EVALUATION OF FRAMES FOR PROBABILISTIC RISK-BASED SIMULATION OF LONG-TERM COVER SYSTEMS

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ABSTRACT
Current guidelines for landfill cover designs at mill tailing and other hazardous-waste sites are not risk-based and do not consider long-term site specific influences, such as climate and soil type. A probabilistic risk-based performance assessment method has been developed in order to address these site-specific influences that may contribute to groundwater contamination and human exposure. A probabilistic software tool (FRAMES) was evaluated and used in the performance-assessment simulations.

For this study, the performance assessment method was tested using data from the Lakeview Repository in Oregon. The Lakeview Repository is used to isolate uranium mill tailings. Contaminants of concern at the repository include arsenic, cadmium, radium 226, uranium 234, uranium 238, as well as radium and uranium progeny (bismuth 210, lead 210, polonium 210, radon 222, thorium 230, and thorium 234). Performance metrics of this study include water percolation to the contaminants, contaminant concentrations in groundwater, and dose to receptor. The performance assessment is based on computer models that incorporate flux through the cover, source-term release, vadose-zone transport, saturated-zone transport, and exposure pathways. The total system uses sensitivity/uncertainty distributions of important input parameters sampled using the Latin Hypercube Sampling method, resulting in quantification of uncertainty and identification of the parameters most influential to performance. Results show that the dose and water concentration are primarily sensitive to the uncertainties in the adsorption coefficients.

A second aspect of this study was to validate the individual computer modules in FRAMES by comparing the results to analytical solutions of contaminant transport. Analytical predictions were compared to simplified models in FRAMES of advective-dispersive transport. This exercise revealed some bugs in the code that were fixed in the version that was used for the Lakeview simulations.

INTRODUCTION

Background
Through the 2006 Accelerated Cleanup Plan [1], the Department of Energy (DOE) intends to clean up more than 90 percent of the contaminated sites in its Environmental Management Program. Long-term landfill covers are considered a vital option to assist in this cleanup plan because they help isolate contaminants from the biosphere at near-surface landfills, waste-disposal sites, and high-level radioactive waste tanks. DOE Order 435.1 [2] mandates performance assessments be conducted for low-level radioactive waste disposed after September

26, 1988, and performance objectives should be evaluated for a 1,000 year period in order to determine potential health risk impacts to the public and environment. However, current guidelines for landfill cover design are not risk based and do not consider long-term site specific influences such as climate, vegetation, and soil types. Therefore, these design guidelines may not address important long-term features, events, and processes at the site that may contribute to long-term risk of groundwater contamination and human exposure. In addition, traditional design guidelines for covers often rely on deterministic models of flow and transport processes that neglect uncertainty inherent in actual contaminant transport.

Sandia National Laboratories, in association with the Pacific Northwest National Laboratory developed a probabilistic, risk-based performance-assessment methodology to assist in the selection design, and monitoring of long-term cover systems. This methodology follows five steps: 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 uncertain input parameters; 4. Perform calculations and sensitivity/uncertainty analyses; and 5. Document results and provide feedback to previous steps and associated areas to improve calculations, as needed [3].

In this study, the five-step performance-assessment methodology is applied to the Lakeview Mill Site in Oregon. Performance metrics of this study include contaminant concentrations in groundwater and dose to receptor. The performance assessment is based on computer models produced with the FRAMES 1.5 software that incorporate flux through the cover, source-term release, vadose-zone transport, saturated-zone transport, and exposure pathways.

The second aspect of this study is to validate the individual computer modules in FRAMES 1.5 by comparing the results to analytical solutions of contaminant transport. Analytical predictions were compared to simplified models in FRAMES 1.5 of advective-dispersive transport.

The Lakeview Mill Site

The Lakeview Mill site is located in Lake County Oregon approximately 16 miles north of the Oregon-California border. The present climate at the Lakeview Mill site is "semi-arid" with an average annual precipitation of ~40 cm (15.8 inches) and an average annual temperature of 8.3 °C (47 °F). The Lakeview Mill was constructed in 1958 by the Lakeview Mining Company. Although the mill was contracted through 1963, it was closed in November of 1960 due to lack of ore. At closure, the mill had produced approximately 130,000 tons of mill tailings. These mill tailings are sand-like materials that remain after uranium has been extracted from the ore and contain radioactive materials as well as other contaminants. The contaminants of concern at the Lakeview Mill Site include arsenic, cadmium, radium 226, uranium 234, uranium 238, as well as the progeny of radium and uranium. [4]

Remediation History

In 1978 the Uranium Mill Tailings Radiation Control Act (UMTRCA) was passed in order to access and remediate 24 privately owned mill sites, including the Lakeview Mill Site in Oregon. Under Title I of the UMTRCA, the DOE is required to clean up the site to be in compliance with

40 CFR 192 standards set by the U.S. Environmental Protection Agency (EPA), with concurrence of the U.S. Nuclear Regulatory Commission.

Remediation of the Lakeview Mill Site began in June 1986 and was completed in October 1989. At the Lakeview Mill Site, there were three separate units addressed by the UMTRCA project: mill tailing and contaminated materials at the mill site, vicinity properties contaminated by material from the mill site, and contaminated surface and groundwater down gradient of the mill site. [5]

The remediation of the first unit included excavation and relocation of approximately 716,680 cubic meters (943,000 cubic yards) of mill tailings and contaminated soil to the Collins Ranch Disposal Site, located approximately seven miles northwest of the mill. Relocation of the contaminated materials was necessary because of possible naturally occurring geothermal and seismic instabilities at the Lakeview site. [5]

The second unit included eight off-site properties, identified by the DOE, contaminated with tailings from the Lakeview Mill Site. These vicinity properties included residences and commercial buildings where mill tailings were used in construction materials, as well as open lands contaminated by mill tailings that were transported by wind and water erosion. Cleanup of these vicinity properties began in May 1987 and was completed in August 1988. The remediation of these properties included removing all residual radioactive material (RRM) and transporting it to the Collins Ranch Disposal site. [5]

Once the Collins Ranch Disposal cell was filled, a three-foot, multilayered, earthen cap was constructed. The contaminated material is covered with a 1.5 ft thick radon barrier/infiltration layer composed of compacted fine-grained, clayey soil. Overlying this layer is a 6-inch thick layer of crushed coarse stones designed to facilitate drainage and prevent erosion. A one-foot thick layer of crushed basalt covers the drainage layer. The topmost layer is a vegetation layer composed of native grasses. Annual monitoring of groundwater near the Collins Ranch Disposal site is done to demonstrate the integrity of the cell structure. [5]

Approximately 2.8 billion liters (727 million gallons) of groundwater has been contaminated by materials generated from uranium ore processing at the Lakeview Mill Site, including molybdenum, radium, and arsenic. The contaminated plume covers approximately 116 acres and affects the shallow aquifer in the stream and lakebed strata beneath the mill site. Groundwater outside the contaminated plume is generally poor quality because of naturally occurring hydrothermal processes in the area that produce high mineral content. The DOE is in the process of developing a Programmatic Environmental Impact Statement pertaining to all 24 UMTRCA sites. Options include: (1) no remediation at sites where milling-related contamination of groundwater is not considered a risk to human health; (2) natural cleansing of the milling-related groundwater contamination (natural flushing); (3) active programs to clean groundwater using engineered systems. Once this statement is complete, site-specific National Environmental Policy Act documentation will be developed to propose an appropriate groundwater compliance strategy and reasonable alternatives for the Lakeview Mill Site. [5]

Regulations

Although groundwater in the vicinity of the Lakeview Repository is naturally below drinking water standards, the metrics used in this study are based on drinking water standards set forth by the EPA. The regulatory limits used in this study are for: arsenic (0.01 mg/L) [6], cadmium (0.005) mg/L [6, 7], radium-226 (5 pCi/L) [6, 7], and uranium-238 (0.03 mg/L) [6, 7]. The cumulative dose regulatory limit used in this study is 100 mrem/yr [8].

Model Validation: Simple Deterministic and Probalistic Models

Before probabilistic models of the Lakeview Mill Site were run with the FRAMES 1.5 software, validation models for the FRAMES 1.5 software were created to insure the integrity of the study method. Two types of model validation were done by comparing simple FRAMES 1.5 models of vadose zone transport with analytical solutions of the one-dimensional advection-dispersion equation. The two types of model validation are: (1) one-dimensional deterministic advection-dispersion models, and (2) one-dimensional probabilistic advection-dispersion models.

The analytical solutions are based on the one-dimensional advection-dispersion equation:

$$\frac{D_x \partial^2 C}{R_f \partial x^2} - \frac{\upsilon_p \partial C}{R_f \partial x} = \frac{\partial C}{\partial t} \qquad \text{(Eq. 1)}$$

The solution to this equation solves the contaminant concentration at a specific location (x) over time, when given the longitudinal dispersivity (α_x), the pore velocity (υ_p), the retardation factor (R_f), and the source concentration (C_o) of the system:

$$\frac{C}{C_o} = 0.5 erfc\left[\frac{\left(R_f x - \upsilon_p t\right)}{2\left(\alpha_x \upsilon_p t R_f\right)^{\frac{1}{2}}}\right] \qquad \text{(Eq. 2)}$$

The one-dimensional deterministic analytical models use both arsenic and cadmium as constituents because they do not degrade or decay over time and both are contaminants of concern at the Lakeview Mill Site. Input values are based on data from the Lakeview Mill Site. The one-dimensional analytical models solve Equation 2 using expressions for the parameters based on the MEPAS software formulations [9]. The analytical model assumes a constant contaminant source.

The deterministic FRAMES model used in this study has three modules: a constituent module, a source module, and a vadose zone module. The constituent module defines both arsenic and cadmium as constituents for the model. The majority of the input parameters for the source module and vadose zone module are based on data from the Lakeview Mill Site, but several variables were changed to simplify the models. For example, the source inventories for both constituents were increased significantly to represent a continual contaminant source, and the Darcy infiltration rate through the source was calculated based on infiltration rate through the cover and the hydraulic conductivity of the source. Table I summarizes the input parameters for the deterministic models.

Table I. Input Parameters for One-Dimensional Deterministic Model Validation

Source Module			Vadose Zone		
Depth of clean soil above source	0.9144	m	Percent Sand	20	%
Thickness of waste source	4.8768	m	Percent Silt	20	%
Length of waste source	228.6	m	Percent Clay	60	%
Width of waste source	304.8	m	Percent Organic Matter	0	%
Bulk density of soil at source	1.3	g/cm^3	Percent Iron and Aluminum	0	%
Total porosity of soil at source	50	%	Soil Type Coefficient	11.4	
Moisture content of soil at source	30	%	pH of pore water	7	
Ave. Air temperature	20	C	Total Porosity	45	%
Kd. Arsenic	148	ml/g	Field Capacity	9.1	%
Kd. Cadmium	19.76	ml/g	Hydraulic Conductivity	0.001	cm/s
Water solubility – Arsenic	$3.7*10^4$	mg/L	Thickness of vadose zone	6.096	m
Water solubility – Cadmium	$1.67*10^6$	mg/L	Longitudinal dispersivity	0.06096	m
Inventory – Arsenic	10^{20}	Kg	Bulk density	1.44	g/cm^3
Inventory – Cadmium	10^{20}	Kg	Kd – Arsenic	148	ml/g
Darcy Infiltration Rate	$3.5*10^{-7}$	cm/s	Kd – Cadmium	19.76	ml/g
			Water solubility – Arsenic	$3.7*10^4$	mg/L
			Water solubility – Cadmium	$1.67*10^6$	mg/L

The second type of model validation performed was a one-dimensional probabilistic model that again uses cadmium and arsenic as constituents. The analytical probabilistic models follow the same formulations as the analytical deterministic models. However, both the longitudinal dispersivity (α_x) and the distribution coefficient (Kd) vary with uniform distributions to represent uncertainty in those parameters. The uniform distribution for the longitudinal dispersivity has a maximum value of 10 ft and a minimum value of 0.1 ft. The uniform distribution of the arsenic distribution coefficient ($Kd_{arsenic}$) has a maximum value of 50 ml/g and a minimum value of 0 ml/g. The uniform distribution for the cadmium distribution coefficient ($Kd_{cadmium}$) has a maximum value of 70 ml/g and a minimum value of 10 ml/g.

Latin Hypercube Sampling was applied to each distribution and 100 values were sampled. The sampled values were then applied to the formulations used in the deterministic runs to calculate a range of concentrations at specific times for each constituent. These ranges were used to calculate cumulative distribution functions (CDF) of each contaminant at 1000 years, 1500 years, and 2000 years. All other input values used were the same as used in the deterministic models. Once the CDFs were plotted for each constituent, sensitivity/uncertainty analyses were conducted to determine influences of the stochastic parameters.

The probabilistic FRAMES model has four modules: the constituent module, the source module, the vadose zone module, and a sensitivity/uncertainty module. The first three modules are identical to those used in the deterministic runs. The sensitivity/uncertainty module allows distributions of input parameters to be defined. The distributions defined in this module are the same as those used in the probabilistic analytical model. From the sensitivity/uncertainty module, the concentration of each constituent is output at 1000 years, 1500 years, and 2000 years and CDFs were created to compare with the analytical results.

Once the analytical model validation was complete, probabilistic models for the Lakeview Repository were created in FRAMES 1.3 and FRAMES 1.5. Each model consisted of nine modules with site specific parameters: a constituent module, a source term module, a repository liner module, a vadose zone module, a saturated zone module, an exposure pathway module, a receptor intake module, a health impact module, and a sensitivity/uncertainty module. Each model ran 100 simulations using site-specific distributions for 42 input parameters. For each model, the sensitivity/uncertainty module iterates through 100 runs using the Latin Hypercube Sampling Method to sample distributions of 42 different input parameters. The sensitivity/uncertainty module outputs water concentration peak for each contaminant, as well as the summed radiation peak dose. The CDF for each output parameter was calculated and plotted against current regulatory limits.

Model Validation Results

The deterministic comparisons revealed a problem within the FRAMES software. When cadmium was used as the constituent, the FRAMES model produced a good representation of the concentration curve determined by the analytical model, with only a slight amount of numerical dispersion (Fig. 1a). However, when arsenic was used as the constituent the analytical model showed the system reaching contaminant saturation at 17,000 years while the concentration curve produced by the FRAMES model plateaued at 10,000 years (Fig. 1b). It was discovered that the FRAMES software had a built-in termination time of 10,000 years, and all calculations after this final time were inaccurate. The software has been revised to correct this problem.

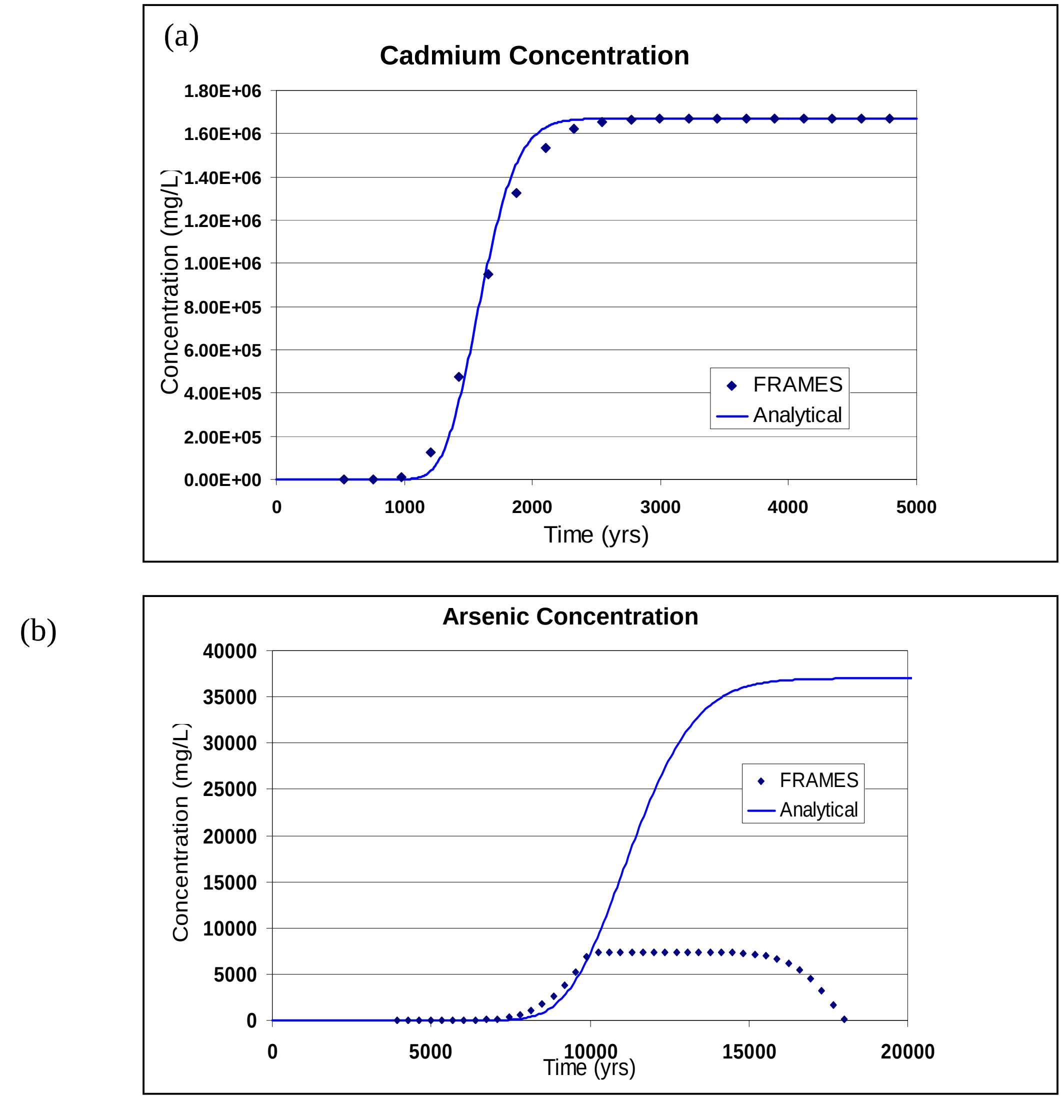


Fig. 1. (a.) Deterministic results for cadmium concentration as a function of time. (b) Deterministic results for arsenic as a function of time; the FRAMES concentration curve plateaus at 10,000 years as a result of a bug in the software. The concentrations are recorded at the exit of the vadose zone (x=6 m).

Once the software was revised, two probabilistic models were created using the different stochastic variables. First, the models were run using only a single stochastic variable, the distribution coefficient (Kd). The second simulation used a varying Kd and a varying longitudinal dispersivity (α). The cumulative probabilities are quite similar between the analytical results and the FRAMES simulations. When both the Kd value and the α value vary, there are minor differences in the CDFs from the two models, but overall the models represent the same trend (Fig 2). When Kd was the only stochastic variable, the concentrations were largely bimodal (either zero or the solubility limit). When the longitudinal dispersivity was added as a stochastic parameter, the simulations resulted in a more uniform concentration distribution as a result of the increased effective dispersion. Results are similar for both constituents; therefore for simplicity in this report, only the cadmium plots are shown (Fig 2).

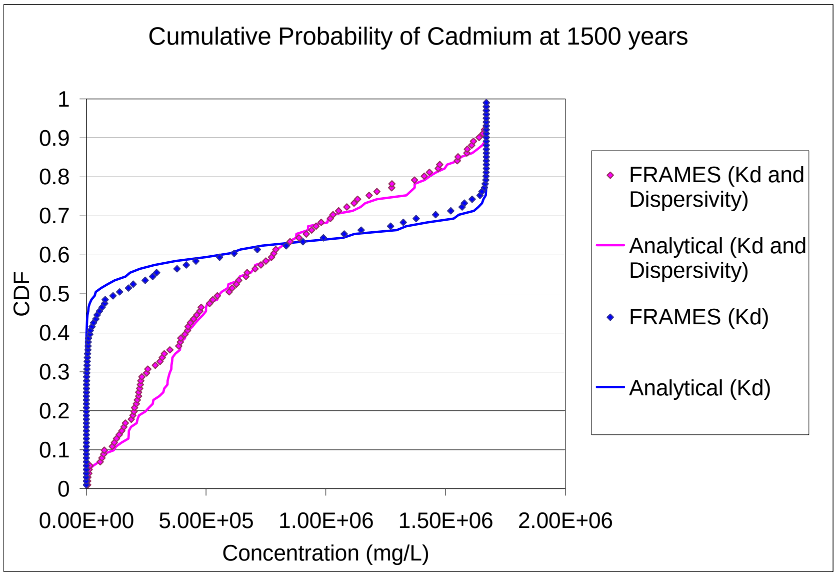


Fig. 2. CDF of cadmium concentration at the exit of the vadose zone (x=6 m).

Sensitivity analyses of percolation through the vadose zone were performed using the stepwise regression method with rank-transformed input and output variables. Both stochastic parameters used in the validation models were influential to the resulting contaminant concentration. The most important variable for both constituents was the constituent Kd value (Table II) (Figure 3c).

Table II. Parameter Sensitivity for Simulated Cadmium Concentration Based on Stepwise Linear-Regression Analysis (One-Dimensional Probabilistic Model)

Step	Variable	R^2	ΔR^2
1	Distribution Coefficient	94.32	94.32
2	Longitudinal Dispersivity	96.52	2.2

Model Validation: LakeView MILL Repository

A final validation study was performed by comparing FRAMES 1.5 models with FRAMES 1.3 models. FRAMES 1.3 has previously been tested on the Monticello Mill Tailings Repository, Utah [3]. The models created for this validation study were more complex then those used in the previous validation studies.

Multiple probabilistic models for the Lakeview Repository with simulations up to 10,000 years were created with the FRAMES 1.5 software at the Pacific Northwest National Laboratory. Two of these models are used as a comparison for models created with FRAMES 1.3. These two models use site-specific input representing present-day conditions and predicted-future conditions at the Lakeview Mill Repository. Each model has nine modules with site specific parameters: a constituent module, a source term module, a repository liner module, a vadose zone module, a saturated zone module, an exposure pathway module, a receptor intake module, a health impact module, and a sensitivity/uncertainty module. Stochastic parameters used in these modules are summarized in Table III. Water percolation through the cover was calculated with the Hydrologic Evaluation of Landfill Performance (HELP) [10] module as a separate aspect of this study prior to model creation.

Table III. Stochastic Parameters Based on Present-Day Conditions Used in the Lakeview Mill Site Repository Models.

Source Term data					
			Distribution Information		
Description	**Value**	**Units**	**Distribution Type**	**Max**	**Min**
Thickness of waste source	16	ft	uniform	32	1
Length of waste source	750	ft	uniform	750	3
Width of waste source	1000	ft	uniform	1000	3
Bulk density of soil at source	1.3	g/cm^3	equation: (1-total porosity)*2.51	N/A	N/A
Total Porosity of soil at source	0.5	fraction	uniform	0.51	0.46
Moisture Content of soil at source	0.3	fraction	uniform	0.38	0.25
Kd - Arsenic	148	ml/g	uniform	229	148
Kd - Cadmium	19.76	ml/g	uniform	162	19.76
Kd - Ra226	100	ml/g	uniform	1000	100
Kd - U234	0.27	ml/g	uniform	10.26	0.27
Kd - U238	0.27	ml/g	uniform	10.26	0.27
Water Solubility - U234	0.2574	mg/L	uniform	23.4	0.2574
Water Solubility - U238	0.2618	mg/L	uniform	23.8	0.2618
Darcy Infiltration rate	5.9	in/yr	uniform	9.50	2.30
Compacted Soil Layer data (Vadose Zone 1)					
			Distribution Information		
Description	**Value**	**Units**	**Distribution Type**	**Max**	**Min**
Field Capacity	0.27	fraction	uniform	0.34	0.2
Hydraulic Conductivity	5.00E-07	cm/sec	uniform	1.00E-06	1.00E-07
Vadose Zone data (Vadose Zone 2)					
			Distribution Information		
Description	**Value**	**Units**	**Distribution Type**	**Max**	**Min**
Field Capacity	0.091	fraction	uniform	0.351	0.091
Hydraulic Conductivity	1.00E-03	cm/s	uniform	1.00E-03	1.00E-07
Thickness of vadose zone	20	ft	uniform	50	20
Bulk Density	0.87	g/cm^3	uniform	1.44	0.87
Saturated Zone data					
			Distribution Information		
Description	**Value**	**Units**	**Distribution Type**	**Max**	**Min**
Darcy velocity	10.00	ft/yr	uniform	20.00	10.00
Longitudinal travel distance to well	31	ft	equation: dist = ((0.5)*source length)+1	N/A	N/A
Longitudinal Dispersivity	3.1	ft	equation: travel distance*0.1	N/A	N/A
Lateral Dispersivity	1.023	ft	equation: travel distance*0.033	N/A	N/A
Vertical Dispersivity	0.00775	ft	equation: travel distance*0.00025	N/A	N/A

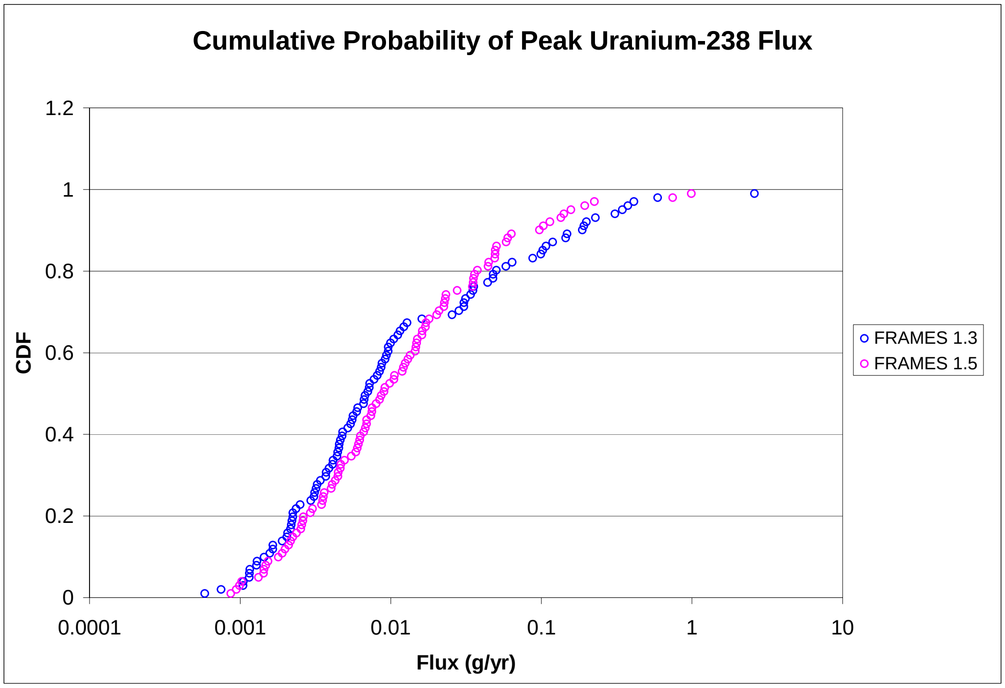
For each model, the sensitivity/uncertainty module iterates through 100 runs using the Latin Hypercube Sampling Method to sample distributions of 42 different input parameters. The sensitivity/uncertainty module outputs water concentration peak for each contaminant, as well as the summed radiation peak dose. The CDFs created with the two software versions for the summed radiation peak dose and constituent concentration in the aquifer are plotted. Finally, input parameter importance is determined by a step-wise regression. Although this statistical analysis is not used directly in comparing the two software versions of FRAMES, it demonstrates the effectiveness of the FRAMES software to produce site-specific models.

Lakeview Repository Model Validation Results

Although the CDFs for each constituent were created for comparison in this study, only the plots for uranium-238 are presented for illustration (Fig. 3a). Plots of the summed radiation dose are also presented in this report (Fig. 4a). The statistical analyses results for all constituents are presented in Table IV.

The models using different versions of FRAMES yield very similar results (Fig. 3a). Minor differences are expected do to the complexity of the overall model and improvements in the newer software. Results for future conditions are very similar to present-day conditions. The statistical analyses indicate the width of the source zone is the most influential parameter on uranium-238 concentration in the aquifer for both present and future conditions. The distribution coefficient for uranium-238 in the source is the next most influential parameter on the system followed by the length and thickness of the source zone. Other parameters have only minor influences in the system (Fig. 3b).

(a)



(b)

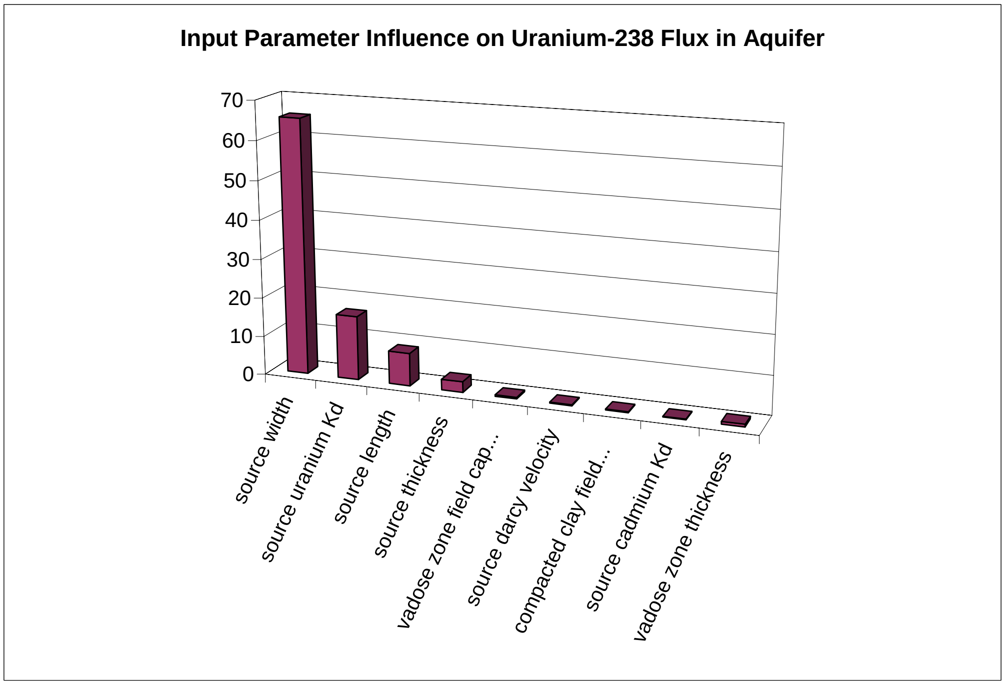


Fig. 3. (a) Comparison of CDFs for uranium-238 flux in the aquifer using FRAMES 1.3 and FRAMES 1.5. (b) Input parameter importance on flux of uranium-238 in the aquifer using both present-day and future conditions. Flux was recorded approximately 115 meters below the source zone.

The most influential parameters on all constituent concentrations for present conditions are summarized in Table IV.

Table IV. Summary of Parameters Important to the Contaminant Concentration in the Aquifer for Present Day Conditions

	Arsenic	**Cadmium**	**Radium-226**	**Uranium-238**
Parameter	**ΔR^2**	**ΔR^2**	**ΔR^2**	**ΔR^2**
source arsenic Kd	6.3	0.35	2.3	0
source cadmium Kd	0	55.87	0	0.23
source radium Kd	0.81	0	30.24	0
source uranium Kd	0.91	0	0	16.42
source length	1.8	0	0	8.43
source thickness	0	0	0	2.72
source width	0	0.73	0	65.45
source bulk density	0	0	0	0.69
source Darcy velocity	6.65	4.63	0	0
source total porosity	0	0	0	0.16
Compacted clay hydraulic conductivity	27.08	6.62	3.52	0
Compacted clay field capacity	0	0.46	0	0.21
vadose zone bulk density	5.97	0	2	0
vadose zone field capacity	0	0	0	0.4
vadose zone hydraulic conductivity	0	1.54	1.42	0
vadose zone thickness	27.6	12.8	5.93	0
aquifer travel distance	0	2.67	0	0
Total	**77.12**	**85.67**	**45.41**	**94.71**

Regarding the summed radiation dose, the models using different versions of FRAMES yielded very similar results (Fig. 4a). The statistical analyses indicate the source width is the most influential parameter for both the present and future conditions, followed by the distribution coefficient of uranium-238 in the source and the source length and thickness (Fig. 4b). The other parameters have only minor influences on the system (Table V).

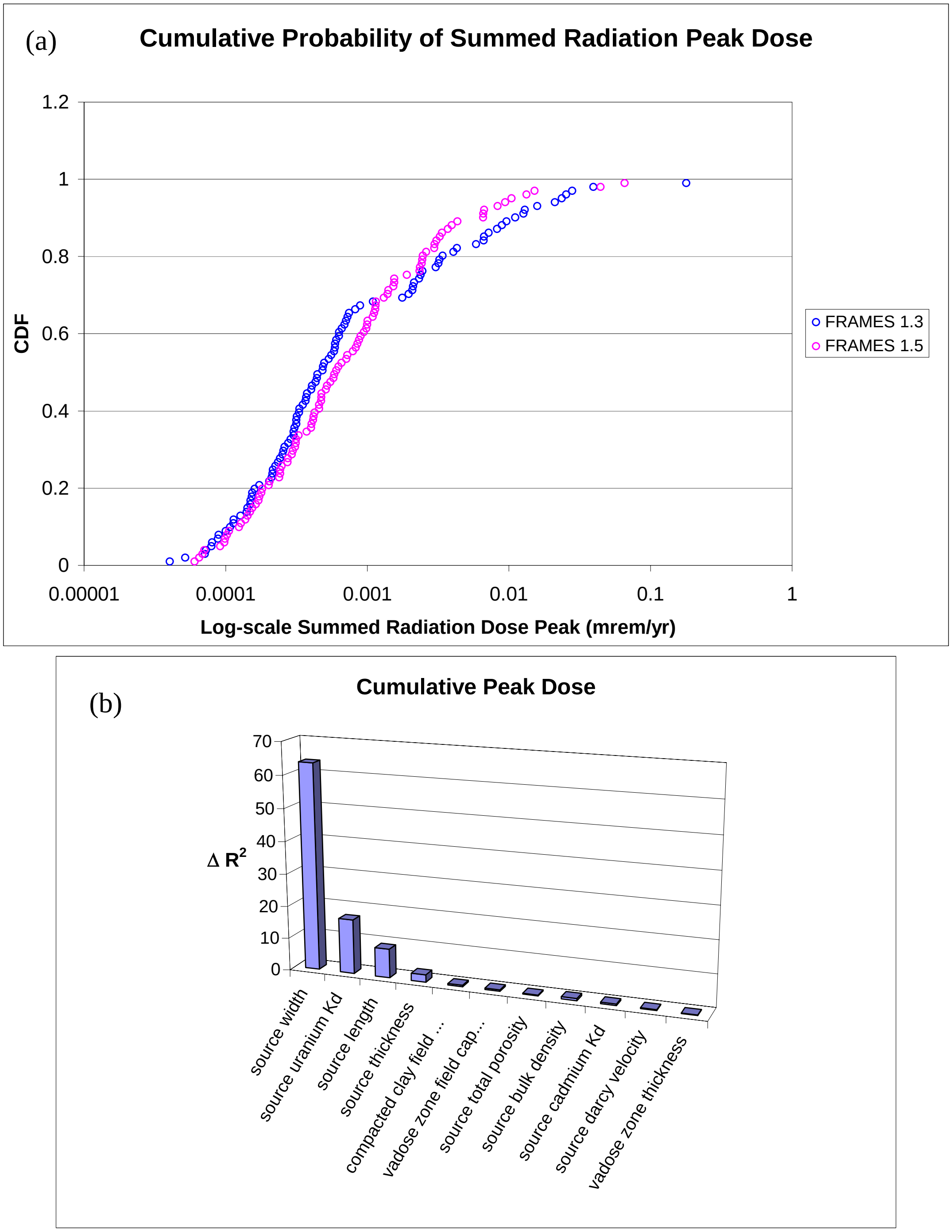


Fig. 4. (a) Comparison of FRAMES 1.5 and FRAMES 1.3 summed radiation dose. (b) Input parameter influence on summed radiation dose for present-day conditions. Peak dose was recorded approximately 115 meters below the source zone.

Table V. Summary of Parameters Important to Simulated Cumulative Peak Dose Based on Stepwise Linear-Regression Analysis

Variable	**Δ R^2**
	63.63
source uranium Kd	16.8
source length	8.93
source thickness	2.29
compacted clay field capacity	0.44
vadose zone field capacity	0.44
source total porosity	0.28
source bulk density	0.66
source cadmium Kd	0.48
source Darcy velocity	0.26
vadose zone thickness	0.18
vadose zone hydraulic conductivity	0
Total	**94.39**

CONCLUSIONS

Probabilistic risk-based performance assessment of landfills is important in order to address site-specific influences that may contribute to groundwater contamination and human exposure. This study provided a detailed evaluation of a software tool (FRAMES) that can be used to created probabilistic risk-based models. By comparing results from simplistic FRAMES models with analytical solutions of one-dimensional contaminant transport, a major bug within the FRAMES software was detected and corrected. Probabilistic analytical models were also compared to the FRAMES stochastic results, and good comparisons were found. The new version of FRAMES (v. 1.5) was also compared to an older version of FRAMES (v. 1.3) to ensure that there were no significant differences or errors. As the software tool improves and new versions become available, it is essential to make sure these newer versions maintain the integrity of the models. The Lakeview Repository was used as a benchmark in these comparisons. A detailed report of risk-based probabilistic simulations of the performance of that site using the revised FRAMES software will be forthcoming.

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