A COMPARISON STUDY OF DIFFERENT SOURCE MODELS FOR HIGH-LEVEL WASTE

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ABSTRACT

The release rate of radionuclides from the engineered facility of a repository can be described with a source model. This paper presents a comparison of the effects of different source models on the performance of a repository. A constant, an exponential and a time- and temperature-dependent leach rate model were analyzed for a basalt repository system. Integrated discharges at the accessible environment were calculated for each model and normalized by the release limits prescribed by EPA. Results were obtained for each of the three models with and without radionuclide solubility-limit constraints. Analyses were also performed for cases where backfilled regions in the repository were modeled as a "mixing cell" where leached materials are mixed uniformly. Results from these analyses indicated that (1) the difference in the repository performance was minor between the constant and the exponential leach models, (2) the leach rate averaged over time appears to adequately simulate the more detailed time-dependent leaching behavior, and (3) the use of a mixing cell model has a significant impact on the far-field consequences.

INTRODUCTION

The leaching behavior of waste forms has been under extensive study in recent years. It is generally concluded that leach rate is a complex function of temperature, time, groundwater chemistry and groundwater flow rate. It is, therefore, reasonable to expect that the leach rate of radionuclides from waste forms in a repository would be time-dependent. In fact, a cyclic leaching pattern for glass waste form has been reported in the literature[1,2]. A number of modeling efforts have been made in estimating the potential releases of the radionuclides from high-level waste (HLW) repositories[3,4,5]. The results of these analyses strongly indicate that the source term, i.e., the release rate at the engineered facility, is a very important parameter. In previous analyses, canisters have typically been assumed to fail simultaneously at the end of a specified lifetime, and the leach rate of the waste form has been assumed to be a constant fraction of the initial inventory. The Environmental Protection Agency (EPA) has published a report on the assessment of long-term repository performance[6]. In EPA's analysis it was assumed that the leach rate is a constant fraction of the inventory "remaining" at any given instant. Recently, Chu, et al.[7] performed an analysis using an empirical time- and temperature-dependent model for leach rate of radionuclides, in addition to considering a time-dependent canister failure rate. In that analysis, however, the effect of geochemical retardation in the far-field and the solubility limits of radionuclides were not considered in the calculation of radionuclide releases to the accessible environment.

EPA has issued a draft Standard (40 CFR 191)[8] which specifies permissible limits for cumulative releases of radionuclides from a HLW repository to the accessible environment. It is the purpose of this paper to examine the importance of the "time-varying" leaching behavior on compliance with the EPA Standard by comparing cumulative releases of radionuclides from several different leach models that have different time-dependence behaviors yet similar average leach rates.

ANALYSES

The three leach models used for this study are the following:

A. A constant leach rate model where the leach rate of the mass of radionuclides is a constant fraction (λ_L) of the initial mass (M_o):

$$dM/dt = \lambda_L M_o \qquad (1)$$

Here, canisters are assumed to fail simultaneously.

B. An exponential leach rate model, similar to that used by EPA, where the leach rate of

mass at time t is a constant fraction (λ_L) of the remaining mass (M(t)):

$$dM/dt = \lambda_L M(t) = \lambda_L M_o e^{-\lambda_L t} \quad (2)$$

Here, canisters are again assumed to fail simultaneously.

C. An empirically defined leach rate model that is both time- and temperature-dependent[7,9]:

$$dM/dt = nK(T)t^{n-1} \quad (3)$$

where n = constant ($o<n<1$) that is characteristic of the reaction mechanism
K = temperature-dependent rate constant with the following empirical expression log K(T) = 3.18-2424/T

T = absolute temperature (Kelvin)

t = time (days) elapsed since onset of leaching

In addition, the canister failure rate is defined by a Weibull distribution resulting from penetration of the canister wall by corrosion[7] (Fig. 1):

$$f(t) = a\,\delta(t) + B\,\lambda\,e^{-\lambda t} + c\,\frac{m}{t}\left[\frac{\mu\Gamma(1+1/m)}{t}\right]^m * e^{-[\mu/t\,\Gamma(1+1/m)]^m} \quad (4)$$

where

δ = Dirac delta function
λ = rate constant for early failures from initial defects
μ = canister design life
t = time
m = Weibull modulus, a distribution shape parameter that governs the relative standard deviation
f(t)dt = fraction of canisters which fail between t and t+dt

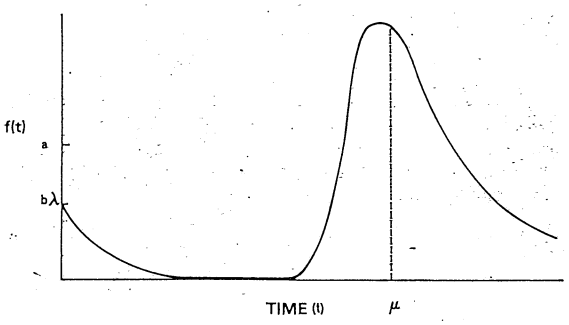

Fig. 1 Canister Failure Rate (f) as a function of time. μ is the design life of the canister.

The first term on the right hand side of the equation represents initial failures (i.e., canisters which have failed prior to repository closure). The second term represents early failures which result from defective canisters. The third term represents failure resulting from penetration of the canister wall by a single corrosion pit.

We require that

$$\int f(t)dt = 1$$

which gives

$$a+b+c = 1$$

The capability of Model C is very versatile. It can be used to simulate many types of leach rate behavior by adjusting the parameters in Eq. (3). In this analysis, a cyclic leach rate was simulated by using Model C. Cyclic leaching behavior has been observed for glass waste forms in laboratory experiments, and is thought to be due to the formation and peeling of siliceous gel layers.

To assess the impact of these three source models on compliance with the Draft EPA Standard, consequence is presented in terms of a "Release Ratio" (RR). The ratio is defined as

$$RR = \sum_i Q_i/(RL)_i \quad (5)$$

where, Q_i is the integrated discharge of radionuclide i to the accessible environment over 10,000 years and (RL) is the release limit allowed for radionuclide i from 1,000 Metric Tons Heavy Metal (MTHM) as specified in the EPA Draft Standard. For releases occurring with a probability greater than or equal to 0.1 in 10,000 years, compliance with the EPA Draft Standard requires that RR be less than unity for each 1,000 MTHM.

Sandia National Laboratories has developed a radionuclide transport code (NWFT/DVM)[10] which incorporates decay/production of radionuclide chains and geochemical retardation. The NWFT/DVM code assumes a constant leach rate of the waste form; it also allows the backfilled rooms in the repository to be modeled as a "mixing cell" where leached materials are mixed uniformly[3]. Solubility limits may also be defined, if desired. For the present study, source models B and C have been added to NWFT/DVM. Radionuclide releases to the accessible environment can therefore be calculated with NWFT/DVM using one of the three source models above.

For this study, analyses were performed for a repository in basalt[3]. The inventory assumed consists of 46,800 MTHM[3]. The accessible environment was assumed to be located one mile from the repository in an overlying aquifer. A 1400-year groundwater travel time from the repository to the accessible environment was used. This groundwater travel time is the mean value used for the fractured-dense-basalt scenario in an earlier analysis[3]. Geochemical retardation factors and solubility limits used for each radionuclide are given in Table I[11]. A dispersivity of 50 ft. was assumed in the far field for all calculations. In addition, a dispersivity of 500 feet was assumed in one of the analyses using the cyclic leach rate model.

To be consistent with the requirements in 10 CFR 60[12] all canisters were assumed to fail at 300 years for source Models A and B, and a mean canister life of 300 years was used for Model C. The temperature history in the very-near-field[13] used to define the temperature-dependent leach rate in Model C is given in Table II. Other parameters from Eqs. (3) and (4) are given in Table III.

TABLE I

Geochemical Parameters

Radionuclide	Retardation Factors	Solubility Limit (g/g)
Cm	500	1.E-9
Am	500	1.E-10
Pu	500	1.E-9
Np	100	1.E-9
	50	1.E-9
Th	5,000	1.E-9
Pa	500	No Limit
Ac	500	No Limit
Pb	50	1.E-7
Ra	500	1.E-8
Sn	1,000	1.E-9
Tc	5	1.E-9
I	1	No Limit
Sr	200	No Limit
Cs	1,000	No Limit
C	1	No Limit

TABLE II

Temperature History of Canister Surface in Basalt HLW Repository

(Thermal Loading = 50 kW/Acre)

Time[a] (Year)	Temperature (°C)
0	210
25	170
75	120
275	75
475	68
1,000	60

Ambient Temperature = 34°C

[a]Time zero = 25 years after emplacement of waste.

TABLE III

Parameters Used in the Time- and Temperature-dependent Leach Rate Model

Parameter	Value
n	0.67
a	0.001
b	0.01
λ	0.1
m	3.0
μ	300

Four sets of calculations were performed. The first set involved calculating integrated discharges and release ratios at the accessible environment for a 10^{-5} parts/year constant leach rate (Model A, $\lambda_L = 10^{-5}$), an exponential leach rate (Model B, $\lambda_L = 10^{-5}$) and a time-and temperature-dependent leach rate (Model C) using data from Tables II and III. The second set of calculations was a repeat of the first set except with the solubility limits of radionuclides imposed. In the third set of calculations, the outflow of groundwater from the repository was assumed to be through a small conduit, e.g., a borehole. Here, the backfilled region of the entire repository was modeled as a mixing cell in which flowing groundwater was assumed to mix with radionuclides in the volume of the mixing cell. The rate of outflow of radionuclides from the repository is then given by the product of the uniform concentration in the mixing cell and the rate of outflow of groundwater through the small conduit. As a result of the small outflow rate compared with the large mixing cell volume, the leached radionuclides typically reside in the mixing cell for a relatively long time before exiting the repository. Source Model A and Model C were compared for this calculation. Finally, calculations were performed for a cyclic leach rate source model where the average leach rate was 10^{-5} parts/year. This cyclic leach rate was simulated by using source Model C with a cyclic temperature history data.

RESULTS

Figure 2 shows the fractional leach rates of waste forms as a function of time for the three leach models. Two leach rate curves (n = 0.67 and n = 0.75) are shown for the time- and temperature-dependent leach model. For the n = 0.67 case, the average leach rate for the first 10,000 years was calculated to be 9.4×10^{-6} per year. As a result, this case of Model C (i.e., n = 0.67) was used to compare with the 10^{-5} constant and 10^{-5} exponential leach models. Release Ratios were computed for each 10,000-year period during the first 100,000 years for each of the three leach models. The results are shown in the first three columns for the leach limited cases in Table IV. These results indicated that the difference in the repository performance was minor between the constant and the exponential leach models, especially for the first 50,000 years. The time- and temperature-dependent leach model gave higher consequences than the other two models in the first 10,000 years and lower consequences afterwards. This is a direct result of the leach rate history as shown in Fig. 2. When the solubility limits of radionuclides were imposed, the Release Ratios calculated were somewhat lower; these values are also shown in Table IV. The discharge rates of the main contributing isotopes for the constant and the time-varying leach models are shown in Fig. 3.

A cyclic leach rate model (Fig. 4) with average leach rate of 10^{-5} parts/year was then used to compare with far-field consequences from a constant 10^{-5} parts/year model (Model A). The discharge rates of the fission products from this cyclic leach model are shown in Fig. 5. Figure 6 shows the results of a similar analysis when the dispersivity was increased to 500 ft. It can be seen that with larger dispersivity the cyclic pattern in the discharge rate was smoothed out significantly. The

TABLE IV

Release Ratios for Different Leach Models

Time Interval (Yrs)	Leach Model	10^{-5} Constant	10^{-5} Exponential	time- and temperature-dependent+ n = 0.67	++ Cyclic
0 - 1.E4	LCH*	0.36	0.35	0.51	0.50
	Sol**	0.34	0.33	0.45	
1.E4 - 2.E4	LCH	0.24	0.23	0.10	0.20
	Sol	0.16	0.15	0.09	
2.E4 - 3.E4	LCH	0.13	0.13	0.05	0.15
	Sol	0.08	0.07	0.05	
3.E4 - 4.E4	LCH	0.13	0.10	0.04	0.13
	Sol	0.05	0.05	0.04	
4.E4 - 5.E4	LCH	0.12	0.08	0.03	0.11
	Sol	0.05	0.04	0.04	
5.E4 - 6.E4	LCH	0.14	0.07	0.06	0.13
	Sol	0.06	0.05	0.05	
6.E4 - 7.E4	LCH	0.47	0.27	0.30	0.39
	Sol	0.24	0.10	0.12	
7.E4 - 8.E4	LCH	1.2	0.66	0.50	0.77
	Sol	0.62	0.20	0.19	
8.E4 - 9.E4	LCH	1.6	0.85	0.43	0.99
	Sol	0.86	0.28	0.17	
9.E4 - 1.E5	LCH	1.8	0.87	0.37	1.1
	Sol	0.97	0.30	0.16	

* LCH = Leach limited only
** Sol = Solubility limits imposed also

\+ Average leach rate = 9.4×10^{-6}/year for the first 10,000 years
++ Average leach rate = 1.0×10^{-5}/year for the first 100,000 years

TABLE V

Release Ratios With Mixing Cell Assumption

Time Interval (Yrs)	Leach Model	10^{-5} Constant	time- and temperature-dependent n = 0.67
0 - 1.E4		1.3E-4	2.3E-4
1.E4 - 2.E4		2.7E-4	2.4E-4
2.E4 - 3.E4		3.1E-4	2.0E-4
3.E4 - 4.E4		3.7E-4	2.0E-4
4.E4 - 5.E4		4.5E-4	2.1E-4
5.E4 - 6.E4		5.5E-4	2.3E-4
6.E4 - 7.E4		8.0E-3	3.9E-4
7.E4 - 8.E4		1.6E-3	8.3E-4
8.E4 - 9.E4		3.1E-3	1.3E-3
9.E4 - 1.E5		4.9E-3	1.7E-3

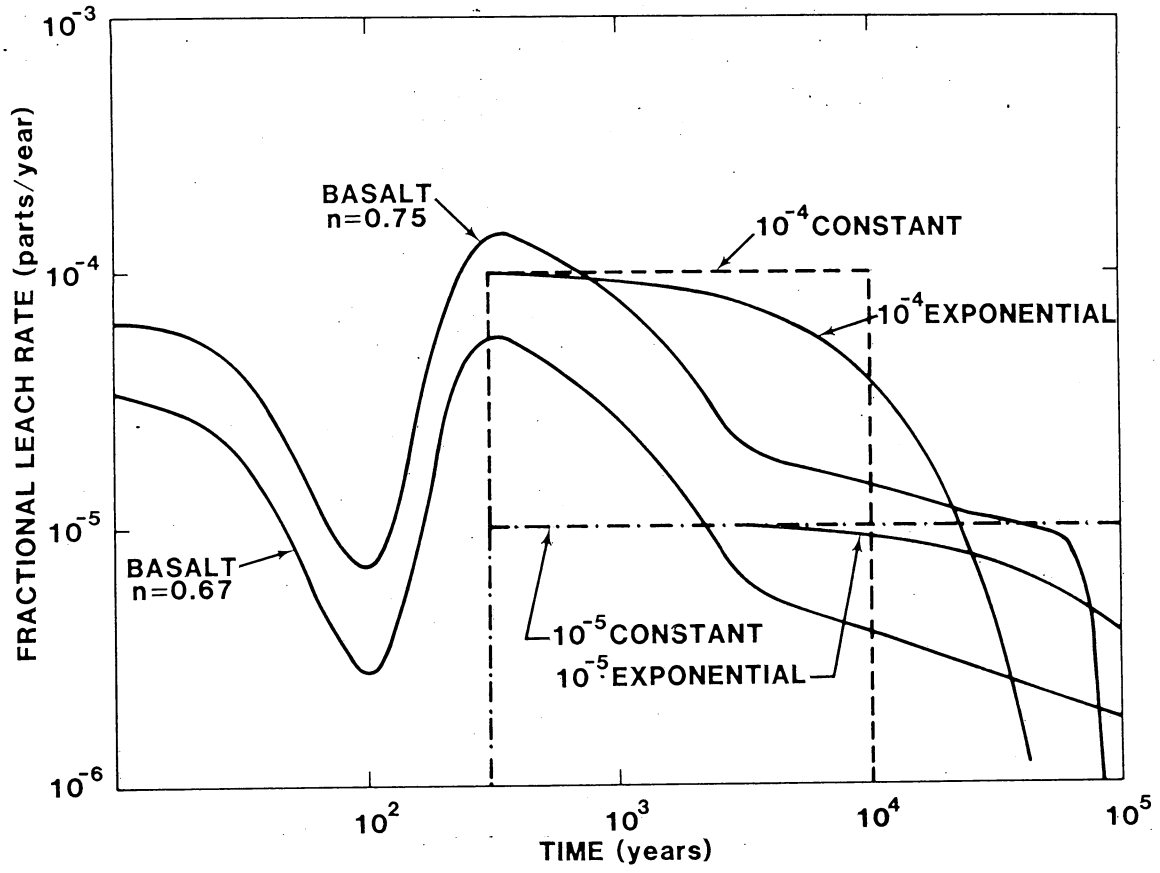

Fig. 2 Fractional leach rate of radionuclides from waste form as a function of time for several leach models.

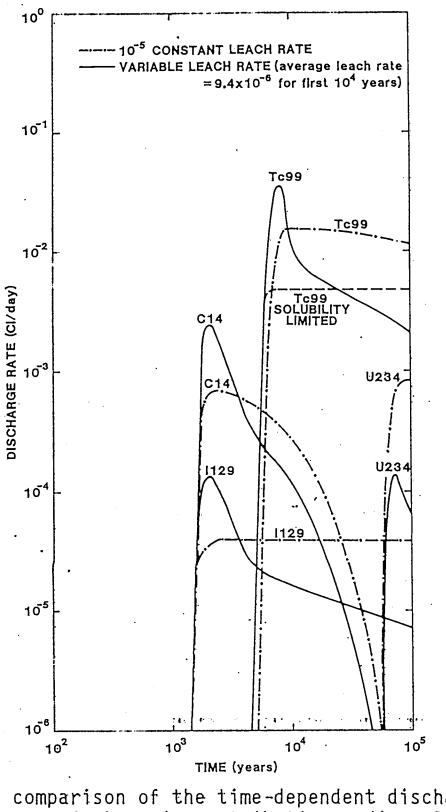

Fig. 3 A comparison of the time-dependent discharge rates of the main contributing radionuclides at the accessible environment for the constant leach rate model and the time- and temperature-dependent leach rate model.

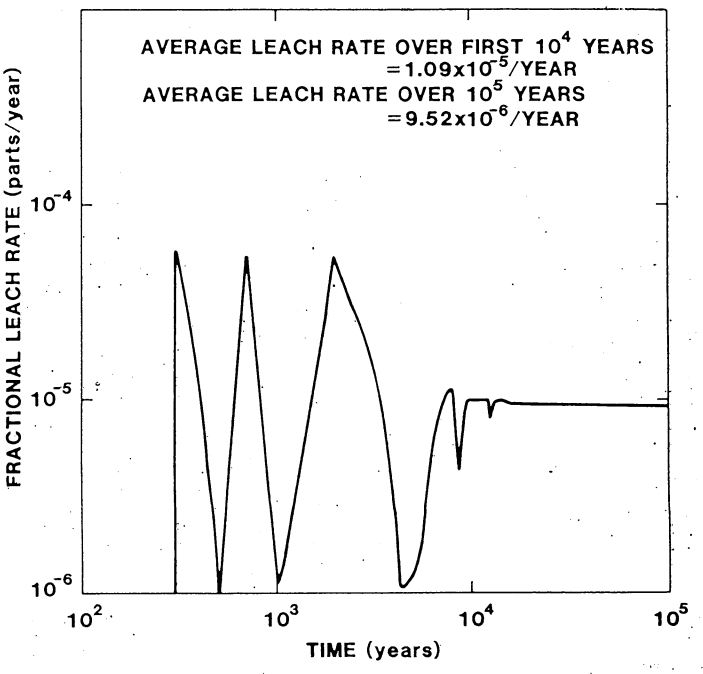

Fig. 4 Fractional leach rate of radionuclides form for the cyclic leach rate model.

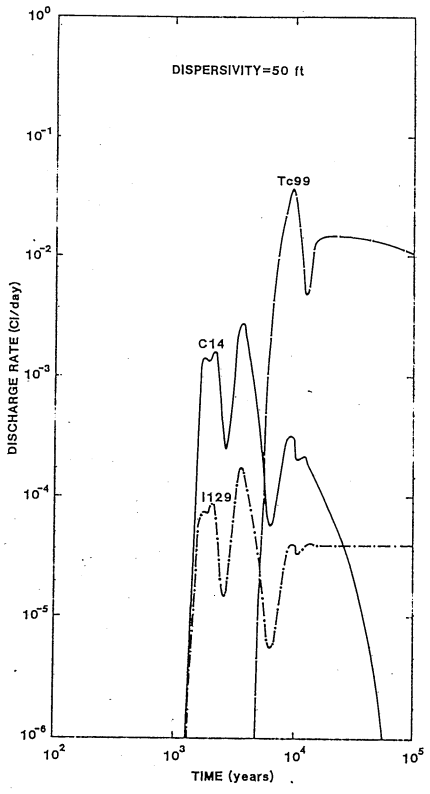

Fig. 5 Time-dependent discharge rates of fission-product radionuclides at the accessible environment for the cyclic leach rate model. Dispersivity = 50 ft.

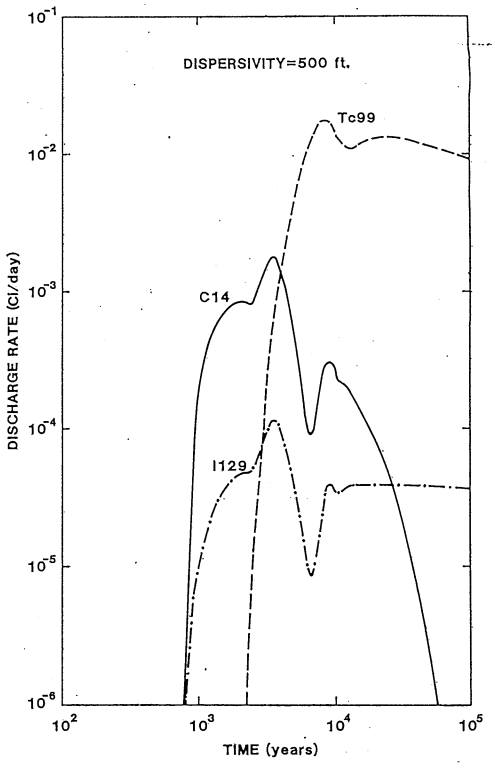

Fig. 6 Time-dependent discharge rates of fission-product radionuclides at the accessible environment for the cyclic leach rate model. Dispersivity = 500 ft.

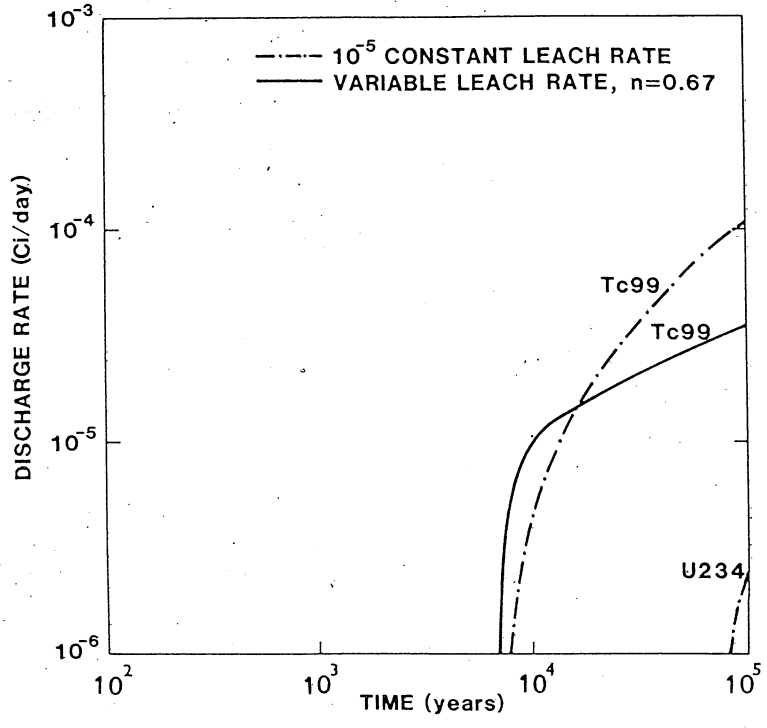

Fig. 7 A comparison of the time-dependent discharge rates of the main contributing radionuclides at the accessible environment for the constant leach rate model and the time- and temperature-dependent leach rate model when mixing cell condition was used.

Release Ratios calculated for the cyclic leach model (d = 50 ft) are shown in the last column of Table IV. It is seen that the consequences from the cyclic leach model are very similar to that of the constant model.

In the borehole scenario where the mixing cell model was assumed, the majority of the leached radionuclides remained inside the repository for a relatively long time before exiting through the borehole. The volume of the mixing cell used for this calculation was 10^6 ft^3. Figure 7 shows the lowered discharge rate curves of the main contributing radionuclides. The small Release Ratios resulting from this fourth set of calculations are shown in Table V.

CONCLUSIONS

The effect on the performance of hypothetical high-level waste repositories of using different source models was studied in this analysis. It was found that, for similar average leach rates, the differences in the leach models had a minor impact as far as compliance with EPA limits is concerned. On the other hand, if the "mixing-cell" effect was included, the impact on far field consequence was significant. These preliminary analyses, therefore, indicate that an "average" leach rate can adequately simulate the more detailed, time-dependent leaching behavior. In addition, the significant impact of the mixing-cell effect indicates that further research is needed in the near field area.

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