SENSITIVITY OF RADIONUCLIDE TRANSPORT IN GROUNDWATER TO SOURCE AND SITE CHARACTERISTICS

John C. Walton
Idaho National Engineering Laboratory
Idaho Falls, Idaho 83415

Martin S. Bensky, Mark E. Mease, Tom J. Bander
Westinghouse Hanford Operations
Richland, Washington 99352

ABSTRACT

A methodology for identification of parameter combinations which can lead to significant degradation of total system performance of a waste repository is described. An analytical solution with dimensionless variables is utilized. The technique is intended to serve a screening function by identifying scenarios which may require more detailed modeling.

INTRODUCTION

The sensitivity study discussed in this paper analyzes overall performance of a waste isolation system using a lumped parameter advection-dispersion equation. Transport of dissolved radionuclides through the geoshpere is envisioned as travel along a pathline or flow tube to the accessible environment. Cumulative radionuclide flux at the accessible environment, defined as any distance of environmental or regulatory concern, is utilized as the principal measure of system performance.

Radionuclide transport in the flow tube is subject to advection, dispersion, and retardation processes. Lumped parameters, derived from field tests or from analyses with subsystem models, are used to estimate the impacts of the three processes. This simple approach is justified by a) lack of detailed site characterization data and b) the need to identify key parameters and processes prior to conducting more sophisticated studies.

The sensitivity study also provides a basis for identifying disruptive event scenarios that warrant detailed study. The importance of any disruptive scenario is dependent upon the level of probability of the scenario occurring and the consequences associated with that scenario. That is, only scenarios with sufficiently high probability and consequences need be considered in the assessment of repository performance. This sensitivity study provides a useful screening tool by quickly identifying combinations of key system parameters which can lead to unacceptable degradation of total system performance, i.e., unacceptable consequences. These combinations can be related to specific scenario classes which can then be identified for more detailed investigation.

BASIC GOVERNING EQUATION

The general advection-dispersion equation for saturated convective transport of a single dissolved substance in porous or highly fractured (i.e., equivalent porous) media is (1):

$$\frac{\partial c}{\partial t} + \frac{U}{R_d}\frac{\partial c}{\partial x} = \frac{D_x}{R_d}\frac{\partial^2 c}{\partial x^2} + \frac{D_y}{R_d}\frac{\partial^2 c}{\partial y^2} + \frac{D_z}{R_d}\frac{\partial^2 c}{\partial z^2} - \lambda c \qquad (1)$$

where:

c = concentration in liquid phase

U = x component of groundwater pore velocity

R_d = retardation coefficient

t = time

D_x, D_y, D_z = dispersion coefficients in the x, y, and z directions

λ = radioactive decay coefficient = ln 2 / half-life

For an impulse release that travels unidirectionally in an infinite homogeneous medium, the solution to the equation is:

$$c = \frac{M}{n_e R_d A (4 \pi D t / R_d)^{0.5}} \exp\left(- \frac{(x - U t / R_d)^2}{4 D t / R_d} - \lambda t \right) \qquad (2)$$

where:

M = amount of radionuclide released (e.g., Curies, grams)

n_e = effective porosity

A = cross-sectional area of flow tube

D = dispersion coefficient in the direction of travel

A continuous, time variant release can be simulated by superposition of the solution for individual pulses:

$$c = \sum_{i=1}^{k} \frac{M_i}{n R_d A (4 \pi D t_i / R_d)^{.5}} \exp\left(- \frac{(x - U t_i / R_d)^2}{4 D t_i / R_d} - \lambda t_i \right) \quad (3)$$

t_i = age of pulse = (t - tp)

t_p = time when pulse was released

t = total time

M_i = amount of material released in pulse # i

k = total number of pulses

That is, uniform release is simulated by a series of overlapping pulses.

Different assumptions concerning the boundary conditions (e.g., semi-infinite domain) lead to different solutions of the equation. The differences between the solutions are more important for short time frames with high dispersion than for long time frames with low dispersion.

One measure of water flow is the groundwater travel time (t_{gw}), which is the average time for a hypothetical particle of water to travel along the flow tube to the accessible environment. For dissolved radionuclides, the average transport time is given by the product of groundwater travel time and the retardation factor. Dimensionless time is expressed as:

$$T = \frac{t}{t_{gw} R_d} \quad (4)$$

where:

t = time frame of interest (years)

t_{gw} = groundwater travel time (years)

R_d = retardation factor (dimensionless)

Retardation reflects the fact that dissolved constituents frequently travel more slowly than the bulk water flow. Many conceptual models are possible to describe retardation in fractured crystalline rock. The one given here is based upon the assumption of porous flow within secondary-mineral-filled fractures. If sorption of radionuclides is assumed to be linear and reversible, then the retardation factor can be derived as a function of distribution coefficient (K_d), porosity of materials within the fractures, and density, related by the expression (2):

$$R_d = \frac{n_c}{n_e} + \frac{\rho}{n_e} K_d = 1 + \frac{n_s}{n_e} + \frac{\rho}{n_e} K_d \quad (5)$$

where:

K_d = distribution coefficient between fluid and secondary minerals (ml/g)

n_c = total connected porosity in fractures

n_e = effective porosity in fractures

n_s = stagnant porosity in fractures = $n_c - n_e$

ρ = bulk density of secondary minerals within fractures (g/cm^3)

The three terms on the right represent the contribution from a) moving fluid, b) fluid in dead end fractures or isolated pores, and c) sorption onto secondary minerals in the fractures, respectively. Diffusion into dead end fractures may be quite important for slowing the transport of radionuclides such as I-129 which do not sorb strongly. The porosity of secondary minerals in fractures (n_c) is expected to be much higher than the overall porosity of crystalline rocks.

If the simplifying assumption is made that $n_e = n_c$, then ns/nc approaches zero and

$$R_d = 1 + \frac{1}{n_c} K_d \quad (6)$$

Dispersion of radionuclides results from spatial variability in water flow rates and retardation. The advection-dispersion equation accounts for dispersion through analogy with Fick's Law for diffusion. In practice, dispersion is estimated using tracer tests. The value has been shown to be adequately represented by the relation $D = \alpha U$ where is the dispersivity in meters (1). A measure of the role of dispersion in a system is given by the dimensionless Peclet number which represents the ratio of advection to dispersion:

$$Pe = \frac{U L}{D} = \frac{U L}{\alpha U} = \frac{L}{\alpha} \quad (7)$$

where L is equal to the distance to the accessible environment (4). Defined this way, the Peclet number effectively removes advection from the equation and represents a measure of dispersion that takes place in a length, L. Varying the Peclet number means varying only the value for dispersivity.

DIMENSIONLESS CUMULATIVE FLUX

Equation (1) can be expressed in terms of the dimensionless variables (2):

$$\frac{\partial C}{\partial T} + \frac{\partial C}{\partial X} = \frac{1}{Pe}\frac{\partial^2 C}{\partial X^2} - G\,C \qquad (8)$$

where: $X = \frac{x}{L}$, $C = \frac{c\,A\,L\,R_d\,n}{M}$, and $G = t_{gw}\,R_d\,\lambda$

Rearrangement of the equations allows solution for a dimensionless concentration given as:

$$C = \frac{1}{2\,(\pi\,T / Pe)^{0.5}} \exp\left(-\frac{(1 - T)^2}{4\,T / Pe} - G\,T\right) \qquad (9)$$

The use of dimensionless groups allows rapid evaluation of combinations of parameters. The influence of decay, dispersivity, and time on concentration is illustrated in Figs. 1 and 2. When decay is absent, a low Peclet number results in large plume spread and earlier arrival time, but does not impact steady state concentration. In the presence of radionuclide decay, the Peclet number assumes a greater importance. A low Peclet number allows some material to arrive prior to decay.

The influence of release rate peaks on flux and concentration at the accessible environment is important from both a safety and an analysis perspective. Representation of release as a series of impulses is ideally suited to this determination. When a series of instantaneous impulses cannot be distinguished from a continuous release, when measured at the accessible environment, then spikes in release rate of a duration less than or equal to the spacing between the impulses are unimportant to performance. Since spikes in release may be more difficult to control than long term averages it is important to evaluate under what conditions such spikes are important. One measure of the spread of the impulse function is its standard deviation as measured from a cumulative arrival curve at the accessible environment. When impulses are spaced at intervals of less than one standard deviation, the superposition of their individual contributions gives a fairly constant result. The standard deviation for dimensionless concentration is given by (2):

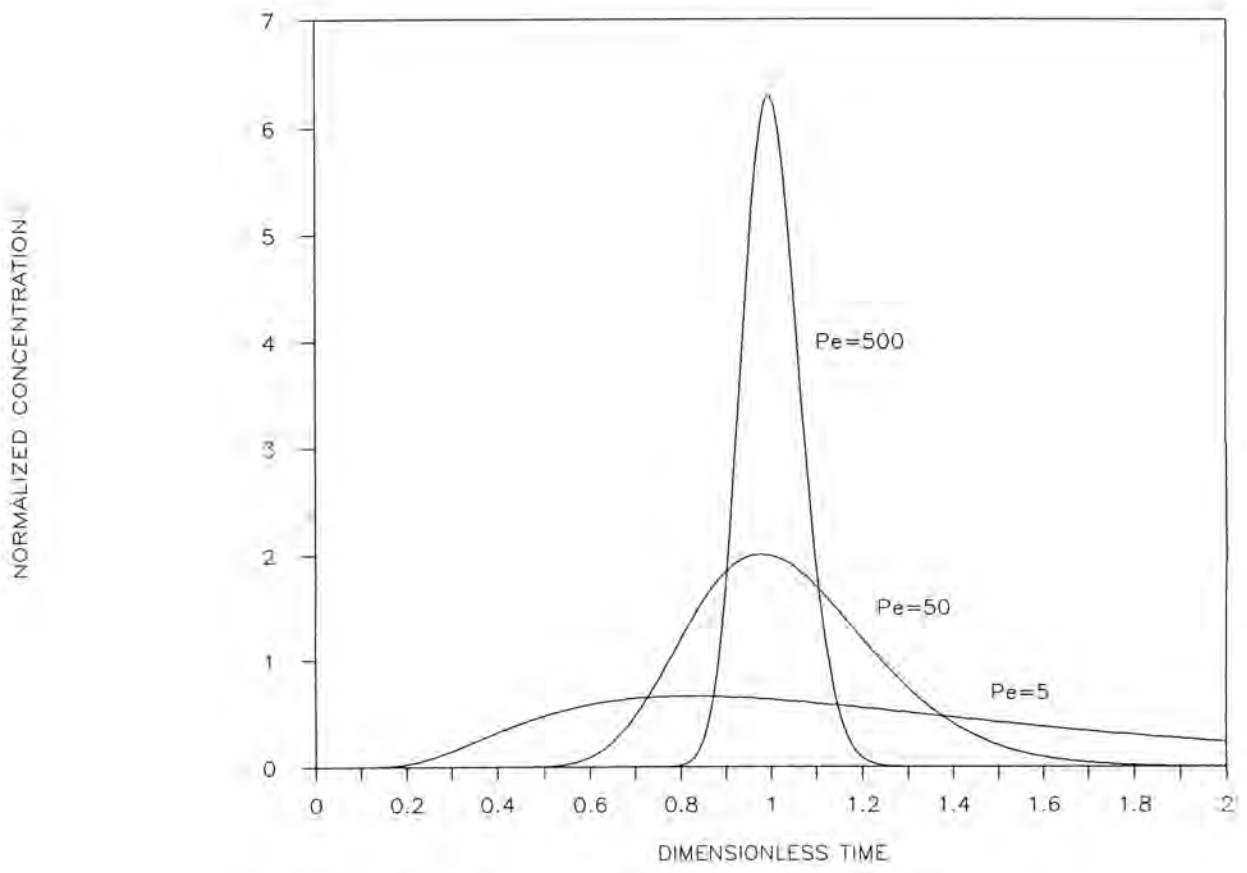

Fig. 1. Influence of Peclet Number on Transport When Decay is Absent.

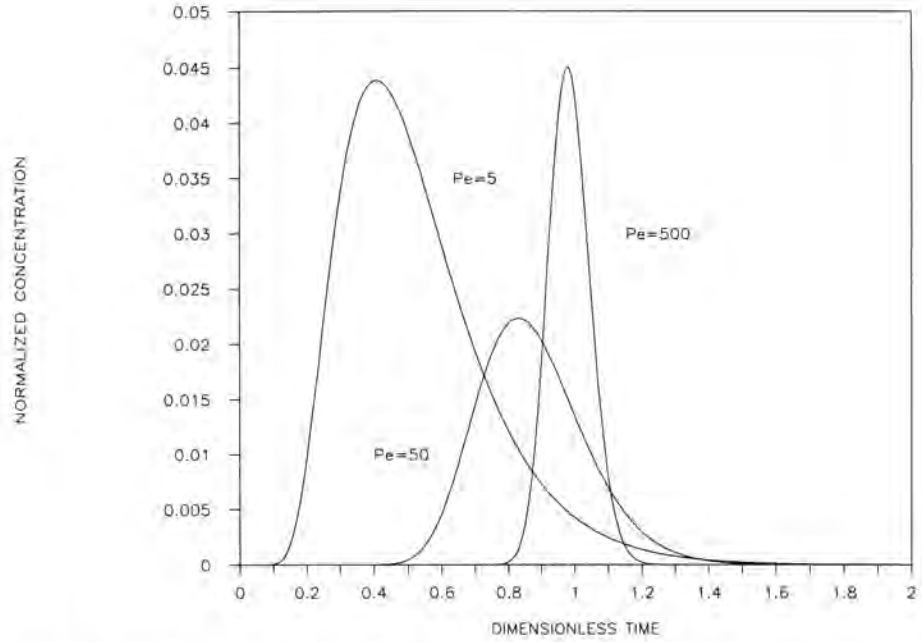

Fig. 2. Influence of Peclet Number on Transport When Dimensionless Decay Parameter is 5.

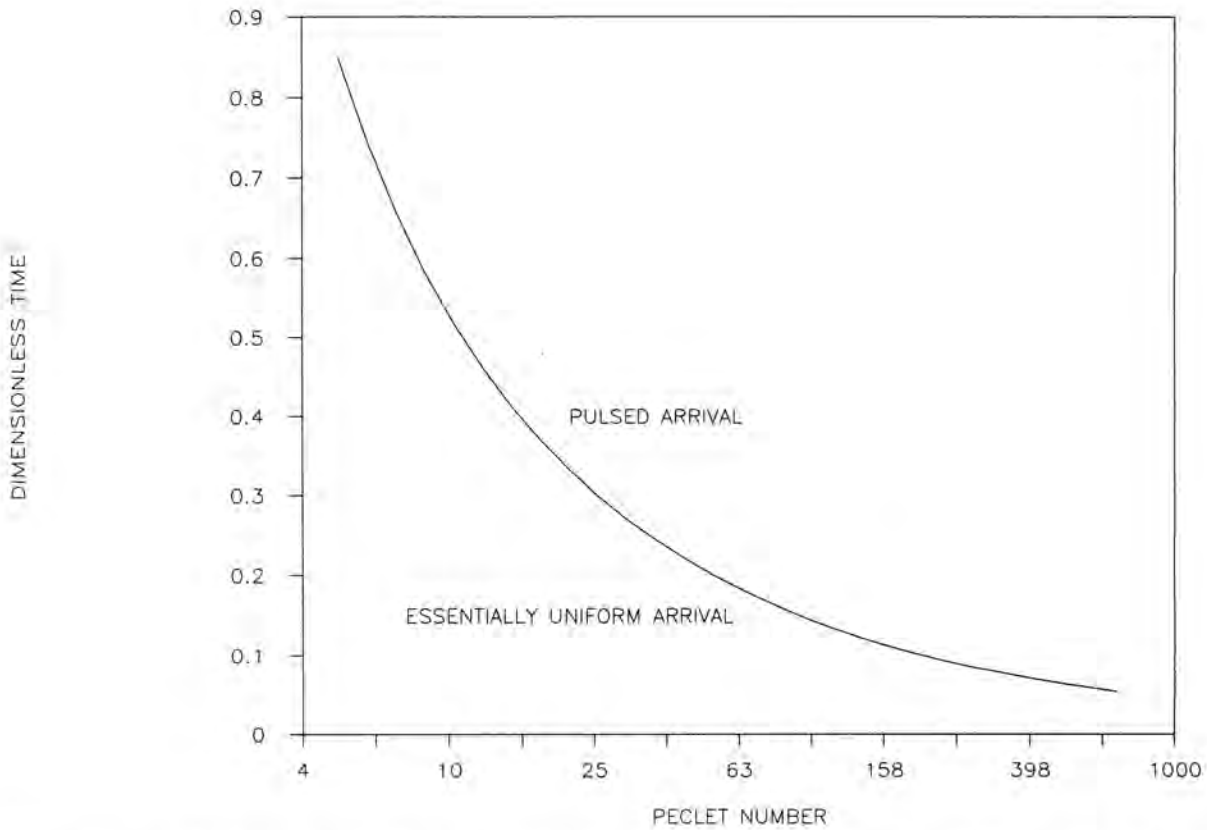

Fig. 3. Averaging Time for Instantaneous Peaks in Release Rate. Peaks of Shorter Duration Will Have Little Influence on Total System Performance.

$$\sigma = \left(\frac{2}{Pe} + \frac{8}{Pe^2}\right)^{0.5} \qquad (10)$$

Figure 3 shows the standard deviation in terms of dimensionless variables. Expressing release rate as a constant representing the cumulative release in an interval divided by the duration of the interval is reasonable if the individual impulses occur within one standard deviation of each other.

Fig. 3. Averaging time for instantaneous peaks in release rate. Peaks of shorter duration will have little influence on total system performance.

Flux is given by:

$$F = n U C = n D \frac{dC}{dx}$$

$$= \sum_{i=1}^{k} \frac{M_i (1 + T_i)}{t_{gw} R_d 4 T_i (\pi T_i / Pe)^{0.5}} \exp\left(\frac{(1 - T_i)^2}{4 T_i / Pe} - G T_i\right) \qquad (11)$$

where:

T_i = age of pulse = $(T - T_p)$

T_p = dimensionless time when pulse was released

T = total dimensionless time

M_i = amount of material released in pulse # i

Cumulative flux is obtained by integration over time:

$$CF = \sum_{i=1}^{k} \int_0^{T_i} \frac{M_i (1 + T)}{4 T (\pi T / Pe)^{0.5}} \exp\left(\frac{(1 - T)^2}{4 T / Pe} - G T\right) dT \qquad (12)$$

The cumulative flux at the accessible environment can be normalized by dividing by the cumulative release from the engineered barriers. To provide ease of interpretation, and because closely spaced peaks in release rate were previously shown to be unimportant, a constant engineered barriers release rate is assumed, leading to the following expression for dimensionless cumulative flux:

$$DCF = \frac{s}{T} \sum_{i=1}^{k} \int_0^{T_i} \frac{(1 + T)}{4 T (\pi T / Pe)^{0.5}} \exp\left(\frac{(1 - T)^2}{4 T / Pe} - G T\right) dT \qquad (13)$$

where:

s = spacing between impulses (years)

Figures 4 - 6 illustrate the importance of site Peclet number, radionuclide decay and dimensionless time upon the cumulative flux to cumulative release ratio measured at the accessible environment. Cumulative flux is obtained by multiplication of the flux ratio by cumulative release from the engineered barriers. The flux ratio can be interpreted as the proportion of the source material which has reached the accessible environment. With zero decay, the curves asymptotically approach one as time increases. Low Peclet numbers cause earlier arrival of the contaminant plume, but have little impact at longer time periods. A low Peclet number allows a significant amount of material to arrive ahead of the groundwater travel time.

When radionuclide decay in introduced, the dimensionless cumulative flux asymptotically approaches a limit less than one as time increases; the value of the asymptotic limit is dependent upon the decay rate and site Peclet number. In the presence of radionuclide decay, the Peclet number assumes a greater importance. Low values of the Peclet number result in greater cumulative flux at the accessible environment at all time periods. The high dispersion of the plume at low Peclet numbers results in some material arriving at the accessible environment rather rapidly, before it has time to fully decay. Therefore, when highly dispersive flow is present, radionuclide decay is less effective in removal of material during transport.

IMPORTANCE OF SENSITIVITY ANALYSIS IN ASSESSING COMPLIANCE WITH REGULATIONS

An example of the usefulness of the approach is illustrated using the NRC (subsystem) and EPA (total system) regulations for the US High Level Nuclear Waste Program. The relevance of the graphs of dimensionless cumulative flux to the regulations can be most easily grasped if the regulatory requirements are cast into the same dimensionless format as the graphs. This can be done in a variety of manners. One of the most useful is to normalize the total system allowable cumulative flux (EPA) requirements by the engineered barrier subsystem allowable release (NRC) requirements. This allows one to display the relationship between requirements imposed at the total systems level and requirements imposed on subsystem performance. For example, an allowable cumulative flux / release limit ratio less than one indicates that the engineered barrier subsystem requirements are not as stringent as those for the total system.

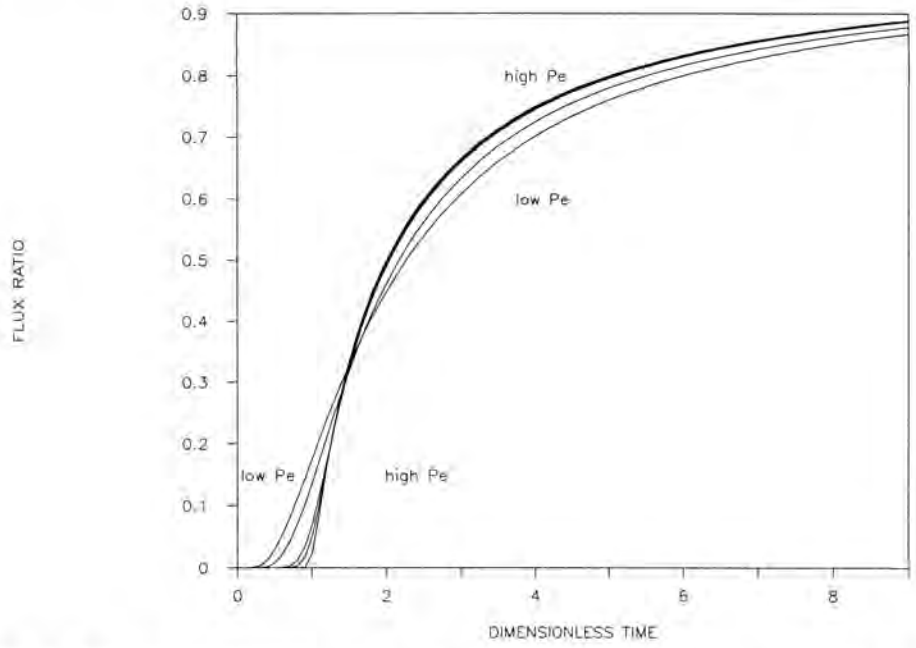

Fig. 4. Behavior of Dimensionless Flux Ratio in the Absence of Radionuclide Decay.

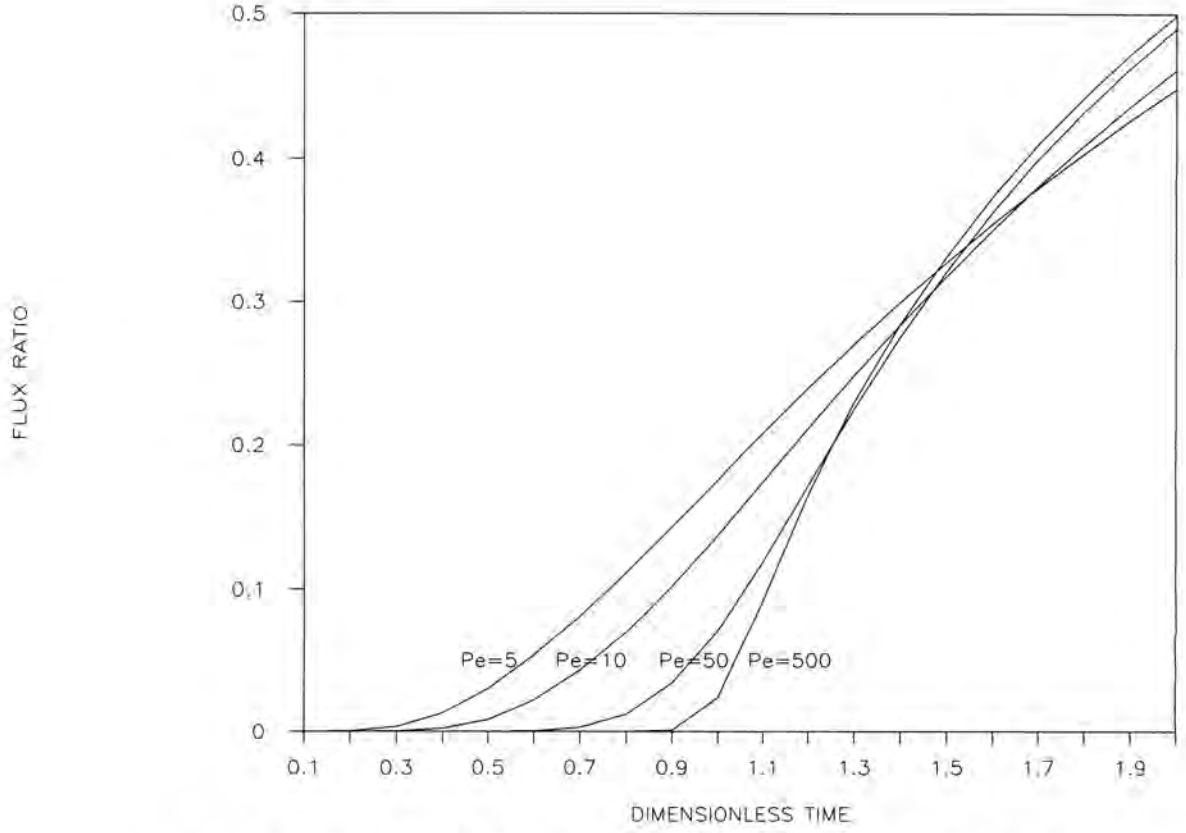

Fig. 5. Influence of Peclet on Dimensionless Flux Ratio in the Absence of Decay.

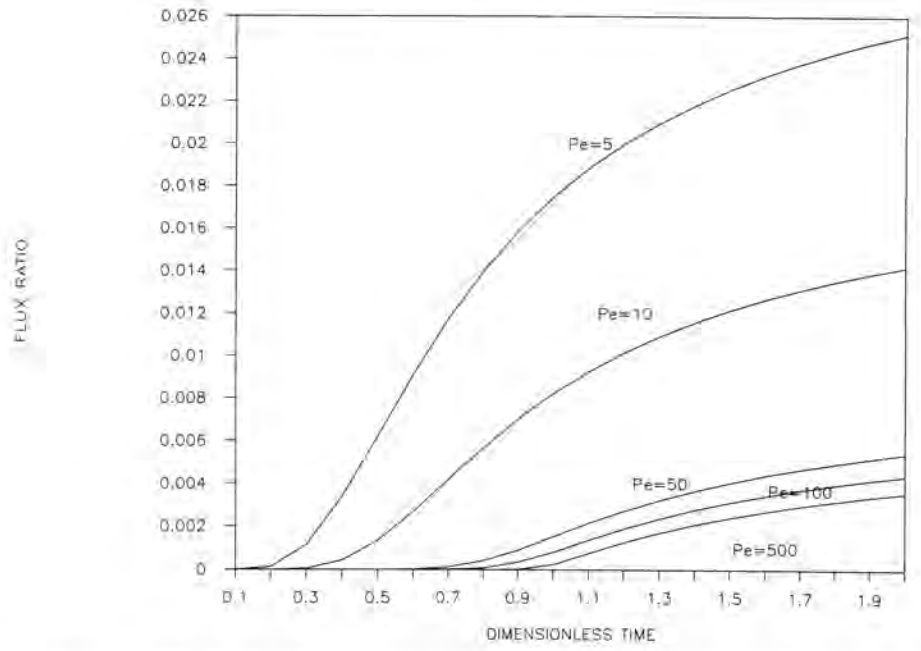

Fig. 6. Influence of Peclet Number on Dimensionless Flux Ratio at Dimensionless Decay of 5.

It makes clear the fact that the geological system must provide additional restrictions to radionuclide movement or the total system cumulative flux limit will be exceeded.

Table I is a list of key radionuclides with the EPA allowable limit normalized by the NRC limit for release rate from the engineered barriers. In particular, the table identifies radionuclides which have the potential for contributing significantly to the EPA limit.

The NRC allowable release rate and a zero year container lifetime are assumed throughout for the subsystem requirements. The important radionuclides subject to inventory limited release during the 10,000 year regulatory period are tin-126, selenium-79, and iodine-129.

Of the radionuclides listed, Technetium-99, Tin-126, Zirconium-93, and Selenium-79 all have an EPA allowable cumulative flux which exceeds the allowable cumulative release from the engineered barriers; even if these radionuclides were released from the repository at the NRC maximum permitted rate and radioactive decay were ignored, the EPA cumulative flux standard would not be exceeded. Additionally, Tin-126, Selenium-79 and Iodine-129 have estimated inventories which are below the EPA limit. If the entire inventory of any 2 of these 3 radionuclides were released immediately, the EPA standard could not be surpassed; if the total inventory of all 3 reached the accessible environment in less than 10,000 years, the EPA standard would be violated. In order of increasing EPA/NRC ratio, Americium-241, Plutonium-240, Plutonium-239,

TABLE I

Characteristics of Key Radionuclides

Nuclide	Actual Inventory (Ci/KMT)†	NRC Release Limit (Ci/KMT /yr)	NRC Release Limit in 10,000 yr (Ci/KMT)	EPA Limit (Ci/KMT)	EPA/NRC Ratio
Am-241	831000	8.31	83100	100	0.0012
Pu-240	412000	4.12	41200	100	0.0024
Pu-239	284000	2.84	28400	100	0.0035
Tc-99	12700	0.127	1270	10000	7.87
Zr-93	1650	0.017	170	1000	5.88
Np-237	1100	0.016	160	100	0.625
C-14	657	0.016	160	100	0.625
Sn-126	481	0.016	160	1000*	6.25
Se-79	347	0.016	160	1000*	6.25
I-129	33	0.016	160	100*	0.625

† Ci/KMT = Curies Per 1,000 metric tons
* Note that allowable limit exceeds actual inventory

Neptunium-237, and Carbon-14 have inventories which could contribute to violating the EPA standard.

Performance of a potential repository with respect to each of the radionuclides in Table I by comparison of the EPA/NRC ratio with the dimensionless flux ratio in figures such as Fig. 4 - 6. The methodology provides a rapid and convenient consequence screening tool for disruptive scenario analysis.

REFERENCES

1. CODELL, R. B. and J. D. DUGUID, "Transport of Radionuclides in Groundwater," in J. E. Till and H. R. Meyer, ed., Radiological Assessment: A Textbook on Environmental Dose Analysis, NUREG/CR-3332, ORNL-5968, (1983).

2. LEVENSPIEL, O. and K. B. BISHOFF, "Patterns of Flow in Chemical Process Vessels", in T. B. Drew, J. W. Hoopes, and T. Vermeulen, ed., Advances in Chemical Engineering, Academic Press, New York, (1963).