

RADIONUCLIDE MIGRATION TO THE AQUIFER THROUGH THE POROUS MEDIUM

Y. Hwang, M.C. Lee, H.S. Shin, C.L. Kim and J. Kim
Korea Atomic Energy Research Institute

ABSTRACT

Time dependent relative dose rates of radionuclides from a waste container to the aquifer through the porous medium are analyzed for 1. congruently released radionuclides such as Np-237 1. and 2. highly soluble radionuclides such as Cs-135 and I-129 1). Radioactive dose rates in the well water of the aquifer estimated from the analytical analyses are compared with the maximum allowable limits 2) in the water. Results show that all dose rates are below the limits 2). Among the radionuclides considered in this paper Cs-135 and I-129 are proven to be the most dangerous radionuclides.

INTRODUCTION

To estimate the radionuclide migration from a waste container to the aquifer down through the porous medium as shown in Fig. 1, following radionuclide release scenario is developed:

1. A waste package which contains the spent fuel is emplaced horizontally in the repository.
2. Conservatively at time zero, a waste package is assumed to be failed by corrosion or other mechanisms such as cracking.
3. Release rates of low-soluble radionuclides such as Pu-239 and Np-237 are governed by the congruent release model(1). The release rates of highly-soluble radionuclides such as I-129 and Cs-135 are estimated by the gap release model(1).
4. Radionuclides released from the waste container surface are diffused into the surrounding porous medium. Inside the porous medium radionuclide migration is retarded by the physicochemical sorption and desorption processes.
5. Radionuclides disscused down through the porous medium reach the porous rock/aquifer interface.
6. Radionuclides migrate in the aquifer by advection and dispersion.

7. A well is emplaced into the aquifer and the water in the aquifer which contains the radionuclides migrating up to that point is taken out.

In our analysis we do not analyze the biological effect of intake of the water from the well to the humanbeings. Instead our final goal is limited to estimate the dose rates of the radionuclides in the well water.

The above scenario can be applicable to the current U.S. Yucca Mountain project. Yucca mountain repository is located above the water table so that the surrounding porous tuff is not fully saturated. But conservatively, the repository is assumed to be fully saturated by the possible flooding in the future. Then once-released radionuclides from the repository are diffused through the fully saturated porous tuff layer and then finally reach the aquifer located underneath the repository and then migrate by the advection and dispersion.

The most severe threat against the safety of the potential repository is caused by possible human intrusion to the repository. We can assume that the future humanbeings who live near the repository drill a well into the aquifer which is contaminated by the released radionuclides. Then it is necessary to estimate the release rates of radionuclides into the well water to assure the safety of the repository. In the following section we develop the analytical approaches to estimate the possible radionuclide does rates in the well water.

MATHEMATICAL FORMULATIONS

To estimate the radionuclide release from the waste package into the aquifer through the porous medium for soluble and congruently released radionuclides respectively, following Eq. (1) and (2) are used to describe the diffusional mass fluxes from the porous medium into the aquifer L distance away from the waste container (3):

The release rate of the congruent released radionuclide such as Np-237 as a function of time since the container failure and position away from the waste container surface is described as (3):

$$J(t) = \epsilon \sqrt{DR_m} \frac{M_{10}}{M_{m0}} C_{m0} e^{-\lambda_1 t} \frac{1}{\sqrt{\pi t}} e^{-\frac{z^2}{4Dt}}, \quad t \geq 0, \quad (\text{Eq. 1})$$

The release rate of the highly-soluble radionuclide such as I-129 is expressed as (3):

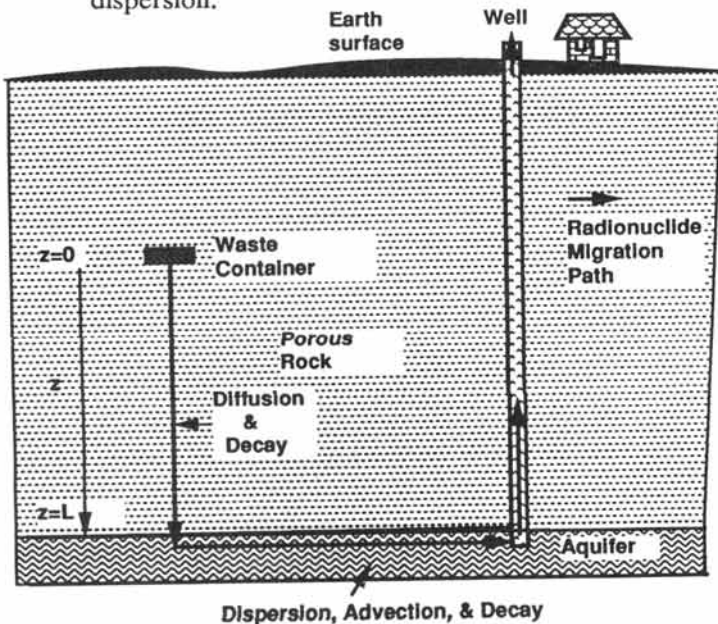


Fig. 1. Schematic of radionuclide transport from the single container in the repository to the aquifer through the fully saturated porous media.

$$J(t) = \epsilon N^o \sqrt{R_i D} \left[\frac{1}{\sqrt{\pi t}} - \beta \operatorname{erf} l(\beta \sqrt{t} + \frac{1}{2} \sqrt{\frac{R_i}{D t}} (z - a)) \right] \times e^{-\lambda_i t} e^{-\frac{R_i}{4 D t} (z - a)^2}, \quad t \geq 0 \quad (\text{Eq. 2})$$

where

$$\operatorname{erf} l(x) = e^{x^2} \operatorname{erf} c(x) \quad (\text{Eq. 3})$$

R_m is the retardation coefficient of the uranium matrix, (-),

R_i is the retardation coefficient of the i th radionuclide, (-),

$C_m(z, t)$ is the concentration of the uranium matrix in surrounding porous media, (g/m^3),

C_{m0} is the solubility of the uranium matrix in pore water (g/m^3),

D is the diffusion coefficient of the uranium matrix in pore water of porous media, (m^2/yr),

$$D_m = \frac{D}{R_m} \quad (\text{Eq. 4a})$$

$$D_i = \frac{D}{R_i} \quad (\text{Eq. 4b})$$

M_{i0} is the initial inventory of the i th radionuclide in a single waste package, (g),

M_{m0} is the initial inventory of the uranium matrix in a single waste package, (g),

R_i is the retardation coefficient of the i th radionuclide, (-)

C_i is the i th radionuclide concentration (g/m^3),

N^o is the concentration of the soluble radionuclide dissolved in the void gap water volume inside the container at the time of the container failure, (g/m^3),

a is the gap width, (m),

$$\beta = [D R_i \epsilon^2 / a^2]^{1/2}, \quad [-] \quad (\text{Eq. 5})$$

and λ_i is the decay constant of the i th radionuclide, ($1/\text{yr}$).

Here one percent of initial inventory of the soluble nuclide is assumed to be already dissolved in the void volume filled with water inside the waste container (1).

Equations (1) and (2) are theoretically valid for the semi-infinite one-dimensional rectangular domain. Therefore, mathematically Eqs. (1) and (2) are not valid to describe the mass release rate in the finite porous medium which interfaces with the aquifer. Fortunately, the release rates expressions at a certain point away from the source for the semi-infinite medium are proven to be acceptable to describe those in the finite medium.

To analyze the transient dispersive and advective radionuclide migration in the aquifer the following Eq. is used (4):

$$K_i \frac{\partial N_i(x, t)}{\partial t} + v \frac{\partial N_i(x, t)}{\partial x} = D^* \frac{\partial^2 N_i(x, t)}{\partial x^2} + \quad (\text{Eq. 6})$$

$$t > 0, \quad -\infty < x < \infty$$

Side conditions are:

$$N_i(x, 0) = -\infty < x < \infty \quad (\text{Eq. 7a})$$

$$N_i(\infty, t) = N(-\infty, t) = 0, \quad t > 0 \quad (\text{Eq. 7b})$$

where K_i is the retardation coefficient of the i th radionuclide in the fracture, (-),

$N_i(x, t)$ is the i th radionuclide concentration in the aquifer, (g/m^3),

g is the conversion factor defined in Fig. 2, (-),

v is the pore water velocity, (m/yr),

D^* is the dispersion coefficient of radionuclide in the aquifer, (m^2/yr), and $\delta(x)$ is the delta function, ($1/\text{m}$).

Then the solution is given as(4):

$$N_i(x, t) = g \int_0^t J(t - \tau) e^{-\lambda \tau} \frac{1}{\sqrt{\frac{4 \pi D^* \tau}{K_i}}} e^{-\frac{(x - \frac{v \tau}{K_i})^2}{\frac{4 D^* \tau}{K_i}}} d\tau, \quad -\infty \leq x \leq \infty, \quad t \geq 0 \quad (\text{Eq. 8})$$

The relative dose rate is defined as:

$$F(x, t) = \frac{\lambda_i N_i(x, t)}{P} f \quad (\text{Eq. 9})$$

where P is the maximum allowable dose rate in the water (2), [$\frac{\mu\text{Ci}}{\text{ml}}$],

and f is the unit conversion factor.

In the following section the radionuclide relative dose rates are calculated based on Eq. (9).

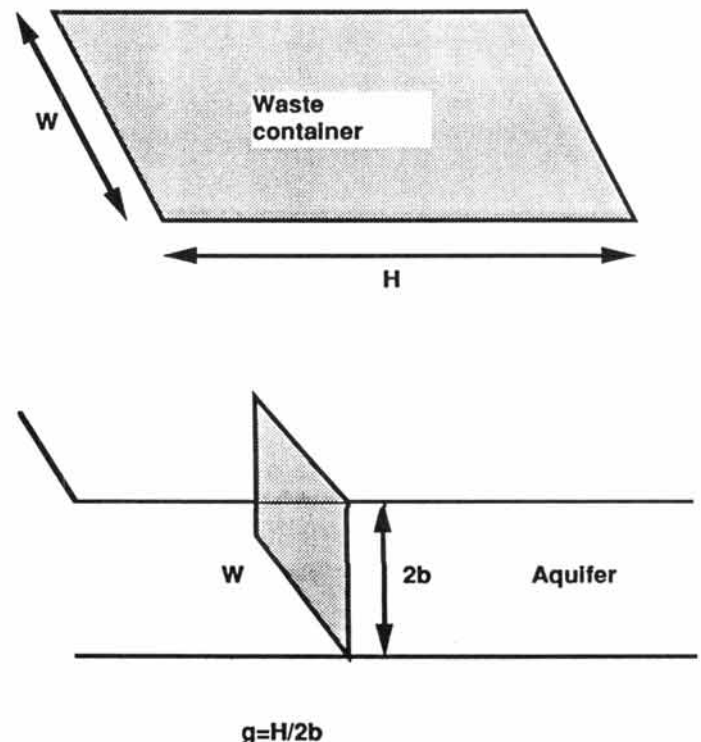


Fig. 2. Conversion factor between the porous rock and the aquifer.

NUMERICAL ILLUSTRATIONS

Pu-239 is chosen to represent the relative dose rates of congruently released radionuclides in the well water and Cs-135 and I-129 are chosen for the highly soluble radionuclides. Tables I, II, and III list the parameter values (1,4) used in our analysis.

TABLE I

Isotope Parameter Values Used in the Calculations

Isotope	Decay Constant (I/yr)	Gap Inventory (g/m ³)	Initial Inventory (g)	Retardation Coefficient (-)
Pu-239	2.88x10 ⁻⁵	not used	23.51	500
Np-237	1.74x10 ⁻⁵	not used	1.39x10 ³	100
I-129	4.077x10 ⁻⁸	11.8	not used	1
Cs-135	2.31x10 ⁻⁷	66.4	not used	10

TABLE II

Parameter Values Used in the Calculations

D*	D	v	ε	R _m	K _i	M _m	C _m	a
(m ² /yr)	(m ² /yr)	(m/yr)	(-)	(-)	(-)	(g)	(g/m ³)	(cm)
0.315	3.15x10 ⁻²	1	0.01	50	1	4.6x10	10 ⁻³	10

TABLE III

Maximum Allowable Dose Rates of Pu-239, I-129, and Cs-135

Isotope	Pu-239	I-129	Cs-135	Np-237
Maximum Allowable Dose Rate, [μCi/ml]	5x10 ⁻⁶	6x10 ⁻⁸	2x10 ⁻⁵	3x10 ⁻⁵

Figure 3 shows the mass release rates of the Pu-239 into the porous rock/aquifer interface as a function of time since emplacement for difference distances between the waste container surface and aquifer. At early time radionuclides do not migrate to the interface yet. As time increases more radionuclides enters the interface. But after it reaches the maximum the flux begins to decrease and finally decays out. This trend can be verified from Eq. (1) which is repeated as:

$$J(t) = \epsilon \sqrt{DR_m} \frac{M_{i0}}{M_{m0}} C_{m0} e^{-\lambda_i t} \frac{1}{\sqrt{\pi t}} e^{-\frac{z^2}{4D_i t}}, \quad t \geq 0, \text{ (Eq. 1)}$$

At t = 0 the $\frac{1}{\sqrt{t}}$ term goes to infinity. But the exponential term $e^{-\frac{K_i}{4Dt} (z-a)^2}$ becomes zero. Since the exponential function is stronger at t = 0 the flux is zero. As time increases the flux becomes increasing and reaches the maximum. But as time increases, the flux begins to decrease and as predicted by Eq. (1), the flux finally decays out. In Fig. 3, as diffusional migra-

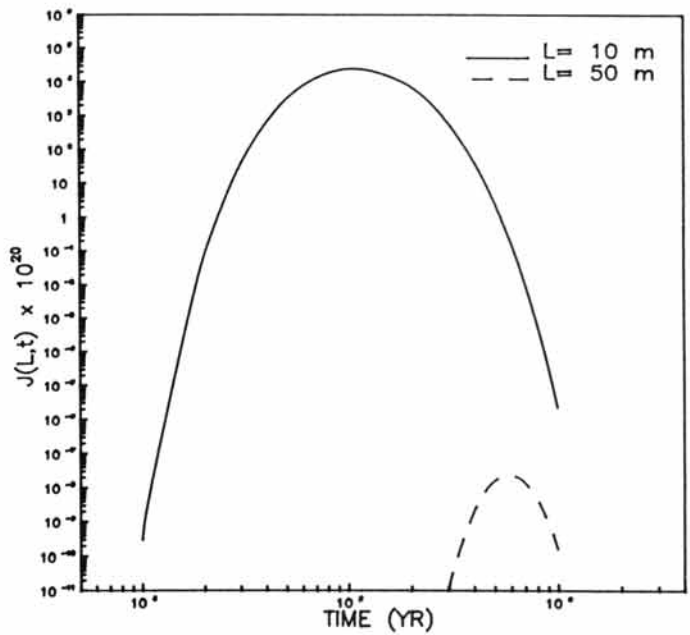


Fig. 3. Pu-239 diffusional fluxes into the aquifer as a function of time for different L's.

tion distance which is the distance between the waste container and the aquifer increases, less radionuclides are delivered to the aquifer. This proves that the thicker geosphere acts as the stronger barrier against the migration of the radionuclides.

Figure 4 illustrates the fluxes of I-129 into the aquifer. Even though the trend is exactly the same as that of Pu-239 into the aquifer. Even though the trend is exactly the same as that of Pu-239 is much higher than that of Pu-239.

Figure 5 illustrates the fluxes of Cs-135 into the aquifer. Since the void water concentration N_0 and the retardation coefficient of Cs-135 are higher than those of I-129, at early time, when the contribution from the erfl function in Eq. (2)

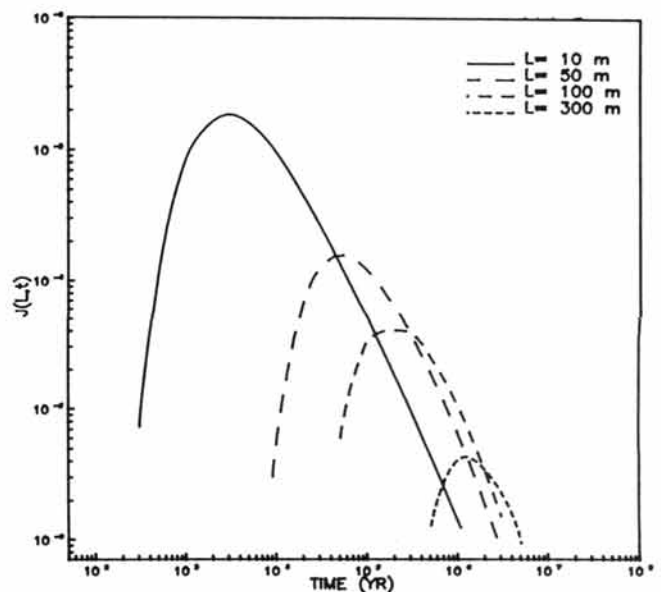


Fig. 4. Cs-135 diffusional fluxes into the aquifer as a function of time for different L's.

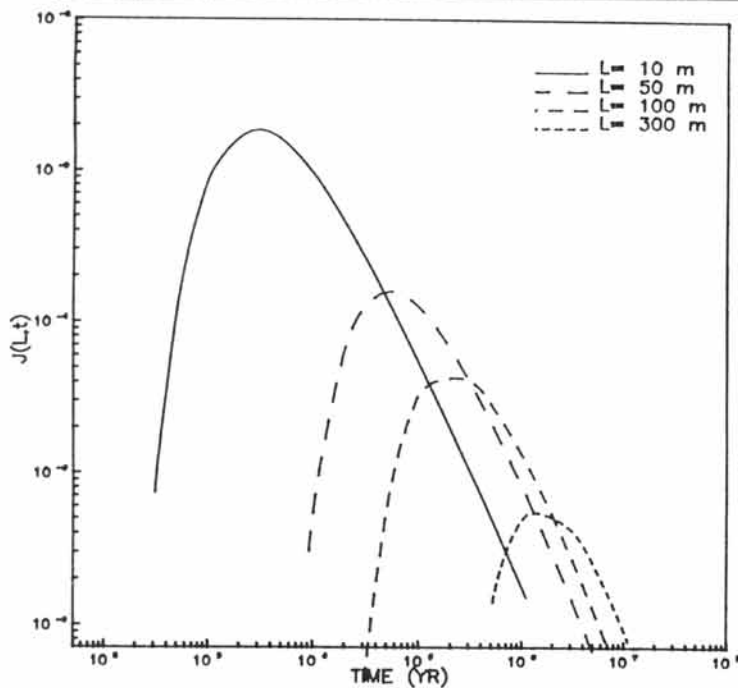


Fig. 5. I-129 diffusional fluxes into the aquifer as a function of time for different L's.

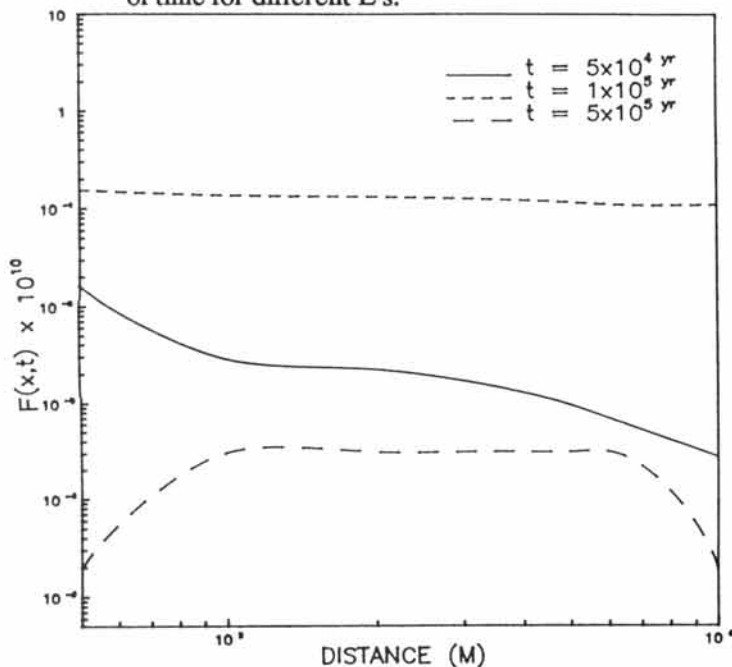


Fig. 6. Relative dose rates of Pu-239 in the aquifer as a function of distance for fixed times.

and effect from the decay is negligible, the fluxes of Cs-135 is higher than those of I-129.

Figure 6 shows the relative dose rate of Pu-239 as a function of the distance for fixed times. At early time when the flux into the aquifer is constantly increasing, the dose rate in the aquifer decreases as the distance increases. But at around $t = 5 \times 10^5$ year due to the decreasing $J(t)$ the Pu-239 concentration in the near field is lower than that of intermediate range. But in the far-field range the dose rate decreases as the distance increases. The results show that the relative dose rates in the aquifer are considerably lower than one for given data set used in our calculations.

Figure 7 shows the similar trend for Np-237.

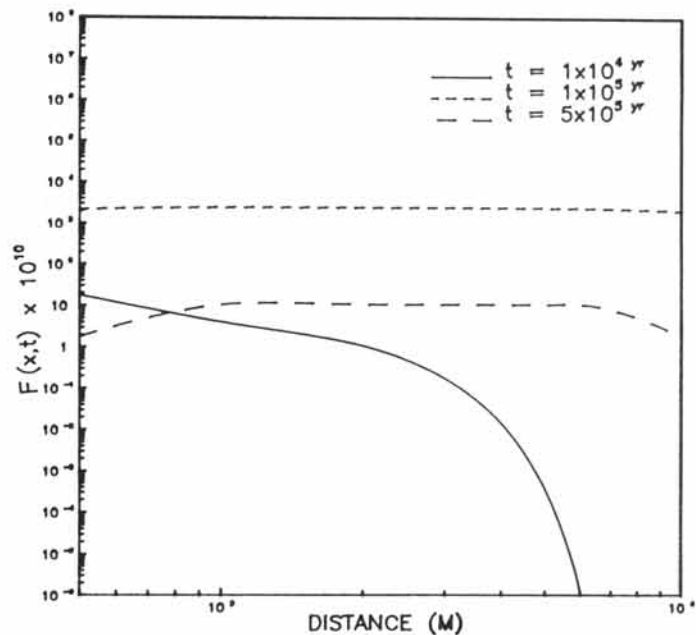


Fig. 7. Relative dose rates of Np-237 in the aquifer as a function of distance for fixed times.

Figure 8 illustrates the relative dose rates of Cs-135 as a function of distance for fixed times. Since the retardation coefficient of Cs-135 as a function of distance for fixed times. Since the retardation coefficient of Cs-135 is lower than those of uranium matrix and Pu-239 the migration of Cs-135 is faster than Pu-239 which is congruently released. Even though the solubility of Cs-135 is very high the relative dose rate of Cs-135 is still lower than one. Figure 9 shows the same trend for I-129.

Figures 10, 11, and 12 show the relative dose rates as a function of time for given positions for Pu-239, Cs-135, and I-129 respectively.

Even though all relative dose rates of three radionuclides are lower than one, we can notice that fission products such as Cs-135 and I-129 are more dangerous than actinides.

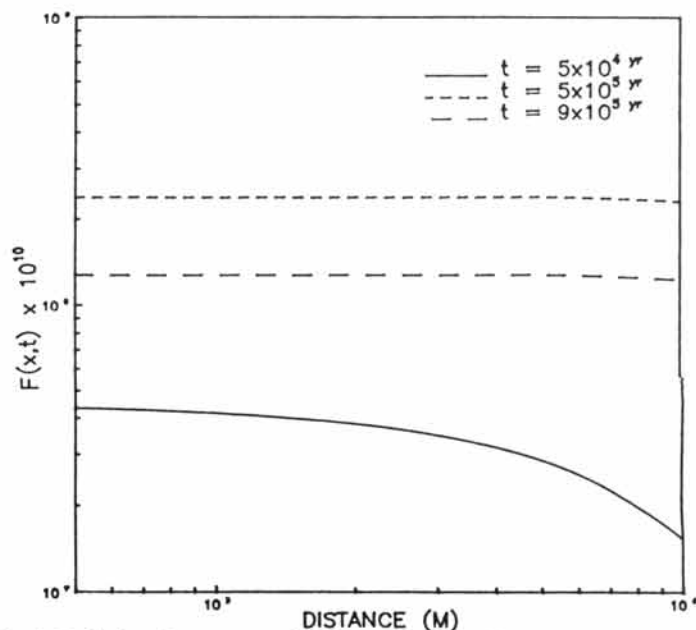


Fig. 8. Relative dose rates of Cs-135 in the aquifer as a function of distance for fixed times.

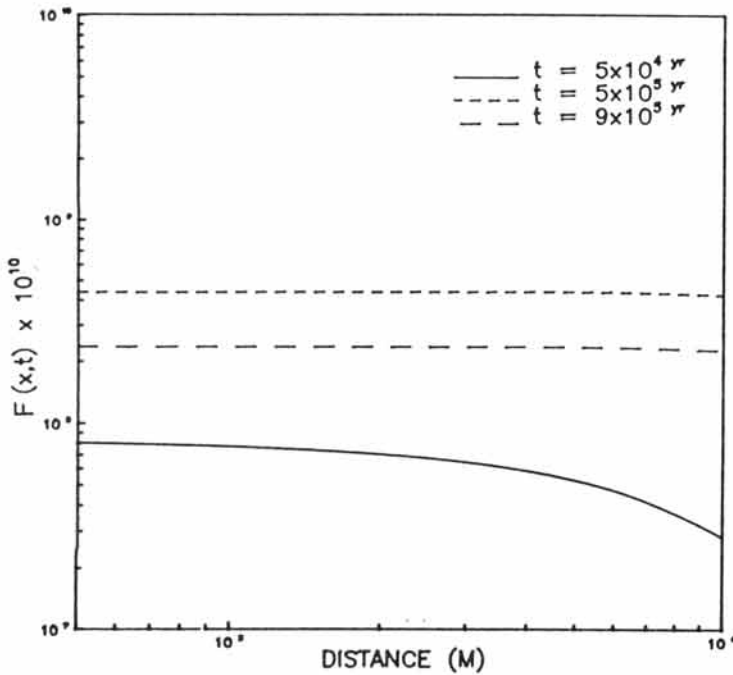


Fig. 9. Relative dose rates of I-129 in the aquifer as a function of distance for fixed times.

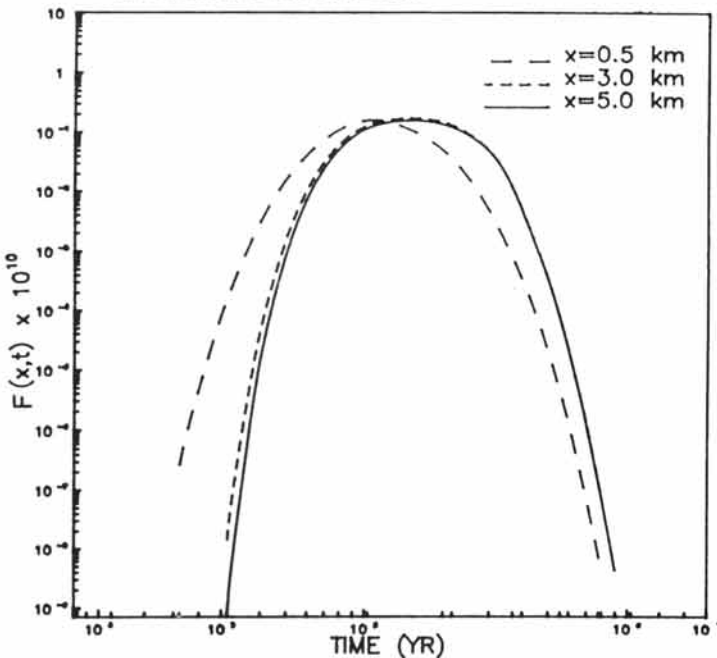


Fig. 10. Relative dose rates of Pu-239 in the aquifer as a function of time for different distances.

CONCLUSION

We develop the analytical solutions for the relative dose rates of the radionuclides released from the waste container to the aquifer down through the porous medium. Numerical illustrations indicate that dose rates of all radionuclides studied in this paper are below those of maximum permissible dose rate limits. But the relative dose rates of high-soluble fission products such as I-129 and Cs-135 are considerably higher than those of transuranic actinides such as Pu-239. Our study can be easily applicable to the U.S. Yucca Mountain repository not only to estimate the release rates in the aquifer but also to find out which isotope is the most dangerous against the safety of the repository.

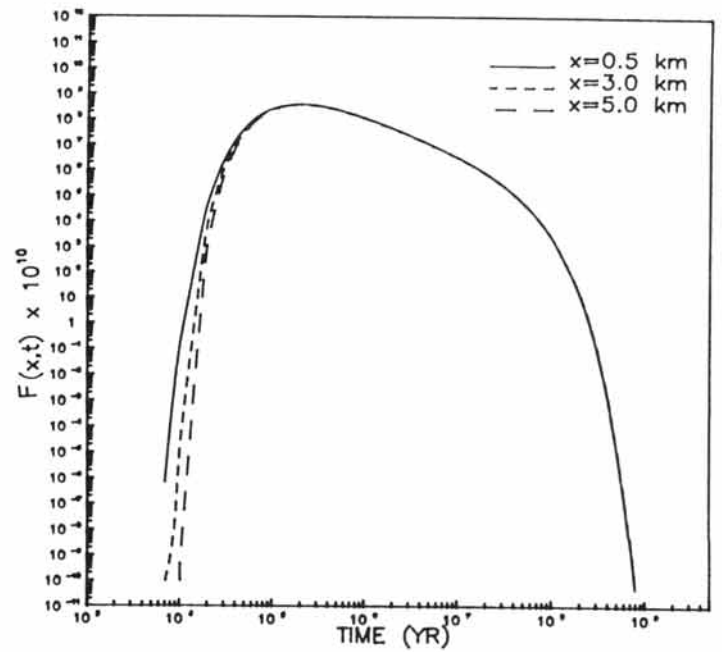


Fig. 11. Relative dose rates of Cs-135 in the aquifer as a function of time for different distances.

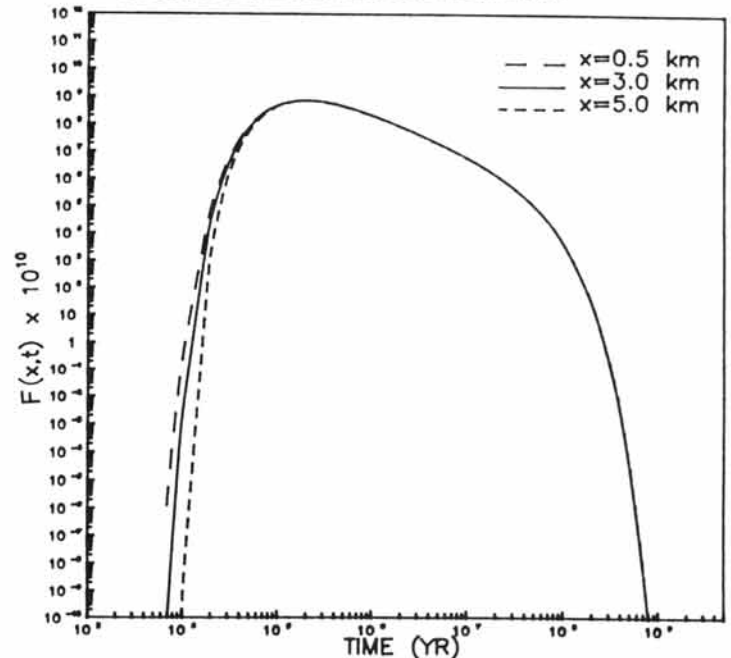


Fig. 12. Relative dose rates of I-129 in the aquifer as a function of time for different distances.

We are currently extending our study to analyze the dose rate of all important radionuclides. Also we study the additional effect of advective transport in the porous medium.

REFERENCES

1. T.H. PIGFORD et al., "A Review of Near-Field Mass Transfer in Geologic Disposal Systems", LBL-27045, UC-802, University of California Berkeley, 1990.
2. M. BENEDICT et al., Nuclear Chemical Engineering, 2nd Edition, McGrawHill, N.Y., 1981.
3. Y. HWANG, submitted to *Waste Management*, September 1992.
4. C.L. KIM, Ph.D. Dissertation, University of California, Berkeley, 1987.