Groundwater Calculations for Depleted Uranium Disposed of as Uranium Tetrafluoride (UF$_4$)

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GROUNDWATER CALCULATIONS FOR DEPLETED URANIUM DISPOSED OF AS URANIUM TETRAFLUORIDE (UF$_4$)

by

D. Tomasko

ABSTRACT

This report discusses calculations performed to estimate the impacts on groundwater from the long-term disposal of depleted uranium in the form of uranium tetrafluoride (UF$_4$) in a trench, vault, and mined cavity. The calculations were done for a deep groundwater system, typical of conditions in the western United States. They were performed for two initial inventories of UF$_4$: 500,000 and 630,000 metric tons. Disposal was in either 30- or 50-gal drums. All of the contaminant and radioactivity concentrations at the water table are predicted to be very low, even for a fairly mobile compound. In general, concentrations after 1,000 years at the water table would be about an order of magnitude greater for disposal in a deep mine than for disposal in a trench or vault. The largest activity concentration at the water table after 1,000 years would be derived from a failed mine that releases a fairly mobile and very soluble uranium complex; it would be about 8.6×10^{-6} pCi/L for 500,000 metric tons of UF$_4$ disposed of in 30-gal drums. The smallest activity concentrations at the water table after 1,000 years would, in general, be derived from a failed trench (1.2×10^{-9} pCi/L), if the contaminant reached the water table as schoepite. Although all the activity concentrations at the water table after 1,000 years are predicted to be small, maximum activity concentrations could still be large, even after dilution. Maximum activity concentrations of uranium would exceed 900 pCi/L for a failed disposal facility if a very soluble and mobile uranium complex formed. If the solubility of the uranium compound was small (2.4×10^{-3} g/L), the resulting activity concentrations would be small, less than 2.5 pCi/L. The estimated time for these maximum concentrations would range from 65,000 to 2,000,000 years.

1 INTRODUCTION

This report discusses calculations performed to estimate the impacts on groundwater produced from the long-term disposal of depleted uranium in the form of uranium tetrafluoride (UF$_4$)

in an engineered trench, a vault, and a mined cavity. The calculations were done for a deep groundwater system, typical of conditions in the western portion of the country. Calculations were performed for two initial inventories of UF_4: 500,000 and 630,000 metric tons (t). These values are consistent with 560,000 and 705,000 t of disposed uranium hexafluoride (UF_6), respectively, discussed in Folga and Kier (2001). Disposal was in either 30- or 50-gal drums. Where possible, the calculations follow the same format as those reported in the *Programmatic Environmental Impact Statement for Alternative Strategies for the Long-Term Management and Use of Depleted Uranium Hexafluoride* (PEIS) published by the U.S. Department of Energy (DOE 1997).

2 SOURCE TERM

Solid UF_4 in storage poses no direct threat to groundwater resources. However, UF_4 is an unstable compound that can dissolve and react with oxygen and water to form more complex compounds. These compounds can then migrate into the groundwater system and be transported by existing hydraulic gradients.

In order to perform transport calculations for the groundwater, a source term is needed. This source term requires an estimate of the length of time that uranium is being introduced into the system and its initial concentration at the discharge point. For simplicity, the source term is assumed to behave like a unit step function (Hildebrand 1976) that has a value C_0 for a time Δt and then returns to zero. This initial concentration, C_0, is assumed to be equal to the solubility of either UF_4 or the solubility of one of the many possible uranium compound complexes. The duration of the source term, Δt, can be estimated as the dissolution time for the entire inventory of UF_4 in the disposal area.

After exposure to water, UF_4 dissolves and then undergoes hydrolysis. Dissolution to a solubility limit is assumed to be instantaneous. The time for hydrolysis, however, can be significant. If the hydrolysis time is long compared to the travel time to a potential receptor, the dissolved form of uranium at the receptor will be UF_4. However, if the hydrolysis time is short compared to the travel time, other uranium compounds are possible.

In the presence of oxygen and water, UF_4 is unstable and dissolves and chemically reacts. The reported solubility of UF_4 ranges from about 0.03 to 0.1 g/L at $25\,^\circ$C, per information from Oak Ridge National Laboratory in 1960 on Atomic Energy Commission Report AECD-3412 and from Kozak et al. (1992). UF_4 can also chemically react with oxygen and water to form schoepite ($UO_3 \cdot 2H_2O$) via the following chemical reaction:

$$UF_4 \;+\; \frac{1}{2}O_2(g) \;+\; 4H_2O \;\rightarrow\; 4HF(aq) \;+\; UO_3 \cdot 2H_2O \;. \tag{1}$$
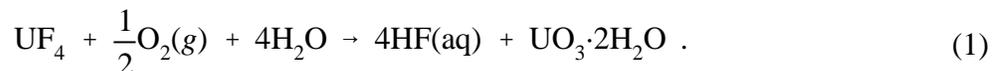
In addition, the following reaction can occur, producing triuranium octaoxide (U_3O_8):

$$3UF_4 \;+\; O_2(g) \;+\; 6H_2O \;\rightarrow 12HF(aq) \;+\; U_3O_8 \;. \tag{2}$$
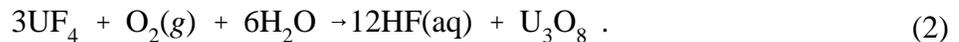
The U_3O_8 produced in the reaction given by Equation 2 can then form schoepite under oxidizing conditions. The solubility of schoepite is approximately 2.4×10^{-3} g/L (Kozak et al. 1992).

Because of its corrosive properties, the HF released by the above reactions is assumed to react with material such as Portland cement grout and carbonate in limestone formations, releasing

carbonates into solution. Because of such reactions, the HF is assumed to be removed from the groundwater pathway and is not considered to be a risk at the location of potential receptors.

Dissolved carbonate released by reactions involving HF can have a significant effect on the aqueous chemistry and solubility of uranium (U[VI]) through the formation of strong anionic carbonato complexes (DOE and U.S. Environmental Protection Agency [EPA] 1999). Some of these compounds include dioxouranium carbonate species: UO_2CO_3, $UO_2(CO_3)_2$, and $UO_2(CO_3)_3$. The predominant species formed depends on the pH of the system. Under neutral conditions (pH = 7.0), the predominant form is $UO_2(CO_3)_2$. For slightly acidic conditions (pH = 6.0), such as those expected in the presence of the released HF, the predominant form is UO_2CO_3 (Langmuir 1978). The solubility of UO_2CO_3 is reported as 0.028 g/L (Gmelin 1983). If nitrogen is also present in solution, additional uranium complexes can form. One of these, an ammonium carbonate uranium complex that has the chemical formula $(NH_4)_4(UO_2(CO_3)_3$, has a solubility of 23 g/L (Gmelin 1983).

As the above discussion indicates, the final form of uranium in solution at the location of a potential receptor is a complex function of the reaction time of the dissolved phase present relative to the transport time to a potential receptor, the physical and chemical attributes of the disposal facility, and the geochemistry of the receiving water. Because the reaction rate for UF_4 hydrolysis is not well known (Kozak et al. 1992), and because the geochemistry of the groundwater is site-specific, a range of solubilities was selected to bracket the calculations. At the low end of the solubility range, the solubility is assumed to be equal to that of schoepite (2.4×10^{-3} g/L). Because groundwater uranium concentrations produced by schoepite during long-term disposal are very small (Tomasko 1997a), a smaller solubility was not needed for the present analyses. At the high end of the solubility range, the solubility is assumed to be four orders of magnitude greater (24 g/L). This value is similar to the solubility of the ammonium carbonate uranium complex mentioned above.

An intermediate set of calculations could also be performed for this study. Here, the solubility would be set to equal the solubility of UF_4 (approximately 0.1 g/L). For this calculation, the hydrolysis of UF_4 would be assumed to be slow when compared with the transport time to a potential receptor. For this condition, UF_4 would remain in solution and not react significantly. Its initial concentration at the point of discharge to the environment would be equal to its solubility (0.1 g/L). Because this value lies within the range selected for the analyses, a separate analysis was not performed for the intermediate solubility.

Because the disposed radioactive material is assumed to be soluble, it will act as a source of contamination for a number of years. The duration time for the source can be estimated by using the following expression:

$$\Delta t = \frac{M_t}{VASol_i} , \tag{3}$$

where *A* is the area of the disposal facility footprint, M_t is the total initial inventory of contaminant (500,000 and 630,000 t of UF_4), Sol_i is the solubility of the ith radioactive material (i.e., 2.4×10^{-3} or 24 g/L), and *V* is the infiltration velocity of water.

For a dry climate, the quantity of water infiltrating a soil column is approximately 1% of the annual precipitation (Rice et al. 1989). For an average annual precipitation of 10 in. in an arid climate (Tomasko 1997a), the net rate of water infiltration is 0.1 in./yr (2.54×10^{-3} m/yr). The range of the solubilities of the potential radioactive dissolution products for Equation 3 is 2.4×10^{-3} to 24 g/L, as discussed above. Footprint areas for the different disposal options are listed in Table 1 (Folga and Kier 2001).

A list of duration times for the various sources of contamination estimated with Equation 3 are given in Table 2 for disposal in a trench, vault, and deep mine. Except for the highly soluble uranium complex, all source durations are very long, exceeding 33 million years. The shortest source duration is 3,320 years for disposing of 500,000 t of UF_4 in 30-gal drums in a mine in which water reacts with the radioactive material to form a very soluble (24 g/L) uranium complex.

TABLE 1 Footprint Areas (in hectares) for the Long-Term Disposal of UF_4

Disposal Method	500,000 t		630,000 t	
	30-gal Drums	50-gal Drums	30-gal Drums	50-gal Drums
Trench	20.6	17.4	25.5	21.6
Vault	25.4	31.6	31.6	37.9
Mine	247	187	310	234

TABLE 2 Duration Times (in years) for Contaminant Sources

Facility	Solubility (g/L)	500,000 t		630,000 t	
		30-gal Drums	50-gal Drums	30-gal Drums	50-gal Drums
Trench	2.4×10^{-3}	398,000,000	471,000,000	501,000,000	593,000,000
	24	39,800	47,100	50,100	59,300
Vault	2.4×10^{-3}	323,000,000	260,000,000	407,000,000	328,000,000
	24	32,300	26,000	40,700	32,800
Mine	2.4×10^{-3}	33,200,000	43,800,000	41,200,000	54,700,000
	24	3,320	4,380	4,120	5,470

3 CALCULATIONS

A one-dimensional analytical solution of the advection/dispersion equation was used to calculate the concentrations of the various radioactive contaminants (Tomasko 1997b). The calculations were performed for a time of 1,000 years, to be consistent with calculations done for the PEIS (DOE 1997), and for groundwater directly beneath the footprint of the disposal facility. Longitudinal transport was not included because the times required for transport to a point 1,000 ft from the disposal facility are much shorter (about 50 to 500 years for a groundwater velocity of 110 ft/yr and for transport velocities that are 5 to 50 times slower, respectively, than the velocity of the groundwater [retardations of 5 and 50]) than the times required for transport through the unsaturated zone (60,000 to 300,000 years for a deep mine with a thickness of 100 ft and for a trench/vault with a thickness of 500 ft, respectively, and a retardation of 5. Because of this shorter travel time, concentrations at a distance of 1,000 ft from the facility would be essentially the same as those calculated for a point directly below the facility footprint. In addition to groundwater calculations performed at 1,000 years, the calculations were extended to determine the maximum concentration that would be achieved and the time of this maximum value.

Longitudinal dispersion was calculated by using the following expression (Bear 1972):

$$D = \alpha V \ , \tag{4}$$

where α is the dispersivity of the formation and V is the infiltration velocity of the water (0.1 in./yr). Dispersivity was assumed to be scale-dependent (Lallamand-Barres and Peaudecerf 1978), i.e.,

$$\alpha = 0.1L \ ,$$

where L is the separation distance between the source release point and the point of interest (the water table directly below the disposal facility footprint). For the trench/vault, longitudinal dispersion was 0.4 ft^2/yr; for the mine, the dispersion was 0.083 ft^2/yr. A summary of parameters for the disposal calculations is given in Table 3.

As a contaminant is transported by groundwater, it may have an apparent velocity that is less than the velocity of the water because of sorption. This process is known as retardation (Freeze and Cherry 1979). Because retardation of a dissolved radionuclide is a site-specific parameter that has a high degree of uncertainty, two values were used to bound the calculations, 5 and 50. These values represent a fairly mobile contaminant (retardation of 5) and a contaminant that undergoes substantial sorption along its flow path (retardation of 50).

Figure 1 shows breakthrough curves of the dimensionless concentration (concentration at a time t divided by the initial concentration of the contaminant, i.e., C/C_0) for schoepite at the water

TABLE 3 Summary of Hydrological Parameters Used for the Disposal Calculations

| Parameter	Trench	Vault	Mine
Depth to water table (ft)	500	500	100
Infiltration velocity of water (ft/yr)	8.33×10^{-3}	8.33×10^{-3}	8.33×10^{-3}
Saturated zone water velocity (ft/yr)	110	110	110
Longitudinal dispersion in unsaturated zone (ft^2/yr)	0.4	0.4	0.083
Longitudinal dispersion in saturated zone (ft^2/yr)	11,000	11,000	11,000
Retardation	5 and 50	5 and 50	5 and 50
Thickness of saturated zone (ft)	100	100	100
Porosity	0.1	0.1	0.1
Hydraulic conductivity of saturated zone (ft/yr)	11,000	11,000	11,000
Hydraulic gradient in saturated zone	0.001	0.001	0.001
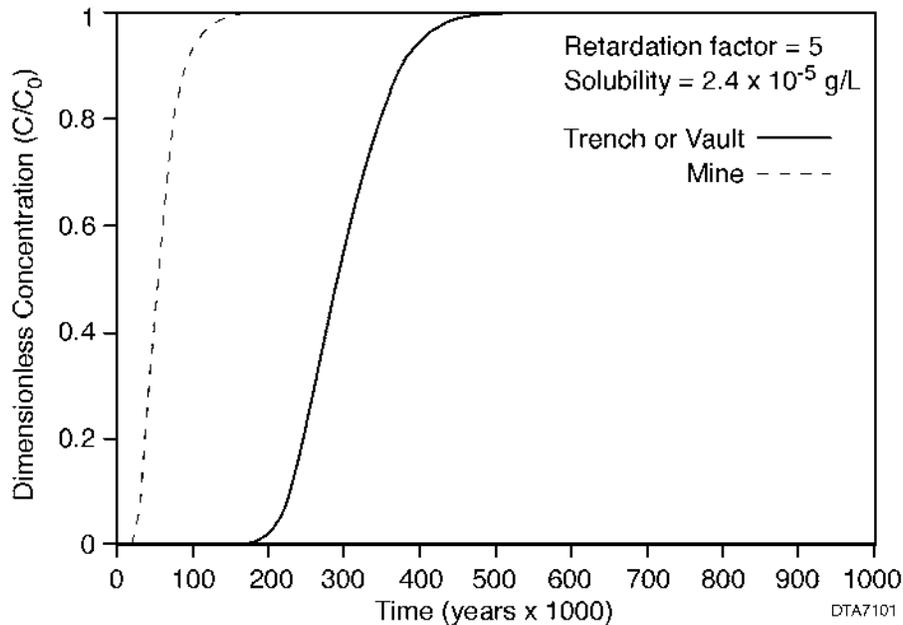
FIGURE 1 Breakthrough Curves for a Trench, Vault, and Mine for a Fairly Mobile, Low-Solubility Uranium Complex

table as a function of time for disposal in a vault or trench and in a mine for fairly mobile conditions (retardation of 5). For the time scale shown, there are no differences between the breakthrough curves for disposal in 30- or 50-gal drums. Because of the long durations of the source possible with schoepite (Table 3), the maximum concentration at the water table can equal the initial concentration (i.e., $C/C_0 = 1.0$). In a trench or vault, the maximum concentration would be reached after about 550,000 years (Table 4). In a mine, the maximum concentration would be reached after about 200,000 years. Maximum concentrations for a mine occur earlier because of the shorter travel distance from the mine to the water table (100 versus 500 ft). For more sorption (retardation of 50), the peak concentration ($C/C_0 = 1$) for a trench or vault would be reached after approximately 5 million years (Table 4) as shown in Figure 2. For a mine, this concentration would be reached after about 2 million years.

The dimensionless concentrations for schoepite released from a trench or vault after 1,000 years are 2.1×10^{-9} and 6.1×10^{-21} for retardations of 5 and 50, respectively (Table 5). For a mine, the dimensionless concentrations after 1,000 years would be 3.9×10^{-9} and 1.7×10^{-9} for retardations of 5 and 50, respectively. As shown in Table 4, for a specific activity of 2.4×10^{-7} Ci/g (Tomasko 1997a), the maximum uranium activity concentration from a failed trench, vault, or mine would be 576 pCi/L (1 pCi or picocurie = 1×10^{-12} Ci or curies).

By multiplying the dimensionless concentrations shown in Figures 1 and 2 by the initial concentration of the source (2.4×10^{-3} g/L), the uranium concentrations after 1,000 years for a failed trench or vault would be 5.0×10^{-12} and 1.5×10^{-23} g/L for retardations of 5 and 50, respectively (Table 5). For a failed mine, the uranium concentrations would be 9.5×10^{-12} and 4.0×10^{-12} for retardations of 5 and 50, respectively.

By multiplying the uranium concentration in g/L by the specific activity of the compound (Ci/g), the uranium activity concentration for the compound can be found. For a specific activity of 2.4×10^{-7} Ci/g for schoepite (Tomasko 1997a), the uranium activity concentrations from a failed trench or vault at 1,000 years would be 1.2×10^{-6} and 3.6×10^{-18} pCi/L, for retardations of 5 and 50, respectively. For a failed mine, the activity concentration of the uranium would be 2.3×10^{-6} and 9.6×10^{-7} pCi/L for retardations of 5 and 50, respectively.

If the solubility of the uranium were increased by four orders of magnitude to 24 g/L, the duration time of the source from a failed disposal facility would be decreased by a factor of 10,000 (Table 2). Figures 3 through 5 show the breakthrough curves for a highly soluble uranium complex from a trench, vault, and mine, respectively. For the higher solubility cases, the maximum dimensionless concentration (C/C_0) reached is less than 1.0, and the maximum occurs much earlier in time (Table 4). A maximum dimensionless uranium concentration of 0.39 occurs for a failed trench in which 630,000 t of UF_4 are disposed of in 50-gal drums. This maximum would occur after about 315,000 years for an assumed retardation of 5. For an initial source term concentration of 24 g/L, the maximum concentration would be 9.4 g/L. The smallest dimensionless maximum

TABLE 4 Maximum Concentrations and Occurrence Times for Long-Term Disposal of UF$_4$

Solubility (g/L)	R[a]	Facility	500,000 t in 30-gal Drums C/C_0	Concentration (g/L)[b]	Activity Concentration (pCi/L)[c]	500,000 t in 50-gal Drums C/C_0	Concentration (g/L)[b]	Activity Concentration (pCi/L)[c]
2.4×10^{-3}	5	Trench	1.0 550,000 yr	2.4×10^{-3}	576	1.0 550,000 yr	2.4×10^{-3}	576
		Vault	1.0 550,000 yr	2.4×10^{-3}	576	1.0 550,000 yr	2.4×10^{-3}	576
		Mine	1.0 200,000 yr	2.4×10^{-3}	576	1.0 200,000 yr	2.4×10^{-3}	576
	50	Trench	1.0 5,000,000 yr	2.4×10^{-3}	576	1.0 5,000,000 yr	2.4×10^{-3}	576
		Vault	1.0 5,000,000 yr	2.4×10^{-3}	576	1.0 5,000,000 yr	2.4×10^{-3}	576
		Mine	1.0 2,000,000 yr	2.4×10^{-3}	576	1.0 2,000,000 yr	2.4×10^{-3}	576
24	5	Trench	0.27 310,000 yr	6.5	1.0×10^{6}	0.32 315,000 yr	7.7	1.2×10^{6}
		Vault	0.22 305,000 yr	5.3	5.3×10^{5}	0.18 300,000 yr	4.3	6.9×10^{5}
		Mine	0.06 60,000 yr	1.4	2.2×10^{5}	0.08 65,000 yr	1.9	1.6×10^{5}
	50	Trench	0.043 2,900,000 yr	1.0	1.6×10^{5}	0.036 2,850,000 yr	0.9	1.4×10^{5}
		Vault	0.022 2,850,000 yr	0.5	8.0×10^{4}	0.017 2,800,000 yr	0.4	6.4×10^{4}
		Mine	0.006 450,000 yr	0.14	2.2×10^{4}	0.0082 445,000 yr	0.2	3.2×10^{4}

Solubility (g/L)	R[a]	Facility	630,000 t in 30-gal drums C/C_0	Concentration (g/L)[b]	Activity Concentration (pCi/L)[c]	630,000 t in 50-gal drums C/C_0	Concentration (g/L)[b]	Activity Concentration (pCi/L)[c]
2.4×10^{-3}	5	Trench	1.0 550,000 yr	2.4×10^{-3}	576	1.0 550,000 yr	2.4×10^{-3}	576
		Vault	1.0 550,000 yr	2.4×10^{-3}	576	1.0 550,000 y	2.4×10^{-3}	576
		Mine	1.0 200,000 yr	2.4×10^{-3}	576	1.0 200,000 yr	2.4×10^{-3}	576
	50	Trench	1.0 5,000,000 yr	2.4×10^{-3}	576	1.0 5,000,000 yr	2.4×10^{-3}	576
		Vault	1.0 5,000,000 yr	2.4×10^{-3}	576	1.0 5,000,000 yr	2.4×10^{-3}	576
		Mine	1.0 2,000,000 yr	2.4×10^{-3}	576	1.0 2,000,000 yr	2.4×10^{-3}	576

TABLE 4 (Cont.)

Solubility (g/L)	R^a	Facility	630,000 t in 30-gal drums			630,000 t in 50-gal drums		
			C/C_0	Concentration (g/L)[b]	Activity Concentration (pCi/L)[c]	C/C_0	Concentration (g/L)[b]	Activity Concentration (pCi/L)[c]
24	5	Trench	0.34 310,000 yr	8.2	1.3×10^6	0.39 315,000 yr	9.4	1.5×10^6
		Vault	0.28 310,000 yr	6.7	1.1×10^6	0.23 308,000 yr	5.5	8.8×10^5
		Mine	0.075 65,000 yr	1.8	2.9×10^5	0.1 75,000 yr	2.4	3.8×10^5
	50	Trench	0.033 2,800,000 yr	0.8	1.3×10^5	0.028 2,700,000 yr	0.7	1.1×10^5
		Vault	0.028 2,900,000 yr	0.7	1.1×10^5	0.0225 2,870,000 yr	0.5	8.0×10^4
		Mine	0.0075 452,000 yr	0.2	3.2×10^4	0.01 452,000 yr	0.2	3.2×10^4

[a] R = retardation factor.

[b] Concentration is found by multiplying the dimensionless concentration, C/C_0, by the compound's solubility.

[c] Activity concentration is found by multiplying the concentration of a compound by its specific activity.
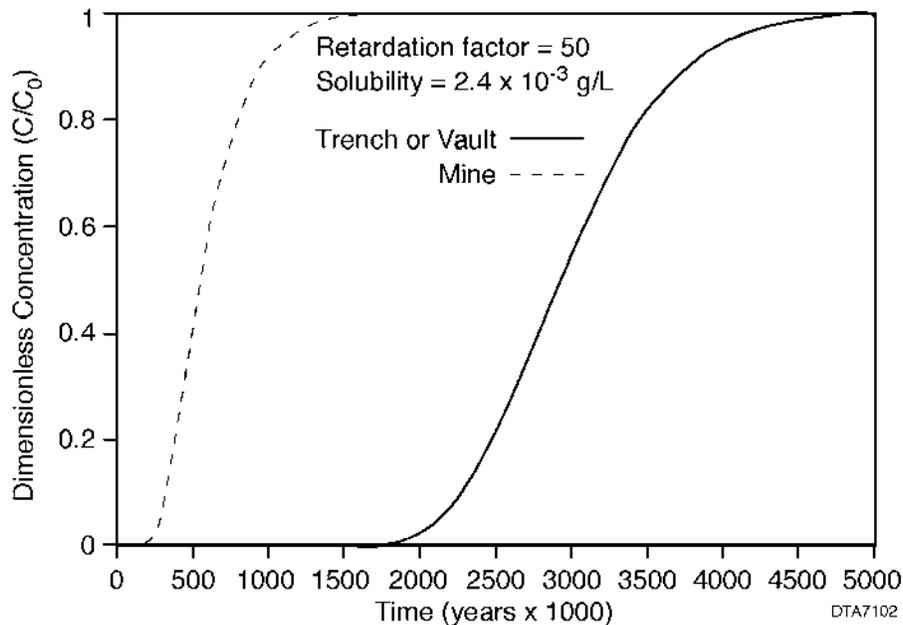

FIGURE 2 Breakthrough Curves for a Trench, Vault, and Mine for a Fairly Immobile, Low-Solubility Uranium Complex

TABLE 5 Summary of Concentrations at 1,000 Years at the Water Table for Long-Term Disposal of UF$_4$

Solubility (g/L)	Facility	500,000 t in 30-gal Drums			500,000 t in 50-gal Drums		
		C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)
2.4×10^{-3}	Trench	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}
	Vault	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}
	Mine	3.9×10^{-9} 1.7×10^{-9}	9.5×10^{-12} 4.0×10^{-12}	2.3×10^{-6} 9.6×10^{-7}	3.9×10^{-9} 1.7×10^{-9}	9.5×10^{-12} 4.0×10^{-12}	2.3×10^{-6} 9.6×10^{-7}
24	Trench	1.9×10^{-11} 3.4×10^{-21}	4.5×10^{-10} 8.1×10^{-20}	7.2×10^{-5} 1.3×10^{-14}	1.9×10^{-11} 3.6×10^{-21}	4.6×10^{-10} 8.7×10^{-20}	7.3×10^{-5} 1.4×10^{-14}
	Vault	2.2×10^{-11} 3.0×10^{-21}	5.3×10^{-10} 7.2×10^{-20}	8.5×10^{-5} 1.2×10^{-14}	1.1×10^{-11} 2.7×10^{-21}	2.5×10^{-10} 6.4×10^{-20}	4.0×10^{-5} 1.0×10^{-14}
	Mine	5.6×10^{-10} 5.5×10^{-12}	1.4×10^{-8} 1.3×10^{-10}	2.2×10^{-3} 2.1×10^{-5}	5.8×10^{-10} 7.3×10^{-12}	1.4×10^{-8} 1.8×10^{-10}	2.2×10^{-3} 2.8×10^{-5}

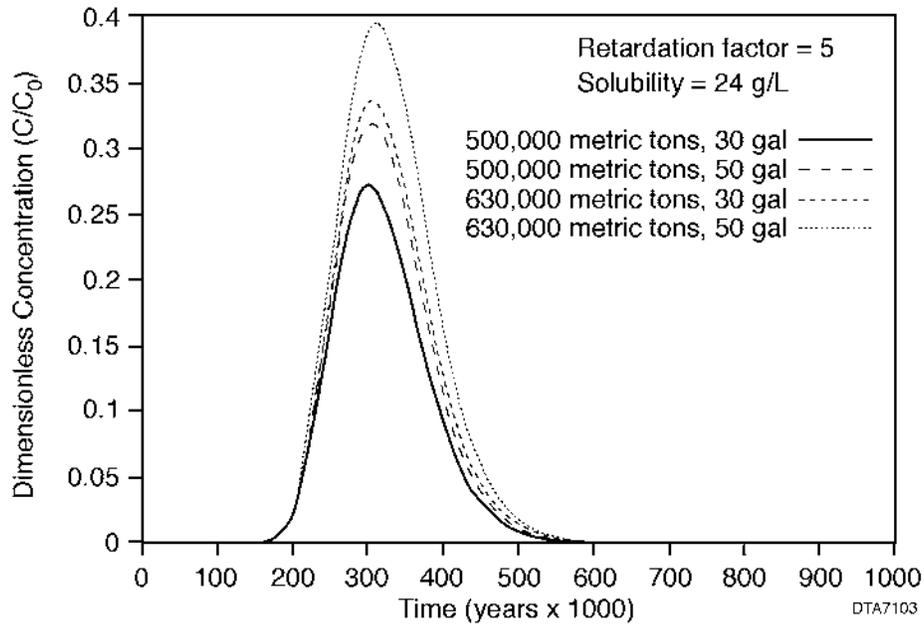| Solubility (g/L)	Facility	630,000 t in 30-gal Drums			630,000 t in 50-gal Drums		
		C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration	Activity Concentration
2.4×10^{-3}	Trench	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}
	Vault	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}	2.1×10^{-9} 6.1×10^{-21}	5.0×10^{-12} 1.5×10^{-23}	1.2×10^{-6} 3.6×10^{-18}
	Mine	3.9×10^{-9} 1.7×10^{-9}	9.5×10^{-12} 4.0×10^{-12}	2.3×10^{-6} 9.6×10^{-7}	3.9×10^{-9} 1.7×10^{-9}	9.5×10^{-12} 4.0×10^{-12}	2.3×10^{-6} 9.6×10^{-7}
24	Trench	1.8×10^{-11} 3.8×10^{-21}	4.4×10^{-10} 9.0×10^{-20}	7.1×10^{-5} 1.4×10^{-14}	2.4×10^{-11} 4.0×10^{-21}	5.6×10^{-10} 9.7×10^{-20}	9.0×10^{-5} 1.6×10^{-14}
	Vault	2.0×10^{-11} 3.4×10^{-21}	4.7×10^{-10} 8.2×10^{-20}	7.6×10^{-5} 1.3×10^{-14}	2.0×10^{-11} 3.0×10^{-21}	4.8×10^{-10} 7.3×10^{-20}	7.6×10^{-5} 1.2×10^{-14}
	Mine	5.4×10^{-10} 6.8×10^{-12}	1.3×10^{-8} 1.6×10^{-10}	2.1×10^{-3} 2.6×10^{-5}	7.4×10^{-10} 9.3×10^{-12}	1.8×10^{-8} 2.2×10^{-10}	2.9×10^{-3} 3.6×10^{-5}

FIGURE 3 Breakthrough Curves for a Trench for a Fairly Mobile, Very Soluble Uranium Complex
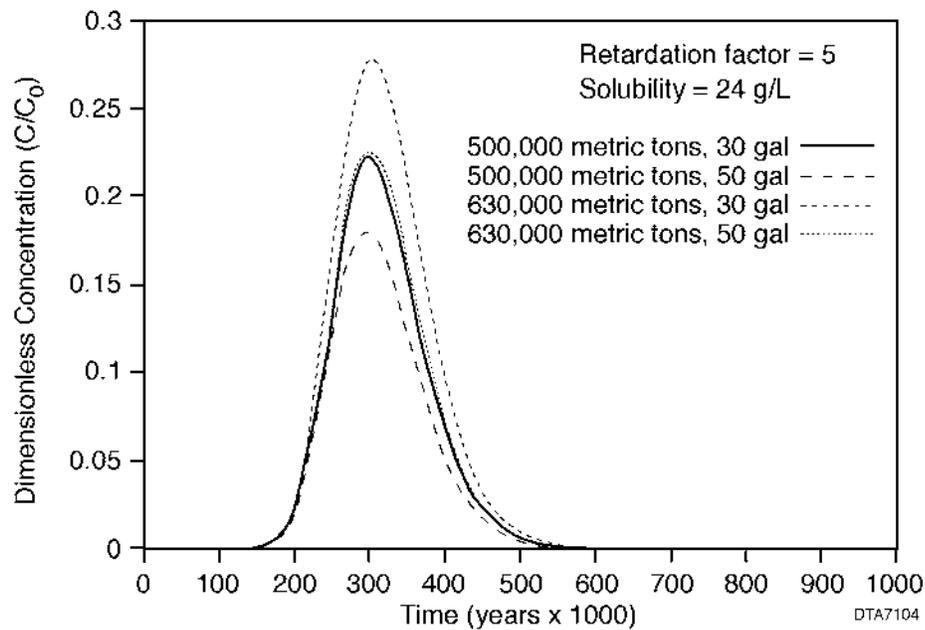

FIGURE 4 Breakthrough Curves for a Vault for a Fairly Mobile, Very Soluble Uranium Complex

FIGURE 5 Breakthrough Curves for a Mine for a Fairly Mobile, Very Soluble Uranium Complex

concentration, 0.06, would occur for a mine after about 60,000 years. For an initial source term concentration of 24 g/L, the maximum concentration would be 1.4 g/L.

In order to estimate the activity concentration for the highly soluble uranium complex, its chemical composition must be known. The actual chemical composition and also the solubility of this compound are site-specific parameters that depend on the geochemistry present. For the present calculations, the highly soluble uranium complex is assumed to be $(NH_4)_4[UO_2(CO_3)]_3$.

The specific activity, *SA*, of a carrier-free radionuclide (i.e., one that does not contain any other isotopes of the radionuclide) can be estimated by using the following relationship (Cember 1989):

$$SA = \frac{4.18 \times 10^{23}}{At_{1/2} \; 3.3 \times 10^{10}} \; , \tag{6}$$

where *A* is the molecular weight of the compound and $t_{1/2}$ is its half life in seconds. For a molecular weight of 492 g/mole, the specific activity of the highly soluble uranium complex is approximately 1.6×10^{-7} Ci/g. For this specific activity, the maximum uranium activity concentrations at the water table for a retardation of 5 would all be very large, exceeding 160,000 pCi/L (Table 4).

If the highly soluble uranium complex is significantly retarded (retardation of 50), the maximum uranium concentrations at the water table would be smaller than those discussed above (Table 4 and Figures 6 through 8). For this case, the largest dimensionless concentration, 0.043, would occur after 2.9 million years for a failed trench in which 500,000 t of UF_4 were disposed of in 30-gal drums. The smallest maximum dimensionless concentration, 0.006, would occur after 450,000 years for a failed mine in which 500,000 t of UF_4 were disposed of in 30-gal drums. Although these dimensionless concentrations are less than 1.0, the uranium activity concentrations would still be very large, exceeding 22,000 pCi/L.

Although the maximum uranium activity concentrations at the water table would be very large for any of the scenarios, dimensionless concentrations, actual concentrations, and activity concentrations after 1,000 years would be very small (Table 5). For a fairly mobile form of highly soluble uranium (retardation of 5), the maximum activity concentration would be 2.9×10^{-3} pCi/L for a failed mine in which 630,000 t of UF_4 were disposed of in 50-gal drums. For a retardation of 50, the maximum activity concentration of uranium after 1,000 years would be 3.6×10^{-5} pCi/L, once again for a failed mine in which 630,000 t of UF_4 were disposed of in 50-gal drums.

If the form of uranium released was fairly mobile schoepite, the maximum activity concentration at the water table after 1,000 years would be 2.3×10^{-6} pCi/L for disposing of the UF_4 in a mine. This value would be independent of the size of the storage container or the total mass that was disposed of because of the very long duration of the source.

All of the above estimated concentrations and activity concentrations were values for water within the unsaturated soil column. At the water table, additional dilution will occur when the contaminated water from the unsaturated enters the zone of saturation and mixes with initially clean water. The amount of dilution, D_f, can be estimated with the following expression (Tomasko 1997a):

$$D_f = \frac{V_d T}{IL\Phi_v} + 1 \, ,$$
(7)

where

I = rate of water infiltration,

L = length of the disposal cell footprint parallel for the direction of groundwater flow,

T = thickness of the saturated zone of groundwater, and

Φ_v = porosity or water content of the unsaturated zone.
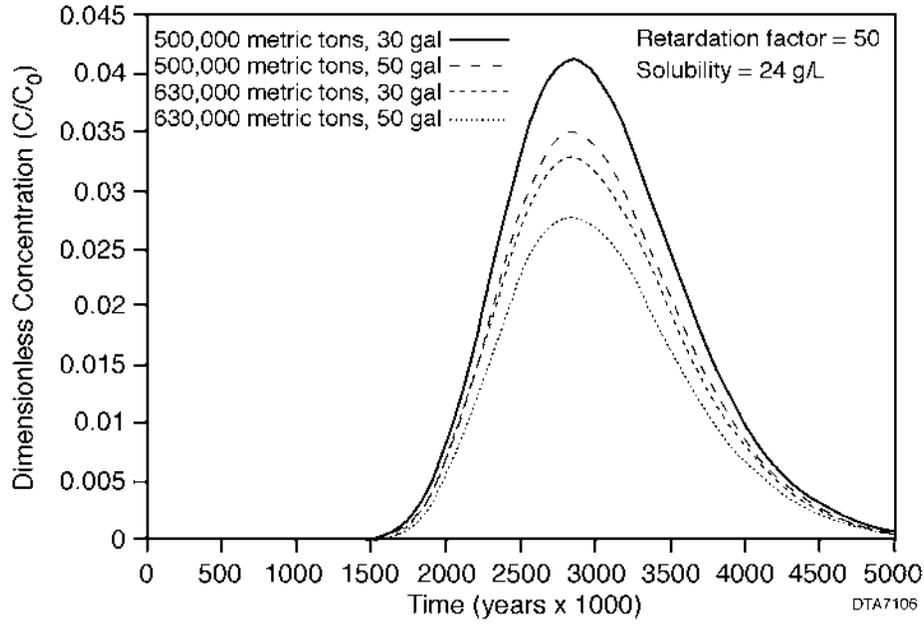
FIGURE 6 Breakthrough Curves for a Trench for a Fairly Immobile, Very Soluble Uranium Complex
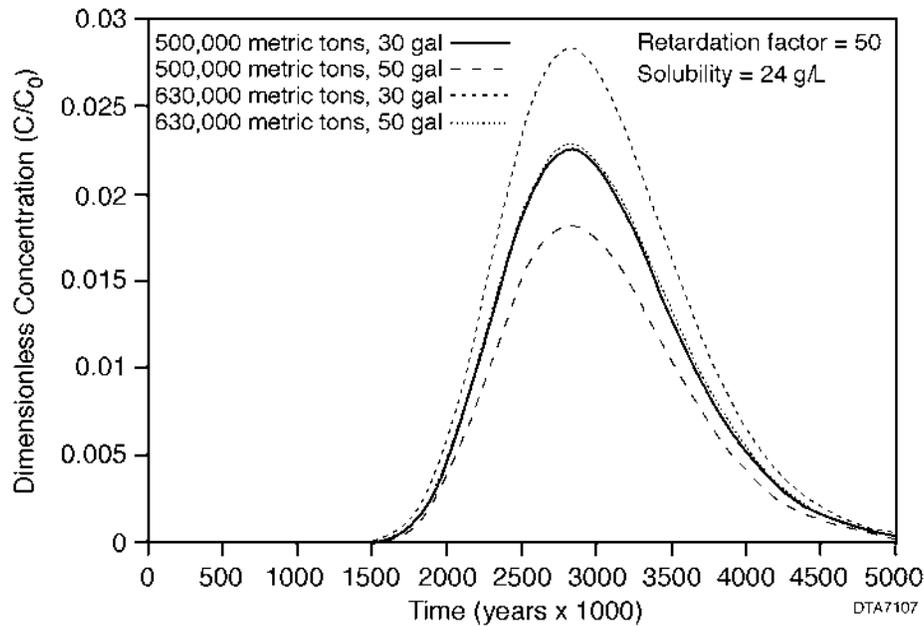
FIGURE 7 Breakthrough Curves for a Vault for a Fairly Immobile, Very Soluble Uranium Complex
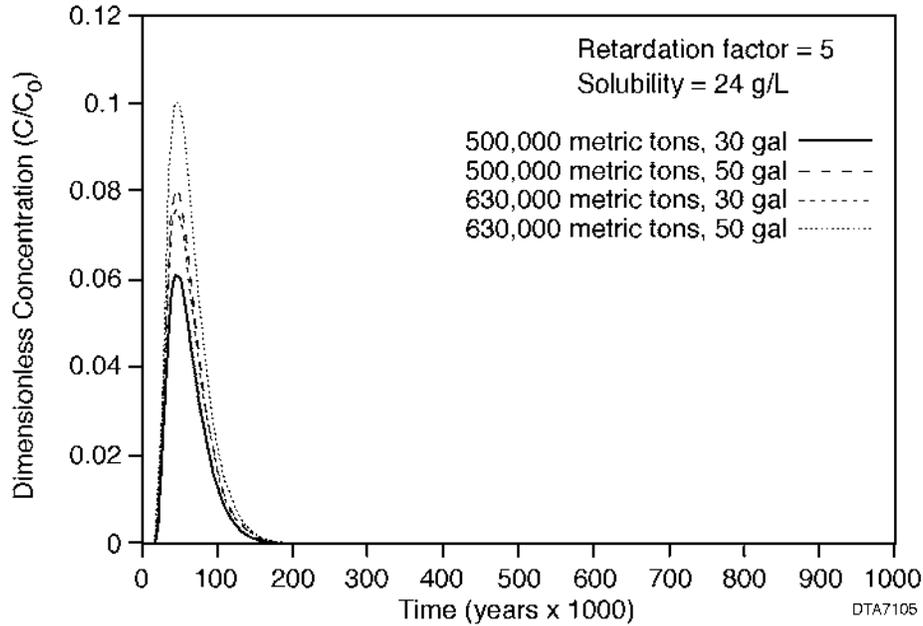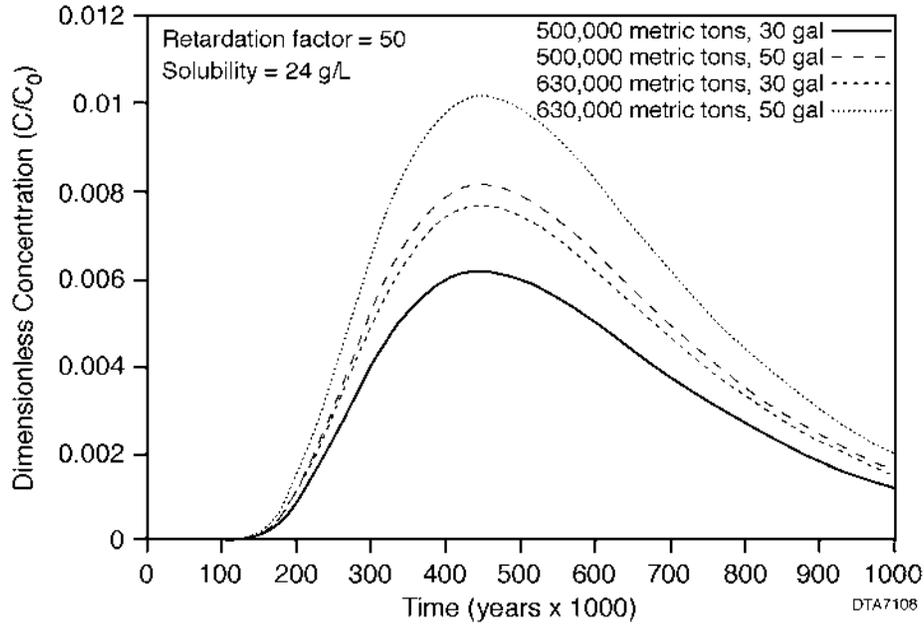
FIGURE 8 Breakthrough Curves for a Mine for a Fairly Immobile, Very Soluble Uranium Complex

The magnitude of the Darcy velocity, V_d, in Equation 7 is given by the following expression (Freeze and Cherry 1979):

$$V_d = K\nabla h \ , \tag{8}$$

where K is the hydraulic conductivity of the saturated zone of groundwater and ∇h is the local hydraulic gradient present (change in water level with distance).

In order to estimate the amount of dilution at the water table, the length of the disposal facility parallel to the direction of groundwater flow is required. As indicated in Table 1, each of the alternative disposal facilities (trench, vault, and mine) has a footprint area that is a function of the number of drums required for disposing of the UF_4. For the present calculations, the footprints are assumed to be square and oriented with two of their opposite sides parallel to the direction of groundwater flow. The length of the facility parallel to the direction of groundwater flow is then equal to the square root of the footprint area (Table 6).

In addition to the lengths of the disposal facilities parallel to the direction of groundwater flow, the magnitude of the Darcy velocity is required. For a hydraulic conductivity of 11,000 ft/yr and a hydraulic gradient of 0.001 (Table 3), the magnitude of the Darcy velocity is 11 ft/yr. For a

TABLE 6 Lengths of Disposal Cell Footprint (in meters) Parallel to the Direction of Groundwater Flow

	500,000 t		630,000 t	
Facility	30-gal Drums	50-gal Drums	30-gal Drums	50-gal Drums
Trench	454	417	505	465
Vault	504	562	562	616
Mine	1,570	1,370	1,760	1,530

TABLE 7 Dilution Factors for Mixing at the Water Table

	500,000 t		630,000 t	
Facility	30-gal Drums	50-gal Drums	30-gal Drums	50-gal Drums
Trench	885	964	797	865
Vault	798	716	716	653
Mine	256	294	229	262

saturated zone thickness of 100 ft, an infiltration velocity of 0.1 in./yr, and a porosity of 0.1 (Table 3), the dilution factors for the three disposal facilities are given in Table 7. The dilution factor ranges from 229 for disposing of 630,000 t of UF_4 in a mine to 964 for disposing of 500,000 t of UF_4 in a trench in 50-gal drums. In general, the larger the facility (i.e., length of the disposal facility in a direction parallel to the direction of groundwater flow), the smaller the amount of dilution. A summary of concentration and activity concentrations after 1,000 years after mixing with initially clean water in the saturated zone is given in Table 8; maximum concentrations and activity concentrations after dilution are given in Table 9.

TABLE 8 Summary of Concentrations at 1,000 Years after Dilution at the Water Table

Solubility (g/L)	Facility	500,000 t in 30-gal Drums			500,000 t in 50-gal Drums		
		C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)
2.4×10^{-3}	Trench	2.4×10^{-12}	5.8×10^{-15}	1.4×10^{-9}	2.2×10^{-12}	5.2×10^{-15}	1.2×10^{-9}
		6.9×10^{-24}	1.7×10^{-26}	4.1×10^{-21}	6.3×10^{-24}	1.6×10^{-26}	3.7×10^{-21}
	Vault	2.6×10^{-12}	6.3×10^{-15}	1.5×10^{-9}	2.9×10^{-12}	7.0×10^{-15}	1.7×10^{-9}
		7.6×10^{-24}	1.9×10^{-26}	4.5×10^{-21}	8.5×10^{-24}	2.1×10^{-26}	5.0×10^{-21}
	Mine	1.5×10^{-11}	3.7×10^{-14}	9.0×10^{-9}	1.3×10^{-11}	3.2×10^{-14}	7.8×10^{-9}
		6.6×10^{-12}	1.6×10^{-14}	3.8×10^{-9}	5.8×10^{-12}	1.4×10^{-14}	3.3×10^{-9}
24	Trench	2.2×10^{-14}	5.1×10^{-13}	8.1×10^{-8}	2.0×10^{-14}	4.7×10^{-13}	7.5×10^{-8}
		3.8×10^{-24}	9.2×10^{-23}	1.5×10^{-17}	3.5×10^{-24}	8.5×10^{-23}	1.4×10^{-17}
	Vault	2.8×10^{-14}	6.6×10^{-13}	1.1×10^{-7}	3.1×10^{-14}	7.4×10^{-13}	1.2×10^{-7}
		3.8×10^{-24}	9.0×10^{-23}	1.5×10^{-17}	4.2×10^{-24}	1.0×10^{-22}	1.6×10^{-17}
	Mine	2.2×10^{-12}	5.5×10^{-11}	8.6×10^{-6}	1.9×10^{-12}	4.6×10^{-11}	7.4×10^{-6}
		2.2×10^{-14}	5.1×10^{-13}	8.2×10^{-8}	1.9×10^{-14}	4.5×10^{-13}	7.2×10^{-8}

Solubility (g/L)	Facility	630,000 t in 30-gal drums			630,000 t in 50-gal drums		
		C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration	Activity Concentration
2.4×10^{-3}	Trench	2.6×10^{-12}	6.3×10^{-15}	1.5×10^{-9}	2.4×10^{-12}	5.8×10^{-15}	1.4×10^{-9}
		7.7×10^{-24}	1.9×10^{-26}	4.5×10^{-21}	7.1×10^{-24}	1.7×10^{-26}	4.2×10^{-21}
	Vault	2.9×10^{-12}	7.0×10^{-15}	1.7×10^{-9}	3.2×10^{-12}	7.7×10^{-15}	1.8×10^{-9}
		7.7×10^{-24}	5.0×10^{-21}	1.2×10^{-15}	9.3×10^{-24}	2.3×10^{-26}	5.5×10^{-21}
	Mine	1.7×10^{-11}	4.1×10^{-14}	1.0×10^{-8}	1.5×10^{-11}	3.6×10^{-14}	8.8×10^{-9}
		7.4×10^{-12}	1.8×10^{-14}	4.2×10^{-9}	6.5×10^{-12}	1.5×10^{-14}	3.7×10^{-9}
24	Trench	2.3×10^{-14}	5.5×10^{-13}	8.9×10^{-8}	2.1×10^{-14}	5.0×10^{-13}	8.0×10^{-8}
		4.8×10^{-24}	1.1×10^{-22}	1.8×10^{-17}	4.4×10^{-24}	1.1×10^{-22}	1.8×10^{-17}
	Vault	2.8×10^{-14}	6.6×10^{-13}	1.1×10^{-7}	3.1×10^{-14}	7.4×10^{-13}	1.2×10^{-7}
		4.8×10^{-24}	1.2×10^{-22}	1.8×10^{-17}	5.2×10^{-24}	1.3×10^{-22}	2.1×10^{-17}
	Mine	2.4×10^{-12}	5.7×10^{-11}	9.2×10^{-6}	2.1×10^{-12}	5.0×10^{-11}	8.0×10^{-6}
		3.0×10^{-14}	7.0×10^{-13}	1.1×10^{-7}	2.6×10^{-14}	6.2×10^{-13}	9.9×10^{-8}

TABLE 9 Maximum Concentrations and Occurrence Times for Long-Term Disposal of UF$_4$ after Dilution

Solubility (g/L)	R[a]	Facility	500,000 t in 30-gal Drums			500,000 t in 50-gal Drums		
			C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)
2.4×10^{-3}	5	Trench	1.1×10^{-3} 550,000 yr	2.7×10^{-6}	0.7	1.0×10^{-3} 550,000 yr	2.4×10^{-6}	0.6
		Vault	1.3×10^{-3} 550,000 yr	3.0×10^{-6}	0.7	1.4×10^{-3} 550,000 yr	2.4×10^{-3}	0.8
		Mine	3.9×10^{-3} 200,000 yr	9.4×10^{-6}	2.3	3.4×10^{-3} 200,000 yr	8.2×10^{-6}	2.0
	50	Trench	1.1×10^{-3} 5,000,000 yr	2.7×10^{-6}	0.7	1.1×10^{-3} 5,000,000 yr	2.4×10^{-6}	0.6
		Vault	1.3×10^{-3} 5,000,000 yr	3.0×10^{-6}	0.7	1.3×10^{-3} 5,000,000 yr	2.4×10^{-3}	0.8
		Mine	3.9×10^{-3} 2,000,000 yr	9.4×10^{-6}	2.3	3.9×10^{-3} 2,000,000 yr	8.2×10^{-6}	2.0
24	5	Trench	3.1×10^{-4} 310,000 yr	7.3×10^{-3}	1,170	3.3×10^{-4} 315,000 yr	8.0×10^{-3}	1,270
		Vault	2.8×10^{-4} 305,000 yr	6.6×10^{-3}	1,060	2.5×10^{-4} 300,000 yr	6.0×10^{-3}	965
		Mine	2.3×10^{-4} 60,000 yr	5.6×10^{-3}	900	2.7×10^{-4} 65,000 yr	6.5×10^{-3}	1,040
	50	Trench	4.9×10^{-5} 2,900,000 yr	1.2×10^{-3}	187	3.7×10^{-5} 2,850,000 yr	9.0×10^{-4}	143
		Vault	2.8×10^{-5} 2,850,000 yr	6.6×10^{-4}	106	2.8×10^{-5} 2,800,000 yr	5.7×10^{-4}	91
		Mine	2.3×10^{-5} 450,000 yr	5.6×10^{-4}	90	2.8×10^{-5} 445,000 yr	6.7×10^{-4}	107

Solubility (g/L)	R[a]	Facility	630,000 t in 30-gal Drums			630,000 t in 50-gal Drums		
			C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)
2.4×10^{-3}	5	Trench	1.3×10^{-3} 550,000 yr	3.0×10^{-6}	0.7	1.2×10^{-3} 550,000 yr	2.8×10^{-6}	0.7
		Vault	1.4×10^{-3} 550,000 yr	3.4×10^{-6}	0.8	1.5×10^{-3} 550,000 yr	3.7×10^{-6}	0.9
		Mine	4.4×10^{-3} 200,000 yr	1.1×10^{-5}	2.5	3.8×10^{-3} 200,000 yr	9.2×10^{-6}	2.2
	50	Trench	1.3×10^{-3} 5,000,000 yr	3.0×10^{-6}	0.7	1.2×10^{-3} 5,000,000 yr	2.8×10^{-6}	0.7
		Vault	1.4×10^{-3} 5,000,000 yr	3.4×10^{-6}	0.8	1.5×10^{-3} 5,000,000 yr	3.7×10^{-6}	0.9
		Mine	4.4×10^{-3} 2,000,000 yr	1.1×10^{-5}	2.5	3.8×10^{-3} 2,000,000 yr	9.2×10^{-6}	2.2

TABLE 9 (Cont.)

Solubility (g/L)	R^a	Facility	630,000 t in 30-gal Drums			630,000 t in 50-gal Drums		
			C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)	C/C_0	Concentration (g/L)	Activity Concentration (pCi/L)
24	5	Trench	4.3×10^{-4} 310,000 yr	1.0×10^{-2}	1,640	4.5×10^{-4} 315,000 yr	1.1×10^{-2}	1,730
		Vault	3.9×10^{-4} 310,000 yr	9.4×10^{-3}	1,500	3.5×10^{-4} 308,000 yr	8.5×10^{-3}	1,350
		Mine	3.3×10^{-4} 65,000 yr	7.9×10^{-3}	1,260	3.8×10^{-4} 75,000 yr	9.2×10^{-3}	1,470
	50	Trench	4.1×10^{-5} 2,800,000 yr	9.9×10^{-4}	159	3.2×10^{-5} 2,700,000 yr	7.8×10^{-4}	124
		Vault	3.9×10^{-5} 2,900,000 yr	9.4×10^{-4}	150	3.5×10^{-5} 2,870,000 yr	8.3×10^{-4}	132
		Mine	3.3×10^{-5} 452,000 yr	7.9×10^{-4}	126	3.8×10^{-5} 452,000 yr	9.2×10^{-4}	147

[a] R = retardation factor.

4 DISCUSSION

As indicated in Table 8, all of the contaminant concentrations and activity concentrations at the water table would be very low, even for a fairly mobile compound (retardation of 5). In general, the concentrations after 1,000 years at the water table would be about an order of magnitude greater for disposal in a deep mine than for disposal in a trench or vault.

Out of all the options and potential contaminants considered in this study, the largest activity concentration at the water table after 1,000 years would be derived from a failed mine that released a fairly mobile and highly soluble uranium complex. This concentration would be about 8.6×10^{-6} pCi/L for 500,000 t of UF_4 disposed of in 30-gal drums. The smallest activity concentrations at the water table after 1,000 years would, in general, be derived from a failed trench (1.2×10^{-9} pCi/L), if the contaminant reached the water table as schoepite.

Although all the activity concentrations at the water table after 1,000 years are predicted to be small, maximum activity concentrations could still be large, even after dilution (Table 9). Maximum activity concentrations of uranium would exceed 900 pCi/L for a failed disposal facility if a highly soluble uranium complex formed and if this compound was fairly mobile in the groundwater system. If the solubility of the uranium compound was small (2.4×10^{-3} g/L), the resulting uranium activity concentrations would be small, less than 2.5 pCi/L.

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