WM'01 Conference, February 25-March 1, 2001, Tucson, AZ

PERFORMANCE ASSESSMENT OF CLASS A LOW LEVEL RADIOACTIVE WASTE DISPOSAL USING THE DISCRETE-DISPERSED SOURCE METHOD FOR FATE AND TRANSPORT IN GROUNDWATER

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ABSTRACT

Disposal of Low Level Radioactive Waste (LLRW) in a proposed new cell at the Envirocare of Utah disposal facility requires demonstration that ground water protection levels can be met for a period of 500 years. To demonstrate compliance, the leaching and transport of Class A nuclides have been evaluated using the PATHRAE-RAD Performance Assessment Code for the Land Disposal of Radioactive Wastes (Merrell, et. al, 1995).

For the Envirocare demonstration, a unique method has been developed to overcome the limitations of the closed form analytical solution used by PATHRAE, which is not capable of incorporating two different dispersive components simultaneously. To overcome this limitation, transport is modeled in two steps. First, vertical dispersion in the unsaturated zone is simulated using the code algorithms for saturated dispersive transport. Then a new technique, referred to as the discrete-dispersed source method, is employed to decompose the results of the vertical dispersive solution into a number of starting concentrations and unsaturated zone lengths, which are used as input to the horizontal (saturated zone) simulations. The horizontal (saturated zone) model is run for the entire concentration array, and the results of numerous simulations are summed using superposition to determine the concentrations at the compliance point over time. This technique fully accounts for dispersion of nuclides that potentially leach from the disposal cell, migrate vertically to the water table, and move horizontally toward a compliance well.

The performance assessment includes an evaluation of all Class A nuclides and their potential impact to groundwater. The critical factors for achieving compliance include cell design (which affects infiltration through the cell after closure) and waste characteristics. Key components of the disposal cell design include slope length, angle, layer thickness, and hydraulic properties of cover materials. Significant waste characteristics include waste density, source concentration, half life, and sorption coefficient (K_d) of each nuclide. Sensitivity analyses have been performed for these critical parameters.

The combination of new and existing methods to assess the performance of the LLRW disposal cell results in a successful demonstration of compliance with ground water protection levels for 500 years. All of the 261 Class A nuclides evaluated will be acceptable for disposal in the cell. 257 of the nuclides will be acceptable at the Class A limits or at the maximum theoretical concentration (specific activity). The methods used to achieve these results can be applied to assess the performance of other disposal facilities.

INTRODUCTION

Disposal of Low Level Radioactive Waste (LLRW) in a proposed new cell at the Envirocare of Utah disposal facility requires demonstration that ground water protection levels can be met for a period of 500 years. To demonstrate compliance, the leaching and transport of Class A nuclides have been evaluated using the PATHRAE-RAD Performance Assessment Code for the Land Disposal of Radioactive Wastes (Merrell, et. al, 1995). This performance assessment code was developed from the PRESTO-EPA and PATHRAE-EPA family of codes, and can be used to calculate the maximum annual effective dose equivalents or concentrations resulting from the land disposal of radioactive wastes.

For the Envirocare LLRW performance assessment, the groundwater-to-river pathway module is used to determine groundwater concentrations a compliance well located 90 feet from the edge of the waste. Although the physical situation involves only transport in groundwater (including the saturated and unsaturated zones), the groundwater-to-river pathway module is used because it calculates an undiluted concentration at the compliance well when the user sets the river flow rate equal to the infiltration rate through the disposal cell. PATHRAE-RAD solves for radionuclide transport in groundwater using the following equation (Merrell, et. al, 1995):

$$C = \frac{Q \cdot e^{(-l \cdot (\mathrm{tv}+\mathrm{twc}))} \cdot l_L \cdot f_o}{q_w} \qquad \text{(Eq. 1)}$$

where C = nuclide concentration in water at user-specified output location (Ci/m^3)
Q = inventory of the isotope available in a given year (pCi)
l = radioactive decay constant (1/yr)
q_w = infiltration rate through the cell (or flow rate in the river) (m^3/yr)
f_o = fraction of inventory arriving at the well (river) through the aquifer
t_v = vertical travel time of contaminants to the aquifer (yr)
t_{wc} = waste container lifetime (yr)
l_L = fraction of each nuclide leached from the inventory in a year (1/yr)

The term f_o can be calculated for dispersive or non-dispersive transport in groundwater. The fraction of inventory arriving at the well (or river) is obtained by solving the partial differential equation and factoring out the effect of radioactive decay. The radioactive decay term is included implicitly in the nuclide inventory (Q). For dispersive groundwater transport, f_o is calculated using a band dispersive model, and is given by:

$$f_o = \frac{1}{N}\sum_{\mathrm{j=o}}^{N-1}\left[\mathrm{F_j}(\mathrm{t_j}) - \mathrm{F_j}\left(\mathrm{t_j} - \frac{1}{l_L}\right)\right] \qquad \text{(Eq. 2)}$$

where N = number of mesh points in numerical integration
$F_j(t)$ = $0.5U(t)[\mathrm{erfc}\,(z-) + e^{dj}\,\mathrm{erfc}\,(z+)]$
$U(t)$ = unit step function
$z\pm$ = $\{(d_j)^{0.5}[1 \pm t/(Rt_{wj})]\}/\{2(t/Rt_{wj})^{0.5}\}$
t_j = $t - t_v - t_{wc} + t_{op} - (j+1/2)\, t_{op} / N$

t = time from facility closure (yr)
t_{op} = time of operation of facility (yr)
d_j = distance from center to compliance point, divided by longitudinal dispersivity
t_{wj} = water travel time from center to compliance point (yr)

The analytical solution accounts for dispersion in the horizontal groundwater pathway from the cell to the compliance point. However, vertical dispersion, which occurs as leachate migrates from the base of the cell to the water table, is neglected in the basic PATHRAE-RAD code.

Dispersion causes a portion of the nuclide mass to migrate faster than the advective flow of groundwater, in both the vertical transport path to the water table and the horizontal path to the compliance well. State regulators and other parties were concerned that vertical dispersion could cause constituents to exceed groundwater protection levels within 500 years, when those constituents might not fail using horizontal dispersion alone. Since the PATHRAE code has been used previously for performance assessment at the Envirocare site, it is desirable to use the same program without significant recoding and recompilation that might alter the fundamental code.

DISCRETE DISPERSED SOURCE METHOD

For the Envirocare performance assessment modeling, a unique method has been developed to overcome the limitations of the closed form analytical solution used by PATHRAE-RAD, which is incapable of simultaneously incorporating vertical (unsaturated) and horizontal (saturated) dispersion along the flow path. To overcome this limitation, the code has been recompiled to allow up to 256 output times (instead of the original limit of 10 output times) and a procedure has been developed to model the vertical and horizontal pathways separately. The vertical path from the base of the waste to the top of the aquifer is run first, and vertical dispersion is included by setting the saturated aquifer parameters equal to the vadose zone properties and specifying a value for dispersivity. As such, the model applies dispersion to the vertical path and produces a time and concentration array for the arrival of nuclides at the water table.

In this performance assessment, the vertical model produces a concentration array for 100 radionuclides and 116 output times. The concentration at the water table provides the basis for the source term input to the horizontal model. The arrival time at the water table is converted to a distance by which to shorten (or lengthen) the vertical path as a result of positive (or negative) dispersion and retardation (Figure 1).

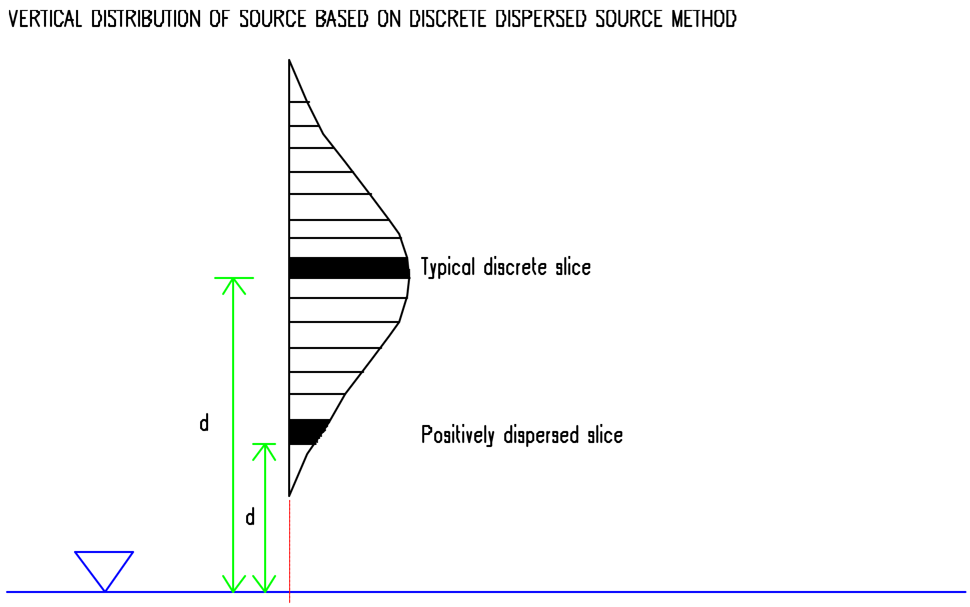

Fig. 1. Vertical Distribution of Source Term Based on the Discrete Dispersed Source Method

The concentration source term for the horizontal model is calculated from the output from the vertical model by calculating the mass (or activity) in each of the 116 "slices" using the following equation:

$$M = \frac{(C_t + C_{t+n})}{2} \cdot ((t+n) - t) \cdot (q_{in}) \cdot A \qquad \text{(Eq. 3)}$$

where:
- M = Mass (or activity) of nuclide in given time slice [Ci]
- C_t = Output concentration at time t [Ci/m^3]
- C_{t+n} = Output concentration at time t+n [Ci/m^3]
- t = Time at beginning of "time slice" [years]
- $t+n$ = Time at end of "time slice" [years]
- n = Duration of "time slice" [years]
- q_{in} = Infiltration rate [m/yr]
- A = Unit area over which infiltration occurs [1 m^2]

The modeling is performed on a concentration basis by using a unit (1 m^3) source. Making the assumption that the entire mass (or activity) is ascribed to one cubic meter of aquifer soil, the mass (or activity) is converted from Ci to Ci/m^3, as the source term for the horizontal modeling.

The horizontal model is then run 116 times, using the calculated initial concentration and distance for each time "slice". The resulting concentrations at the compliance point are summed (using superposition), to determine total concentration, which is then compared to the groundwater protection levels established for the LLRW cell monitoring wells.

FACTORS AFFECTING PERFORMANCE

The performance assessment includes an evaluation of all Class A nuclides and their potential impact to groundwater. Input parameters affecting the PATHRAE-RAD model results include:

- Infiltration Rate – Infiltration through the cell is modeled using the EPA HELP model (Schroeder and Peyton, 1995). Infiltration rate is one of the most significant factors affecting cell performance, because it directly affects radionuclide leach rates and vadose zone velocities. Higher infiltration rates result in faster radionuclide travel times and greater likelihood of exceeding aquifer protection levels. Cell cover design (including slope angle, slope length, and permeability of the engineered cover) and climatic factors have considerable effect on infiltration rates.
- Vadose Zone Velocity – The aquifer velocity in the vertical path is calculated according to the equation for average linear velocity in the vadose zone (Stephens, 1996):

  $$v = q / \theta_e \qquad \text{(Eq. 4)}$$

  where v = average linear velocity (L/T)
  q = infiltration rate (L/T)
  θ_e = effective water content that participates in carrying the flow (L^3/L^3)

  The infiltration rate (q) used in this equation is determined using the EPA HELP3 model. Moisture contents (θ_e) are determined for each vadose zone material in the profile, using the Pacific Northwest Laboratories UNSAT-H model (Fayer and Jones, 1990; Fayer, 1999). The moisture content (θ_e) of the entire vertical profile is calculated as a thickness-weighted average of the clay liner and natural soils below (the equivalent porous media for the materials underlying the waste), and is calculated at 8.1%. Taken alone, lower moisture contents result in higher vadose zone velocities. However, since moisture content is inversely dependent on infiltration rates, better performance (lower vadose zone velocity) is generally associated with lower infiltration rates and lower moisture contents in the subsurface.

- Saturated Aquifer Moisture Content – In the horizontal model, the aquifer moisture content is set equal to the saturated porosity of 29%. This variable has no significant effect on performance, within the reasonable range of values for saturated aquifer porosity at the Envirocare site.
- Waste Source Term Concentrations – The waste source term concentrations for 261 Class A nuclides have been developed from data supplied by the Manifest Information Management System (MIMS) or by specific activity. The MIMS database is managed by the Department of Energy (DOE), and contains a summary of national low-level radioactive waste disposal information. The maximum Class A concentrations from the MIMS database are used in the modeling. For Class A radioisotopes not listed in the MIMS database, the waste source term in the model is set at the specific activity, which is calculated using the following formula:

$$SA = \frac{\ln(2)}{t_{1/2}}\left(6.02x10^{23}\frac{molecules}{Mole}\right)\left(\frac{1}{GMW_{g/m}}\right)\left(3.7x10^{-2}\right) \quad \text{(Eq. 5)}$$

where SA = Specific activity in pCi/g
$t_{1/2}$ = Half life in seconds
GMW= Gram molecular weight in grams per mole

Depending on the partitioning coefficient (K_d) of a nuclide, the source term concentration may have a significant effect on performance of the disposal cell. Nuclides having high K_d values may be accepted in essentially any concentration.

- Waste Bulk Density – The average density of Class A waste disposed has been determined by data supplied by MIMS. The total volume (cubic feet) of waste by class and the total weight (pounds) by class have been calculated from disposal manifests for waste disposed from 1986 to present. The calculated density of the Class A waste is 1.11 g/cm^3. Waste bulk density had a slight effect on cell performance.
- Partitioning Coefficients (K_ds) – The partitioning coefficients (a.k.a. distribution coefficients, or K_ds) are set at the most conservative (lowest) K_d values found in the literature (Sheppard and Thibault, 1990; Looney et. al, 1987; Baes, et. al, 1984) for all nuclides except those having site-specific values(1) for the Envirocare site. The performance assessment modeling preferentially uses 1) site-specific K_d values, 2) the lowest measured soil K_d values published in the literature, and 3) published K_d values calculated from the soil:plant ratio. K_d values significantly affect the model results. Nuclides having high K_d values are more likely to meet the performance criteria (groundwater protection levels at the compliance well.)
- Fractional Release Rate – The annual fractional release rate (or "leach rate") for the vertical model is calculated using the following equation (Kozak 1990):

$$L = \frac{q_{in}}{d\theta\left(1+\frac{\rho K_d}{\theta}\right)} \quad \text{(Eq. 6)}$$

where L = fractional annual contaminant release rate (yr^{-1})
q_{in} = water infiltration rate (m/yr)
θ = volumetric moisture content of waste
d = waste layer thickness (meters)
ρ = waste density (g/cm^3)
K_d = waste distribution coefficient (ml/g)

This method of determining the leachate concentration is environmentally-conservative for several reasons. First, PATHRAE assumes that the release rate is constant throughout time. The constituent is leached from the waste at a constant rate, until the initial source concentration is totally mobilized. In reality, the leach rate will decrease as the source concentration decreases. Second, the use of K_d to determine contaminant release rates assumes that all of the constituent is adsorbed and will eventually be completely desorbed (or leached out) by percolating water. In reality, some of the constituent may occur in the refractory phase, which would render it less mobile.

The contaminant release rate for the horizontal simulation is set to 1/yr for all constituents modeled. In this manner, the entire waste concentration in each "time slice" is released "instantaneously". The K_d-limited leach rate is already accounted for in the vertical simulation and the resulting time offset for the "time slices" which are input to the horizontal model.

Fractional release rates are affected by infiltration rate and K_d values, which have already been identified as significant to cell performance. Part of their significance is due to the effect on fractional release rate. Lower release rates contribute to a greater likelihood of meeting performance criteria.

- Vertical Transport Distance – The vertical path length is set to the distance from the bottom of the waste to the aquifer, including the 2-foot thick clay liner and excluding the capillary fringe, in this case. The thickness of the capillary fringe is determined using the UNSAT-H model, and is primarily affected by the soil properties near the water table. A greater vertical transport distance contributes to better cell performance.
- Horizontal Transport Distance – The horizontal path length is set to the distance from the edge of the waste to the nearest compliance well, a distance of 90 feet (27.4 m). Horizontal distance is not considered a variable for sensitivity analysis in the Envirocare modeling, due to regulatory constraints. However, transport distance would significantly affect the modeling results; greater distances between the cell and the compliance monitoring well would result in better performance.
- Dispersivity – The longitudinal dispersivity is set to 10 cm in the vertical pathway, and 10% of the transport distance in the horizontal pathway, in this performance assessment. Within a reasonable range of values, dispersivity has a moderate affect on the model results.
- River Flow Rate – The river flow rate in both the vertical and horizontal modeling are set equal to the infiltration rate, in order to prevent any dilution of concentrations in the aquifer. This extremely conservative assumption has a significant affect on model results. Natural mixing in the aquifer and at the compliance well would result in decreased nuclide concentrations in groundwater, and therefore better cell performance.
- Aquifer Average Linear Velocity – The aquifer velocity in the horizontal path is calculated based on the Darcy equation, such that:

$$\bar{v} = \frac{Ki}{n_e} \qquad \text{(Eq. 7)}$$

where
$\bar{v}$ = average linear velocity in the aquifer (L/T)
K = hydraulic conductivity (L/T)
i = hydraulic gradient (L/L)
n_e = aquifer effective porosity (L^3/L^3)

Aquifer velocity significantly affects the cell performance.

- Hydraulic Conductivity – The saturated hydraulic conductivity have been determined from slug tests performed in 96 wells at the site. The 90% upper confidence level (UCL) about the geometric mean (7.7×10^{-4} cm/sec) is used. The effects are significant, since hydraulic conductivity affects the saturated aquifer velocity.
- Hydraulic Gradient – The hydraulic gradient has been calculated from water level measurements from 20 wells located near the LLRW disposal cell. Its affect on performance is comparable to the effects of hydraulic conductivity, as these factors are equally weighted in the calculation of saturated aquifer velocity.

In summary, the critical factors for achieving compliance include cell design (which affects infiltration through the cell after closure) and waste characteristics. Key components of the disposal cell design include slope length, angle, layer thickness, and hydraulic properties of cover materials. Significant waste characteristics include waste density, source concentration, half life, and sorption coefficient (K_d) of each nuclide. Sensitivity analyses have been performed for these critical parameters.

PERFORMANCE ASSESSMENT RESULTS

The results of the performance assessment for the proposed new cell at the Envirocare facility indicate that all of the 261 Class A nuclides evaluated will be acceptable for disposal in the cell. 257 of the nuclides will be acceptable at the Class A limits or at the maximum theoretical concentration (specific activity), and the remaining four nuclides (Bk-247, Ca-41, Cf-249, and Cl-36) will be acceptable in limited concentrations (Table I).

Table I. Nuclide Concentrations Acceptable for Disposal

		PATHRAE-RAD MODEL RESULTS Side Slope, 0.28 cm/yr Infiltration	
ELEMENT	NUCLIDE	Concentration (pCi/gm)	Concentration (Ci/m3)
Berkelium	Bk-247	0.000159	1.76E-10
Calcium	Ca-41	527.8	5.86E-04
Californium	Cf-249	0.000296	3.28E-10
Chlorine	Cl-36	0.44426	4.93E-07
257 Other Nuclides		Class A Limit or Specific Activity	Class A Limit or Specific Activity

The arrival of four constituents at the water table at the water table are shown in Figure 2. These example nuclides were modeled using low K_d values, and therefore exhibit early arrival times at the water table. Modifications to the PATHRAE-RAD code to allow greater than 10 output times allows detailed definition of the time and concentration curves. Each concentration point shown in Figure 1 serves as input to the subsequent horizontal modeling.

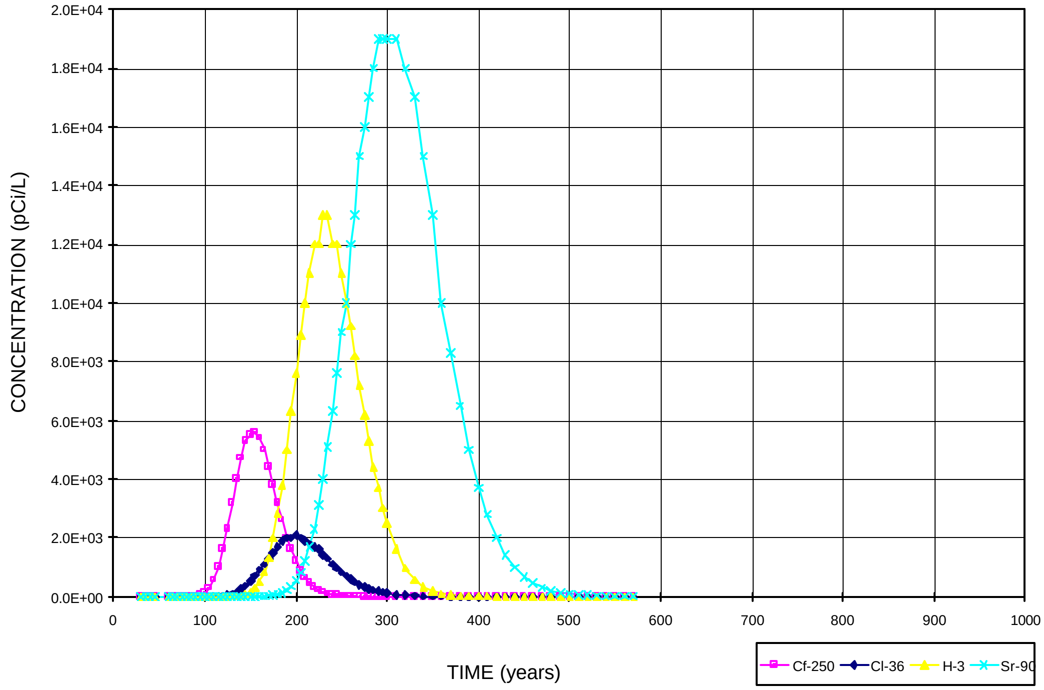

Fig. 2. Nuclide Concentrations at the Water Table (Vertical Model Results)

The arrival of selected constituents at the compliance well is shown in Figure 2. Dispersion, retardation, and radioactive decay have reduced the peak concentrations. Chlorine-36, with the lowest K_d and one of the longer half lives, arrives with the highest concentration among the early-arriving nuclides. The model results for these nuclides are shown on a logarithmic scale in Figure 3.

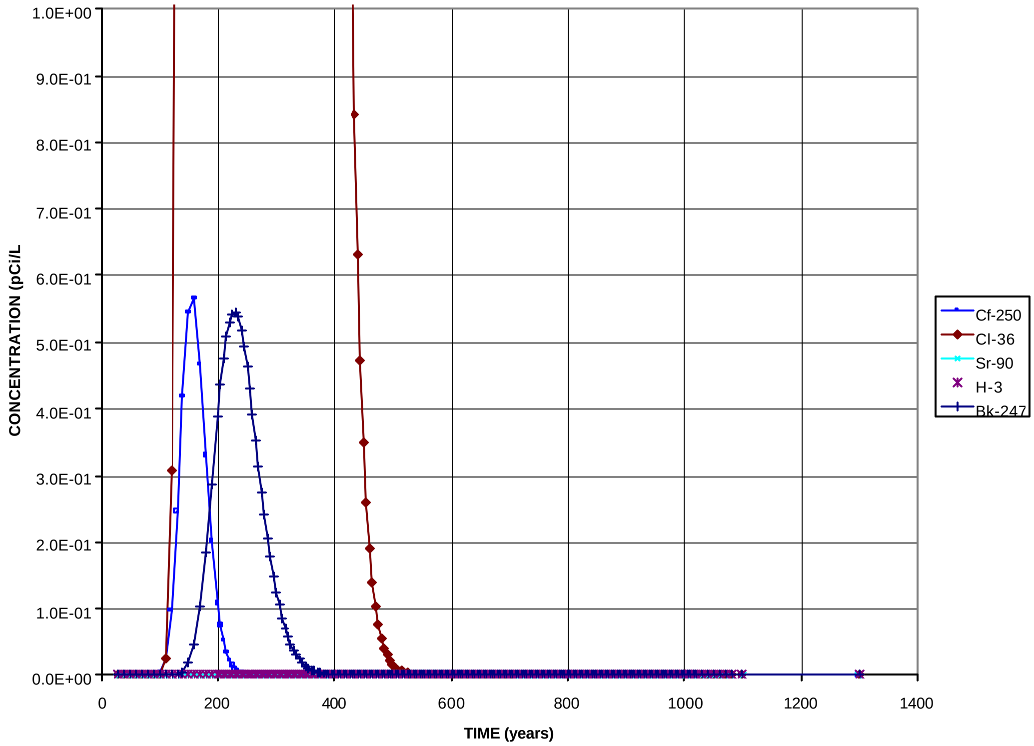

Fig. 3. Nuclide Concentrations at the Compliance Well (Horizontal Model Results)

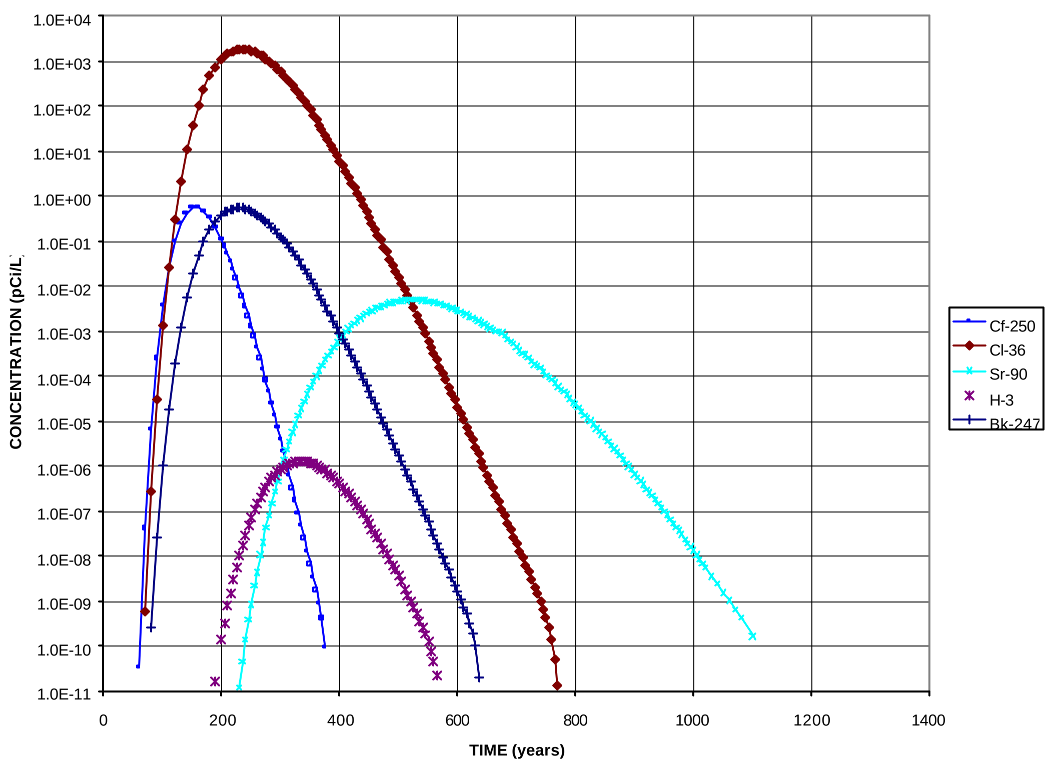

Fig. 4. Nuclide Concentrations at the Compliance Well (Horizontal Model Results), Logarithmic Scale

SUMMARY AND SIGNIFICANCE

The combination of new and existing methods to assess the performance of the Envirocare Class A LLRW disposal cell results in a successful demonstration of compliance with ground water protection levels for 500 years. All of the 261 Class A nuclides evaluated will be acceptable for disposal in the cell. 257 of the nuclides will be acceptable at the Class A limits or at the maximum theoretical concentration (specific activity). The methods used to achieve these results can be applied to assess the performance of other disposal facilities.

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FOOTNOTES

(1) Site-specific K_d values were available for Cs, Co, C, I, Np, Tc, and U