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Modeling One-Dimensional Radionuclide Transport Under Time-Varying Fluid-Flow Conditions

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ABSTRACT

An exact solution is derived for one-dimensional radionuclide transport under time-varying fluid-flow conditions including radioactive decay but with the approximation that all radionuclides have identical retardation factors. The solution is used to obtain exact expressions for the cumulative radionuclide mass transported past a fixed point in space over a given time period, and to assess the effects of a periodic perturbation and a step change on the fluid-flow velocity and dispersion coefficient.

CONTENTS

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1. INTRODUCTION

In assessing the long-term safety of nuclear-waste repositories, the validity of assuming steady-state geological conditions has not been established. For example, over the regulatory time frames of interest, typically thousands of years, one may expect changes in the fluid-flow rates through the repository due to changes in infiltration and recharge caused by intermittent rainfall. Such changes are difficult to predict, but that does not eliminate the need to assess how such changes may affect results from models that rely on constant conditions.

To address part of this problem, a new exact solution is developed to analyze the effects of time-varying conditions on the transport of a decaying radionuclide chain in one-dimensional flow. Few exact solutions are available for time-varying conditions [1, 2, 3, 4, 5], and no exact solutions with time-varying flow rates have been found in the literature that considers radioactive decay. Furthermore, for time-varying conditions, even without radioactive decay, no exact analysis has been found on the cumulative release of radionuclide past a fixed point in space. Such an analysis is important because the performance measure in the containment requirements of the Environmental Protection Agency's standard for the disposal of high-level, spent fuel, and transuranic wastes is given in terms of cumulative radionuclide releases over 10,000 years [6]. The analysis in this work extends available exact solutions to include the release of decaying radionuclides from a repository of arbitrary length under time-varying conditions, and may be used to calculate the cumulative release of radionuclides past a given fixed point in space.

The solution is developed for arbitrary time-varying fluid-flow velocities and dispersion coefficients. However, the solution is constrained to radionuclides that have identical adsorption distribution coefficients or retardation factors. Such an approximation may be used as a conservative estimate of the fastest transport from the repository by assuming that no radionuclide adsorbs on the porous media. Thus, the retardation factors are all unity. In addition, a less conservative, but more realistic approximation is that each radionuclide has a retardation factor equal to the minimum retardation factor of all the radionuclides. This approximation may also be used for the solution presented in this work.

The solution is presented and discussed in Section 2 in four subsections. The governing equation is presented in the first subsection. In the second subsection, the derivation begins with an existing exact solution of the migration of radionuclides from a single instantaneous injection point in a time-varying flow, without radioactive decay [2]. In the third subsection, a general method is derived for extending transport solutions, such as those given in the second subsection, to include radioactive decay. In the last subsection, exact expressions and asymptotic limits for the cumulative mass of radionuclide

past an arbitrary point are presented. The new contributions of this study are a unified approach to existing exact transport solutions with time-varying flow, a general method for extending transport solutions to include radioactive decay, and an analysis for the cumulative radionuclide mass transported past a fixed point.

Four special cases of the solution are discussed in Sections 3 and 4. The first case is for time-invariant flow conditions and serves as a base case for the three time-varying flow cases. A periodic perturbation of the fluid-flow velocity is used in the second and third cases, and a step change in this velocity is used in the fourth case. In Section 4 two representative examples are used to demonstrate applications of the time-varying solution. Finally, in Section 5 the results of this work are summarized.

2. ANALYTICAL SOLUTION

2.1 Governing Equation

For time-varying one-dimensional convective and dispersive transport of a radionuclide chain through an adsorbing porous media, the governing equation for a chain of m radionuclides is [1, 7, 8, 9, 10],

$$R_i \frac{\partial C_i}{\partial t} + U(t) \frac{\partial C_i}{\partial x} = D(t) \frac{\partial^2 C_i}{\partial x^2} - R_i \lambda_i C_i + R_{i-1} \lambda_{i-1} C_{i-1} \tag{1}$$

where $-\infty < x < \infty$,
$i = 1, \ldots, m$,
$\lambda_o = 0$,
t is time,
C_i is the molar concentration in solution of the i-th radionuclide,
R_i is the retardation factor of the i-th radionuclide, and
λ_i is the radioactive decay rate of the i-th radionuclide.

To model time-varying flow, the fluid-flow velocity U(t), and dispersion coefficient D(t) are assumed to be time dependent. It is also assumed that the adsorbed radionuclide concentration on the porous media is proportional to C_i. The so-called retardation factor is then given by

$$R_i = 1 + \frac{\rho_s K_{di}}{\phi} \tag{2}$$

where ϕ is the porosity, ρ_s is the bulk density and K_{di} is the adsorption distribution coefficient for the i-th radionuclide.

2.2 Exact Solution Without Radioactive Decay

The initial and boundary conditions for a point source of radionuclide i of mass per unit area given by M_i, released at $t = 0$ and $x = 0$ into an infinite domain initially containing no radionuclide are

$$C_i(x,0) = \delta(x) M_i \tag{3}$$

$$\int_{-\infty}^{\infty} C_i(x,t)\, dx = M_i \tag{4}$$

$$C_i(x \to \pm\infty, t) = 0 \tag{5}$$

where $\delta(x)$ is the Dirac delta function defined by

$$\delta(0) = \infty \tag{6}$$

$$\delta(x) = 0 \quad \text{for } x \neq 0 \tag{7}$$

$$\int_{-\infty}^{\infty} \delta(x)\, dx = 1 \tag{8}$$

Equation (4) constrains the solution such that all radionuclides in the initial point source remain for all time in the domain $-\infty < x < \infty$.

The solution to Equation (1) without radioactive decay, satisfying Equations (3), (4), and (5) is given by the Green's function [2],

$$C_i(x,t) = \frac{M_i}{\sqrt{4\pi \bar{D}_i t}} \exp\left\{ - \frac{(x-\bar{U}_i t)^2}{4\bar{D}_i t} \right\} \tag{9}$$

where the time-averaged species velocity and retarded dispersion coefficient are given respectively by

$$\bar{U}_i = \frac{\int_o^t U(\tau)\, d\tau}{R_i t} \tag{10}$$

and

$$\bar{D}_i = \frac{\int_o^t D(\tau)\, d\tau}{R_i t} \tag{11}$$

The solution for arbitrary initial conditions may be constructed from Equation (9) by summing point sources over the region of nonzero initial radionuclide concentration. For a repository of length h releasing radionuclide in the region $-h \le x \le 0$, the limit of the summation process results in

$$C_i(x,t) = \int_x^{x+h} \frac{C_{i,s}}{\sqrt{4\pi \bar{D}_i t}} \exp\left[\frac{-(\xi-\bar{U}_i t)^2}{4\bar{D}_i t} \right] d\xi \tag{12}$$

where $C_{i,s}$ is the initial concentration of the i-th radionuclide due to a point source located at a distance ξ from x. $C_{i,s}$ is assumed to have a constant value in the repository over the region $-h \le x \le 0$. Integrating Equation (12) results in [4],

$$\frac{C_i}{C_{i,s}} = \frac{1}{2}\left\{\text{erf}\left[\frac{x+h-\bar{U}_i t}{\sqrt{4\bar{D}_i t}}\right] - \text{erf}\left[\frac{x-\bar{U}_i t}{\sqrt{4\bar{D}_i t}}\right]\right\} \tag{13}$$

where erf is the error function defined by

$$\text{erf}(\zeta) = \frac{2}{\sqrt{\pi}}\int_0^{\zeta} e^{-\beta^2}\, d\beta \tag{14}$$

As $h \to \infty$ Equation (13) reduces to

$$\frac{C_i}{C_{i,s}} = \frac{1}{2}\left\{1 - \text{erf}\left[\frac{x-\bar{U}_i t}{\sqrt{4\bar{D}_i t}}\right]\right\} \tag{15}$$

This special limit for a repository of infinite length was obtained previously by using a Fourier Transform in x [1, 5].

Although the solution is for an infinite domain, the solution may also be used to model the semi-infinite domain $x \geq 0$. For a semi-infinite domain that initially does not contain any radionuclide, the initial and boundary conditions are,

$$C_i(x,0) = 0 \qquad x > 0 \tag{16}$$

$$C_i(x \to \infty, t) = 0 \qquad t \geq 0 \tag{17}$$

$$\frac{C_i(0,t)}{C_{i,s}} = \frac{1}{2}\left\{1 + \text{erf}\left[\frac{\bar{U}_i t}{\sqrt{4\bar{D}_i t}}\right]\right\} \qquad t > 0 \tag{18}$$

From Equation (18) we see that the concentration at the boundary $x = 0$ increases asymptotically to $C_{i,s}$ as $t \to \infty$. The solution to Equation (1) for these boundary and initial conditions is given by Equation (15). This solution may be used as a test case for numerical solutions of Equation (1) since numerical solutions are better suited for semi-infinite domains, than for infinite domains.

2.3 Exact Solution With Radioactive Decay

If the adsorption distribution coefficient of all radionuclides is approximated as being equal, then $K_{di} = K$ for all i, the time-averaged species velocities and dispersion coefficients are independent of the radionuclide, and the subscript i may be dropped from these variables. Then the solutions for different radionuclides given by Equation (13), differ only in the initial radionuclide concentrations. To construct what will be called in this work the fundamental transport solution, which is not radionuclide specific, C_{i_s} in Equation (13) is replaced by C_o, a unit measure of concentration. With this replacement, the fundamental transport solution is the solution for the problem of an instantaneous release resulting in an initial unit radionuclide concentration. This fundamental transport solution is given by

$$C_f(x,t) = \frac{C_o}{2}\left\{\operatorname{erf}\left[\frac{x+h-\bar{U}t}{\sqrt{4\bar{D}t}}\right] - \operatorname{erf}\left[\frac{x-\bar{U}t}{\sqrt{4\bar{D}t}}\right]\right\} \tag{19}$$

for a repository of finite length, and by

$$C_f(x,t) = \frac{C_o}{2}\left\{1 - \operatorname{erf}\left[\frac{x-\bar{U}t}{\sqrt{4\bar{D}t}}\right]\right\} \tag{20}$$

for a repository of infinite length.

The general solution to Equation (1) may be obtained by assuming that it is a product of the fundamental transport solution and an unknown time dependent factor that is radionuclide specific, $\bar{C}_i(t)$, Thus

$$C_i(x,t) = \bar{C}_i(t)C_f(x,t) \tag{21}$$

Substituting Equation (21) into Equation (1) results in the Bateman equations [11], given by the following coupled set of ordinary differential equations for $\bar{C}_i(t)$:

$$\frac{d\bar{C}_1}{dt} = -\lambda_1\bar{C}_1 \tag{22}$$

$$\frac{d\bar{C}_i}{dt} = \lambda_{i-1}\bar{C}_{i-1} - \lambda_i\bar{C}_i \qquad i > 1 \tag{23}$$

For $C_i(x,t)$, given by Equation (21), to reduce to Equation (13) without radioactive decay, the initial conditions for Equations (22) and (23) are

$$\bar{C}_i = C_{i,s} \qquad t = 0, \quad i \geq 1 \tag{24}$$

Since each radionuclide has a distinct decay rate, the i-th eigenvalue of the system of equations is equal to the decay rate of the i-th radionuclide, λ_i. The solution to Equations (22) and (23) is

$$\bar{C}_i = \sum_{j=1}^{i} \alpha_j b_i^{(j)} \exp(-\lambda_j t) \qquad i \geq 1 \tag{25}$$

where the eigenvectors are given by

$$b_i^{(j)} = \begin{cases} 0 & i < j \\ 1 & i = j \\ \prod_{k=j}^{i-1} \dfrac{\lambda_k}{\lambda_{k+1} - \lambda_j} & i > j \end{cases} \tag{26}$$

and

$$\alpha_1 = C_{1,s} \tag{27}$$

$$\alpha_i = C_{i,s} - \sum_{j=1}^{i-1} \alpha_j b_i^{(j)} \qquad i > 1 \tag{28}$$

Therefore, the general solution is given by substituting Equations (19) and (25) into Equation (21) to give

$$C_i(x,t) = \frac{1}{2}\left\{\operatorname{erf}\left[\frac{x+h-\bar{U}t}{\sqrt{4\bar{D}t}}\right] - \operatorname{erf}\left[\frac{x-\bar{U}t}{\sqrt{4\bar{D}t}}\right]\right\} \sum_{j=1}^{i} \alpha_j b_i^{(j)} \exp(-\lambda_j t) \tag{29}$$

where C_o has been dropped from Equation (29) since by definition it is unity.

Equation (29) is the new general solution for instantaneous releases of a decaying radionuclide chain transported by time-varying convection and dispersion processes, but with uniform retardation factors. The

repository length, initial release concentrations of each radionuclide, and the chain length are arbitrary.

2.4 Cumulative Release of Radionuclides Past a Fixed Point

As discussed earlier, a primary concern for nuclear-waste repositories is the cumulative radionuclide mass reaching the accessible environment. This quantity for one-dimensional transport of the i-th radionuclide past the point x = L is given by the cross-sectional area for flow times

$$f_i(t) = \int_o^t \frac{U(\tau)C_i(L,\tau)}{R_i} - \frac{D(\tau)}{R_i}\frac{\partial C_i(L,\tau)}{\partial x}\, d\tau \qquad (30)$$

where L is taken as the location of the accessible environment, and $f_i(t)$ is the cumulative sum of the convective and dispersive mass fluxes of radionuclide i. Since $U(\tau)$ and $D(\tau)$ are arbitrary functions of time, the integral in Equation (30) can not be evaluated until these functions are specified. Furthermore, numerical integration may be required since $C_i(L,\tau)$, $U(\tau)$, and $D(\tau)$ may be given in terms of complicated functions that are not explicitly integrable.

However, the cumulative mass of radionuclide i past a point L, per unit cross-sectional area may be evaluated explicitly and is given by,

$$F_i(t) = \int_L^\infty C_i\, dx = \bar{C}_i(t) \int_L^\infty C_f(x,t)\, dx \qquad (31)$$

Without radioactive decay, $f_i(t)$ and $F_i(t)$ are equal, and the cumulative activity reaching the accessible environment may be computed using either expression. With radioactive decay, $f_i(t)$ may not be equal to $F_i(t)$. $f_i(t)$ accounts for the radionuclide mass in the region $x \geq L$ due to convection and dispersion, but not due to radioactive decay. However, $F_i(t)$ does account for radioactive decay in this region.

$F_i(t)$ may be evaluated explicitly by substituting Equation (29) into Equation (31), and using the integral representation for the error function given by Equation (14) to give

$$F_i(t) = \frac{\bar{C}_i}{\sqrt{\pi}} \int_L^{\infty} \int_{\frac{x-\bar{U}t}{\sqrt{4\bar{D}t}}}^{\frac{x+h-\bar{U}t}{\sqrt{4\bar{D}t}}} e^{-\beta^2} \, d\beta \, dx \tag{32}$$

The double integral in Equation (32) is over an upward sloping semi-infinite strip in the (x,β) plane. This region may be integrated in two parts by reversing the order of integration to give

$$F_i = \frac{\bar{C}_i}{\sqrt{\pi}} \int_{\frac{L+h-\bar{U}t}{\sqrt{4\bar{D}t}}}^{\infty} \int_{\beta\sqrt{4\bar{D}t} - h + \bar{U}t}^{\beta\sqrt{4\bar{D}t} + \bar{U}t} e^{-\beta^2} \, dx \, d\beta + \frac{\bar{C}_i}{\sqrt{\pi}} \int_{\frac{L-\bar{U}t}{\sqrt{4\bar{D}t}}}^{\frac{x+h-\bar{U}t}{\sqrt{4\bar{D}t}}} \int_{L}^{\beta\sqrt{4\bar{D}t} + \bar{U}t} e^{-\beta^2} \, dx \, d\beta \tag{33}$$

Since the integrands are independent of the inner integration variable, the inner integrals may be evaluated to give

$$F_i = \frac{\bar{C}_i}{\sqrt{\pi}} \int_{\frac{L+h-\bar{U}t}{\sqrt{4\bar{D}t}}}^{\infty} h e^{-\beta^2} \, d\beta + \frac{\bar{C}_i}{\sqrt{\pi}} \int_{\frac{L-\bar{U}t}{\sqrt{4\bar{D}t}}}^{\frac{L+h-\bar{U}t}{\sqrt{4\bar{D}t}}} \left[\beta\sqrt{4\bar{D}t} + \bar{U}t - L\right] e^{-\beta^2} \, d\beta \tag{34}$$

Evaluating the single integrals in Equation (34) results in

$$F_i = \frac{\bar{C}_i}{2}\left\{h[1-\mathrm{erf}(z)] + (L-\bar{U}t)[\mathrm{erf}(y)-\mathrm{erf}(z)] + \left[e^{-y^2} - e^{-z^2}\right]\sqrt{\frac{4\bar{D}t}{\pi}}\right\} \quad (35)$$

where

$$y = \frac{L-\bar{U}t}{\sqrt{4\bar{D}t}} \quad (36)$$

and

$$z = \frac{L+h-\bar{U}t}{\sqrt{4\bar{D}t}} \quad (37)$$

For an infinitely long repository, F_i in Equation (35) reduces to

$$F_i = \frac{\bar{C}_i}{2}\left\{(L-\bar{U}t)[\mathrm{erf}(y) - 1] + e^{-y^2}\sqrt{\frac{4\bar{D}t}{\pi}}\right\} \quad (38)$$

The asymptotic values of $F_i(t)$ are given in Table 1. These asymptotic limits may be obtained by using the following approximation for the error function [12],

$$\mathrm{erf}(y) \rightarrow 1 - \frac{e^{-y^2}}{y\sqrt{\pi}} \qquad (y \gg 1) \quad (39)$$

As $t \rightarrow \infty$ for finite values of L of a finite repository, all the radionuclides must pass $x = L$. Thus, in this limit, $F_i(t)$ must be equal to all the radionuclide mass per unit area formed or decayed by nuclear reactions. As given in column one and row one of Table 1, this quantity is the decayed initial radionuclide concentration times the length of the repository. For an infinite repository, as $t \rightarrow \infty$, the radionuclides transported past a fixed point are given by the effective travel distance past $x = L$, times the decayed initial radionuclide concentration. This quantity approaches infinity for an infinite repository, as given in Table 1. Also shown in Table 1 is that the asymptotic limits for $F_i(t)$ as $L \rightarrow -\infty$ are identical to those limits given for $t \rightarrow \infty$. This is because in both asymptotic limits, all the radionuclides are contained in the region of integration of Equation (31). Notice from Table 1 that for long times, F_i is independent of the dispersion coefficient. This long time behavior will be demonstrated in Section 4 with an example problem.

Table 1

Asymptotic Values of $F_i(t)$, the Cumulative

Radionuclide Mass Per Unit Area for $x \geq L$.

Limiting Conditions	Finite Repository ($h > 0$)	Infinite Repository ($h \rightarrow \infty$)
($t \rightarrow \infty$) (L finite)	$h\bar{C}_i$	$\bar{C}_i(\bar{U}t - L)$
($L \rightarrow -\infty$) ($t \geq 0$) (t finite)	$h\bar{C}_i$	$\bar{C}_i(\bar{U}t - L)$

$\bar{C}_i$ is given by Equation (25),

$\bar{U}$ is the time-averaged species velocity,

h is the length of the repository,

t is time, and

L is the point beyond which the cumulative radionuclide mass per unit area is determined.

3. SPECIAL CASES

One can not predict with absolute certainty future geological conditions that may influence flow through a repository. The sensitivity of models to changing geological conditions is therefore of interest. To study this sensitivity for one-dimensional models, the solutions given in the previous section were developed for arbitrary time-dependent functional forms of the fluid-flow velocity and dispersion coefficient. We may now use these solutions to determine how much the constant flow solution differs from a solution obtained using a time-varying perturbation or a step change on the parameters. To evaluate this difference, four special cases are considered in this section. Table 2 summarizes the conditions for each case.

First, a base case is defined in terms of a constant species velocity u, and constant retarded dispersion coefficient $d_o+d_1|u|$. These parameters are defined such that d_o and d_1 are nonnegative constants.

The second and third cases in Table 2 are for a periodic perturbation of the species velocity given by

$$U(t)/R = u + \varepsilon\cos(\omega t) \tag{40}$$

where ω is a nonnegative constant. For $\varepsilon = 0$, the species velocity for the second and third cases reduce to that for the first case in which the flow conditions are constant. To maximize the early time difference between the base case and the time-varying case, the cosine function was chosen instead of the sine function in Equation (40). As will be shown in section 4, even with this maximum difference in the fluid-flow velocity at t=0, the time-varying solution rapidly approaches the base case solution.

For the fourth case, the species velocity will change from u_o to u_1 at time t_1. This case may be used to model an abrupt change in geological conditions.

The retarded dispersion coefficient is often related to the species velocity. For the second and fourth cases this relationship is given by [13],

$$D(t)/R = d_o + d_1|U(t)/R| \tag{41}$$

and for the third case the relationship is given by

$$D(t)/R = d_o + (d_1/u)[U(t)/R]^2 \tag{42}$$

Table 2

Species Velocities and Retarded
Dispersion Coefficients for Special Cases

Case	U/R	D/R	$\bar{U}$	$\bar{D}$	$\bar{U}$ ($\omega t \to \infty$)	$\bar{D}$ ($\omega t \to \infty$)
1	u	d_o+d_1u	u	d_o+d_1u	u	d_o+d_1u
2	$u+\varepsilon\cos(\omega t)$	$d_o+d_1[u+\varepsilon\cos(\omega t)]$	$u + \frac{\varepsilon\sin(\omega t)}{\omega t}$	$d_o+d_1u + \frac{d_1\varepsilon\sin(\omega t)}{\omega t}$	u	d_o+d_1u
3	$u+\varepsilon\cos(\omega t)$	$d_o + \frac{d_1}{u}[u+\varepsilon\cos(\omega t)]^2$	$u + \frac{\varepsilon\sin(\omega t)}{\omega t}$	$d_o+d_1u + \frac{d_1\varepsilon^2}{2u} + \frac{d_1\varepsilon}{4u\omega t}[\varepsilon\sin(2\omega t)+8u\sin(\omega t)]$	u	$d_o+d_1u + \frac{d_1\varepsilon^2}{2u}$
4 ($t < t_1$)	u_o	$d_o+d_1u_o$	u_o	$d_o+d_1u_o$		
4 ($t \geq t_1$)	u_1	$d_o+d_1u_1$	$\frac{u_o t_1 + u_1(t-t_1)}{t}$	$\frac{(d_o+d_1u_o)t_1 + (d_o+d_1u_1)(t-t_1)}{t}$		

Note that u, u_o, u_1, d_o and d_1 are nonnegative constants and for the second case $u \geq |\varepsilon|$.

Note that the constants u, d_o and d_1 are chosen such that as $\varepsilon \to 0$, the perturbed cases reduce to the constant condition case.

The new general solutions for the concentration profile in Equations (29) and the cumulative mass per unit area in Equations (35) and (38) are expressed in terms of time-averaged quantities given by Equations (10) and (11). The time-averaged quantities for the four cases are given in Table 2.

From Table 2 we see that for cases 2 and 3 in the limit of long times (i.e. $\omega t \to \infty$), the time-averaged species velocity approaches the constant value of u.

For case 2 the long time time-averaged retarded dispersion coefficient approaches a constant value of d_o+d_1u, which is identical to that for case 1. However, due to the quadratic model used in case 3, the long time time-averaged retarded dispersion coefficient is not equal to that for case 1.

4. EXAMPLE PROBLEMS

Two example problems are used to demonstrate the significance of time-varying flow conditions for an infinite repository. Table 3 lists the parameters for the example problems. In the first example, a periodic fluid-flow velocity is used with the linear and the quadratic models for the retarded dispersion coefficent. The parameter ε was chosen arbitrarily such that the fluid-flow velocity would oscillate with a 100% variation about u for the first example, as shown in Figure 1. The frequency of oscillation was also chosen arbitrarily, but for specific sites one may wish to use a different value of ω [14]. In the second example, an order of magnitude step change in the fluid-flow velocity half way through the simulation is used. For both examples d_o = 0.03 m^2/year, d_1 = 10 m and L = 5,000 m. From these values, the base-case species velocity and dispersion coefficient are 1 m/year and 10.03 m^2/year, respectively.

The cumulative mass of radionuclide past L = 5,000 m for time-varying conditions relative to that for constant conditions is shown in Figure 2 for Example 1, and in Figure 3 for Example 2. This ratio is independent of the radionuclide decay rate and the initial radionuclide concentration. Deviations from unity of this ratio indicate deviations of the time-varying solution from the constant flow solution. The lines in the Figures were generated using Equation (38) for both the time-varying and constant flow conditions. Also plotted in Figure 2 as discrete points is the same ratio calculated based on the asymptotic formula given in Table 1. For a periodic fluid-flow velocity given by Equation (40), the asymptotic value of the ratio is given by

$$F_{\text{time-varying}}/F_{\text{constant}} \rightarrow 1 + \frac{\varepsilon \sin(\omega t)}{u\omega t} \tag{43}$$

The solid and dashed lines in Figure 2 are for the linear and the quadratic models of the dispersion coefficient, respectively. Notice that little difference was found between using a linear or a quadratic model for the dispersion coefficient as given by Equations (41) and (42), respectively. As can be seen from Figure 2, the asymptotic expression in Equation (43) provides an excellent approximation at long times. Furthermore, as expected from the asymptotic analysis given in section 2.4, F_i is not sensitive to the model used for the dispersion coefficient. Notice that although there is a 100% variation in the flow conditions, the oscillations dampen quickly after one or two cycles in the fluid-flow rate. Thus, although the analysis in section 3 shows that for long times the solution should approach that for constant conditions, this example demonstrates that the constant flow solution may be a good approximation in this case after only one cycle in the fluid-flow velocity.

Table 3

Parameters in Example Problems

No.	Fluid-Flow	Parameters	Time Period (years)	Figures
1	Periodic	$\omega = 2\pi/10{,}000$ year^{-1}	100,000	1, 2
		$u = 1$ m/year		
		$\varepsilon = 1$ m/year		
2	Step Change	$u_o = 0.1$ m/year	10,000	3
		$u_1 = 1$ m/year		
		$u = 0.55$ m/year		
		$t_1 = 5{,}000$ years		

For both examples, $d_o = 0.03$ m^2/year, $d_1 = 10$ m and $L = 5{,}000$ m.

In Figure 3 the cumulative mass ratio is shown for Example 2. In this example, for constant conditions, the fluid-flow velocity is 0.55 m/year, which is the average fluid-flow velocity over 10,000 years. Thus, for the first 5,000 years, the fluid-flow velocity for constant conditions greatly exceeds the initial fluid-flow velocity of 0.1 m/year for time-varying conditions. Therefore, the ratio shown in Figure 3 is much less than unity for about the first 8,500 years. However, the cumulative fluid-flows are equal for the constant and time-varying cases at 10,000 years. At that time the ratio shown in Figure 3 is unity, which indicates for this example that at 10,000 years the cumulative radionuclide release is not affected significantly by the step change in the fluid-flow velocity.

5. SUMMARY AND CONCLUSIONS

An exact solution has been obtained for radionuclide transport under time-varying fluid-flow velocities and dispersion coefficients, including radioactive decay. The solution was based on a unified treatment of previously reported transport solutions without radioactive decay. New exact expressions were obtained for the cumulative radionuclide mass per unit area past a fixed point in the flow. These new expressions were used to determine the effects of a periodic perturbation and a step change of the fluid-flow rate on the cumulative radionuclide mass per unit area past a fixed point.

For the example presented of a periodic variation in the fluid-flow rate, the time-varying solution for the cumulative radionuclide mass past a fixed point dampened rapidly, and approached the constant flow solution regardless of the model for the dispersion coefficient.

For the example presented of a step change in the fluid-flow velocity, the cumulative radionuclide mass past a fixed point reached that for the constant flow solution when the cumulative fluid-flows were identical.

The examples demonstrated that the solutions presented in this work are useful for assessing the effects of time-varying flow, but are limited to radionuclide chains with uniform retardation factors. Numerical solutions may be required to account for nonuniform retardation factors. These numerical solutions may be tested by using the exact solutions in this work for cases when the retardation factors are uniform.

REFERENCES

1. Bear, J. and Todd, D. K., "The Transition Zone between Fresh and Salt Waters in Coastal Aquifers," Hydraulic Laboratory, University of California Berkeley, Water Resources Center Contribution No. 29, September 1, 1960.

2. Bischoff, K. B., "Axial Dispersion with Time Variable Flow," Chemical Engineering Science, pp. 989-990, Vol. 19, 1964.

3. Turner, J. C. R., "A Note on Axial Dispersion with Time Variable Flow," Chemical Engineering Science, pp. 65-66, Vol. 20, 1965.

4. Gill, W. N., "Analysis of Axial Dispersion with Time Variable Flow," Chemical Engineering Science, pp. 1013-1017, Vol. 22, 1967.

5. Bear, J., Dynamics of Fluids in Porous Media, American Elsevier, Environmental Science Series, New York, p. 627, 1972.

6. Environmental Protection Agency, 40 CFR Part 191, Environmental Standard for the Management and Disposal of Spent Nuclear Fuel, High-Level Waste and Transuranic Radioactive Waste; Final Rule, Environmental Protection Agency, Federal Register, Washington, DC., Vol. 50, No. 182, 1985.

7. Rogers, V. C., "Migration of Radionuclide Chains in Groundwater," Nuclear Technology, Vol. 40, pp. 315-320, October 1978.

8. Pigford, T. H., Chambre, P. L., Albert, M., Foglia, M., Harada, M., Iwamoto, F., Kanki, T., Leung, D., Masuda, S., Muraoka, S., and Ting, D., "Migration of Radionuclides Through Sorbing Media: Analytical Solutions--II," Lawrence Berkeley Laboratory, Earth Sciences Division, LBL-11616, UC-70, Volumes 1 and 2, October 1980.

9. Gureghian, A. B. and Jansen, G., "One-Dimensional Analytical Solutions for the Migration of a Three-Member Radionuclide Decay Chain in a Multilayered Geologic Medium," Water Resources Research, Vol. 21, No. 5, pp. 733-742, May 1985.

10. van Genuchten, M. Th. and Alves, W. J., "Analytical Solutions of the One-Dimensional Convective-Dispersive Solute Transport Equation," U. S. Department of Agriculture, Agricultural Research Service, Technical Bulletin Number 1661, 1982.

11. Bateman, H., "The Solution of a System of Differential Equations Ocurring in the Theory of Radioactive Transformations," Proceedings of the Cambridge Philosophical Society, Vol. 15, Cambridge University Press, New York, 1910.

12. Abramowitz, M. and Stegun, I. A., (Editors), Handbook of Mathematical Functions, Dover Publications, Inc., New York, p. 298, 1972.

13. Bear, J., Dynamics of Fluids in Porous Media, American Elsevier, Environmental Science Series, New York, p. 606, 1972.

14. Bartlein, P. J., Webb, T., Hostetler, S., "Climatology," in "Techniques for Determining Probabilities of Events and Processes Affecting the Performance of Geological Repositories," Hunter, R. L. and Mann, C. J. (Editors), Sandia National Laboratories, Albuquerque, NM, NUREG/CR-3964, SAND86-0196, Vol. 1, June 1989.

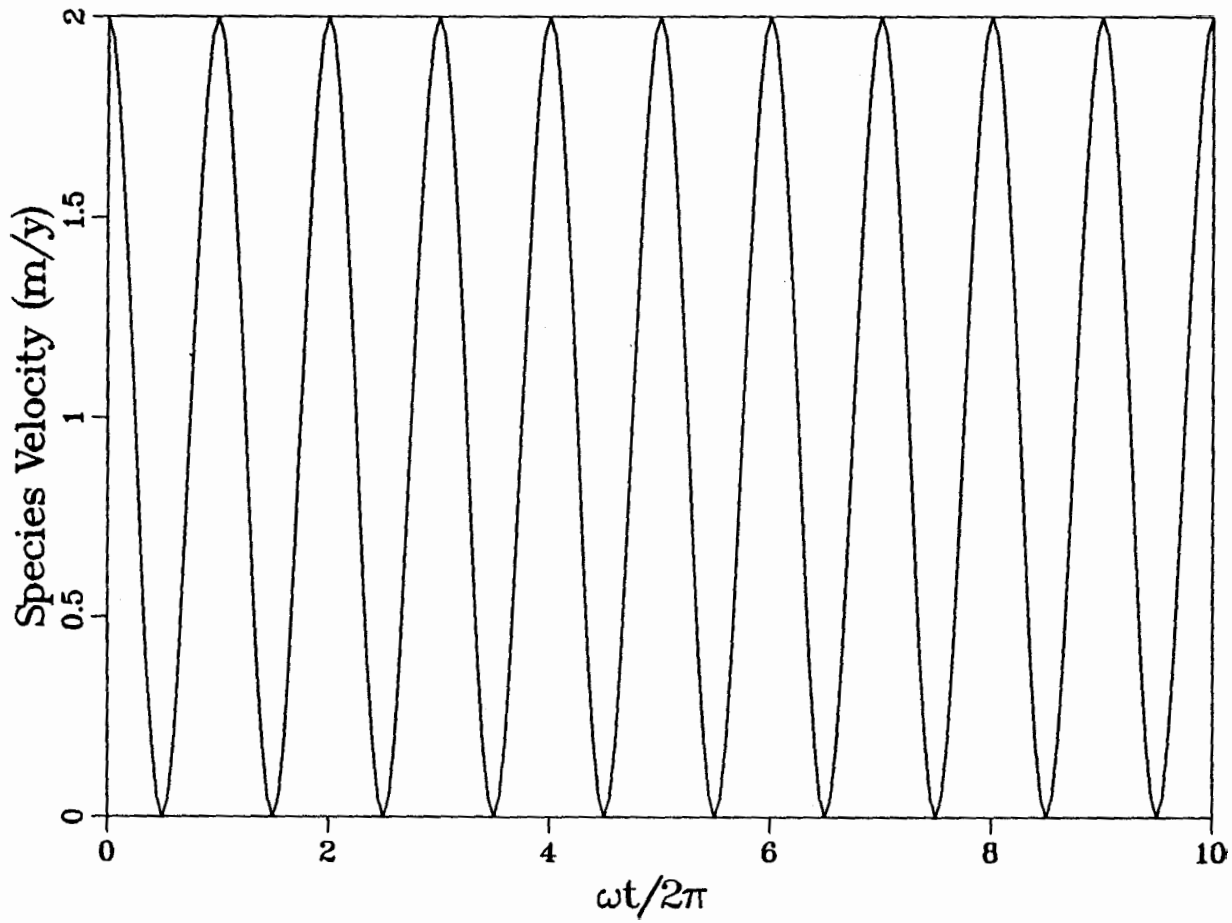

Figure 1. Species velocity for first example problem given by $u+\varepsilon\cos(\omega t)$, where $u = 1$ meter/year, $\varepsilon = 1$ meter/year, and t is time.

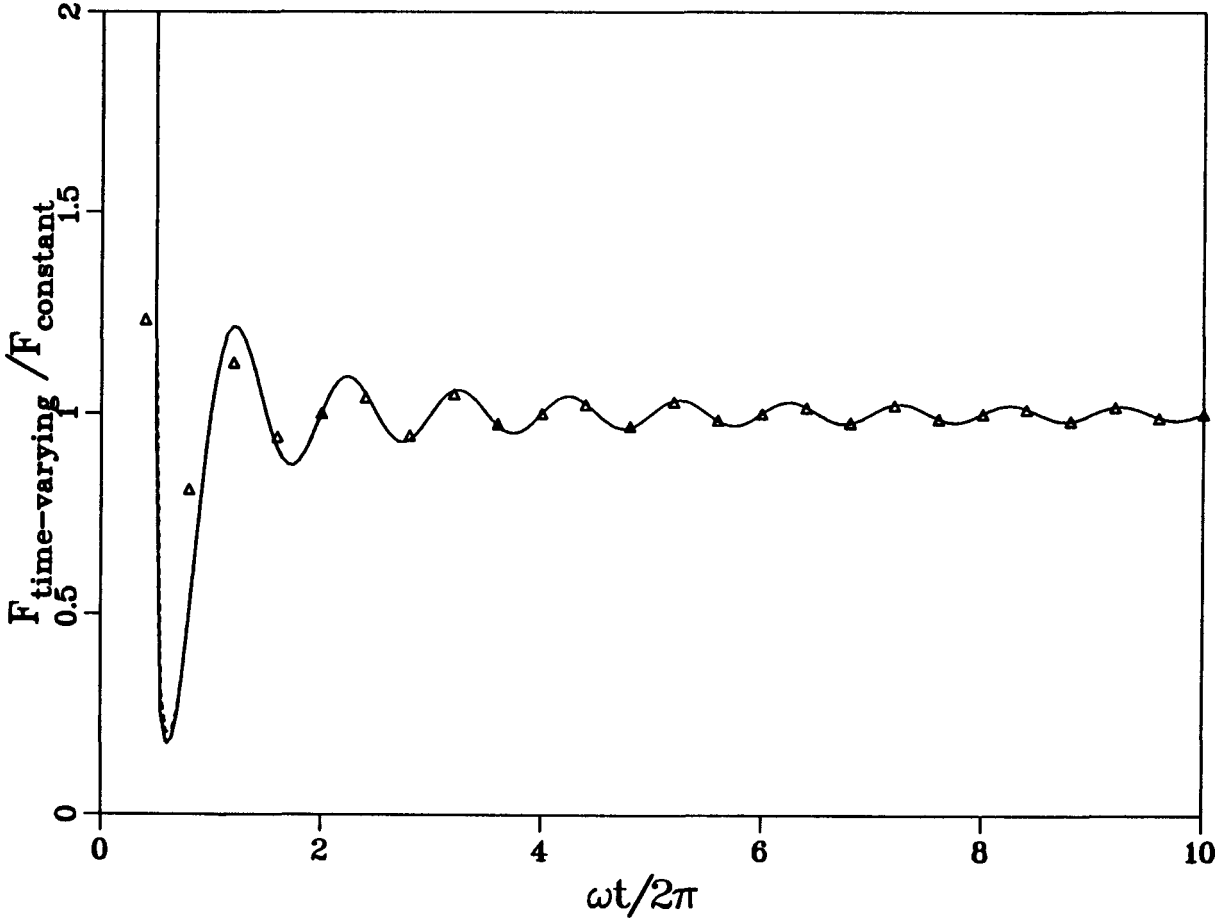

Figure 2. Ratio of cumulative radionuclide mass past L = 5,000 meters for the first example problem, where the solid and dashed lines are for the linear and quadratic models of the dispersion coefficients, respectively, as given by Equations (41) and (42), respectively. The discrete points were calculated using the asymptotic approximation given by Equation (43).

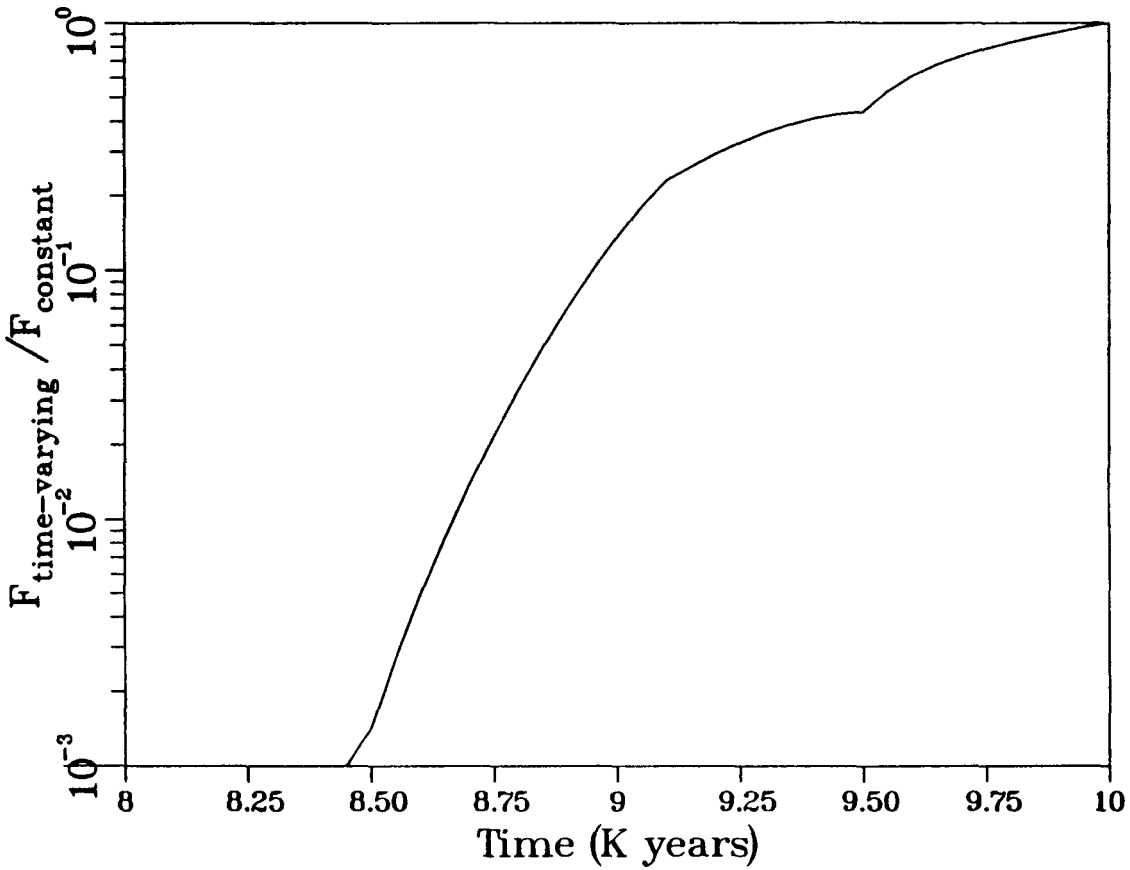

Figure 3. Ratio of cumulative radionuclide mass past L = 5,000 meters for the second example problem. At 5,000 years the fluid-flow velocity changed from 0.1 meter/year to 1.0 meter/year. The constant flow conditions were for a fluid-flow velocity of 0.55 meter/year.

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