Development of Probabilistic Fate and Transport Models for the Mixed Waste Landfill At Sandia National Laboratories

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ABSTRACT

A probabilistic performance assessment has been conducted to evaluate the fate and transport of radionuclides (Am-241, Cs-137, Co-60, Pu-238, Pu-239, Ra-226, Rn-222, Sr-90, Th-232, H-3, U-238), heavy metals (lead and cadmium), and volatile organic compounds at the Mixed Waste Landfill (MWL). Probabilistic analyses were performed to quantify uncertainties inherent in the system and models for a 1,000-year period, and sensitivity analyses were performed to identify parameters and processes that were most important to the simulated performance metrics. Comparisons between simulated results and measured values at the MWL were made to gain confidence in the models and perform calibrations when data were available. In addition, long-term monitoring requirements and triggers were recommended based on the results of the quantified uncertainty and sensitivity analyses.

INTRODUCTION

The Mixed Waste Landfill (MWL) is located approximately five miles southeast of Albuquerque International Sunport and four miles south of Sandia National Laboratories' (SNL) central facilities. The landfill is a fenced, 2.6-acre (10,500 m²) area with a mean elevation of 5381 feet (1640 m). The climate is semi-arid with an average annual precipitation of 9 inches (23 cm) and an average annual temperature of $56^{\circ}F(13^{\circ}C)$. The MWL was established in 1959 as a disposal area for low-level radioactive and mixed waste that was generated at SNL research facilities. The MWL accepted low-level radioactive waste and minor amounts of mixed waste from March 1959 through December 1988. Approximately 100,000 cubic ft (2,800 m³) of low-level radioactive waste containing approximately 6,300 curies of activity was disposed of at the landfill. The MWL consists of two distinct disposal areas: the classified area, occupying 0.6 acres (2400 m²), and the unclassified area, occupying 2.0 acres (8100 m²) (Fig. 1). Low-level radioactive and mixed waste has been disposed of in each area. Wastes in the classified area were buried in unlined, vertical pits. Wastes in the unclassified area were buried in unlined, shallow trenches. Contaminants of concern at the site that were evaluated include radionuclides, heavy metals (lead and cadmium), and tetrachloroethylene (PCE) as a proxy for other VOCs.

MODELING APPROACH

Probabilistic Performance Assessment Method

Previous studies have looked at individual components of the landfill performance, and nearly all of the studies relied on deterministic evaluations. This study uses a probabilistic performance-assessment approach that captures the inherent uncertainties in the system while honoring site-specific features, processes, and parameters. Sensitivity analyses are also introduced that utilize the probabilistic results to identify the parameters and processes that are most important to the simulated performance metrics.



Fig. 1. Mixed waste landfill (looking southwest, 1987)

A performance assessment is defined in DOE M 435.1-1 [1] as "an analysis of a radioactive waste disposal facility conducted to demonstrate there is a reasonable expectation that performance objectives established for the long-term protection of the public and the environment will not be exceeded following closure of the facility." In addition, DOE M 435.1-1 states that the method used for the performance assessment must include uncertainty analyses. A method that addresses these requirements has been used for the Waste Isolation Pilot Plant [2] and the Yucca Mountain Project [3] to assess the long-term performance of nuclear waste repositories. Probabilistic performance assessments have also been used for sites with uranium mill tailings [4]. A similar systematic approach has been used here to conduct a performance assessment of the MWL. The approach is outlined as follows: (1) Develop and screen scenarios based on regulatory requirements (performance objectives) and relevant features, events, and processes; (2) Develop models of relevant features, events, and processes; (3) Develop values and/or uncertainty distributions for input parameters; (4) Perform calculations and sensitivity/uncertainty analyses; (5) Compare results to performance objectives, identify important parameters and processes, and provide feedback to improve calculations, as needed.

A period of 1,000 years was selected for the probabilistic analysis to be consistent with DOE Order 435.1. DOE Order 435.1 requires that performance assessments be conducted for low-level radioactive waste disposed after September 26, 1988, and that performance objectives be evaluated for a 1,000-year period to determine potential risk impacts to the public and environment. Although most of the MWL wastes were disposed of prior to September 26, 1988, a 1,000-year period was nonetheless determined to be appropriate for assessment of regulatory performance metrics. In this study, 100 realizations were simulated for each scenario. A preliminary sensitivity analysis was performed using 100 vs. 200 realizations, and results showed that 100 realizations were sufficient to adequately represent the parameter uncertainty and distribution of the simulated performance metrics.

Sensitivity Analyses

The sensitivity of the performance-assessment model to uncertain input variables can be determined from the Monte Carlo probabilistic realizations using regression analysis. Multiple regression analysis involves construction of a linear regression model of the simulated output (the dependent variable) and the stochastic

input variables (independent variables) using a least-squares procedure. Stepwise linear regression is a modified version of multiple regression that selectively adds input parameters to the regression model in successive steps [5]. In this method, a sequence of regression models is constructed that successively adds the most important input parameters to the regression to improve the overall correlation. In the end, the sensitivity analysis identifies those parameters that are significantly correlated to the performance metric, and omits those parameters that are not. This study uses a stepwise linear rank regression to perform sensitivity analyses on simulated performance metrics that are at risk of being exceeded.

Scenarios and Performance Objectives

Table I summarizes the specific contaminants, scenarios, and performance objectives that were considered in this study. In general, the two pathways of concern include transport of volatile or gas-phase contaminants from the MWL to the atmosphere, and migration of aqueous-phase or vapor-phase contaminants through the vadose zone to the groundwater. For each of these primary pathways, relevant performance objectives and metrics were identified for each of the contaminants of concern. The chosen scenarios represent the most likely releases of contaminants from the MWL based on estimated inventories, contaminant properties, and previous studies.

Performance-Assessment Models

Infiltration Through Cover

Infiltration of water through a proposed soil cover for the MWL was modeled using the onedimensional, numerical code UNSAT-H [6]. UNSAT-H is a Richards' equation-based model that simulates infiltration, unsaturated flow, redistribution, evaporation, plant transpiration, and deep infiltration of water. The modeling was conducted in 2003 and 2004 using site-specific climate, hydrologic, and vegetation input parameters. Complete modeling input parameters, boundary conditions, and results are discussed in [6].

Leaching and Aqueous Transport

The transport of heavy metals (lead and cadmium) and the radionuclides (except for radon) were simulated using the probabilistic simulation tools FRAMES¹ (Framework for Risk Analysis in Multimedia Environmental Systems; [13]) and MEPAS² (Multimedia Environmental Pollutant Assessment System; Whelan et al., [14]), developed by Pacific Northwest National Laboratory. The FRAMES system, which integrates the fate and transport models comprising MEPAS, allows for a holistic approach to modeling in which models of different type (i.e., source, fate and transport, exposure, health impact), resolution (i.e., analytical, semi-analytical, and numerical), and operating platforms can be combined as part of the overall assessment of contaminant fate and transport in the environment. The FRAMES system employs a graphical user interface for integrating computer models, an extensive contaminant database, a probabilistic sensitivity/uncertainty module, and textual and graphical viewers for presenting modeling outputs.

¹ http://mepas.pnl.gov/FRAMESV1

² http://mepas.pnl.gov/earth/mepasmain.html

Scenario	Description	Performance Objectives ^a
1	Water percolates through the cover to the waste	• Infiltration through the cover shall be less than 10 ⁻⁷ cm/s (a unit-gradient flow is assumed to equate infiltration to hydraulic conductivity) [7]
2	Tritium diffuses to the atmosphere and migrates via gas and aqueous phases through the vadose zone to the groundwater	 Dose to the public via the air pathway shall be less than 10 mrem/yr (excludes radon) [8] Dose from beta particles and photon emitters in drinking water shall be less than 4 mrem/yr [9, 10] Tritium concentrations in groundwater shall not exceed 20,000 pCi/L [9, Table A; tied to 4 mrem/yr]
3	Radon steadily diffuses to the atmosphere and migrates via gas and aqueous phases through the vadose zone to the groundwater	 The average flux of radon-222 gas shall be less than 20 pCi/m²/s at the surface of the landfill [11] Radon concentrations in groundwater shall not exceed 300 pCi/L [12]
4	One or more radionuclides migrate via the aqueous phase through the vadose zone to the groundwater	 Maximum concentrations in groundwater of gross alpha particle activity (including Ra-226 but excluding radon and uranium) is 15 pCi/L [9, 10] Uranium concentrations in groundwater shall not exceed EPA MCL of 30 g/L [9, 10] Dose from beta particles and photon emitters shall be less than 4 mrem/yr [9, 10]
5	Lead and cadmium migrate via the aqueous phase through the vadose zone to the groundwater	 Lead concentrations in groundwater shall not exceed the EPA action level of 15 g/L [10] Cadmium concentrations in groundwater shall not exceed the EPA MCL of 5 g/L [10]
6	PCE migrates through the vadose zone to the groundwater	• PCE concentrations in groundwater shall not exceed the EPA MCL of 5 g/L [9, 10]

Table I.	Summary of Scenarios and Performance	Objectives used in the	Performance	Assessment of the	
MWL					

MCL = Maximum Contaminant Level; PCE = tetrachloroethylene

^aThe point of compliance is taken at the boundary of the waste site.

Existing models in FRAMES include those derived from MEPAS [14]. MEPAS is a physics-based environmental analysis code that integrates source-term, transport, and exposure models for endpoints such as concentration, dose, or risk. MEPAS is capable of computing contaminant fluxes for multiple routes, which include leaching to groundwater, overland runoff, volatilization, suspension, radioactive decay, constituent degradation, and source/sink terms. In this study, only the source-term and vadose-zone models were implemented. The source-term model conservatively simulates leaching from the waste zone (assuming no containment) based on either the solubility or the inventory-limited concentration [15]. The transport of the contaminant through the vadose-zone is then simulated assuming liquid-phase advection, dispersion, adsorption, and decay of the contaminant [14]. In this study, the aquifer concentration of the constituent in the groundwater at the interface of the vadose-zone and the water table (e.g., dilution caused by transport in the saturated zone was ignored). Uncertainty analyses are performed in FRAMES using the sensitivity module, which utilizes the Latin Hypercube Sampling [16] technique to minimize the number of modeling runs that must be performed to accurately represent distributions selected by the user.

Transient Gas- and Liquid-Phase Transport

A separate model was used to model the transient transport of tritium at the MWL. Tritium, in the form of tritiated water, is volatile and can be transported via both the gas and liquid phases. Regulatory metrics exist for dose caused by exposure to tritium (a beta particle emitter) in both the air and groundwater pathways (see

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Table I). Also, because the half-life of tritium is relatively short (12.3 years), a transient analysis was required. Therefore, the transport of tritium was modeled using a transient model that accounts for advective liquid-phase transport, diffusive gas-phase transport, decay, and adsorption (if applicable) in the vadose zone [17,18]. This same model was also used to model the transport of tetrachloroethylene (PCE), which was used as a proxy for other VOCs based on its relatively high mobility. In this model, a contaminated zone is assumed to initially exist with a defined thickness and concentration. Over time, the contaminant migrates and decays (if applicable) assuming a flux boundary condition at the surface, defined by an atmospheric boundary layer thickness [17] and a zero concentration boundary beneath the waste zone at a location infinitely far away from the source. Superposition is used to account for a clean overburden (cover) above the waste zone [18]. The analytical solution to this model was implemented in Mathcad,[®] and a Monte Carlo analysis was implemented with the uncertain variables using 100 realizations.

Steady-State Gas- and Liquid-Phase Transport

Rn-222 is generated from the decay of Ra-226, which is a decay product of U-238. Because these parent constituents have long half-lives, the source of Rn-222 production is assumed to last indefinitely. Therefore, the transient model described in the previous section that accounts for a finite source of contaminant is not appropriate. Instead, a steady-state model of radon transport was developed to account for steady generation of Rn-222, advective liquid-phase transport, diffusive gas-phase transport, and decay. Mathcad[®] was used to provide a Monte Carlo analysis of the analytical solution using 100 realizations.

Input Parameters and Distributions

The constituents that were included in the performance assessment of the MWL are summarized in Table II. The parameter values and distributions that were used are also summarized in the table. The adsorption coefficient (K_d) was assumed to be an uncertain parameter, so a range of values was obtained from the literature for the constituent and soil type (sandy loam) at the MWL. The inventory of each constituent was also assumed to be an uncertain variable. The estimated inventory from previous reports and studies was used as the lower bound in a uniform distribution for each constituent. The maximum solubility obtained from the literature for each constituent was used. All other parameters were obtained from site-specific reports, scientific literature, or EPA recommendations.

Table III summarizes the contaminated waste-zone (source term) parameters in the models. The wastezone length, width, and thickness are based on the size of the pits, trenches, and dimensions of the MWL. The maximum thickness of the cover is based on the design specifications given in [6]. The minimum thickness of the cover is set equal to zero as a bounding value to account for the possibility that complete erosion of the cover may occur in the future. This is a conservative bounding assumption since the intent is to maintain the integrity of the cover at the MWL.

Table IV summarizes vadose-zone parameters in the models. Uncertainty was included for a number of variables including thickness of the vadose zone, infiltration rate, hydraulic conductivity, and site-specific transport parameters. The distributions used for the various vadose-zone parameters were derived from site-specific data or literature pertaining to the constituents and scenarios evaluated in this study. The liquid- and gas-phase tortuosity coefficients are used to calculate effective diffusion coefficients in porous media. The minimum value is based on formulation by Millington [19], and the maximum value is assumed to be equal to one (the upper bound), which yields the maximum diffusion. Studies of enhanced vapor diffusion have shown that large values of the tortuosity coefficient (yielding diffusion rates equivalent to those in free space) are possible in unsaturated porous media because of evaporation and condensation mechanisms across liquid islands in pores [20].

Constituent and Molecular Weight	Inventory ^a	Half-Life ^b	Specific Activity (Ci/g) ^c	Adsorption Coefficient, K _d (mL/g) ^d	Max Solubility (mg/L) ^e	Liquid-Phase Diffusion Coefficient (m²/s) ^f	Gas-Phase Diffusion Coefficient (m ² /s) ^f	Henry's Constant (C _g /C _l) ^g	Dose Conversion Factor (rem/pCi) ^h
Am-241	<u>Uniform</u> : 0.04 - 0.08 Ci	433 yrs	3.43	<u>Log-Uniform</u> : 1900 – 9600	2.4x10 ⁴	6x10 ⁻¹⁰	N/A	N/A	3.64x10 ⁻⁶
Cs-137	<u>Uniform:</u> 410 – 820 Ci	30.2 yrs	86.4	<u>Log-Uniform</u> : 30 – 4600	137,000	6x10 ⁻¹⁰	N/A	N/A	5.0x10 ⁻⁸
Co-60	<u>Uniform:</u> 3500 – 7000 Ci	5.27 yrs	1130	<u>Log-Uniform</u> : 60 – 1300	600	6x10 ⁻¹⁰	N/A	N/A	2.69x10 ⁻⁸
Pu-238	<u>Uniform</u> : 0.0012 - 0.0024 Ci	87.7 yrs	17.1	<u>Log-Uniform</u> : 80 – 520	0.24	6x10 ⁻¹⁰	N/A	N/A	3.2x10 ⁻⁶
Pu-239	<u>Uniform</u> : 0.0012 - 0.0024 Ci	2.41x10 ⁴ yrs	0.0621	<u>Log-Uniform</u> : 80 – 470	0.24	6x10 ⁻¹⁰	N/A	N/A	3.54x10 ⁻⁶
Ra-226	<u>Uniform</u> : 6-12 Ci	1,600 yrs	0.989	<u>Log-Uniform</u> : 500 – 36,000	0.45	6x10 ⁻¹⁰	N/A	N/A	1.32x10 ⁻⁶
Rn-222	Constant generation from Ra-226	3.82 days	1.54x10 ⁵	0	N/A	0.07exp[-4(S where S=liqui =port	$-S^{2}+S^{5}$] ad saturation, rosity	0.26 ⁻¹	1.44x10 ⁻⁸ (inhalation)
Sr-90	<u>Uniform</u> : 410 -820 Ci	29.1 yrs	137	<u>Log-Uniform</u> : 15 – 20	90,000	6x10 ⁻¹⁰	N/A	N/A	1.42x10 ⁻⁷
Th-232	<u>Uniform</u> : 1 – 2 Ci	1.4x10 ¹⁰ yrs	1.10x10 ⁻⁷	<u>Log-Uniform</u> : 20 – 2000	23	6x10 ⁻¹⁰	N/A	N/A	2.73x10 ⁻⁶
Tritium H-3	<u>Uniform</u> : 2400 – 4800 Ci	12.3 yrs	9690	0	N/A	2.3x10 ⁻⁹	2.6x10 ⁻⁵	1.7x10 ⁻⁵	6.4x10 ⁻¹¹ (inhalation; x1.5 to include dermal absorption)

 Table II. Summary of Input Parameters and Distributions for Constituents used in the Models

Constituent and Molecular Weight	Inventory ^a	Half-Life ^b	Specific Activity (Ci/g) ^c	Adsorption Coefficient, K _d (mL/g) ^d	Max Solubility (mg/L) ^e	Liquid-Phase Diffusion Coefficient (m ² /s) ^f	Gas-Phase Diffusion Coefficient (m ² /s) ^f	Henry's Constant (C _g /C _l) ^g	Dose Conversion Factor (rem/pCi) ^h
U-238	<u>Uniform</u> : 9.3 – 18.6 Ci	4.47x10 ⁹ yrs	3.35x10 ⁻⁷	<u>Log-Uniform</u> : 0.4 – 15	24	6x10 ⁻¹⁰	N/A	N/A	2.55x10 ⁻⁷
Cadmium 112.41	<u>Uniform</u> : 1350 – 2700 kg	stable	N/A	<u>Log-Uniform</u> : 8 – 80	$1.4 \mathrm{x} 10^{6}$	6x10 ⁻¹⁰	N/A	N/A	N/A
Lead 207.2	<u>Uniform</u> : 128,000 – 256,000 kg	stable	N/A	<u>Log-Uniform</u> : 270 – 4360	4.43x10 ⁵	6x10 ⁻¹⁰	N/A	N/A	N/A
PCE 165.83	<u>Uniform</u> : 5 – 70 kg	<u>Log-Uniform</u> : 9 mos – 10 ¹⁰ yrs	N/A	<u>Log-Uniform</u> : 0.038 - 2	N/A	9.2x10 ⁻¹⁰	9.5x10 ⁻⁶	0.42	N/A

N/A-Not Applicable or not used in the model; for solubility, this indicates that the value is not limiting

Alpha particle; Beta particle

^aMinimum inventory of all constituents except cadmium and PCE was estimated from values in SNL [21]; maximum value was assumed to be twice the minimum value. Cadmium inventory was estimated from measured soil concentrations (Peace et al., 2002) and maximum simulated penetration depth (120 feet) of coolant water potentially carrying the cadmium (Wolford, 1997). PCE inventory is estimated from measured soil-gas concentrations [22]; the maximum measured gas concentration (5,900 ppb) was used as a minimum value in a uniform distribution increasing to ten times this value (calibrated to available data). The maximum areal extent of the MWL was used (430 feet x 300 feet) along with an uncertain thickness ranging from 10-27 feet (see Table III for waste-zone description).

^b[23]; half-life of PCE is assumed to range from 9 months (EPA fact sheet: www.epa.gov/WGWDW/dwh/t-voc/tetrachl.html) to 10¹⁰ yrs (no degradation)

^cSpecific activity is calculated as 3.575x10⁵/(half-life (yrs) x molecular weight)

^d[24,25],26], EPA fact sheet: www.epa.gov/WGWDW/dwh/t-voc/tetrachl.html

^e[26,27,28,29,30], and EPA Online Fact Sheets (www.epa.gov/safewater/dwh/t-ioc). Based on the maximum inventory and minimum waste volume possible, the solubility may potentially limit the maximum aqueous source concentration for Ra-226, Th-232, U-238, and lead; all other constituents are not limited by the solubility.

^f[14,31,32,33]

^g[32,31], steam tables, and EPA's online Henry's Constant calculator (www.epa.gov/athens/learn2model/part-two/onsite/esthenry.htm)

^h[34]

Input Parameter	Value or Distribution	Basis and Comments		
Waste-Zone Length	Uniform	Minimum value determined by size of individual pit (10'). Maximum value determined by extent of Mixed Waste Landfill.		
[m]	3.05 - 131			
Waste-Zone Width	Uniform	Minimum value determined by size of individual pit (10'). Maximum		
[m]	3.05 - 91.4	value determined by extent of Mixed Waste Landfill.		
Waste-Zone Thickness [m]	Uniform 3.05 – 8.23	The thickness of the waste zone for all constituents except for cadmium is based on the depth of the trenches and pits, which range from $3 - 8$ m (10 - 27 feet). The thickness of the cadmium contamination zone is assumed to be equal to 36.6 m (120 feet), which is the maximum simulated penetration depth of the coolant water that may have carried the cadmium [35].		
Thickness of Cover and Clean Overburden [m]	Uniform 0 – 4.88	Minimum value is assumed to be zero due to erosion. ^a Maximum value is based on maximum thickness of the cover at various locations [6].		

Table III. Summary of Input Parameters and Distributions for the Waste Zone

^aThe intent is to maintain the integrity of the cover at the MWL. Complete erosion of the cover is a conservative bounding assumption for modeling purposes.

Table IV. Summary of Input Parameters and Distributions for the Vadose Zone

Input Parameter	Value or Distribution	Basis and Comments	
Thickness of Vadose Zone ^a [m]	Uniform 133 - 148	Thickness of the vadose zone for all constituents except for cadmium is based on measured depths to the water table. The depth to the water table from the surface ranges from $141 - 151$ m (461 - 495 feet) [36]. The range of vadose-zone thicknesses accounts for the waste-zone thickness. For cadmium, the thickness is assumed to be 104 m (461 - 120 = 341 feet).	
Infiltration Rate [m/s]	Uniform 1.18x10 ⁻¹¹ – 6.12x10 ⁻¹¹	Minimum value based on infiltration through 2 ft of engineered cover under current climate [6]; maximum value based on two times the current maximum precipitation in a natural analog vegetative cover to account for future climates [37].	
Saturated Hydraulic Conductivity [cm/day]	Log-Normal Mean log: 1.039 S.D. log: 0.705 Upper bound: 173 Lower bound: 0.38	[38]	
Porosity [-]	Uniform 0.302 – 0.445	[6]	
Volumetric Moisture Content [-]	Uniform 0.053 – 0.225	[6]	
Longitudinal dispersivity [m]	0.1 times the travel distance (vadose-zone thickness)	Based on field data reported in Gelhar et al. [39]. This is used in the FRAMES/MEPAS models for liquid transport to the groundwater.	
Liquid-Phase Tortuosity Factor [-]	Uniform 0.001 – 1	Lower bound based on formulation of Millington [19]; upper bound is physical limit. This is used in the tritium and PCE models.	
Gas-Phase Tortuosity Factor [-]	Uniform 0.1 – 1	Lower bound based on formulation of Millington [19]; upper bound is physical limit. This is used in the tritium and PCE models.	

^aUsed only in FRAMES/MEPAS. For all other models, the depth to the water table (141-151 m) is used.

Finally, Table V summarizes the parameters and distributions used to estimate dose due to exposure via the atmospheric (e.g., inhalation) or groundwater pathway. Dose via inhalation and dermal adsorption of

gas-phase tritium was calculated based on the surface flux (pCi/m²/s) of tritium determined in the models. The length and width of the waste zone was used to determine the flux rate of tritium at the surface (pCi/s), and the average wind speed and vertical mixing height was used to determine the average concentration above the landfill. The inhalation rate was then used to estimate the human intake of gas-phase tritium, and the dose-conversion factor (Table II) was used to determine the dose. For groundwater exposure, a conservative estimate for water ingestion (10 L/day) was used together with the simulated groundwater concentrations to determine intake. The assumed water ingestion rate of 10 L/day is five times greater than the EPA drinking-water standard of 2 L/day and is intended to account for indirect sources of water ingestion and absorption such as consumption of vegetables and fruits irrigated by contaminated water. The dose-conversion factor was used to estimate dose via the groundwater pathway.

Key Assumptions

The key assumptions regarding the models and input parameters used in the performance assessment of the MWL are summarized below:

- Receptor located adjacent to MWL
 - Tritium dose caused by continuous inhalation and exposure of tritium flux directly above MWL.
 - Groundwater dose calculated based on concentrations in aquifer directly beneath MWL. Water intake assumed to be 10 L/day (five times EPA standard of 2 L/day for drinking water).
- Maximum waste inventory set equal to twice estimated values based on historical records.
- Sealed sources of Ra-226 allowed to degrade in 1,000 years (emanation factor for radon-222 allowed to increase).
- Cover allowed to completely erode in 1,000 years.
- 1-D model: yields maximum transport to surface and groundwater.
- Bounding tortuosity coefficients: yields maximum diffusion rates.

Input Parameter	Value or Distribution	Basis and Comments		
Atmospheric Boundary Layer	Uniform	Minimum is based on values reported by [17]. Maximum is a		
Thickness [m]	0.001 - 1	conservative upper value.		
Vertical Atmospheric Mixing Length [m]	2	Conservative value to encompass volume occupied by a human [40].		
Average Wind Speed [m/s]	3.63	Average value based on seven years of site data (SNL Site Environmental Monitoring Reports 1990-1996).		
Inhalation Rate [m ³ /day]	20	[41]		
Water Intake [L/day]	10	Conservative estimate to account for drinking water and indirect ingestion or absorption via plants, animals, showering, etc. Recommended value for drinking water is 2 L/day [42].		
Distance to Receptor [m]	0	The point of compliance for groundwater concentrations is assumed to be at the boundary of the landfill. Receptor is assumed to be located adjacent to landfill for inhalation, and water used for drinking, irrigation, etc. is assumed to be drawn from the aquifer directly beneath the MWL.		

Table V. Summary of input parameters and distributions for the biosphere.

MODEL RESULTS

Water Infiltration through the Cover

One of the objectives of the modeling was to assess whether the proposed 3-ft cover will meet the EPAprescribed technical equivalency criteria. The EPA performance-based, technical equivalency criteria used in this study are 31.5 millimeter (mm)/year (yr), or less, for net annual infiltration and 1 x 10^{-7} centimeter (cm)/second (s) average infiltration rate, based on a hydraulic conductivity of 1 x 10^{-7} cm/s and the assumption of constant unit gradient conditions. The modeling results demonstrate that the proposed 3-ft vegetated soil cover will meet the EPA-prescribed technical equivalency criteria for RCRA landfills under both present and future conditions. Predicted average infiltration rates through the MWL cover are expected to range from 1.18 X 10^{-9} cm/s for present conditions to 6.12 X 10^{-9} cm/s for future conditions, under the assumption of significantly higher precipitation.

Fate and Transport of Tritium

In 1990 and 1993, measurements of tritium at the surface and at locations in the subsurface were performed at the MWL [43]. These measurements were used as a reference to check the simulated results of the model. Fig. 2 shows the simulated tritium surface flux as a function of time for 100 realizations. The minimum and maximum measured tritium surface flux values taken in 1993 are also shown in the figure. The measured values are shown spanning 5 to 33 years because the actual time elapsed since the tritium was emplaced is uncertain. Emplacement of tritium at the MWL began in 1960 and ended in 1988; therefore, the measured values sampled in 1993 could have occurred between 5 and 33 years after emplacement. Results show that the simulated results during this span of time are either within or above the measured bounding values. Similar comparisons were made for different locations in the subsurface. In most cases, the simulated fluxes and concentrations are higher than the measured values. These results and comparisons provide evidence that the model is producing conservatively high results for surface fluxes and subsurface concentration because of the conservative values and distributions used for the model parameters.



Fig. 2. Left: Comparison of simulated tritium surface flux as a function of time for 100 realizations with range of measured values in 1993. Right: Cumulative probability for simulated peak tritium dose via the air pathway for 100 realizations

The simulated tritium concentrations reaching the groundwater were all well below 20,000 pCi/L. The cumulative probability for the simulated peak tritium dose via groundwater was calculated based on the simulated aquifer concentrations and a conservative water intake of 10 L/day (accounts for drinking water, indirect ingestion via plants and animals, absorption and inhalation via showering, etc.). The results shows that all simulated groundwater dose values were well below the EPA metric of 4 mrem/year.

Fig. 2 also shows the cumulative probability for the simulated peak tritium dose via the air pathway for 100 realizations. The simulated dose due to inhalation (and skin absorption) is based on continuous exposure to the concentration of gas-phase tritium immediately above the MWL. The average wind velocity, vertical mixing length, and surface flux of tritium are used to calculate the air concentration above the MWL, and the inhalation rate is used to calculate the intake (Table V). The dose conversion factor (Table I) is then used to calculate the dose rate. Because the simulated surface flux of tritium for several realizations was quite high (Fig. 2), a small percentage ($\sim 2\%$) of the realizations yield a dose via the air pathway that exceeds the EPA metric of 10 mrem/year.

Inhalation dose was found to be sensitive to upward diffusion through the liquid phase as well as the gas phase. The cover thickness and atmospheric boundary-layer thickness were also found to be significant. Finally, although not included as an uncertain parameter, the location and disposition of the receptor played an important role in the simulated inhalation dose. In this study, the receptor was assumed to be located adjacent to the MWL, continuously inhaling air directly above the MWL (24 hours a day, 365 days a year). If the receptor were located further away from the site, or if the exposure were not continuous, the simulated dose via the air pathway would be considerably less.

Fate and Transport of Radon

The potential sources of Rn-222 (Ra-226) were sealed and contained before disposal in the MWL. Therefore, the containment is assumed to be generally intact at present, but defects or breaks may still be present. The minimum emanation factor, which accounts for present-day emissions, was adjusted to yield a radon flux between 0.1 and 1 pCi/m²/s (equivalent to the difference in maximum measured and background fluxes). The resulting minimum emanation factor used in the probabilistic simulations was 10^{-6} . The maximum emanation factor was estimated based on the possibility that the sealed containers may degrade in the future. The integrity of the containers is expected to last well beyond 1,000 years, but an upper value of the emanation factor was set equal to 0.01 to represent the possibility that 1% of the containers will completely degrade within 1,000 years. An evaluation was also performed assuming that the maximum emanation factor was equal to one, which is equivalent to complete degradation of the containment of all the radon sources within 1,000 years. A log-uniform distribution between 10^{-6} and the maximum value was used for the emanation factor.

Fig. 3 shows the cumulative probability for the simulated peak Rn-222 surface flux for 100 realizations. For the scenario with a maximum emanation factor of 0.01 (1% of the radon-source containers degrades completely), the results show that 97% of the simulated radon surface fluxes are below the design standard of 20 pCi/m²/s (3% of the realizations yield radon surface fluxes that exceed the design standard). In the bounding scenario, where we allow all of the containment of the sealed sources to completely degrade, nearly 30% of the realizations exceed the design standard of 20 pCi/m²/s. The large uncertainty in the emanation factor allowed significant variations in the simulated radon surface flux. The waste volume, cover thickness, and effective diffusion coefficient were also shown to be statistically correlated to the simulated radon surface flux, but to a much lower degree.

It is unlikely that the sealed sources and containers for Ra-226 will degrade significantly over the next few hundred years, but because the half-life of Ra-226 and U-238 is extremely long, Rn-222 will continue

to be generated from these parent products indefinitely. Therefore, degradation of the containers may eventually cause the emanation factor for Rn-222 to increase at some point in the future. For a 1,000-year evaluation period, however, the probability of exceeding the radon surface-flux design standard is very small if the sealed sources and containers do not degrade significantly and the emanation factor remains below 0.01. Simulated radon concentrations in groundwater were negligible ($<10^{-20}$ pCi/L). The short half-life of radon (3.8 days) and the large thickness of the vadose zone prohibit radon from migrating significant distances to the water table when the source originates from the landfill.



Fig. 3. Cumulative probability for simulated peak Rn-222 surface flux for 100 realizations using two different maximum values for the emanation factor, E

Fate and Transport of Aqueous Radionuclides

Results of the FRAMES/MEPAS simulations showed that none of the aqueous radionuclides (see Table II) were simulated to reach the groundwater in 1,000 years. All of the radionuclides were retarded sufficiently by adsorption to prevent significant migration in 1,000 years, even with the realistically conservative distributions used for model inputs. Simulations with extended periods showed the uranium could reach the groundwater after approximately 10,000 years, but at concentrations below the regulatory metric of 30 μ g/L. Sensitivity studies showed that the uranium concentrations in the groundwater could exceed the regulatory metric in 1,000 years if the maximum infiltration were increased by two orders of magnitude.

Fate and Transport of Heavy Metals

Simulations showed that neither lead nor cadmium were able to reach the groundwater in all 100 realizations for 1,000 years. Extended simulation periods (>10,000 years) also did not yield any breakthrough of lead or cadmium to the water table. Both lead and cadmium have relatively large adsorption coefficients (see Table II), which retard their transport through the thick vadose zone. Sensitivity analyses showed that cadmium could reach the groundwater in 1,000 years and exceed its regulatory metric if the maximum infiltration were increased by three orders of magnitude. Lead was simulated to reach the water table in 1,000 years if the maximum infiltration were increased by four orders of magnitude over the maximum expected infiltration.

Fate and Transport of PCE

Samples of PCE soil-gas concentrations were taken at the MWL in 1993 [43]. The ranges of measured values at two different depths (10 feet and 30 feet) were compared to simulated soil-gas concentrations using the transient PCE transport model described in the previous section. Fig. 4 shows the comparisons for all 100 simulated realizations at 30 feet (similar comparisons were found at 10 feet). As discussed in previous sections, the measured values in 1993 are shown spanning a time period between 5 and 33 years, which accounts for the uncertainty in the time of emplacement. Results show the majority of simulated soil-gas concentrations during this time period at the two depths are between the maximum and minimum values measured in 1993.



Fig. 4. Left: Simulated PCE gas concentration at a depth of 30 feet as a function of time for 100 realizations with a range of measured values in 1993. Right: Cumulative probability for simulated PCE peak groundwater concentrations for 100 realizations

The cumulative probability of the peak PCE groundwater concentration for all 100 realizations is shown in Fig. 4. The results show that approximately 99% of the realizations yield groundwater concentrations less than the regulatory metric of 5 μ g/L. Only 1% of the realizations yielded groundwater concentrations that exceeded the regulatory metric. The majority of the realizations show the aquifer concentrations peaking before 50 years. Depending on the time of disposal, this corresponds to peak concentrations occurring by 2010 – 2040. So far, no detectable amounts of PCE have been found in the groundwater at the MWL. This is still consistent with the simulations, which show a large amount of variability in the simulated concentrations resulting from uncertainty included in the input parameters.

The uncertainty in the PCE K_d , half-life (degradation), inventory concentration, source thickness, and cover thickness values were found to be the most statistically significant parameters that impacted the variability in the simulated PCE aquifer concentrations. As stated in previous sections, the adsorption coefficient, K_d , plays an important role in the retardation and mobility of the constituent. The half-life and inventory both govern the persistence and availability of the PCE during migration to the groundwater. The source thickness also contributes to the overall inventory of PCE since the inventory concentration is applied to the entire source volume.

RECOMMENDED TRIGGERS FOR LONG-TERM MONITORING

Based on the results of the probabilistic performance-assessment modeling conducted for the MWL, monitoring triggers were proposed for surface emissions of tritium and radon, infiltration through the MWL cover, and groundwater concentrations. Specific triggers include thresholds for radon concentrations in the air, tritium concentrations in surface soil, moisture content in the vadose zone, and uranium and select VOC concentrations in groundwater [44]. The proposed triggers are based on EPA and DOE regulatory standards. If a trigger is exceeded, then SNL/DOE will initiate a trigger evaluation process which will allow sufficient data to be collected to assess trends and recommend corrective action, if necessary.

SUMMARY AND CONCLUSIONS

A probabilistic performance assessment has been conducted to evaluate the fate and transport of contaminants of concern at the Mixed Waste Landfill. The contaminants that were simulated include radionuclides (Am-241, Cs-137, Co-60, Pu-238, Pu-239, Ra-226, Rn-222, Sr-90, Th-232, H-3, U-238), heavy metals (lead and cadmium), and a volatile organic compound (PCE). The current analysis differs from previous analyses in several ways: (1) probabilistic analyses were performed to quantify uncertainties inherent in the system and models; (2) a comprehensive analysis of the performance of the MWL was evaluated and compared against relevant regulatory metrics; (3) sensitivity analyses were performed to identify parameters and processes that were most important to the simulated performance metrics; and (4) long-term monitoring requirements and triggers were recommended based on the results of the quantified uncertainty and sensitivity analyses.

Results showed that exposure to tritium via the air pathway exceeded the regulatory metric of 10 mrem/year in about 2% of the simulated realizations when the receptor was located at the MWL (continuously exposed to the air directly above the MWL). Simulations showed that peak radon gas fluxes exceeded the design standard of 20 pCi/m²/s in about 3% of the realizations if up to 1% of the containers of sealed Ra-226 sources were assumed to completely degrade in the future. If up to 100% of the containers of Ra-226 sources were assumed to completely degrade, 30% of the realizations yielded radon surface fluxes that exceeded the design standard. For the groundwater pathway, simulations showed that none of the radionuclides or heavy metals (lead and cadmium) reached the groundwater during the 1,000 evaluation period. Tetrachloroethylene (PCE) was used as a proxy for other VOCs because of its mobility and potential to exceed maximum contaminant levels in the groundwater relative to other VOCs. Simulations showed that PCE reached the groundwater, but only 1% of the realizations yielded aquifer concentrations that exceeded the regulatory metric of 5 $\mu g/L$.

Based on these results, monitoring triggers have been proposed for the air, surface soil, vadose zone, and groundwater at the MWL. Specific triggers include numerical thresholds for radon concentrations in the air, tritium concentrations in surface soil, infiltration through the vadose zone, and uranium and select VOC concentrations in groundwater. The proposed triggers are based on U.S. Environmental Protection Agency and Department of Energy regulatory standards. If a trigger is exceeded, then a trigger evaluation process will be initiated which will allow sufficient data to be collected to assess trends and recommend corrective actions, if necessary.

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