chert; multicolored in brown, red, orange and yellow; very dense.	1.1 - 4.6	3.5 - 15

Source: Bechtel National, Inc. (1984).

Adjacent to the raffinate pits, the gently rolling terrain slopes and drains naturally toward the west and north across the Army Reserve and National Guard training area and the August A. Busch Wildlife Area (Figure 2). No streams are present onsite, but incipient drainageways convey surface water runoff to offsite streams. Most surface drainage from the area discharges via an intermittent stream in the Army Reserve and National Guard training area or into Ash Pond on the adjacent Weldon Spring Chemical Plant property. Discharges from the intermittent stream and Ash Pond combine near County Road D; flow northward to Schote Creek, which enters Dardennes Creek at a point about 6 km (3.8 mi) northeast of the raffinate pits; and then eventually reach the

Figure 6. Generalized Geologic Cross Section in the Vicinity of the Weldon Spring Raffinate Pits Site. Source: Lomenick (1982).

Mississippi River north and west of St. Louis, Missouri (Natl. Lead Co. Ohio 1977; Bechtel Natl. Inc. 1983a, 1984).

SITE HYDROGEOLOGY AND WATER SUPPLY

Groundwater investigations in the vicinity of the Raffinate Pits site were conducted as early as the mid-1940s by the U.S. Geological Survey (Fisher and Williams 1944). Roberts and Theis (1951) reported that a groundwater divide exists near the site. In addition, it has been noted that the groundwater table contour in the vicinity of the Raffinate Pits site generally follows the land surface topography (Lawrence Berkeley Lab. 1980). Based on this information, a hydraulic gradient of approximately 0.01 can be calculated from U.S. Geological Survey topographic maps. Bechtel National Inc. (1983b, 1984) reports that the groundwater flow was in the northerly direction with a gradient of 0.095 during the winter of 1982-1983. The groundwater table at this site is reported to be in Mississippian limestones, 18 m (60 ft) below the pit bottoms (Lenhard et al. 1967). Surface elevations across the site range from 188 to 200 m (620 to 665 ft) mean sea level (MSL). A recent study indicates that the depth to groundwater may be somewhat less; groundwater elevations in the winter of 1982-1983 ranged from 177 to 187 m (580 to 615 ft) MSL (Bechtel Natl. Inc. 1983b, 1984), about 8 to 18 m (25 to 60 ft) below ground surface. Groundwater in the vicinity of the raffinate pits may also occur under localized perched conditions in the various unconsolidated geologic units (Bechtel Natl. Inc. 1983a).

The clays underlying the pits are highly impermeable, with reported permeabilities ranging from 2.9×10^{-6} cm/s (1.1×10^{-6} in./s) (Lenhard et al. 1967) to 3.2×10^{-7} cm/s (1.3×10^{-7} in./s) (Natl. Lead Co. Ohio 1977) to 1×10^{-8} or 1×10^{-9} cm/s (4×10^{-9} or 4×10^{-10} in./s) (Lomenick 1982). A recent study of the site indicated that the permeability of the clay overburden ranges from 1.6×10^{-8} cm/s (6.3×10^{-9} in./s) to 3×10^{-6} cm/s (1.2×10^{-6} in./s) and that lenses of more permeable materials are not present (Bechtel Natl. Inc. 1983b, 1984).

The residuum underlying the silty clay is slightly more permeable than the clay materials, and the underlying Mississippian limestones have high

permeabilities associated with their solution cavities and fractures (Lenhard et al. 1967). The Mississippian and Ordovician units are hydraulically connected and are unconfined aquifers. A molecular diffusion coefficient of $0.65 \times 10^{-10} \text{ m}^2/\text{s}$ ($7.0 \times 10^{-10} \text{ ft}^2/\text{s}$) and a tortuosity of 0.67 are used to describe the Raffinate Pits site because no other more appropriate data are available (these data were obtained for the nearby Weldon Spring Quarry site [Lawrence Berkeley Lab. 1981--Table 3]). As such data become available for the Raffinate Pits site, they will be incorporated into the analysis. The effective porosity of clayey soils generally ranges from 0.03 to 0.1 (Gilbert et al. 1983). The vertical dispersivity for clay can be expected to range from about 0.01 to 0.1 m (0.03 to 0.3 ft); the longitudinal dispersivity will be about 10 times larger (Gilbert et al. 1983).

Approximately 97% of the total water use in Jefferson, St. Charles, and St. Louis counties is derived from the Missouri and Mississippi rivers, 2% from alluvial aquifers in the Missouri and Mississippi floodplains, and 1% from bedrock aquifers. Most of the public wells in St. Charles County draw from alluvial aquifers (Miller et al. 1974). Major potable groundwater supplies are obtained from the Sulfur Springs (Bushberg sandstone) strata (Figure 6) located at a depth of about 110 to 120 m (350 to 400 ft) below the ground surface. The main source of soft water for the area is the artesian St. Peter Sandstone aquifer located 210 m (700 ft) below the pit bottoms (Natl. Lead Co. Ohio 1977).

The average annual precipitation at St. Charles, Missouri, based on a 40-year period of record, is about 93 cm (37 in.). About 42% occurs during May through August. The mean annual temperature is about 13°C (55°F) (Natl. Lead Co. Ohio 1977). A total of about 10 cm (4 in.) of precipitation per year has been calculated to be available as recharge to the unconsolidated glacial sediments. The rest is lost as runoff or is evaporated (Lawrence Berkeley Lab. 1980). Temperature and precipitation affect the depth of water present in the raffinate pits at any given time. In the summer months, the surface water in Pits 1 and 2 may evaporate, leaving the residues exposed. The level of water in Pits 3 and 4 also varies, but historically there has always been some surface water present (Weidner and Boback 1982; Bechtel Natl. Inc. 1983a). Water has never overflowed from any of the pits.

MODELING OF POTENTIAL GROUNDWATER CONTAMINATION

DESCRIPTION OF MODELING PROCEDURES

The potential contamination of groundwater in the Raffinate Pits area under existing conditions was analyzed by determining the ratio, C/S , of the radionuclide concentration in the groundwater (C) to the radionuclide concentration in the waste (S). The concentration of radionuclides in the water can be subsequently determined by multiplying this ratio by the radionuclide concentration in the waste (S).

The modeling procedures used to determine the C/S ratio involved two principal phases, namely groundwater hydraulics and solute transport. The hydraulic model, which gives the solution to the two-dimensional (longitudinal and vertical) partial differential equation of groundwater flow, generates groundwater flow velocities at various times and locations in the subsurface system. These flow rates and directions are then used in the solute transport model to simulate the migration of radionuclides in time and two-dimensional space. The solute transport model includes the effects of convective transport, molecular diffusion, hydrodynamic dispersion, radioactive decay, and chemical sorption. The flow and solute transport equations have been described in detail by Yeh and Ward (1980, 1981), Bear (1979), and Robertson (1974). For the current study, solutions of the flow and solute transport equations were obtained by using the finite-element models FEMWATER and FEMWASTE developed at Oak Ridge National Laboratory (Yeh and Ward 1980, 1981).

The FEMWATER and FEMWASTE models are designed to simulate contaminant transport in a saturated or unsaturated porous medium subject to variable initial or boundary conditions. These two codes have been verified and are generally considered to be good models for simulating contaminant transport in aquifer systems, especially in the unsaturated zone (Thomas et al. 1982; Oster 1982).

For the finite element computations, the region beneath the raffinate pits was subdivided into an assemblage of small elements with dimensions of

0.2 m $\times$ 2 m (vertical $\times$ horizontal). The procedures used were the finite-element weighted-residual method with quadrilateral bilinear spatial discretization and Gaussian elimination techniques. From these, the flow and solute transport equations were solved for the flow characteristics and contaminant concentrations at all nodal points of the elements.

SOURCE TERM

The source term that defines the boundary conditions at the bottom of the raffinate pits is a major factor in determining the radionuclide concentrations in the aquifer system. In this analysis, the wastes were assumed to be homogeneous and uniformly distributed in the pits. It was assumed that all wastes were emplaced at a single point in time and that leaching and migration begin immediately after placement. This is a reasonable assumption given the 10,000-year time frame for the analysis.*

Analyses of radionuclides in the two decay chains (Figures 4 and 5) were performed. Given the half-lives of the various radionuclides, it was determined that the only radionuclides that needed to be analyzed individually were uranium-238, thorium-232, thorium-230, and radium-226. The migration of uranium-234 was assumed to be comparable to that of uranium-238. Because the half-lives of the radionuclides below thorium-232 and radium-226 in the two decay chains are relatively short, at least with regard to the time periods during which migration occurs, it was assumed that a condition of secular equilibrium of the decay products beneath thorium-232 and radium-226 would be maintained.

*Even though the proposed radiological criteria developed by DOE for the Formerly Utilized Sites Remedial Action Program (FUSRAP) and remote Surplus Facilities Management Program (SFMP) sites require a design lifetime of 1000 years for long-term waste management, the current analysis considered a time frame of 10,000 years to provide additional information regarding use of the Raffinate Pits site.

The release rate of radium-226 per unit area per unit time, $\dot{m}_{Ra}$, is given by

$$\dot{m}_{Ra} = \frac{I \rho_s}{n_e R_d} \left\{ S_{Ra} \cdot e^{-(L_{Ra} + \lambda_{Ra})t} + \frac{\lambda_{Ra}}{L_{Ra} + \lambda_{Ra}} S_{Th} \left[1 - e^{-(L_{Ra} + \lambda_{Ra})t} \right] \right\} \quad (1)$$

in which S_{Ra} and S_{Th} are initial concentrations (pCi/g) of radium-226 and thorium-230 in the waste field; λ_{Ra} and L_{Ra} are decay and leach constants for radium-226; I is the infiltration rate of water; ρ_s is the average bulk density of the porous media; n_e is the effective porosity; $R_d = 1 + \rho_s K_d / n_e$ is the retardation factor; and K_d is the distribution coefficient for the radionuclide. This equation was derived by considering the two sources of radium-226 (i.e., that currently in the pits and that from ingrowth of the thorium-230 in the pits) and the mechanisms by which radium-226 is depleted (i.e., radioactive decay and leaching). It is necessary to consider these two sources of radium-226 because the radionuclides in this decay chain are in a state of disequilibrium as a result of previous processing operations. Furthermore, the derivation of Equation 1 was based on the assumption that the radioactive decay of thorium-230 itself is negligible, i.e., the ingrowth term is taken to be constant. This assumption is acceptable within the study time frame, which is considerably shorter than the half-life of thorium-230.

The total release rate of radium-226, $\dot{M}_{Ra}$, is obtained by multiplying $\dot{m}_{Ra}$ by the pit bottom area. For uranium-238, thorium-232, and thorium-230, the source release rates are similar to $\dot{M}_{Ra}$ except that the second term on the right-hand side of Equation 1, which is the ingrowth term, disappears. The ingrowth of thorium-230 from uranium-234 is negligible because of the long half-life of uranium-234 and the time frame (10,000 years) considered in this analysis. The initial source concentrations for all radionuclides considered are shown in Table 1.

HYDROGEOLOGICAL PARAMETER VALUES

The waste-field dimensions used in this analysis of the raffinate pits are shown in Table 3. Pits 1 and 2 are combined and treated as a single pit. The various hydrogeological parameters used in the current study are listed in Table 4. The sources for the selection of some of the parameter values are discussed in the previous section.

Table 3. Geometric Characteristics of Weldon Spring Raffinate Pits

Pit	Length ^{†1} (m)	Width ^{†1} (m)	Pit Depth (m)	Depth of Radioactive Materials (m)	Overburden Thickness (m)
1 & 2	70	140	2.9	2.7	12
3	140	240	3.7	2.9	9.1
4	180	340	5.6	0.70	7.6

^{†1} Obtained from sketches of the Raffinate Pits site.

In the unsaturated zone, the hydraulic conductivity, K_h , and the moisture content, θ , are functions of the pressure head. The values of K_h and θ generally increase with pressure head. However, the distribution of pressure head in the unsaturated overburden and its functional relationship with K_h and θ have not been uniquely defined for the Raffinate Pits site; therefore, constant values of K_h and θ were assumed for this study.

The values of K_d for specific sites generally vary depending on the soil properties, chemical content, pH of the water, and nature of the radionuclides. The K_d values for the raffinate pits were not available at the time this analysis was performed. In addition, studies providing empirical relations and data that could be used to relate the distribution coefficient to the above-mentioned factors are lacking. Ranges and mean values of K_d for various types of soils and clays of pH 4 to 9 are given in Table 5 for all three elements considered (Baes and Sharp 1983; Gilbert et al. 1983). The mean values of K_d given in Table 5 were used for the current study.

Table 4. Summary of Hydrogeologic and Radiological Parameters for Weldon Spring Raffinate Pits

Parameter	Symbol	Unit	Value† ¹
Infiltration rate	I	m/yr	0.1
Bulk density of soil	ρ_s	kg/m ³	1800
Effective porosity	n_e	-	0.05
Moisture content	θ	m ³ /m ³	0.05
Hydraulic conductivity	K_h	m/s	1.0×10^{-8}
Hydraulic gradient	i	-	0.01
Longitudinal dispersivity	α_L	m	1.0
Vertical dispersivity	α_V	m	0.1
Molecular diffusion coefficient	D_m	m ² /s	0.65×10^{-10}
Distribution coefficients	K_d (U)	m ³ /kg	0.05
	K_d (Ra)	m ³ /kg	0.1
	K_d (Th)	m ³ /kg	60
Radioactive decay constants	λ (U-238)	1/yr	1.55×10^{-10}
	λ (Ra-226)	1/yr	4.33×10^{-4}
	λ (Th-230)	1/yr	9.02×10^{-6}
	λ (Th-232)	1/yr	4.95×10^{-11}
Leach constants	L (U)	1/yr	5.29×10^{-4} (Pits 1 & 2)
			5.92×10^{-4} (Pit 3)
			21.6×10^{-4} (Pit 4)
	L (Ra)	1/yr	$1.38 \times 10^{-4}\dagger^2$
	L (Th)	1/yr	4.42×10^{-7} (Pits 1 & 2)
			4.93×10^{-7} (Pit 3)
			18.3×10^{-7} (Pit 4)

†¹ In analyses such as these, it is common to use ranges of values for the various parameters to bound the contamination potential. Because site-specific data for some parameters are not currently available, representative values for these parameters were used, assuming homogeneous conditions at the site.

†² Same value is used for all pits because of the ingrowth of radium-226 from thorium-230.

Table 5. Range and Mean Values of K_d for Various Radionuclides

Element	K_d (m^3/kg)	
	Range	Mean
Uranium	0.01 - 0.2	0.05
Thorium	10 - 300	60
Radium	0.02 - 0.8	0.1

Sources: Baes and Sharp (1983); Gilbert et al. (1983).

Sorption tests for uranium-238 and radium-226 were later performed on soil samples obtained from the Raffinate Pits site (Kelmars and Seeley 1983). K_d values ranging from 0.6 to 18 m^3/kg were reported for radium-226. For uranium-238, K_d values varied from 0.3 to 1.3 m^3/kg . These values are greater than the K_d value used for the present study. However, examination of Kelmars and Seeley's test results indicated that if initial concentrations in the groundwater were lower than the initial concentration of 100,000 pCi/mL used in the tests, then the K_d value could be significantly lower than the values reported. Because of this uncertainty, the use of low K_d values as shown in Tables 4 and 5 for the current study was considered to be reasonable and conservative.

DISCUSSION OF RESULTS

The time variations of radionuclide concentrations in the subsurface aquifer system beneath the raffinate pits were calculated using the selected geometric and hydrogeologic data given in Tables 3 and 4. The selection of these hydrogeologic parameter values was intended to provide realistic estimates of radionuclide migration under existing conditions at the site using currently available environmental data. As discussed previously, the overburden in the vicinity of the raffinate pits has a depth of 5.2 to 15 m (17 to 50 ft) consisting of unsaturated materials with low permeabilities (Bechtel Natl. Inc. 1983b). The groundwater table in the vicinity of the pits lies at a depth of about 18 m (60 ft) below the raffinate pit bottoms (Lenhard et al.

1967; Lomenick 1982). The only driving force for movement of contaminants downward into the aquifer system is the infiltrating water. The time frame used for studying the contaminant migration was 10,000 years.

The simulation was first performed to determine the values of various hydrodynamic variables. These were computed by the flow model to yield flow characteristics beneath the pits. The results, which are not presented here, indicate that during the time frame used (10,000 years), the infiltrating water flows downward in a vertical direction.

The predicted distributions of the concentration ratio C/S for radium-226 and uranium-238 over the time period of 10,000 years are shown in Figures 7 and 8. The distributions of thorium-232 and thorium-230 are not presented because the extent of thorium migration is minimal due to its high rate of retardation. The results shown in Figures 7 and 8 indicate that over 10,000 years, both radium-226 and uranium-238 would migrate to only a limited depth; also, because of the different elevations of the pit bottoms and their relative locations, the contaminated regions under each pit would be somewhat isolated and would not interact with one another.

The vertical concentration profiles of radium-226 and uranium-238 at the center of the raffinate pits are shown in Figures 9 through 14. Again, the profiles for the distribution of thorium-232 and thorium-230 are not presented because the migration of thorium is minimal in 10,000 years. The concentrations given in Figures 9 through 14 represent the maximum concentrations at a given depth because these profiles are at the centers of the pits. The profiles for both radium-226 and uranium-238 indicate that over a time period of 10,000 years, the maximum depth of radionuclide migration would be less than 2 m (7 ft) and the contamination would remain in the unsaturated area. This small amount of migration is attributed mainly to the assumed low permeability of the clayey soils overlying the limestone bedrock.

Comparison of the concentration distributions in Figures 9 through 14 indicates that even though radium-226 has a higher retardation factor than uranium-238, the depth of radium-226 migration is greater than that of uranium-238. This is attributed to the ingrowth of radium-226 from thorium-230

Figure 7. Distribution of Radium-226 Concentration Ratios, $(\text{pCi/L})/(\text{pCi/g})$, Beneath the Raffinate Pits at $t = 10,000$ Years.

Figure 8. Distribution of Uranium-238 Concentration Ratios, $(\text{pCi/L})/(\text{pCi/g})$, Beneath the Raffinate Pits at $t = 10,000$ Years.

Figure 9. Vertical Concentration Profile for Radium-226 at the Center of Pits 1 and 2 for Selected Times.

Figure 10. Vertical Concentration Profile for Radium-226 at the Center of Pit 3 for Selected Times.

Figure 12. Vertical Concentration Profile for Uranium-238 at the Center of Pits 1 and 2 for Selected Times.

Figure 11. Vertical Concentration Profile for Radium-226 at the Center of Pit 4 for Selected Times.

Figure 13. Vertical Concentration Profile for Uranium-238 at the Center of Pit 3 for Selected Times.

Figure 14. Vertical Concentration Profile for Uranium-238 at the Center of Pit 4 for Selected Times.

in the raffinate pits, which results in a large concentration gradient. At 1,000 and 10,000 years, the uranium-238 concentration profiles for Pit 4 are identical; however, for these same time frames, the uranium-238 concentration profiles are different for Pits 1, 2, and 3. The reason for this difference is due to the much smaller depth of radioactive materials in Pit 4 compared to the depth in the other pits (Table 3). Because of this shallow depth, the uranium-238 in Pit 4 would be quickly leached out and the rate of waste release would taper off to a small value in a short time. This small amount of waste release would essentially result in no further significant migration of uranium-238. Similar behavior was not observed for the radium-226 in Pit 4 (Figure 11) because of the ingrowth of radium-226 from thorium-230 in the pit, which would result in a longer release of radium-226.

The time variations of the concentration ratio C/S in the subsurface region of each raffinate pit where the maximum concentrations were calculated are presented in Table 6. At some point in time, the radionuclide concentration beneath each pit would reach an absolute maximum value; this value, and the time to reach this absolute maximum, are also given in Table 6. From these values, it appears that during the time period of 10,000 years, the maximum concentration ratio C/S reached in the raffinate pit area would be about 10 and 17.6 (pCi/L)/(pCi/g), respectively, for radium-226 and uranium-238. The maximum concentrations for thorium-232 and thorium-230 would occur after 10,000 years. Using the radionuclide concentrations in the waste (S) given in Table 1 for radium-226 and uranium-238 and the maximum concentration ratios presented in Table 6, the maximum concentrations in groundwater (C) of radium-226 and uranium-238 can be determined (Table 7). These concentrations would occur immediately beneath the bottom of the pits.

SUMMARY

Results of the analysis of contaminant migration beneath the raffinate pits at the Weldon Spring Raffinate Pits site indicate that during a 10,000-year time period, the maximum concentrations in the water immediately beneath the pit bottoms would be about 4,600 pCi/L of radium-226 (Pit 3) and about 12,000 pCi/L of uranium-238 (Pit 1); these concentrations would occur at the

Table 6. Predicted Time Variations of Radionuclide Concentration Ratios at the Maximum Concentration Locations Beneath the Weldon Spring Raffinate Pits†¹

Radionuclide	Pit Number	Concentration Ratios at Various Times (pCi/L)/(pCi/g)				Absolute Maximum Concentration Ratio Reached (pCi/L)/(pCi/g)	Time to Reach Absolute Maximum Concentration (yr)
		20 yr	100 yr	1,000 yr	10,000 yr		
Radium-226	1 & 2	4.6	8.2	9.9	10.0	10.0	3,800
	3	4.4	7.7	9.8	9.9	9.9	5,200
	4	8.4	9.9	10.0	10.0	10.0	1,000
Uranium-238	1 & 2	9.9	14.5	17.5	17.5	17.6	1,300
	3	9.9	14.5	17.4	17.4	17.5	1,200
	4	9.8	14.1	15.5	15.5	15.6	400
Thorium-230	1 & 2	0.00046	0.00123	0.0036	0.0076	-† ²	>10,000† ³
	3	0.00046	0.00123	0.0036	0.0076	-	>10,000
	4	0.00046	0.00123	0.0036	0.0075	-	>10,000
Thorium-232	1 & 2	0.00046	0.00123	0.0036	0.0079	-	>10,000
	3	0.00046	0.00123	0.0036	0.0079	-	>10,000
	4	0.00046	0.00123	0.0036	0.0079	-	>10,000

†¹ Values of hydrological parameters used for the calculations are given in Table 4. The maximum concentration locations would occur at the centers of the pit bottoms.

†² A blank entry (-) in the table indicates that the concentrations were not calculated.

†³ A time period of 10,000 years was used for this analysis. The maximum concentrations for thorium-230 and thorium-232 beneath the pits occur after 10,000 years.

Table 7. Maximum Concentrations of Radium-226 and Uranium-238 in Water Beneath the Raffinate Pits from Materials Currently Stored in the Pits

Radionuclide	Pit Number	Maximum Concentration Reached† ¹ (pCi/L)	Time to Reach Maximum Concentration (yr)
Radium-226	1 & 2† ²	4,400	3,800
	3	4,600	5,200
	4	110	1,000
Uranium-238	1 & 2† ²	12,000	1,300
	3	9,100	1,200
	4	9,700	400

†¹ Results are given to two significant figures. The maximum concentrations would occur at the centers of the pit bottoms.

†² The larger value of the concentrations in Pits 1 & 2 given in Table 1 was used.

centers of the pit bottoms. Based on the assumptions used in this study, the radioactive contaminants in the pits would migrate no more than 2 m (7 ft) below the pit bottoms. Because 6 to 12 m (20 to 40 ft) of silty clays underlie the raffinate pits, the radioactive contaminants would take several tens of thousands of years to reach nearby groundwater supplies.

Although the results of these analyses indicate that a high degree of confinement is provided by the four raffinate pits, it should be noted that the validity of such analyses rests on the quality of the parameter values utilized. Due to a lack of current site-specific data for some physical parameters, it has been necessary to use historical and regional data for these values. The values cited are at times inconsistent and contradictory, e.g., the wide range of values indicated for the permeability of clays underlying the pits. However, these were the only data available. The analysis reported herein indicates that within the limitations of the available data, use of the Raffinate Pits site for long-term management of radioactive materials such as those currently being stored in the four pits appears to be feasible.

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