Assessing the Impact of Hazardous Constituents on the Mobilization, Transport, and Fate of Radionuclides in RCRA Waste Disposal Units

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NOTATION

The following is a list of the acronyms, initialisms, and abbreviations (including units of measure) used in this document. Acronyms and abbreviations used only in tables and figures are defined in the respective tables and figures.

ACRONYMS AND ABBREVIATIONS

C	liquid (phase in soil)
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act of 1980
DOC	dissolved organic carbon
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
K_d	distribution coefficient
LHS	Latin hypercube sampling
LLMW	low-level-radioactivity mixed waste
MEI	maximally exposed individual
MSW	municipal solid waste
PNNL	Pacific Northwest National Laboratory
RCRA	Resource Conservation and Recovery Act
S	solid (phase in soil)
TOC	total organic carbon

UNITS OF MEASURE

cm	centimer(s)
cm^3	cubic centimeter(s)
d	day(s)
g	gram(s)
kg	kilogram(s)
L	liter(s)
m	meter(s)
m^2	square meter(s)
m^3	cubic meter(s)
µm	micrometer(s)
pCi	picocurie(s)
ppm	part(s) per million
yr	year(s)

ASSESSING THE IMPACT OF HAZARDOUS CONSTITUENTS ON THE MOBILIZATION, TRANSPORT, AND FATE OF RADIONUCLIDES IN RCRA WASTE DISPOSAL UNITS

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ABSTRACT

This report discusses the impact that hazardous organic chemical constituents could have on the mobilization, transport, and fate of radionuclides in disposal units regulated by the Resource Conservation and Recovery Act (RCRA). The effect on a radionuclide's distribution coefficient (K_d) is used as an indicator. Many factors can affect K_d, including the chemical form of the radionuclide, pH of the leachate, nature of the organic constituents, porosity of the soil, amount of water in the landfill, infiltration rate of the water, presence of a chelating agent or other chemical species, and age of the landfill. A total of 19 radionuclides were studied. Of these, nine (H-3, C-14, Se-79, Sr-90, Tc-99, I-129, U-238, Np-237, and Am-241) were found to have the potential to reach groundwater and cause contamination; the remaining 10 (Co-60, Ni-63, Sb-125, Cs-137, Sm-151, Eu-152, Eu-154, Th-230, Th-232, and Pu-239) were considered less likely to cause groundwater contamination. It was also found that when organic material is in solution, it tends to lower a radionuclide's K_d (and enhance transport), whereas when it is in a solid phase, it tends to increase the K_d. The study introduces a simple model to estimate effective K_d values on the basis of total organic carbon concentrations in landfill leachate. However, given the fact that the effective K_d values of radionuclides in RCRA disposal units can either increase or decrease as the result of many factors, including the form of the organic matter (solid or in solution), the study concludes that whenever they are available, actual (measured) K_d values rather than modeled values should be used to conduct dose and risk assessments of radionuclides in RCRA disposal units.

1 INTRODUCTION

Disposal of low-level-radioactivity mixed waste (LLMW) in a Resource Conservation and Recovery Act (RCRA) waste disposal unit was studied previously by Pacific Northwest National

Laboratory (PNNL 1995, 1997). In one study, the potential impacts of disposal cell failure were studied by assuming a leaking liner (PNNL 1997). However, the potential effect of the waste's hazardous constituents on the mobilization of radionuclides was not addressed.

The leaching of contamination depends on several factors. The most important are the infiltration rate of water and the adsorbing capability of the waste matrix to retain the waste constituents. The adsorbing capability of the matrix relative to a particular constituent is usually gauged by the distribution coefficient (K_d) of that constituent and is influenced by properties of the matrix material and the co-existence of other constituents. The infiltration rate of water depends on the engineering design of the disposal unit and the degree of cover failure.

The main objective of this study is to evaluate the effect of chemical constituents in LLMW on the mobilization, transport, and fate of radionuclides in RCRA waste disposal units. To study this effect, a sensitivity analysis was conducted on the K_d values (K_ds) by using the RESRAD computer code (Yu et al. 1993). The results are presented in Section 2 and Appendix B. The factors that affect the K_d in RCRA disposal units are discussed in Section 3, and a proposed model for the estimation of K_d is presented in Section 4. A discussion and summary of the potential effects of organic materials on radionuclide migration in landfills is given in Section 5. The bounding ranges of K_ds for selected radionuclides are presented in Appendix C.

A total of 19 radionuclides are considered in this study: H-3, C-14, Co-60, Ni-63, Se-79, Sr-90, Tc-99, Sb-125, I-129, Cs-137, Sm-151, Eu-152, Eu-154, Th-230, Th-232, U-238, Np-237, Pu-239, and Am-241. These radionuclides were selected by both the U.S. Department of Energy (DOE) and Argonne National Laboratory (Argonne) as radionuclides potentially found in RCRA waste disposal units.

2 K_d SENSITIVITY ANALYSIS

2.1 BACKGROUND

The RESRAD computer code version 5.95, which incorporates an uncertainty analysis module, was used in this study for dose assessment. Ranges and distributions of K_ds used in the analyses were developed on the basis of a literature search (Biwer et al. 2000) and accounted for different soil types.

Several mechanisms affect the migration potential of a radionuclide in soils. To simplify mathematical modeling, the typical approach applied in risk assessments is to use an empirical parameter, K_d, to lump all the influencing factors into one value. The K_d parameter estimates the distribution of radionuclides between the solid phase (S) and liquid phase (C) in soil. In the K_d model, it is implicitly assumed that the liquid and solid phases in soil are at equilibrium and that there is a linear relationship (linear isotherm) between radionuclide concentration in the solid and liquid phases (Sheppard 1985), as expressed by the equation: $S = K_d C$. This linear isotherm assumption is a good assumption when the concentrations are low, as is the case for LLMW.

A large K_d implies that the degree to which the radionuclide is tightly bound to soil particles in the solid phase is greater than the degree to which it is dissolved in the liquid phase. As a result, when water infiltrates through the disposal unit, fewer radionuclides will dissolve in the water (liquid phase) and leach out to the surrounding environment. In contrast, a small K_d implies that the radionuclide will leach out more quickly from the adsorbed (solid) phase and be transported more quickly in soils. More detailed discussions on mechanisms that affect the K_d values are provided in Section 3.

Once radionuclides leach out from the disposal unit, they will transport downward to deeper (vadose) soils with the infiltration water and eventually reach the groundwater table. The time required for radionuclides to reach the groundwater table depends on the K_d values of the radionuclides in the vadose soils. After reaching the groundwater table, radionuclides will be carried by the groundwater and transported to downgradient locations.

In summary, the K_ds of radionuclides in the contaminated zone (disposal unit), unsaturated zones (vadose soils), and saturated zone (groundwater) affect the transport mechanism and play a role in determining the resulting potential radiation exposure. Because environmental conditions vary from site to site, so do values of the K_d parameter. The range of potential radiation exposure cannot be analyzed without the knowledge of how the K_d parameter and the environmental setting affect the transport mechanism. Mathematical formulations used in the RESRAD code that describe these relationships are discussed in Appendix A. The results of the sensitivity analyses are presented in the following section.

2.2 SENSITIVITY ANALYSES ON THE K_d PARAMETER

Sensitivity analyses to study impacts of the K_ds on potential radiation exposures were conducted by using the RESRAD code, version 5.95, for a hypothetical RCRA waste disposal unit. Information for the hypothetical waste disposal unit was developed on the basis of information about the Beatty, Nevada, site as described in a draft report by PNNL (1997). The hypothetical site has an area of 45,400 m^2 (305 × 149 m) and a thickness of 30.5 m. The length of the contaminated zone parallel to the flow direction of the groundwater aquifer is 305 m. The groundwater aquifer is assumed to be located 76 m below the contaminated zone.

The integrity of the hypothetical RCRA disposal unit was assumed to fail after a certain period of operation. The failure resulted in infiltration of rain water to the disposal cell at a rate of 4.1 cm/yr. The rain water penetrated the cover layer (which was assumed to be composed of 2.4 m of clay soil), percolated through the contaminated zone, and then passed through the bottom liner (0.2-m thick). The leachate collection system was assumed to be nonfunctioning so that all the infiltration water flowed downward to the vadose soils.

A well at the downgradient edge of the contaminated zone was assumed to be a source of drinking water. The exposure parameters considered for the maximally exposed individual (MEI) were 2 L/d of drinking water and 365 d/yr of exposure frequency.

To simplify interpretation of the K_d impacts, radiation exposures were considered from only the drinking water pathway. In this way, the estimated radiation exposures would be proportional to the concentration of radionuclides in well water, which then would be controlled by values of the leach rate, breakthrough time, rise time, and dilution factor. As discussed in Appendix A, the four water transport factors are directly affected by K_ds in different soil layers.

2.2.1 Variation Cases

In addition to the K_ds, other site-specific parameters can affect the potential radiation exposure of a human receptor. Impacts of these parameters on the well water concentrations are described by equations listed in Appendix A. In general, radionuclide concentrations in well water depend on three variables: (1) the distance of transport, (2) the transport speed of the pore water, and (3) the transport speeds of radionuclides relative to the transport speed of water. The impact from the third variable, which can be studied by varying the K_ds in different soil layers, is the focus of this sensitivity analysis. In the analysis, the K_ds were assigned a lognormal distribution, and distribution parameters were obtained from a previous Argonne report (Biwer et al. 2000) that compiled data from various literature sources. In conjunction with the K_ds, the water infiltration rate, thickness of the unsaturated zone, and thickness of the liner were also varied so that impacts on potential radiation exposure from the first two variables could also be studied. Two water infiltration rates

were used in the analysis: 0.041 m/yr to stimulate dry climate conditions and 0.4572 m/yr to simulate wet ones. The two thicknesses of the unsaturated zones used were 76 m for deep aquifer conditions and 3 m for shallow. The disposal unit with and without a liner (or a liner that failed) was simulated by using a liner thickness of 0.2 m and 0 m, respectively.

Eight cases were developed and analyzed by using the RESRAD computer code. Table 1 lists the parameter values used in the different cases. Parameters whose values were varied during the analyses are shaded. Those that were not listed were set to the RESRAD default values. To obtain an infiltration rate of 0.041 m/yr, an evapotranspiration coefficient of 0.55, a precipitation rate of 0.114 m/yr (dry site), an irrigation rate of 0 m/yr, and a runoff coefficient of 0.2 were used. An infiltration rate of 0.4572 m/yr was obtained by assigning a precipitation rate of 1.27 m/yr (wet site). K_d distributions were assigned to the unsaturated and saturated zones for all eight cases. For Cases I to VI, fixed K_d values were used for the contaminated zone and the liner. The fixed values correspond to those measured for a grouted waste and a clay liner, respectively, as specified in Table F.3 of the draft PNNL report (PNNL 1997). When the measured values were not available, the RESRAD default values were used as the fixed values. In Cases VII and VIII, K_d values in the contaminated zone were also assigned a distribution so that impacts of chemical constituents on mobilities of the radionuclides could be simulated. Table 2 lists the fixed K_d values and the distributions used in the RESRAD analyses.

To single out the impact of K_d value of the contaminated zone, a varied Case VIII was studied for Tc-99. In the varied Case VIII, only the K_d value of the contaminated zone was assigned a distribution; K_d values of the unsaturated and saturated zones were set to the RESRAD default value.

2.2.2 Calculation Results and Discussion

Concentrations of radionuclides in the disposal cell vary from site to site. For comparison purposes, a concentration of 100 pCi/g was considered for each radionuclide, and the peak radiation doses within 1,000 years after failure of the disposal facility were compared. The Latin hypercube sampling (LHS) method was used to generate 300 sets of different K_ds for each analytical case according to the specified distribution characteristics. Each set of K_ds was then combined with the other input parameters with fixed values to form an input data set for use by the RESRAD code. The RESRAD calculations were performed for each input data set and were repeated 300 times for each analytical case. With 300 sets of calculation results, statistical analysis on the peak radiation doses were performed to determine the 5-percentile and 95-percentile values.

TABLE 1 RESRAD Input Parameters Used in the Analyses[a]

RESRAD Parameter	Cases Analyzed							
	I	II	III	IV	V	VI	VII	VIII
RCRA Waste Disposal Unit (Cover and Contaminated Zone) Parameters								
Area of contaminated zone (m²)	45,400[b]							
Thickness of contaminated zone (m)	30.5[b]							
Length parallel to aquifer flow (m)	305[b]							
Cover depth (m)	2.4[b]							
Density of cover material (g/cm)	1.5[b]							
Cover depth erosion rate (m/yr)	0.001[b]							
Infiltration rate following liner failure (m/yr)[c]	0.041	0.041	0.041	0.041	0.1143	0.1143	0.041	0.041
Density of contaminated material (g/cm³)	1.6[b]							
Liner Parameters								
Thickness of clay liner	0.2	0.2	0	0	0	0	0	0
Density (g/cm³)	1.5	1.5	NA[d]	NA	NA	NA	NA	NA
Total porosity	0.448	0.448	NA	NA	NA	NA	NA	NA
Effective porosity	0.448	0.448	NA	NA	NA	NA	NA	NA
Soil-specific exponential b parameter	10	10	NA	NA	NA	NA	NA	NA
Hydraulic conductivity (m/yr)	0.041	0.041	NA	NA	NA	NA	NA	NA
Saturated Zone Parameters								
Aquifer velocity (m/yr)	2[b]							
Aquifer porosity	0.4[b]							
Unsaturated (Vadose) Zone Parameters								
Thickness of unsaturated (vadose) zone (m)	76	3	76	3	76	3	76	3
Uncertainty Analysis								
Contaminated zone K_d	No	No	No	No	No	No	Yes	Yes
Liner K_d	No	No	NA	NA	NA	NA	NA	NA
Unsaturated zone K_d	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Saturated zone K_d	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes

[a] Parameters not listed in this table were assigned the RESRAD default values. Parameters whose values were varied during the analyses are shaded.

[b] Values apply to all eight cases.

[c] An infiltration rate of 0.041 m/yr was obtained by assigning a precipitation rate of 0.114 m/yr, a runoff coefficient of 0.2, and an evapotranspiration coefficient of 0.55. An infiltration rate of 0.1143 m/yr was obtained by assigning a precipitation rate of 1.27 m/yr, a runoff coefficient of 0.2, and an evapotranspiration coefficient of 0.55.

[d] NA = not applicable.

TABLE 2 Fixed and Distribution Characteristics of K_d Values (cm³/g) Used in the RESRAD Analyses

Element	Contaminated Zone[a]	Liner[a]	Lognormal Distribution[b]		
			μ	σ	exp(μ)
H	0	0	-2.81	0.5	0.06
C	2,625	0	2.4	3.22	11
Co	125	1,000	5.46	2.53	235
Ni	1,000*	1,000*	6.05	1.46	424
Se	0*	0*	4.73	0.57	113
Sr	125	100	3.45	2.12	32
Tc	2	1	-0.67	3.16	0.51
Sb	0*	0*	5.94	3.22	380
I	0	1	1.52	2.19	4.6
Cs	125	1,000	6.1	2.33	446
Sm	825*	825*	6.72	3.22	825
Th	60,000*	60,000*	8.68	3.62	5884
U	2,625	100	4.84	3.13	126
Np	125	40	2.84	2.25	17
Pu	2,625	2,000	6.86	1.89	953
Am	2,625	1,000	7.28	3.15	1445
Eu	825*	825*	6.72	3.22	825

[a] Values for the contaminated zone and liner are those for grouted waste and clay liner, respectively, listed in Table F.3 of the PNNL report (1997). Values followed by an asterisk (*) are RESRAD default values.

[b] In the RESRAD analyses, lognormal distribution for the K_d values was assumed. The distribution parameters were obtained from a previous Argonne report (Biwer et al. 2000). μ = mean of the underlying normal distribution after taking the natural logarithm of the K_d values; σ = standard deviation of the underlying normal distribution after taking the natural logarithm of the K_d values; exp(μ) = exponential of the mean value.

Figure 1 shows the typical scatter plot of the peak radiation doses versus Tc-99 K_d of the contaminated zone obtained for one of the analytical cases. Figure 2 shows the corresponding cumulative plot for the peak radiation doses versus Tc-99 K_d of the contaminated zone. Figure 3 shows the typical scatter plot of the peak radiation doses versus Np-237 K_d of the unsaturated zone obtained for one of the analytical cases. Figure 4 shows the corresponding cumulative plot for the peak radiation doses versus Np-237 K_d of the unsaturated zone. Numerical values for the 5-percentile and 95-percentile peak radiation doses for all the analytical cases are listed in Table 3. The 5-percentile and 95-percentile values were selected to study the potential range of the peak radiation dose.

It can be seen from Table 3 that for 10 radionuclides (i.e., Co-60, Ni-63, Sb-125, Cs-137, Sm-151, Th-230, Th-232, Pu-239, Eu-152, and Eu-154), the listed 95-percentile dose-source ratios for the eight cases studied are all less than 1 mrem/yr per 100 pCi/g of radionuclide within 1,000 years of the time of calculation. For the other nine radionuclides studied (i.e., H-3, C-14, Se-79, Sr-90, Tc-99, I-129, U-238, Np-237, and Am-241), the dose-source ratios exceeded 1 mrem/yr per 100 pCi/g from drinking the potentially contaminated groundwater for the hypothetical RCRA site. These results may vary from site to site, depending on the K_ds, the water table depth, and whether the climate of the site is dry or wet. Additional discussion on the results for these radionuclides is included in Appendix B.
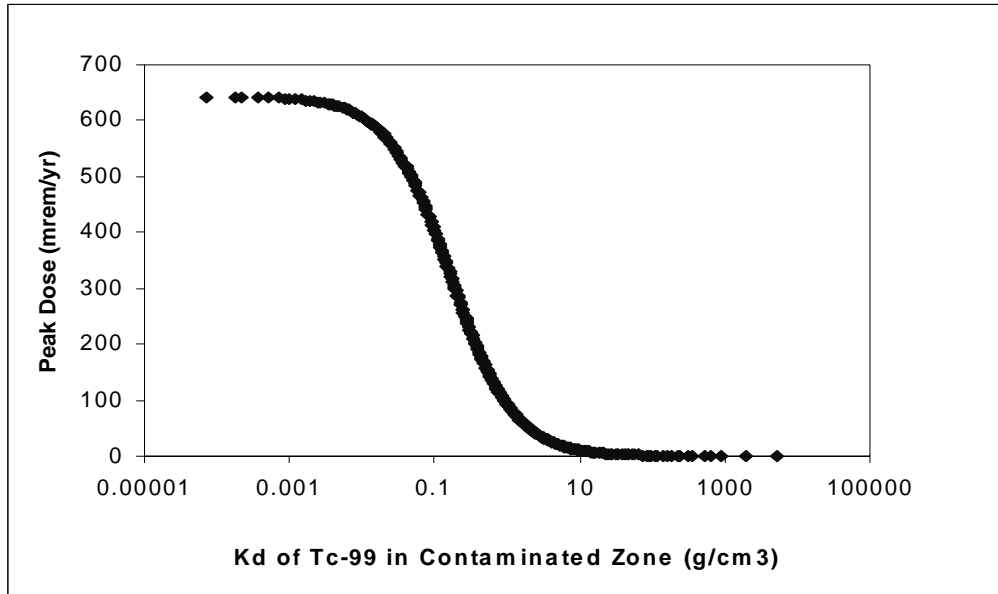
FIGURE 1 Scatter Plot of the Peak Dose versus K_d in the Contaminated Zone for Tc-99 in Varied Case VIII
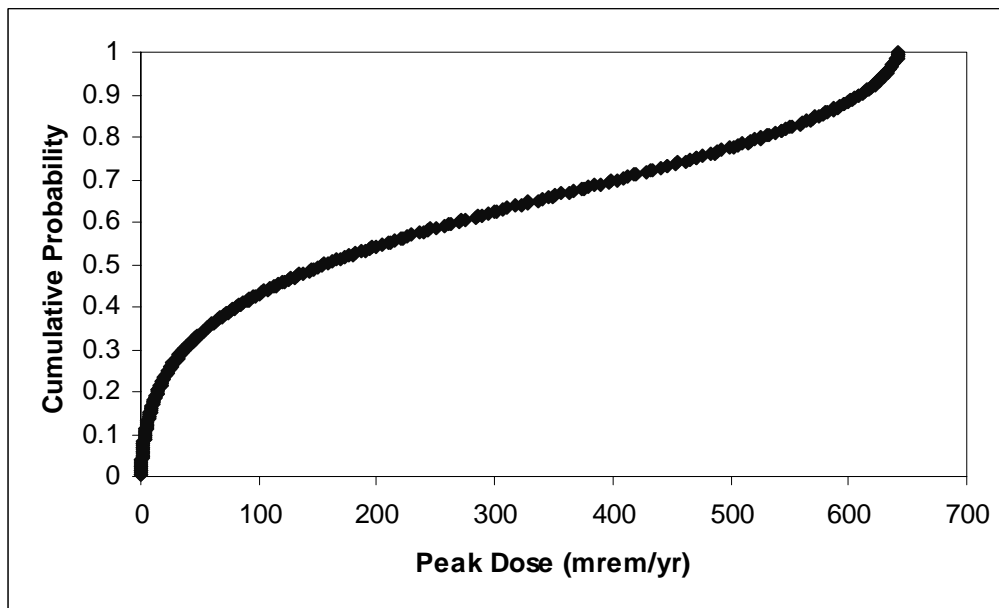
FIGURE 2 Cumulative Probability of the Peak Dose for Tc-99 in Varied Case VIII
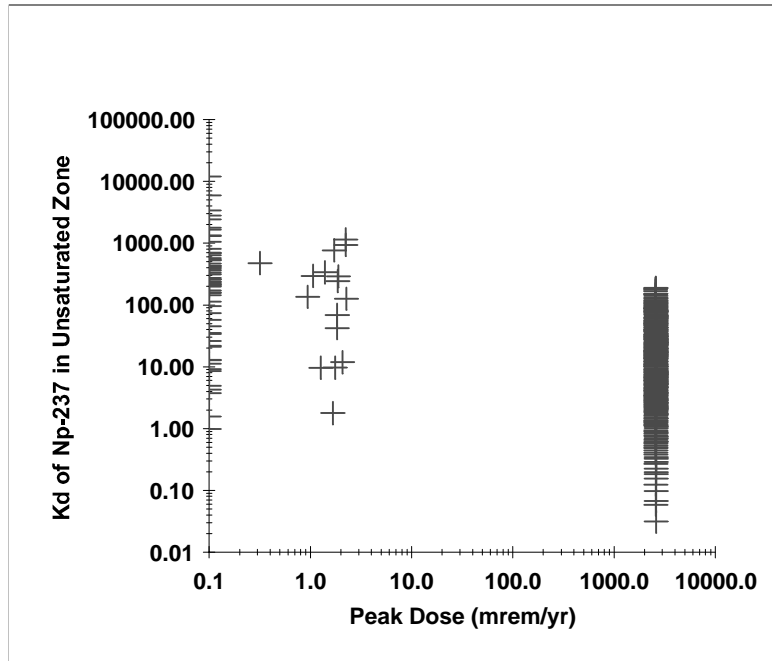
FIGURE 3 Scatter Plot of the Peak Dose versus K_d in the Unsaturated Zone for Np-237 in Case VI
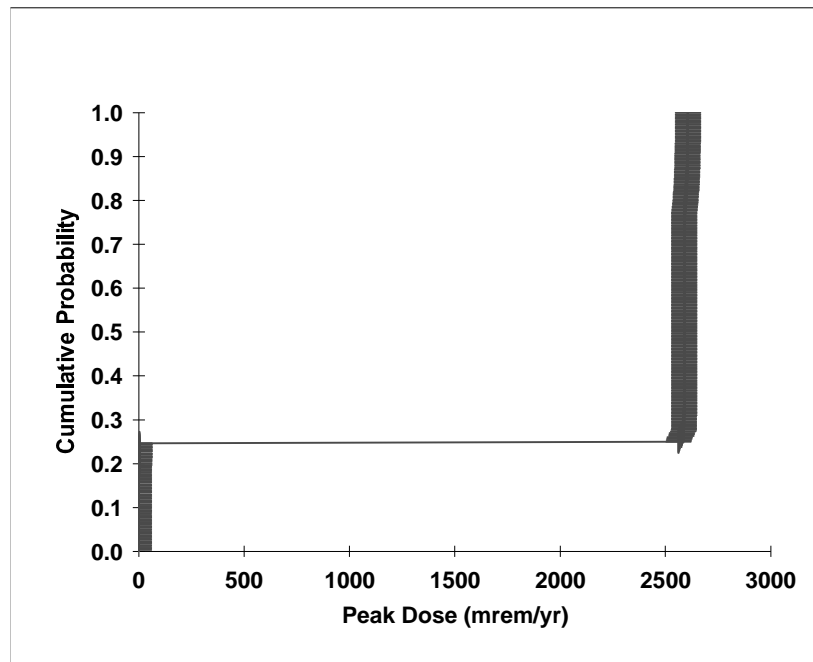
FIGURE 4 Cumulative Probability of the Peak Dose for Np-237 in Case VI

TABLE 3 Summary of the RESRAD Calculation Results: 5-Percentile and 95-Percentile Values of the Peak Doses (mrem/yr) per 100 pCi/g of Radionuclides

Radionuclide	Case I 5%	Case I 95%	Case II 5%	Case II 95%	Case III 5%	Case III 95%	Case IV 5%	Case IV 95%
H-3	3.84E-10	2.52E-06	9.25E+00	1.31E+01	4.35E-10	2.83E-06	1.05E+01	1.48E+01
C-14	0	5.49E-02	0	5.80E-02	0	5.50E-02	0	5.80E-02
Co-60	0	0	0	0	0	0	0	0
Ni-63	0	0	0	0	0	0	0	0
Se-79	0	0	0	0	0	0	0	0
Sr-90	0	0	0	0	0	0	0	1.49E+00
Tc-99	0	4.92E+01	0	4.95E+01	0	4.93E+01	0	4.95E+01
Sb-125	0	0	0	0	0	0	0	0
I-129	0	1.21E+05	0	1.21E+05	0	1.21E+05	0	1.21E+05
Cs-137	0	0	0	0	0	0	0	0
Sm-151	0	0	0	0	0	0	0	0
Th-230	0	0	0	0	0	0	0	0
Th-232	0	0	0	0	0	0	0	0
U-238	0	0	0	0	0	0	0	7.48E+00
Np-237	0	0	0	2.59E+03	0	0	0	2.59E+03
Pu-239	0	0	0	2.59E-05	0	0	0	1.86E-04
Am-241	0	0	0	3.08E-01	0	0	0	3.91E-01
Eu-152	0	0	0	0	0	0	0	0
Eu-154	0	0	0	0	0	0	0	0

TABLE 3 (Cont.)

Radionuclide	Case V		Case VI		Case VII		Case VIII	
	5%	95%	5%	95%	5%	95%	5%	95%
H-3	2.00E+00	4.34E+00	2.12E+01	2.18E+01	2.85E-10	2.07E-06	6.58E+00	1.19E+01
C-14	0	5.79E-02	0	5.85E-02	0	1.17E+01	0	1.44E+02
Co-60	0	0	0	6.17E-02	0	0	0	0
Ni-63	0	0	0	5.83E-04	0	0	0	0
Se-79	0	0	0	3.15E+03	0	0	0	0
Sr-90	0	1.13E+00	0	7.53E+01	0	0	0	1.49E+00
Tc-99	0	4.87E+01	0	4.88E+01	0	5.96E+02	0	6.20E+02
Sb-125	0	0	0	3.28E-02	0	0	0	0
I-129	0	1.01E+05	0	1.01E+05	0	5.56E+03	0	4.73E+04
Cs-137	0	0	0	0	0	0	0	0
Sm-151	0	0	0	0	0	0	0	0
Th-230	0	0	0	5.12E-04	0	0	0	0
Th-232	0	0	0	0	0	0	0	0
U-238	0	7.48E+00	0	7.52E+00	0	0	0	7.92E+01
Np-237	0	2.60E+03	0	2.61E+03	0	0	0	0
Pu-239	0	2.41E-04	0	0	0	0	0	2.44E-05
Am-241	0	3.87E-01	0	4.96E+01	0	0	0	1.16E+01
Eu-152	9.59E-18	2.24E-10	0	4.07E-05	0	0	0	0
Eu-154	0	0	0	8.15E-06	0	0	0	0

3 FACTORS AFFECTING K_d IN RCRA WASTE DISPOSAL UNITS

By definition of the distribution coefficient (K_d), the factors that affect its value for a particular radionuclide are the radionuclide's solubility and the availability of adsorption sites on the solid phase. Higher K_d values will be found in systems with larger numbers of appropriate adsorption sites, and dissolved species with higher solubilities will have lower K_d values. However, if solubility is limited (the species concentration exceeds its solubility product constant), precipitation occurs, raising the K_d value and removing the species from any adsorption-desorption processes.

3.1 SOLUBILITY

The solubility of a species in solution depends primarily on environmental conditions and the chemical nature of the species. The pH and dissolved oxygen content play a large role in species solubility. Some species are more soluble with an increasing pH, and others are more soluble with a decreasing pH. However, most natural systems tend to be near a neutral pH. Thus, the solubility of dissolved species near pH = 7 will be most relevant. The pH at six RCRA hazardous waste landfills was observed to range from 5.8 to 11, while a narrower range of 6.6 to 9.8 was observed at 14 nonhazardous landfills (EPA 2000).

Another important influence on solubility is the dissolved oxygen content. Aerobic conditions promote chemical oxidation, and anaerobic conditions promote chemical reduction. Depending on the radionuclide in question, the oxidized or reduced species may be more soluble. For example, the reduced form of uranium (IV) is highly insoluble, while the oxidized form (VI) can be highly soluble, especially in the presence of carbonate. Anaerobic conditions typically occur as depth from the ground surface increases (distance from the oxygen source increases), since the oxygen is often depleted by microbial action. The level of microbial action depends in part on the amount of digestible organic material available. Under anaerobic conditions, radionuclides can also be precipitated by anaerobic species such as sulfide ions.

The apparent solubility of radionuclides can also be increased by complexation by organic species or by adsorption on colloidal material (Lieser 1995; Silva and Nitsche 1995). Exceptional behavior occurs when particulates bearing surface-adsorbed elements are colloidal in size (e.g., more than 0.003 mm and less than 0.1 mm). Colloidal particles can migrate with groundwater through soil systems that have sufficient porosity or are fractured (Kaplan et al. 1993, 1994).

3.2 ADSORPTION

Adsorption of radionuclides depends on the availability of binding sites and the chemical nature of the radionuclide species and the binding sites (bond strength). The number of available binding sites will decrease with an increase in the aqueous radionuclide concentration because of increased competition for sites. The number of binding sites can also be reduced through competition with other species in solution. Thus, the presence of water-borne agents, organic or inorganic, that can interfere in some way with the uptake of otherwise strongly held species will lower the effective K_d of that species for that particular phase.

3.2.1 General Observations

With the exception of simple monomeric ions resembling sodium (Na^+) and chloride (Cl^-), elements that hydrolyze or form insoluble hydroxides above pH 4 tend to be surface or particle reactive. The ions of those "insoluble" elements are readily immobilized or remain attached to the solid phases in soil materials. An equilibrium distribution of an insoluble or surface-active species (e.g., Th) between a particle phase and the associated aqueous phase can easily exceed a K_d) of 10^7 (i.e., 10 million).

Table 4 lists the observed range in K_d for each radionuclide of interest. These values reflect differences in soil type and composition as well as other environmental factors such as pH and degree of water saturation. The soil types used in Table 4 were categorized by Sheppard and Thibault (1990) according to texture. Sand soils are composed of 70% sand-sized particles; clay soils are composed of 35% clay-sized particles; loam soils are composed of an even distribution of sand-, clay-, and silt-sized particles or are composed of up to 80% silt-sized particles; and organic soils are composed of more than 30% organic matter and are either classic peat or muck soils or the litter horizon of a mineral soil. The resulting oxidation state of the radionuclide has a profound effect on its solubility.

Quadrivalent actinides, including plutonium and thorium, tend to be highly surface active and are strongly retained by particulate materials. If the particulate material is a stationary phase in a groundwater setting, then the above elements are immobilized in a saturated zone (Nelson and Orlandini 1986b).

Trivalent actinides, including americium and curium, tend to be surface or particle reactive but exhibit some variation in their behavior below ground. Americium can have more than one form in groundwater: a particle reactive form and a less reactive species that allows at least some portion of this actinide to be more mobile in a flowing system (Penrose et al. 1990).

TABLE 4 K_d **Values (L/kg)**[a]

Element	Soil Type	No. of Observations	Geometric Mean	Range
Am	Sand	29	1,900	8.2 to 300,000
	Loam	20	9,600	400 to 48,309
	Clay	11	8,400	25 to 400,000
	Organic	5	112,000	6,398 to 450,000
Co	Sand	33	60	0.07 to 9,000
	Loam	23	1,300	100 to 9,700
	Clay	15	550	20 to 14,000
	Organic	6	1,000	120 to 4,500
Eu	NA[b]	NA	825[c]	NA
H	Sand	3	0.06	0.04 to 0.1
	Loam	NA	20	NA
	Clay	NA	30	NA
	Organic	NA	75	NA
I	Sand	22	1.0	0.04 to 81
	Loam	33	5	0.1 to 43
	Clay	8	1	0.2 to 29
	Organic	9	25	1.4 to 368
Ni	Sand	11	400	60 to 3,600
	Loam	NA	300	NA
	Clay	10	650	305 to 2,467
	Organic	6	1,100	360 to 4,700
Np	Sand	16	5	0.5 to 390
	Loam	11	25	1.3 to 79
	Clay	4	55	0.4 to 2,575
	Organic	3	1,200	857 to 1,900
Pu	Sand	39	550	27 to 36,000
	Loam	21	1,200	100 to 5,933
	Clay	18	5,100	316 to 190,000
	Organic	7	1,900	60 to 62,000
				5 to 2,700[d]
Ra	Sand	3	500	57 to 21,000
	Loam	3	36,000	1,262 to 530,000
	Clay	8	9,100	696 to 56,000
	Organic	NA	2,400	NA
Sb	Sand	1	45	NA
	Loam	NA	150	NA
	Clay	NA	250	NA
	Organic	NA	550	NA

TABLE 4 (Cont.)

Element	Soil Type	No. of Observations	Geometric Mean	Range
Se	Sand	3	55	36 to 70
	Loam	1	150	NA
	Clay	14	115	36 to 246
	Organic	4	170	105 to 310
Sm	Sand	NA	245	NA
	Loam	NA	800	NA
	Clay	NA	1,300	NA
	Organic	NA	3,000	NA
Sr	Sand	81	15	0.05 to 190
	Loam	NA	800	NA
	Clay	24	110	3.6 to 32,000
	Organic	12	150	8 to 4,800
				1 to 1,700[d]
Tc	Sand	19	0.1	0.01 to 16
	Loam	10	0.1	0.01 to 0.4
	Clay	4	1	1.16 to 1.32
	Organic	24	1	0.02 to 340
Th	Sand	10	3,200	207 to 150,000
	Loam	NA	3,300	NA
	Clay	5	5,800	244 to 160,000
	Organic	3	89,000	1,579 to 13,000,000
				20 to 300,000[d]
U	Sand	24	35	0.03 to 2,200
	Loam	8	15	0.2 to 4,500
	Clay	7	1,600	46 to 395,100
	Organic	6	410	33 to 7,350
				< 1 to 1,000,000[d]

[a] Data from Sheppard and Thibault (1990) except as noted. Geometric mean was calculated from soil-to-plant concentration ratios if no observations were recorded.

[b] NA = not available.

[c] RESRAD default estimated from using soil-to-plant concentration ratios.

[d] Estimates from calculations over various pHs and clay contents (EPA 1999).

Trivalent rare earths (e.g., Sm, Eu) tend to be particle reactive but appear to have lower reactivity in certain groundwater systems that are carbonate in nature (Banner et al. 1989).

The monovalent and divalent ions of elements such as cesium, strontium, radium, iodine, and technetium form soluble ions and tend to be mobile in groundwater (Lieser and Steinkopff 1989). Cesium is an exception and can be strongly adsorbed by several clay minerals. Technetium is another exception because it will become insoluble under anaerobic conditions (e.g., Tc VII to IV, III).

Multivalent elements such as hexavalent uranium and pentavalent neptunium that are usually soluble in natural groundwater are also strongly adsorbed and retained in natural groundwater that becomes anaerobic (e.g., Np V to IV). Again, the possibility of colloidal transport exists for all adsorbed or precipitated species.

3.2.2 Organic Substrates

While organic species in solution promote increased mobility of metal ions, the adsorption properties of purely organic substrates are at the other extreme. These surfaces are extensions of larger chain organic molecules, such as fulvic and humic acids found in natural systems. These acids are well-known to be strong chelators of metal ions because of their polycarboxylic functional groups (Nelson et al. 1985). The functionality of these groups has been exploited through their use in commercially available exchange resins marketed for the removal of contaminant metals in solution. It is our experience that carbon in a consolidated form is more effective at sequestering radionuclides than carbon in the solution phase. On a normalized basis (the ratio of metal to carbon), the adsorption of metals on the solid phase has been observed to be an order of magnitude greater than adsorption in the solution phase. The adsorption coefficient for the solid phase has been observed to range from approximately 10,000 to 200,000 L/kg, depending on the metal and its speciation in systems dominated by organic systems such as those found in some swamps and bogs (Wahlgren and Orlandini 1982). The data in Table 4 also indicate that organic soils tend to have higher K_ds than clayey, loamy, and sandy soils.

4 K_d ESTIMATION

A brief review of past studies examining the effect of organic material on the migration of metals in soil is first presented, followed by an overview of organic concentrations found in landfill leachates. A simple model is then proposed that relates a radionuclide distribution coefficient to the concentration of organic material present.

4.1 PAST STUDIES

Experience with natural aquatic systems containing varying amounts of soluble organic material has revealed a system of equilibria in which organics compete with the solid phase for metal ions. An inverse relationship has been shown between K_d and the amount of organic material present in solution (Wahlgren and Orlandini 1982; Nelson and Orlandini 1986a; Nelson et al. 1989). As discussed in Section 3, such a relationship results from the complexation of metal ions in solution or from the direct competition for adsorption sites.

An example of a common interference mechanism is complexation by natural humic and fulvic acids or complexation by high concentrations of carbonate ion (Wahlgren and Orlandini 1982; Nelson and Orlandini 1986a; Nelson et al. 1985). Studies of simulated municipal solid waste (MSW) leachate demonstrated the importance of adsorption sites in attenuating metal concentrations in solution (Boyle and Fuller 1987). Leachates high in total organic carbon (TOC) or dissolved salts commonly found in MSW leachate [$CaCl_2$, $Mg(NO_3)_2$, KCl, and NaCl] were shown to significantly reduce metal adsorption through competition for binding sites. Another interference to the retention of particle-active metal ions has been shown to be the formation of a colloidal phase (Kaplan et al. 1993, 1994).

A continuous, inverse relationship between the K_d and the organic concentration (Wahlgren and Orlandini 1982; Nelson and Orlandini 1986a; Nelson et al. 1985) that is independent of the mechanism by which organic material interacts with radionuclide contaminants is developed in Section 4.3.

4.2 CHARACTERIZATION OF ORGANIC SPECIES IN LANDFILL LEACHATES

A recent study conducted by the U.S. Environmental Protection Agency (EPA) has provided information on the median concentrations of various landfill leachate pollutants for both Subtitle C and Subtitle D landfills (EPA 2000). Table 5 summarizes the findings for the organic constituents sampled for both landfill categories. In both cases, the sum of the median organic pollutant concentrations measured is only a few percent of the TOC concentration. This finding

TABLE 5 Median Leachate Concentrations (μg/L) for Hazardous and Nonhazardous Landfills

Pollutant	Subtitle C Hazardous	Subtitle D Municipal
TSS	151,000	137,000
TOC	440,902	376,521
Total phenols	25,004	571
Organics (toxic and nonconventional)		
1,1-Dichloroethane	45.7	NA[a]
1,4-Dioxane	466	10.8
2,4-Dimethylphenol	70	NA
2-Butanone	1,048	1,082
2-Propanone	2,889	992
4-Methyl-2-pentanone	500	101
Alpha-terpineol	95.7	123
Aniline	237	NA
Benzene	36.9	NA
Benzoic acid	2,482	100
Benzyl alcohol	43.6	NA
Diethyl ether	50	NA
Ethylbenzene	44.8	NA
Hexanoic acid	2,703	5,818
Isobutyl alcohol	39.7	NA
Methylene chloride	118	36.8
m-Xylene	29.4	NA
Napthalene	48.9	NA
n,n-Dimethylformamide	NA	10
o+p Xylene	17.1	NA
o-Cresol	78.8	15
Phenol	4,400	102
Pyridine	70	NA
p-Cresol	144	75
Toluene	104	108
trans-1,2-Dichloroethene	74.3	NA
Trichloroethene	44.6	NA
Tripropyleneglycol methyl ether	853	197
Vinyl chloride	42.7	NA
Pesticides/herbicides (nonconventional)		
2,4,5-TP	4.1	NA
2,4-D	5	NA
2,4-DB	7.9	NA
Dicamba	4	NA
Dichloroprop	7.3	6.1
Disulfoton	NA	6.1
MCPA	209	NA
MCPP	870	NA
Picloram	2	NA
Terbuthylazine	14.5	NA
Dioxins/furans (nonconventional)		
1234678-HpCDD	0.00018	0.00014
1234678-HpCDF	0.00013	NA
OCDD	0.00035	0.0018
OCDF	0.0019	NA

[a] NA = not available.

Source: EPA (2000).

suggests that most of the organic content of the landfills is from conventional sources such as the breakdown of clothing, paper products, or other nonhazardous materials. The main organic fraction of leachate has been reported to be free volatile acids (McGinley and Kmet 1984 as cited in EPA 1998). Other constituents found in landfill leachates include carbohydrates, proteins, and humic-like and fulvic-like substances (EPA 2000).

As shown in Table 5, the fact that the TOC concentration is itself well above (by a factor of two or more) the median total suspended solid concentrations measured suggests that more than half the TOC concentration is actually from dissolved material. The EPA (2000) report also states that most of the organics in leachate are soluble. Of the few percent of organic material accounted for in Table 5, most of the organic compounds would not be expected to complex metal radionuclides found in the landfill. However, the organics could occupy binding sites if their solubility limit is exceeded.

Finally, because of the similarities between the Subtitle C and Subtitle D landfill leachates, radionuclide migration within Subtitle C landfills is expected to be similar to that in Subtitle D landfills under the same environmental conditions with respect to organic pollutant concentrations. The median TOC concentrations, the low percentage of hazardous organic species with respect to the TOC concentration, and the absence of strong chelating organics support this conclusion.

Additional data for two RCRA landfills fall within the limits observed in the EPA study (EPA 2000). At the first site, the TOC concentration was observed to range from 44 to 730 parts per million (ppm) over a span of two years, with an average value of 207 ppm from six samples (Schneider 2000). Another site reported a TOC concentration range of 1.8 to 3,400 ppm over a nine-year period, with an average value of 109 ppm from 170 samples (Hu 2000). The respective median TOC values were 84 to 17 ppm.

4.3 PROPOSED MODEL

Contaminants and contaminant concentrations vary widely among landfills. Complex groundwater models requiring detailed thermodynamic input already exist to model the solubility of inorganic species. Yet these models fail if all potential aquifer species are not accounted for and properly characterized. Also, the interaction of radionuclides with organic compounds, with the exception of chelating agents, has received very little study. Thus, the results of the model presented here are intended only to indicate possible outcomes.

A simple model for the partition coefficient is proposed as a first approximation to account for the effect of organic compounds on the mobility of radionuclides in RCRA waste disposal units. This model, as given in Equation 1, is intended only to provide a rough estimate of the effects:

$$\log K_{deff} = \log K_d - m \log \left([TOC] \times (1 - f_{nc})\right), \tag{1}$$

where

K_{deff} = effective distribution coefficient corrected for the organic carbon concentration;

K_d = distribution coefficient for conditions without the presence of organic carbon;

m = slope factor equal to 0.74 for a K_d of ≥ 100 L/kg and 0.4 for a K_d of <100 L/kg;

$[TOC]$ = TOC concentration given in parts per million (ppm), which includes all dissolved, suspended, and colloidal organic material; and

f_{nc} = fraction on noncomplexing organic material.

Note that the quantity $[TOC] \times (1 - f_{nc})$ in Equation 1 should be greater than 1 ppm.

There are a number of uncertainties and conservative assumptions inherent in Equation 1 that must be taken into account when using the model. Such considerations include estimating the K_d value of the radionuclide in the absence of organic effects, the relationship between the radionuclide and dissolved organic material, and the use of TOC rather than the dissolved organic carbon (DOC) concentration as a measure of the organic material present.

As discussed in Section 3, the K_d can vary widely depending on environmental conditions. Thus, Equation 1 requires as input an estimate of an element's K_d value, excluding organic material effects. Table 4 can give an analyst a general idea of what might be expected. The behavior of the radionuclides is not expected to vary among isotopes (e.g., Th-230 versus Th-232) but can depend highly on the oxidation state [e.g., U(VI) versus U(IV)] and range over orders of magnitude.

The second half of Equation 1 was derived from physical-chemical dependencies shown to exist between organic matter and radionuclides in surface and near-surface aquatic systems and modified for use here (Wahlgren and Orlandini 1982; Nelson and Orlandini 1986a,b; Penrose et al. 1990; Nelson et al. 1985, 1987; Boggs and Livermore 1985; Marley et al. 1993). The variable m represents the slope of K_d versus organic concentration. Lack of data preclude a better

characterization of this slope, other than the values of 0.74 for those species that are less mobile and 0.4 for species that are more mobile. Those species that are already mobile will be affected less by organic complexation or covering of adsorption sites than those species that are more tightly bound to surfaces; hence, a smaller slope is used for more mobile species. The choice of a K_d cutoff of 100 for m in Equation 1 is somewhat arbitrary, representing the cutoff between species that are more reactive to adsorption and those that are more mobile. At the two extremes, as discussed further in Section 5, it should be noted that both highly soluble (mobile) and highly insoluble (immobile) radionuclides are not expected to be significantly affected by the aqueous concentration of organic material present (i.e., $m \simeq 0$).

Although the dependency existing between organic matter and radionuclides (i.e., the slope, the decrease of K_d with increasing organic matter content) was derived from surface water data, it has been observed that distribution coefficients for both inorganic and organic species are lowered as the ratio of the solid phase to the aqueous phase increases. In groundwater systems, which more closely resemble RCRA waste disposal units, the soil or solid phase is predominant, and the effective K_ds are significantly lower than those in surface systems, where the aqueous phase is predominant (Honeyman and Santschi 1988; O'Connor and Connolly 1980). This situation was taken into consideration by dividing the observed slopes by a factor of two before using them in Equation 1.

A conservative assumption was made that all organic material in solution will be considered capable of binding radionuclides to adsorption sites (colloids) or complexing the radionuclides of concern, thereby increasing radionuclide solubility (radionuclide mobility). Thus, the measure of organic material in solution was taken to be the TOC content rather than the DOC content. In some cases, the DOC concentration may be more appropriate for use in Equation 1 than the TOC concentration if colloids are not present. Furthermore, use of the DOC concentration itself may be conservative because not all DOC in landfills may be composed of complexing agents that are capable of solvating the radionuclide of concern. In order to reduce the conservativism of such an assumption, the fraction of noncomplexing organic material (f_{nc}) was added to Equation 1 to account for common organic wastes often found in landfills, such as the solvents methylene chloride and TCE. However, additional data on the amount and speciation of both the TOC and DOC found in landfills are required for further refinement of the model proposed in Equation 1. At present, the data on landfill leachate with regard to these parameters are primarily limited to the TOC content.

A direct linear relationship between the distribution coefficients for thorium and Pu(IV) and the DOC concentration was observed in the organic-rich waters found in the Okeefenokee Swamp and Volo Bog, with measured DOC concentrations of 34 ppm and 15 ppm, respectively (Wahlgren and Orlandini 1982). In addition, experience with certain seeps and groundwater from test wells in groundwater systems indicate that the DOC concentration averages below 10 ppm and has a maximum of about 30 ppm (Nelson and Orlandini 1986b; Kaplan et al. 1994; Penrose et al. 1990; Savannah River Site 1990; Golchert and Kolzow 1999). On the other hand, data from eight Subtitle C hazardous landfills indicate TOC concentrations ranging from approximately 0 ppm (i.e.,

below detectable limits) to 3,800 ppm (EPA 2000). A median value of about 441 ppm of TOC (see Table 5) was found for Subtitle C hazardous landfill leachates by using data from EPA sampling, detailed questionnaires, the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) groundwater database, and the Centralized Waste Treatment Industry database (EPA 2000). Similar data for 22 Subtitle D municipal landfills showed a range of TOC concentrations in landfill leachates from 9.4 to 3,400 ppm, with a median value of 377 ppm.

The use of the TOC concentration for this application is conservative. As mentioned above, the highest DOC concentrations for strong complexing agents, such as humic and fulvic acids found in organic-rich waters, were on the order of 10 ppm, well below the median TOC concentration of 441 ppm observed in Subtitle C landfills. It was these DOC complexing agents that were found to have a linear relationship with the radionuclide solubility. More soluble complexing agents could also be present, but such data are not readily available. In addition, the validity of Equation 1 has not been tested for organic complexing agents at concentrations higher than approximately 50 ppm. Thus, this method is only a rough first-order approximation for dissolved organic effects. On the other hand, the high TOC concentrations observed in the landfills compared with the DOC concentrations in the organic-rich waters suggest the presence of colloidal material and/or the presence of more soluble organics in the landfill leachates. In either case, these species may also be capable of binding to adsorption sites and complexing dissolved radionuclides. Thus, with the reservations listed above, the TOC concentration was taken to be the measure for the organic species concentration used in estimating radionuclide mobility in Equation 1.

The use of Equation 1 for landfills with large volumes of organic solids is not recommended. In such cases, radionuclide migration is not expected because the K_d for radionuclides in these areas is expected to be greater than 10,000 L/kg, as discussed in Section 3. If such concentrated conditions of organic materials are found in RCRA landfills, mobile radionuclides passing through such areas are expected to become immobilized. However, such areas are also sources of solution phase organics that might come into contact with a greater percentage of the radionuclides in the landfill. Therefore, it is not possible a priori to ascertain the overall effect of hazardous organic materials on radionuclide migration in a specific landfill.

4.4 MODEL REFINEMENT

This section discusses improved results from the model in Section 4.3 that might be possible given the availability of additional data. Ideally, the effect or organic matter on the solubility of a radionuclide stems directly from the ability of the organic species to bind to the radionuclide ion in solution. If the organic species (dissolved, suspended, or solid) cannot bind to the radionuclide, no effect on the radionuclide K_d will occur.

Radionuclides in any landfill will interact with organic matter. The majority of the organic species in most landfills will eventually be decomposition products from organic wastes, primarily paper or textile (clothing). Any mobility effects of additional RCRA hazardous wastes on radionuclides will be incremental ones, especially because of RCRA waste disposal requirements on leachability. As discussed in Section 3, the major influences on radionuclide solubility and migration are the radionuclide's oxidation state, solution pH, dissolved oxygen content, microbial activity, and available adsorption sites. Thus, the mobility of radionuclides is not expected to be radically altered when they are introduced to a RCRA landfill. In addition, the hazardous organic species (i.e., those with complexing groups) will be competing with naturally occurring organic decomposition products in solution and in the solid phase in their interactions with any radionuclides.

Increased solubility (lower K_d) could occur if a radionuclide were complexed by a water-soluble organic species. However, as mentioned in the previous section, organic compounds such as methylene chloride and TCE are examples of compounds that have no binding groups and do not contribute to the mobility of radionuclides in solution. Thus, better characterization of the organic matter in leachate solution, at least to the degree that functional groups capable of complexing radionuclide ions are determined to be present or not, would greatly reduce the uncertainty in the f_{nc} term in Equation 1. Ideally, the model represented by Equation 1 should have the $[TOC] \times (1 - f_{nc})$ term replaced by the concentration of dissolved organic complexing species.

On the other hand, if the organic species has binding groups but is part of, or adsorbed on, the solid phase, lowered solubility (higher K_d) could occur for a complexed radionuclide. The current model does not account for this possibility unless the adsorbed phase is on colloidal material; it is accounted for in the TOC concentration.

5 DISCUSSION AND SUMMARY

The sensitivity analysis results indicated that many parameters could affect the transport of radionuclides at RCRA waste disposal sites. (The doses calculated in the sensitivity analysis are for a hypothetical site and should not be deemed as representative doses for any particular site.) The results from analyzing 19 radionuclides indicated that 10 of them (i.e., Co-60, Ni-63, Sb-125, Cs-137, Sm-151, Eu-152, Eu-154, Th-230, Th-232, and Pu-239) would be less likely to cause groundwater contamination at a RCRA waste disposal site. The other nine (i.e., H-3, C-14, Se-79, Sr-90, Tc-99, I-129, U-238, Np-237, and Am-241), which tend to form soluble species, could reach the groundwater and cause contamination. Thus, these nine radionuclides would not be expected to be significantly affected by the hazardous organic constituents present in a RCRA disposal unit because they would have dispersed. However, many site-specific parameters could affect the amount of time it would take for a radionuclide to travel to the groundwater. These parameters include soil hydraulic conductivity, precipitation rate, K_d, soil porosity, bulk density, and thickness of the unsaturated (vadose) zone.

The hazardous constituents in a RCRA waste disposal unit could decrease or increase radionuclide K_ds, depending on whether the TOC concentration was in solution or in a solid phase. The change would also depend on the chemical form of the radionuclide, pH of the leachate, and other factors. For example, tritium, iodine, strontium, and radium are generally soluble in an aqueous solution. Thus, these radionuclides can be regarded as persistently mobile species and the least susceptible to changes in organic concentration. Technetium (Tc), neptunium (Np), and selenium (Se) most commonly occur in the hepta, penta, and hexavalent states, respectively. These latter nuclides tend to be mobile in groundwater that has a measurable oxygen content. In a reducing (anoxic) environment, Tc(III,IV), Np(IV), and Sc(IV) tend to become insoluble or adsorbed to the host matrix. Thus, although the organic material content in a solution might enhance the solubility of some of the species in the solution, it would be expected to have very little effect on those species that are highly soluble or insoluble.

Under landfill conditions, other parameters must also be considered. Such parameters include the types of waste accepted, compaction, depth of fill, age, and operating practices (shredding, daily cover, and cap). The primary consideration is the amount of water present in the landfill. Migration will not occur if water is not present. Such ideal dry conditions are approached by landfills that are well above the water table and have a well engineered cap in place, especially landfills in dry environments (i.e., low annual rainfall rates). Most landfills have some water present. The water not only provides a vehicle for radionuclide migration but also influences the sustenance of microbes. Microbial action plays a significant role in the degradation of organic material (the food source) during different phases in the life of a landfill.

Most MSW landfills go through three decomposition phases (O'Leary and Walsh 1991). The first stage is characterized by aerobic decomposition with the production of carbon dioxide, water, and nitrate. As oxygen is used up, facultative and anaerobic microbes that produce volatile acids and carbon dioxide become dominant (second phase). As a result, the pH is lowered, and some inorganic species may go into solution. As time progresses, anaerobic methane-producing bacteria become dominant, and the pH rises to more neutral values (third phase). As a landfill progresses through each phase, the chemical species with which a radionuclide is associated and the organic species present can change, affecting a radionuclide's solubility and therefore its mobility.

In summary, the effect of organic constituents on radionuclide migration in a RCRA waste disposal site depends on a variety of factors. These factors include the chemical form of the radionuclide, pH of the leachate, nature of the organic constituents, and presence of other chemical species. Conditions such as pH and speciation are influenced, in part, by the amount of water present and the age of the landfill. A simple model that considers these factors was introduced to account for organic species' direct effect on a radionuclide's K_d. It is based on the TOC concentration of the landfill leachate corrected for noncomplexing organic material. The model indicates that organic species in leachate may decrease radionuclide K_ds; however, organic material in the solid phase may have the opposite effect, resulting in an increase of radionuclide K_ds. Because a radionuclide's K_d can vary more widely as a result of factors other than TOC, an estimate of the radionuclide's K_d in the absence of organic contamination is also required. A rough approximation to meet this latter requirement can be made if the radionuclide's speciation is known. Finally, organic species in solution may enhance the mobility of partially soluble radionuclides through complexation, but they are not expected to have a significant effect on radionuclides that are already highly soluble or insoluble. It is imperative that whenever feasible, actual (measured) K_d values rather than modeled values be used to conduct dose and risk assessments of radionuclides in RCRA disposal units.

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APPENDIX A:

RESRAD MODEL AND PARAMETER DESCRIPTIONS

The contaminated zone specified in the RESRAD code is where the waste material is placed; in this study, the contaminated zone is the RCRA waste disposal unit. In the RESRAD model, a layer of cover material on top of the contaminated zone can be considered so direct human contact with the radioactive material is avoided. Below the contaminated zone, up to five layers of unsaturated (vadose) zones can be considered, each with its own soil properties. In the saturated zone, radionuclides are considered to mix with uncontaminated groundwater and are collected and pumped out from a well at the downgradient edge of the contaminated zone.

A.1 RADIONUCLIDE RELEASE RATE

The release rate of radionuclides from the contaminated zone is estimated by using a nuclide-dependent, first-order leach rate constant, L_i, which is defined as the fraction of available radionuclide i leached out per unit of time. The radionuclide release rate (source strength, in pCi/yr), can be written as:

$$\dot{R}_i(t) = L_i\, \rho_b^{(cz)}\, A\, T(t)\, S_i(t) , \tag{A.1}$$

where

L_i = leach rate for radionuclide i (yr^{-1}),

$\rho_b^{(cz)}$ = bulk density of the contaminated zone (kg/m^3),

A = area of the contaminated zone (m^2),

$T(t)$ = thickness of the contaminated zone at time t (m), and

$S_i(t)$ = average concentration of the ith principal radionuclide in the contaminated zone available for leaching at time t (pCi/kg).

The first-order leach rate constant used in the current version of RESRAD is a time-independent radionuclide leach rate constant that is estimated on the basis of the soil residence time for the initial thickness of the contaminated zone. A time-independent radionuclide leach rate constant for radionuclide i, L_i, may be written as

$$L_i = \frac{I}{\theta^{(cz)} \, T_0 \, R_{d_i}^{(cz)}} \ , \tag{A.2}$$

where

I = infiltration rate (m/yr),

$\theta^{(cz)}$ = volumetric water content of the contaminated zone,

T_0 = initial thickness of the contaminated zone (m), and

$R_{d_i}^{(cz)}$ = retardation factor in the contaminated zone for radionuclide i (dimensionless).

The infiltration rate is given by

$$I = (1 - C_e)[(1 - C_r)P_r + I_{rr}] \ , \tag{A.3}$$

where

C_e = evapotranspiration coefficient (dimensionless),

C_r = runoff coefficient (dimensionless),

P_r = precipitation rate (annual rainfall), and

I_{rr} = irrigation rate (m/yr).

The runoff coefficient is dependent on the environmental setting and the slope of the contaminated zone.

The volumetric water content of the contaminated zone, $\theta^{(cz)}$, is the product of the saturated water content of the contaminated zone, $\theta_{sat}^{(cz)}$, and the saturation ratio of the contaminated zone, $R_s^{(cz)}$. The saturated water content is the water content when the soil material is saturated. Hence, θ_{sat} equals p_t, where p_t is the total porosity of the soil material, that is,[1]

$$\theta_{sat} = p_t \ . \tag{A.4}$$

---

[1] The superscript is omitted for a general definition. The definition applies for all zones.

The saturation ratio, R_s, is defined as the ratio of θ over θ_{sat}, that is,

$$R_s = \theta/\theta_{sat} = \theta/p_t .$$ (A.5)

When the medium is saturated, R_s equals unity. Under unsaturated infiltration conditions, the saturation ratio is a function of the infiltration rate, the saturated hydraulic conductivity, and the texture of the soil. The saturation ratio can be estimated by using the following equation (Clapp and Hornberger 1978):

$$R_s = \left(\frac{I}{K_{sat}}\right)^{\frac{1}{2b+3}} ,$$ (A.6)

where

I = infiltration rate (m/yr),

K_{sat} = saturated hydraulic conductivity (m/yr), and

b = soil-specific exponential parameter (dimensionless).

The volumetric water content of the unsaturated zone (including the contaminated zone), which is calculated by using Equations A.5 and A.6, is checked against the field capacity (F_c) of the unsaturated soil. The field capacity sets the lower limit of the volumetric water content and is used to replace the calculated value when the calculated value is smaller. Once the volumetric water content is set to the field capacity, the saturation ratio is recalculated by using Equation A.6.

The retardation factor for radionuclide i, R_{d_i}, is the ratio of the average pore water velocity to the radionuclide transport velocity. Assuming that the adsorption-desorption process can be represented with a linear isotherm, the retardation factor can be calculated with the following formula (Yu 1987):

$$R_{d_i} = 1 + \frac{\rho_b K_{d_i}}{\theta} = 1 + \frac{\rho_b K_{d_i}}{p_t R_s} ,$$ (A.7)

where

ρ_b = bulk soil density (g/cm^3),

K_{d_i} = distribution coefficient for the ith principal radionuclide (cm^3/g), and

θ = volumetric water content (dimensionless).

From the above equations, it is known that leach rate of a radionuclide is determined by (1) its K_d value, which decides the relative transport speed of the radionuclide to that of water in the pore space; (2) the water infiltration rate, which determines the capacity of the liquid phase in soil; (3) soil properties such as bulk density, porosity, saturated hydraulic conductivity, and the b parameter; and (4) the extent of contamination, which is described by contaminated zone thickness, area, and radionuclide concentration.

A.2 WATER TRANSPORT PARAMETERS

Radionuclides that leach out from the contaminated zone will transport through the soil column and reach the groundwater table. Once in the groundwater table, the radionuclides will be carried by the groundwater and travel horizontally to a downgradient well. Concentrations of radionuclides in the well water are time-dependent and are functions of the two transport times — breakthrough time (vertical transport time to reach the groundwater table) and rise time (horizontal transport time to reach the well) — and the dilution factor, which accounts for mixing of leachate with clean/uncontaminated groundwater. Therefore, depending on the time frame considered in a risk assessment, contamination in well water may not be observed within the time frame, or, even if it is observed, the concentration may not be large enough to cause significant human exposure.

While transporting in soils, the total amount of radionuclides will change over time as a result of radioactive ingrowth and decay. The decay products (progeny radionuclides) also have the potential of dissolving in pore water and transporting to deeper soils. However, the transport speeds of the progeny radionuclides are not necessarily the same as that of their parent and also likely to vary among themselves. The change in the total amount and the different transport speeds among radionuclides make mathematical simulation extremely difficult.

In the RESRAD modeling, a transfer function, G, is used. It calculates the amount of progeny radionuclide j in groundwater, as observed at time t, caused by the existence of parent radionuclide i in the contaminated zone, at the beginning of the simulation time 0. The transfer function is then convolved with the release rate of the radionuclide, as discussed in Section A.1. Detailed discussions on the development of the transfer function and the convolution can be found in Appendixes E and I in the RESRAD manual (Yu et al. 1993).

Calculations of the breakthrough time, rise time, and dilution factor as well as the parameters used in the calculations are discussed in the following sections.

A.2.1 Breakthrough Time

The well from which water is drawn for various applications can be assumed to be located at the center of the site (MB model in RESRAD) or at the downgradient edge of the contaminated zone (ND model in RESRAD). To be conservative, the MB model was used in the sensitivity analysis. However, equations used for the ND model are also discussed. Radionuclides are assumed to enter the groundwater as soon as they reach the water table; hence, the transport time through the aquifer is zero, and the breakthrough time is equal to the transport time through the unsaturated zones, which is the sum of the times during which a radionuclide is transported through the strata of the unsaturated zone that lie below the contaminated zone:

$$\Delta t_i = \sum_{m=1}^{n+1} \Delta t_{im}^{(uz)} \, , \qquad (A.8)$$

where

n = number of distinct horizontal strata in the unsaturated zone below the contaminated zone at time 0 and

$\Delta t_{im}^{(uz)}$ = transport time for the ith principal radionuclide through the mth stratum (yr).

The upper bound, $n+1$, of the summation in Equation A.8 is for the $(n+1)$th unsaturated stratum created by the decrease in the water table. The thickness of this $(n+1)$th stratum, Δz_{n+1}, is equal to the product of the water table drop rate, v_{wt}, and time, that is,

$$\Delta z_{n+1} = v_{wt} \times t \, , \qquad (A.9)$$

where v_{wt} = water table drop rate (m/yr).

The hydrogeological and hydrogeochemical properties of the $(n+1)$th unsaturated stratum are assumed to be the same as those of the saturated stratum. The RESRAD code allows up to five horizontal strata below the contaminated zone, that is, n is ≤ 5. If $n = 0$, the contaminated zone extends down to the aquifer.

The formula for the transport time is

$$\Delta t_{im}^{(uz)} = \frac{\Delta z_m \, R_{d_{im}}^{(uz)} \, p_{e_m}^{(uz)} \, R_{s_m}^{(uz)}}{I} \, , \qquad (A.10)$$

where

Δz_m = thickness of the *m*th stratum (m),

$R_{d_{im}}^{(uz)}$ = retardation factor of the *i*th principal radionuclide in the *m*th stratum of the unsaturated zone,

$p_{e_m}^{(uz)}$ = effective porosity of the *m*th stratum of the unsaturated zone (dimensionless), and

$R_{s_m}^{(uz)}$ = saturation ratio of the *m*th stratum (dimensionless).

The unsaturated zone retardation factors, $R_{d_{im}}^{(uz)}$, are calculated by the formula

$$R_{d_{im}}^{(uz)} = 1 + \frac{\rho_{b_m}^{(uz)} \, K_{d_{im}}^{(uz)}}{\left(p_{t_m}^{(uz)} \, R_{s_m}^{(uz)} \right)} \, , \tag{A.11}$$

where

$\rho_{b_m}^{(uz)}$ = bulk soil density in the *m*th stratum (g/cm^3),

$K_{d_{im}}^{(uz)}$ = distribution coefficient for the *i*th principal radionuclide in the *m*th stratum (cm^3/g), and

$p_{t_m}^{(uz)}$ = total porosity of the *m*th stratum (dimensionless).

The saturation ratio, $R_{s_m}^{(uz)}$, can be determined by using Equation A.6.

The distance from the ground surface to the water table, $D_{wt}(t)$, at time *t* is

$$D_{wt}(t) = C_d(t) + T(t) + \sum_{m=1}^{n+1} \Delta z_m \, , \tag{A.12}$$

where

$C_d(t) =$ cover depth at time t (m),

$T(t) =$ thickness of contaminated zone at time t (m), and

$\Delta z_m =$ as defined in Equations A.9 and A.10.

In the current version of RESRAD, the water table is assumed to be below the contaminated zone, that is, $D_{wt}(0) \geq C_d(0) + T(0)$.

Similar to the calculation of leach rate, the breakthrough time is determined by K_d values in the unsaturated zones, the infiltration rate of water, and soil properties of the unsaturated zones. However, instead of the extent of the contamination source, the thicknesses of the unsaturated zones determine the length of the breakthrough time.

A.2.2 Rise Time

When the well is located at the center of the site, the rise time is zero, whereas when the well is located at the downgradient edge of the contaminated zone, the rise time is given by the formula

$$\delta t_{i1} = \tau_{\ell_i}, \qquad \zeta \leq d_w, \ D_{wt}(t) \geq C_d(t) + T(t) \qquad (\text{A.13})$$
$$= \tau_{\ell_i}/(\zeta/d_w), \ \zeta > d_w, \ D_{wt}(t) \geq C_d(t) + T(t) ,$$

where

$\zeta = (I/V_{wfr})\ell,$

$=$ distance from the water table to the lower boundary of contamination in the aquifer at the downgradient edge of the contaminated zone (m),

$I =$ infiltration rate (m/yr),

$V_{wfr} = K_s^{(sz)} J_x,$

$=$ water flow rate per unit of cross-sectional area in saturated zone (Darcy velocity) (m/yr),

$K_s^{(sz)}$ = saturated hydraulic conductivity of saturated zone (100 m/yr),

J_x = hydraulic gradient in flow (x) direction (0.02, dimensionless),

ℓ = length of the contaminated zone parallel to the hydraulic gradient (maximum distance from the upgradient edge to downgradient edge parallel to the hydraulic gradient, 100 m),

d_w = distance of the well intake below the water table (10 m),

τ_{ℓ_i} = $p_e^{(sz)} R_{d_i}^{(sz)} \ell / V_{wfr}$

= time for the ith principal radionuclide to be transported from the upgradient edge to the downgradient edge of the saturated zone (yr),

$p_e^{(sz)}$ = effective porosity of the aquifer (0.2, dimensionless), and

$R_{d_i}^{(sz)}$ = retardation factor for the ith principal radionuclide in the saturated zone (dimensionless).

For the ND model (where the well is located at the edge of the contaminated zone), in addition to the K_d value in the saturated zone, the infiltration rate of water, soil properties of the saturated zone, and characteristics of the groundwater table (i.e., the hydraulic gradient) are used to determine the value of rise time.

A.2.3 Dilution Factor

It is assumed that the water flow is vertically downward from the bottom of the contaminated zone to the water table; hence, no dilution will occur in the unsaturated zone. The dilution in the saturated zone is estimated by using the conservative assumption that the dispersivity is zero.

For the MB model, the dilution factor is a radionuclide-independent factor given by the formula

$$f = \frac{AI}{U_w}, \quad AI < U_w, \tag{A.14}$$

$$= 1, \; AI \geq U_w ,$$

where U_w = well pumping rate = annual volume of water withdrawn from the well (m³/yr).

For a well located at the downgradient edge of the contaminated zone, the dilution factor is given by the formula

$$f_{i1} = \frac{\zeta}{d_w} \qquad d_r \leq \frac{A}{\ell} \ , \ \zeta < d_w$$

$$= \frac{AI}{U_w} \qquad d_r > \frac{A}{\ell} \ , \ \zeta < d_w$$

$$= 1.0 \qquad d_r \leq \frac{A}{\ell} \ , \ \zeta \geq d_w$$

$$= \frac{AI \ d_w}{U_w \ \zeta} \qquad d_r > \frac{A}{\ell} \ , \ \zeta \geq d_w,$$

where d_r is the effective pumping width (m) and is calculated as

$$d_r = \frac{U_w}{V_{wfr} \ d_w} \ . \tag{A.15}$$

The definitions of the parameters used in Equation 15 are the same as those used in Equations A.13 and A.14.

The dilution factor of a well that is located off site can be significantly smaller than the dilution factor estimated by Equations A.14 and A.15 for a well located at the center of the site or immediately adjacent to the contaminated zone, respectively. Therefore, radiation exposures estimated with the on-site well assumption are more conservative.

Again, K_d value in the saturated zone plays an important role in determining the dilution factor. Other factors that affect the dilution include the well water pumping rate, well screening depth below the groundwater table, and the infiltration rate of water.

A.3 APPENDIX A REFERENCES

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APPENDIX B:

SELECTED CALCULATIONAL RESULTS

The sensitivity analysis discussed in Section 2 indicated that the potential radiation exposure resulting from drinking groundwater may be of concern for some radionuclides. These radionuclides are H-3, C-14, Se-79, Sr-90, Tc-99, I-129, U-238, Np-237, and Am-241. The results for these radionuclides are summarized below. The initial radionuclide concentration used in the calculations is 100 pCi/g.

Figure B.1 compares the ranges of the peak radiation doses among the eight studied cases for H-3. Because of the small magnitude of the peak radiation doses, the range of Case VII is not shown clearly in the figure. The same situation occurs for other radionuclides in the following discussions. It can be seen that groundwater contamination is not a concern in Cases I, III, and VII, when the unsaturated zone thickness is large (76 m) and the infiltration rate is small (0.041 m/yr). However, contamination of groundwater may become a concern if the water infiltration rate is large (0.4572 m/yr), as it is for Case V. Potential radiation exposure definitely needs to be considered when the thickness of the unsaturated zone is small (Cases II, IV, VI, and VIII), even when the water infiltration rate is also small (0.041 m/yr).

Figure B.2 compares ranges of the peak radiation doses for C-14 among the eight cases studied. Contamination of groundwater would not be an issue of concern if waste in the disposal cell was grouted, resulting in a high K_d value for C-14 (about 2,600 cm^3/g).

Groundwater contamination needs to considered for Se-79 only when both the unsaturated zone thickness is small (3 m) and the infiltration rate is large (0.4572 m/yr). The potential range of radiation exposure from drinking contaminated groundwater is shown in Figure B.3.

As shown in Figure B.4, potential radiation exposures from drinking contaminated groundwater are either zero or less than 2 mrem/yr per 100 pCi/g for Sr-90 in all the cases studied except for Case VI, when both the unsaturated zone thickness is small (3 m) and the infiltration rate is large (0.4572 m/yr).

Because of the high mobility in soils, Tc-99 could leach out from the disposal cell and reach the groundwater table easily. It is also likely that the potential radiation exposure from drinking contaminated groundwater would exceed 40 mrem/yr per 100 pCi/g. The ranges of peak radiation doses for Tc-99 for the eight cases studied are shown in Figure B.5.

Potential radiation exposures to I-129 could be very high (exceeding 1.0×10^5 mrem/yr per 100 pCi/g), as shown in Figure B.6. It is best to maintain the integrity of the cap material so that the amount of water infiltrating to the disposal cell is minimal.
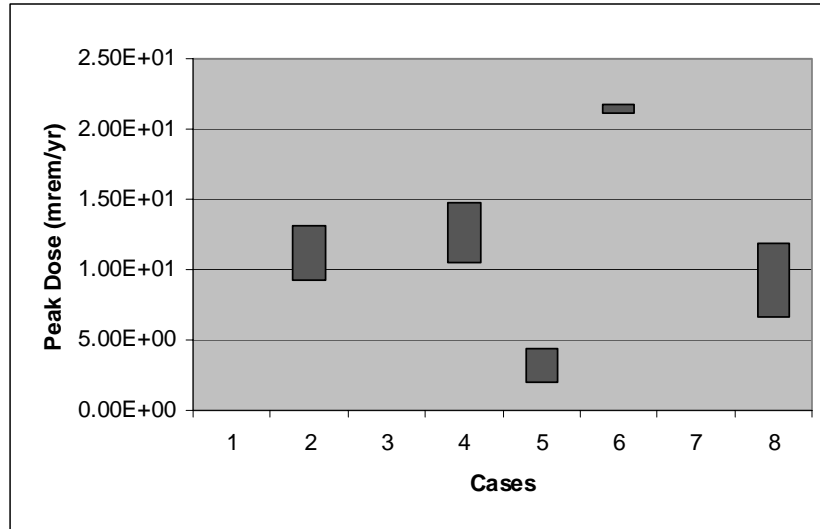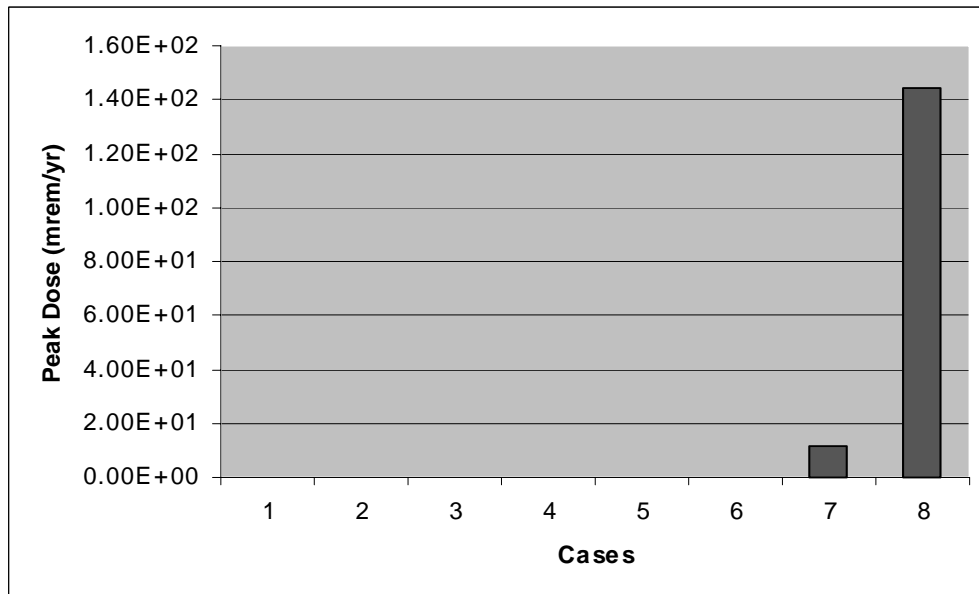
FIGURE B.1 Range of Peak Radiation Doses for H-3

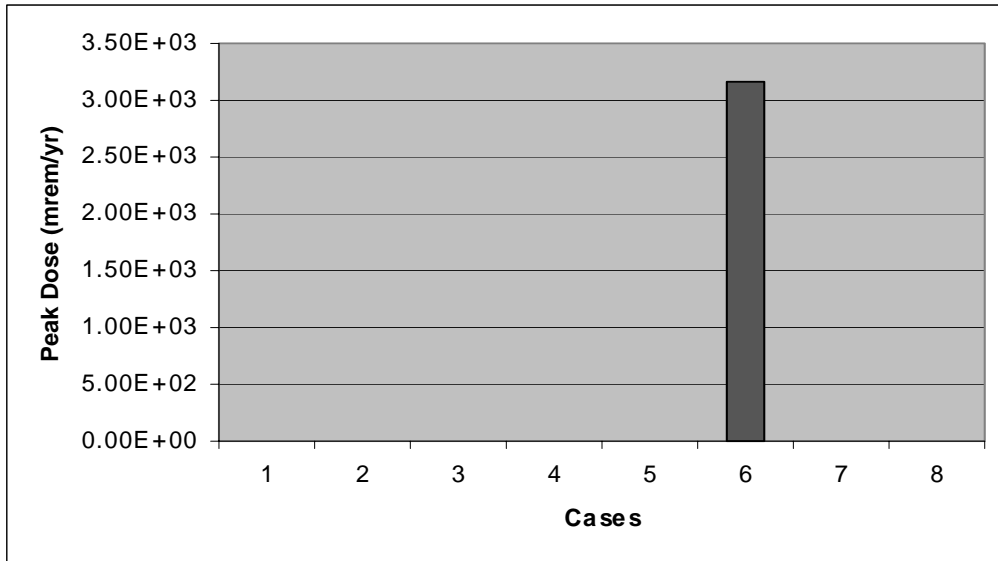**FIGURE B.2 Range of Peak Radiation Doses for C-14**

FIGURE B.3 Range of Peak Radiation Doses for Se-79
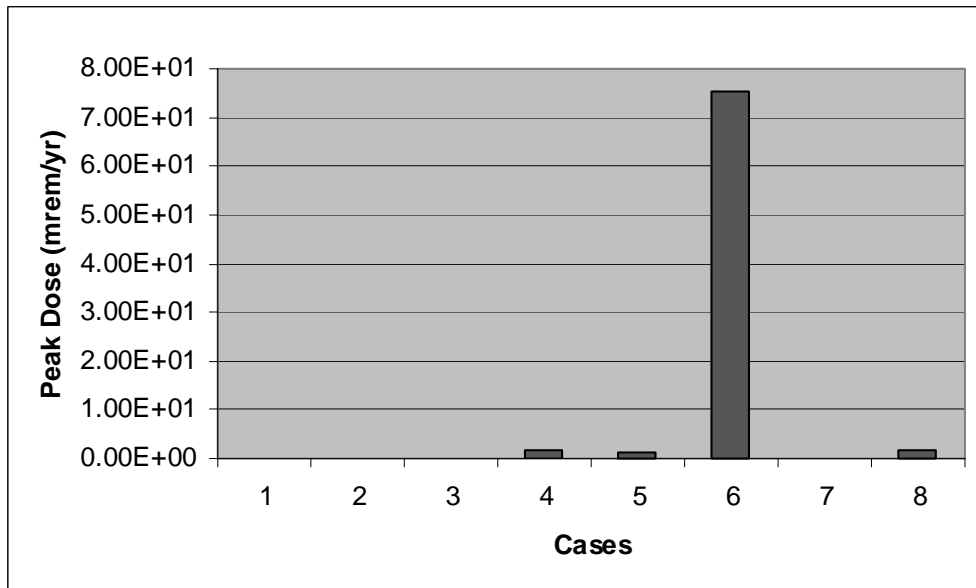

FIGURE B.4 Range of Peak Radiation Doses for Sr-90
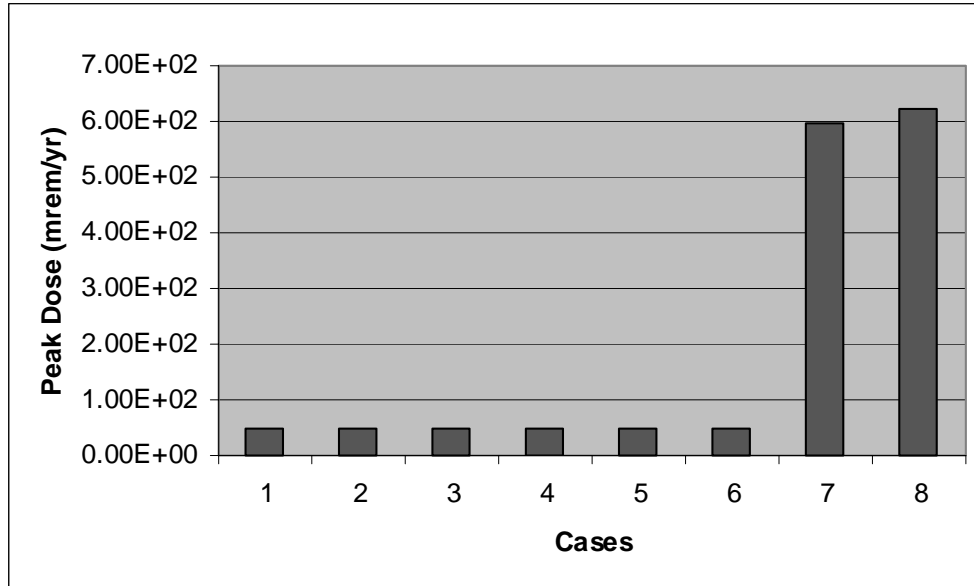
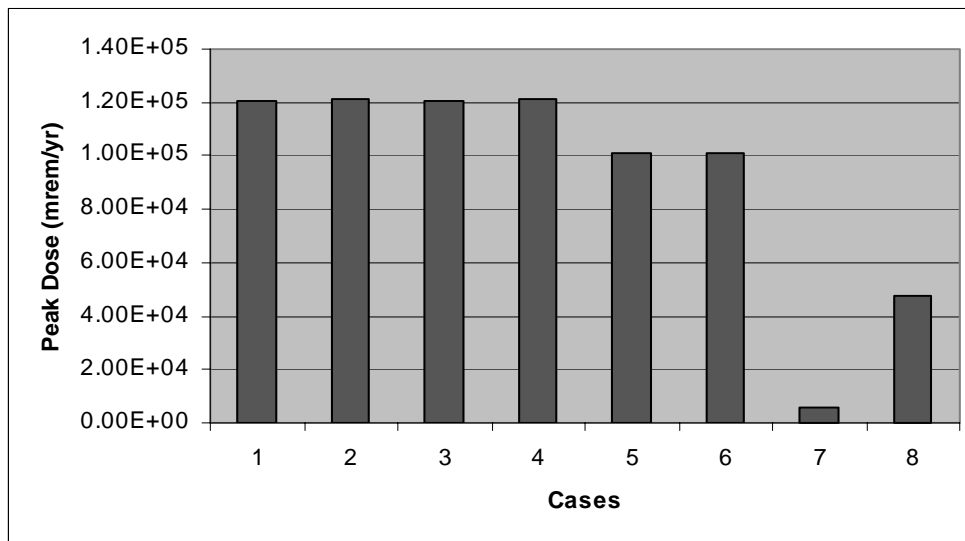**FIGURE B.5 Range of Peak Radiation Doses for Tc-99**

FIGURE B.6 Range of Peak Radiation Doses for I-129

Figure B.7 shows the estimated ranges of potential radiation exposures incurred by drinking groundwater contaminated with U-238. Radiation exposure may occur when either the infiltration rate is large (0.4572 m/yr), or the thickness of the unsaturated zone is small (3 m).

Potential radiation exposure to Np-237 could be large if Np-237 reaches the groundwater table, which could be facilitated by a large infiltration rate (0.4572 m/yr). Figure B.8 shows the ranges of the estimated doses for Cases I to VIII.

Both a large infiltration rate (0.4572 m/yr) and a small thickness of the unsaturated zone would be needed to observe a non-negligible (> 1 mrem/yr per 100 pCi/g) radiation exposure to Am-241, when waste material would be grouted before being placed in the disposal cell. Potential ranges of radiation exposures are shown in Figure B.9.
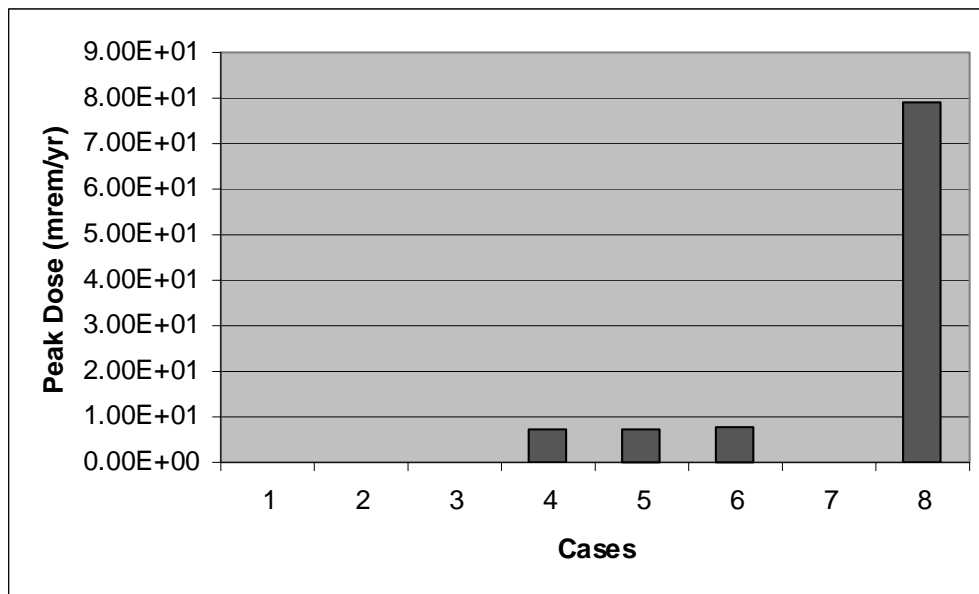

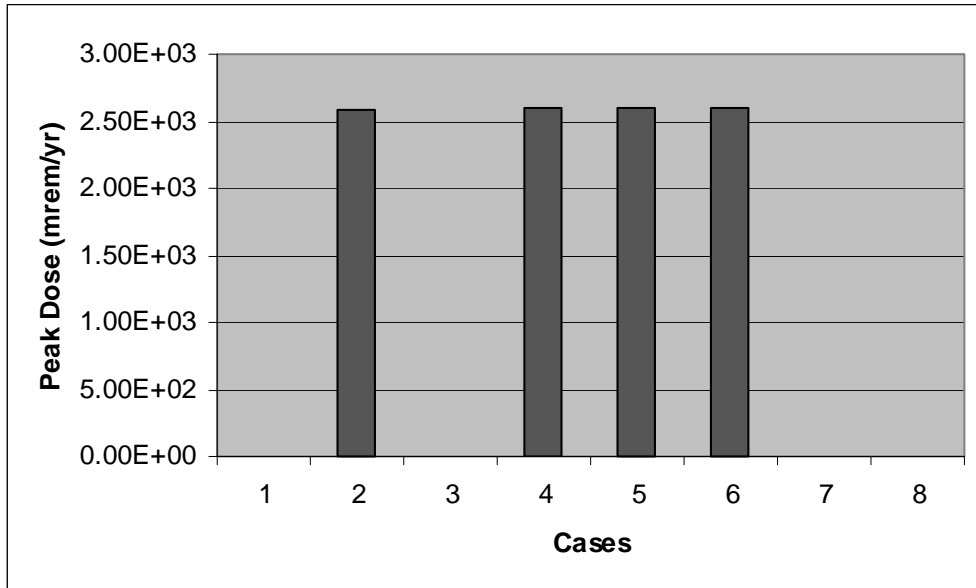**FIGURE B.7 Range of Peak Radiation Doses for U-238**

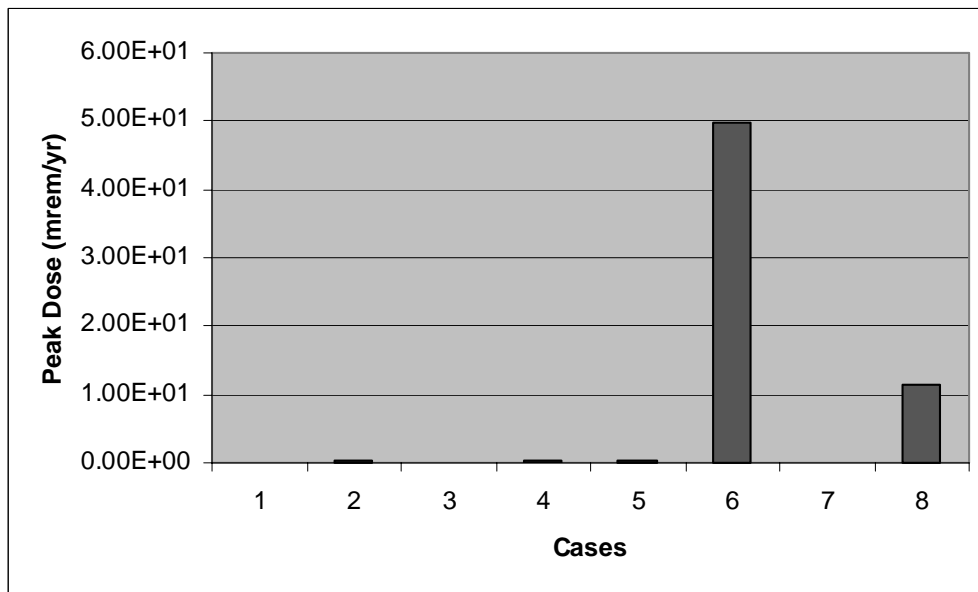**FIGURE B.8 Range of Peak Radiation Doses for Np-237**

FIGURE B.9 Range of Peak Radiation Doses for Am-241

APPENDIX C:

**POTENTIAL ORGANIC SPECIES EFFECT
ON K_d FOR SELECTED RADIONUCLIDES**

Table C.1 lists the estimated range (upper and lower bounds) of distribution coefficients (K_ds) for selected radionuclides when Equation 1 is used. The effective distribution coefficient (K_{deff}) is the adjusted K_d when the median organic concentration observed in existing Resource Conservation and Recovery Act (RCRA) disposal units (total organic carbon [TOC] concentration of 441 parts per million [ppm]) is used with various levels of nonbinding organic constituents ($f_{nc} = 0$, 0.5, and 0.9). For each element with a range of observed K_d values in Table 4, the lowest K_d value listed was used in Equation 1 to obtain the K_{deff}s corrected for organic material as shown in Table C.1. Use of the lowest observed K_d value assured that the widest possible range was considered for each element because the effects accounted for in Equation 1 lower the K_d value. The upper observed K_d values listed in Table C.1 are therefore the largest values for each element taken directly from Table 4. For those elements with only one value listed in Table 4, that value was used to compute the K_{deff}s given in Table C.1.

If all of the TOC in the landfill leachate consisted of organic constituents that strongly chelated (bonded) metal ions in solution, the organic material in solution might lower the observed K_d values (by a factor of 11 to 91 [$10^{0.4 \times \log(441)}$ to $10^{0.74 \times \log(441)}$]) for a median TOC concentration of 441 ppm). However, the magnitude of this effect would probably not be this large because some or all of the radionuclides of concern might already be in solution or be insoluble. Moreover, if the organic material was in a solid phase, its effect on the radionuclide K_d would be the opposite: it would increase the K_d. As Table 4 shows, organic soils tend to have higher K_ds than other types of soils. Therefore, the K_ds of radionuclides in RCRA disposal units can increase or decrease, depending on whether the TOC concentration is in solution or in the solid phase. In addition, a significant fraction of the organic species in solution might be of a noncomplexing nature ($f_{nc} > 0$).

Although the organic concentration in solids will generally increase radionuclide K_ds, there are not enough data to quantify this factor. Therefore, the upper bounds of the estimated K_ds listed in Table C.1 are the same as those observed upper bounds. Most of these upper bounds are for organic soils, while some are for clay and loam soils.

Further work is needed to better characterize the slope variable, m, in Equation 1 for species under groundwater conditions rather than surface water conditions. The current characterization of the slope variable was intended to provide realistic results, but the uncertainty is large because of the lack of data.

As shown in Table C.1, hazardous organic constituents in a RCRA waste disposal site could have a noticeable effect on radionuclide migration. However, as discussed in Sections 3 and 5, other

environmental variables can have a much larger effect on metal mobility. These parameters should be well characterized before an attempt is made to estimate the effects from organic constituents. In addition, many sites, such as the two mentioned in Section 4.2 with median TOC concentrations of 17 and 84 ppm, will have TOC concentrations much lower than the values used in generating Table C.1.

TABLE C.1 Estimated Range of K_d Values (L/kg) for Selected Radionuclides

| | K_{deff}, Corrected for Organics | | | | | | Observed K_d | |
| | Max. (TOC = 3,800 ppm) | | | Median (TOC = 441 ppm) | | | | |
Element	$f_{nc} = 0$	$f_{nc} = 0.5$	$f_{nc} = 0.9$	$f_{nc} = 0$	$f_{nc} = 0.5$	$f_{nc} = 0.9$	Lower	Upper
Am	0.3	0.4	0.76	0.72	0.95	1.8	8.2	450,000
Co	0.0026	0.0034	0.0065	0.0061	0.0081	0.015	0.07	14,000
Eu[a]	1.9	3.1	10	9.1	15	50		825
H[b,c,d]	0.04	0.04	0.04	0.04	0.04	0.04	0.04	75
I[c]	0.04	0.04	0.04	0.04	0.04	0.04	0.04	368
Ni	2.2	2.9	5.6	5.3	6.9	13	60	4,700
Np[e]	0.5	0.5	0.5	0.5	0.5	0.5	0.5	2,580
Pu	0.18	0.24	0.46	0.44	0.58	1.1	5	190,000
Ra[c]	57	57	57	57	57	57	57	530,000
Sb[d]	1.7	2.2	4.2	3.9	5.2	9.9	45	550
Se[e]	36	36	36	36	36	36	36	310
Sm[a,d]	0.55	0.92	3	2.7	4.5	15	245	3,000
Sr[c]	0.05	0.05	0.05	0.05	0.05	0.05	0.05	32,000
Tc[e]	0.01	0.01	0.01	0.01	0.01	0.01	0.01	340
Th	0.74	0.98	1.9	1.8	2.3	4.4	20	13,000,000
U[e]	0.03	0.03	0.03	0.03	0.03	0.03	0.03	1,000,000

[a] K_d value was estimated on the basis of soil-to-plant concentration ratios; no lower and upper K_d values were observed.

[b] Upper K_d value is an estimate for the organic soil type.

[c] No correction was applied. Expected to already be soluble in aqueous solution under most conditions. Lower solubility is not expected to be significantly affected by organic materials in solution.

[d] The lower and/or upper observed K_d values are based on the range of geometric means for different soil types listed in Table 4.

[e] No correction was applied. Lower K_d value is representative of a soluble species that is not expected to be significantly affected by organic materials in solution.