ESTIMATION OF RADIONUCLIDE TRANSPORT IN GROUNDWATER USING NUMERICAL LAPLACE TRANSFORM INVERSION

B. Ganapol and D. Ferrara
Department of Nuclear and Energy Engineering
University of Arizona

ABSTRACT

A numerical Laplace transform inversion was used to solve the one-dimensional advection-diffusion equation for radionuclide transport for infinite, semi-infinite, and finite geometric configurations describing waste migration. Comparsion of the results from the numerical Laplace transform inversion and the analytical solution to the infinite media configuration showed the deviation to be extremely small. This method allows concentrations and outflow rates to be calculated quickly, inexpensively, and without finite differencing in either time or space. A comparison has also been made between a media of five regions each with different Prandtl, retardation/decay, and diffusion numbers and a single region in which these values have been averaged.

INTRODUCTION

An important component in the characterization of a waste disposal site for radioactive nuclides regarding its level of isolation is the potential for radionuclide transport in groundwater. This requirement stems from considerations when the repository containment has been presumed to have failed. Subsequently, radioactive material diffuses out of the failed repository into the far-field environment. Needless to say, the transport process is extremely complicated since it involves radioactive decay, lineal dispersion, advection, sorption, and chemical interactions in a network of intersecting fractures and flow paths in material matrices of varying properties. Obviously, unless one is willing to evaluate the transport process with a detailed monte carlo model and spend enormous amounts of computational resources to obtain meaningful statistics, a simplified modeling approach is required. Such an approach based on radionuclide transport in a flow tube (1,2) has been shown to provide an acceptable model. This model assumes that the lumped parameter advection-dispersion equation is the appropriate mathematical setting. The model includes advection by groundwater, sorption on the rock matrix surfaces, and diffusion through the rock matrix itself.

Here, we consider the advection-diffusion equation without sorption and matrix diffusion primarily from the numerical point of view. We are interested in accurate solutions for a variety of conditions. The approach to obtain the solution will be through a recently developed numerical Laplace transform inversion. This technique is applied to several one-dimensional problems each with an increasing level of modeling detail. We consider the one-dimensional transport of a single radionuclide for the following geometrical configurations:

- infinite homogeneous medium,
- semi-infinite homogeneous medium,
- finite homogeneous medium.

Sensitivity studies are performed for each of the above. In order to determine the influence of transport through distinct material zones, a heterogeneous model is also developed.

ADVECTION/DIFFUSION EQUATION

The general advection/diffusion equation for a single radionuclide can be written as

$$R \frac{\partial}{\partial t} N(x,t) = -\lambda R N(x,t) - v \frac{\partial N}{\partial x}(x,t) + D \frac{\partial^2}{\partial x^2} N(x,t) - \frac{1}{b}\left[\frac{\partial S(x,t)}{\partial t} + \lambda S(x,t)\right] + \frac{D_m}{b} \left.\frac{\partial N_p(x,\omega,t)}{\partial \omega}\right|_{\omega=0} \qquad \text{(Eq.1)}$$

where

N	is the nuclide concentration,
S	is the number of atoms sorbed per square meter,
N_p	is the number of atoms dissolved in micropores,
λ	is the decay constant,
v	is the groundwater velocity,
D	is the lineal dispersivity,
D_m	is the intrinsic rock matrix diffusion coefficient,
R	is the retardation factor,
b	is the average fracture dimension,
x	is the distance,
t	is the time, and
ω	is the perpendicular to flow.

A complete description requires a non-equilibrium equation for S as well as the appropriate diffusion equation for rock matrix diffusion. The inclusion of both sorption and rock matrix diffusion will be the subject of a future effort and, therefore, will not be considered here. Our focus will now be on Eq. (1) (without the terms involving S or N_p), and the data base describing the transport properties will be assumed known.

Equation 1 is rendered dimensionless with the transformation

$t \rightarrow t/t_{gw}R$

$x \rightarrow x/L$

N -- > NALR/M
$G = t_{gw}R$
$p = vL/D$

where t_{gw} is the groundwater travel time,
L is the characteristic length,
A is the cross-sectional flow tube area,
M is the amount of radionuclide released,
G is the retardation/decay number, and
p is the Peclet number.

With these substitutions Eq. (1) becomes

$$\frac{\partial N}{\partial t} + \frac{\partial N}{\partial x} = \frac{1}{p}\frac{\partial^2 N}{\partial x^2} - GN \qquad \text{(Eq.2)}$$

Equation 2 is to be solved subject to boundary conditions appropriate to a particular geometric configuration. A Laplace transform reduces the number of independent variables yielding

$$\frac{d^2\bar{N}(x,s)}{dx^2} = p\,\frac{d\bar{N}(x,s)}{dx} + p(G+s)\,\bar{N}(x,s) = 0 \qquad \text{(Eq.3)}$$

where we have assumed no initial radionuclide present in the medium, $\bar{N}$ is the Laplace transform of N and s is the transform variable. With boundary conditions, this second order ordinary differential equation can easily be solved. The difficulty associated with obtaining the final solution is in the inversion back to time. Because of the complexity of the transform, it cannot be inverted analytically. Thus, a numerical inversion scheme will be employed; and it will be described in some detail in the next section.

THE NUMERICAL LAPLACE TRANSFROM INVERSION

From a casual scan of the applied mathematics literature, one finds that there exists many different methods to invert Laplace transforms numerically. Inversion algorithms have been based on a wide variety of numerical methods including

- limit of an equivalence class,
- expansion in exponential functions,
- Gauss quadrature,
- expansion in orthogonal polynomials,
- Fourier series representation, and
- Pade approximation.

The primary reason for so many approaches is that any purely numerical inversion is ill-conditioned in the sense that a small numerical error in the transform will be amplified in the inverse unless the inversion is performed entirely by analytical means. Each of the methods referred to above has a particular way (choice of a parameter) of addressing this difficulty. In addition, each method is usually best suited to a specific class of functions. For an excellent review article the reader is referred to Ref. 3.

The inversion method developed here is a combination of several of the methods already referred to. With the increased efficiency of today's mainframe and mini-computers, it is now possible to introduce numerical procedures into inversion schemes which were not feasible just a few years ago. The numerical procedure begins with the Fourier cosine integral representation of the inversion integral (4)

$$f(t) = \frac{2e^{\gamma t}}{\pi t}\int_0^\infty d\omega\ \mathrm{Re}\ \bar{f}(\gamma+i\omega/t)\cos\omega \qquad \text{(Eq.4)}$$

where γ is the real part of the Bromwich contour chosen to be greater than the largest real part of any singularity of f. The representation above can be reformulated as an infinite series by decomposing the integral over the half periods of the cosine to yield

$$f(t) = \frac{2e^{\gamma t}}{\pi t}\cdot \left[\int_0^{\pi/2} d\omega\ \mathrm{Re}\ \bar{f}(\gamma+i\omega/t)\cos\omega + \sum_{k=1}^{\infty}(-1)^k\int_{-\pi/2}^{\pi/2} d\omega\ \mathrm{Re}\ \bar{f}\left(\gamma+i\,\frac{\omega+k\pi}{t}\right)\cos\omega\right] \qquad \text{(Eq.5)}$$

This expansion and subsequent evaluation of the integrals seems to have first been suggested by Hurwitz and Zweifel (5) who used a Gauss quadrature. In a modification to this algorithm, a Romberg integration scheme (6) will be used in place of the Gauss quadrature. The advantage of the series representation is that an acceleration scheme can be used to accelerate convergence of the series. The EK transformation essentially replaces a convergent series

$$S = \sum_{k=0}^{\infty}(-1)^k a_k$$

with

where p_x is a parameter to be chosen. Note that for $p_x = 0$, the original series is recovered, while, for $p_x = 1$, we find the

$$S = \frac{1}{1+p_x} \sum_{k=0}^{\infty} (-1)^k \left[\frac{1}{1+p_x}\right]^k \sum_{j=0}^{k} (-1)^j \frac{k!\, p_x^j}{j!(k-j)!} a_{k-j}$$

classical Euler Transformation (7). The EK transformation is supplemented with the van Wijngaarden modification (7) which chooses between adding a term of the original series (first sum) or a term of the EK transformation (second series) depending upon which addition gives the least local error. The acceleration of the series is an essential step in the numerical procedure.

As with any other method of numerical inversion, the proper choice of the Bromwich contour (γ) is crucial for an efficient inversion. To help guide the choice of γ, let us estimate the theoretical dependence of the error associated with truncation of the series representation for f(t) [Eq. (5)]. Thus for truncation at k_0,

$$f(t) = \frac{2\,e^{\gamma t}}{\pi t}\left\{\int_0^{\pi/2} d\omega \,\mathrm{Re}\, \bar{f}(\gamma+i\omega/t) \cos\omega + \right. \tag{Eq.6}$$

$$\left. + \sum_{k=1}^{k_0} \int_{(2k-1)\pi/2}^{(2k+1)\pi/2} d\omega \,\mathrm{Re}\, \bar{f}(\gamma+i\omega/t)\cos\omega + E_L(t)\right\}$$

where the truncation error is given by

$$E_L(t) \equiv \int_{T_L}^{\infty} d\omega \,\mathrm{Re}\, \bar{f}(\gamma+i\omega/t)\cos\omega$$

with

$$T_L \equiv (2k_0+1)\pi/2$$

For a smooth function f(t) where for $\alpha > 0$, and $t^{-\alpha+1}f(t)$ is analytic for all real t, we can write the following asymptotic form (5):

$$\bar{f}(s) \sim \frac{1}{s^\alpha} \sum_{n=0}^{N} \frac{\beta_i}{s^n} \tag{Eq.7}$$

Then substituting the asymptotic form into the error term given by Eq. (6) yields

$$E_L \sim \sum_{n=0}^{N} \beta_j\, t^{(j+\alpha)}\, I_n(\gamma t) \tag{Eq.8}$$

where

$$I_n(\gamma t) = \frac{1}{(\gamma t)^{n+\alpha-1}} \cdot \int_{T_L/\gamma t}^{\infty} du' \frac{\cos[(n+\alpha)\tan^{-1}u']}{(1+u'^2)^{(n+\alpha)/2}} \cos(\gamma t u') \; . \tag{Eq.9}$$

From this expression, we conclude that the error is controlled by the product of γ and t and it will decrease for increasing TL/γt or increasing γt. With this result taken into account, the appropriate contour will be chosen such that

$$\gamma = \bar{\gamma} + a/t \tag{Eq.10}$$

where α and γ are specified by the user with γ greater than γs. The specification of a 1/t dependence for γ is reasonable when one considers that from the Tauberian theorems, f(t) for small t comes primarily from large |s| (large γ) and for large t from small s (small γ).

There is one particular feature of this inversion algorithm that distinguishes it from the others. Theoretically, the actual Bromwich contour is immaterial as long as $\gamma > \gamma_s$. Numerically, however as has been shown by experience, the choice of the contour could be a determining factor regarding accuracy. For this reason, a contour iteration has been imposed. The inverse is obtained at J successive contours approaching γs where the contour is chosen according to

$$\gamma_0 = \gamma \tag{Eq.11}$$

$$\gamma_j = (\gamma_s + \gamma_{j-1})/2, \quad j = 1, 2, \ldots, J$$

At each contour, the dimensionless derivative

$$\left| \frac{f(t;\gamma_j) - f(t;\gamma_{j-1})}{f(t;\gamma_j)} \right| / \left|(\gamma_j - \gamma_{j-1})/\gamma_j\right| \tag{Eq.12}$$

is sampled in order to determine if it is below a specified tolerance ε_γ. Theoretically, of course, the numerator should vanish. The denominator is included to guard against false convergence, e.g., when f(t, γ_j) and f(t,γ j-1) are within the tolerance by virtue of the proximity of γ_j and γ_{j-1}. If the dimensionless derivative is not within ε_γ after J trials then

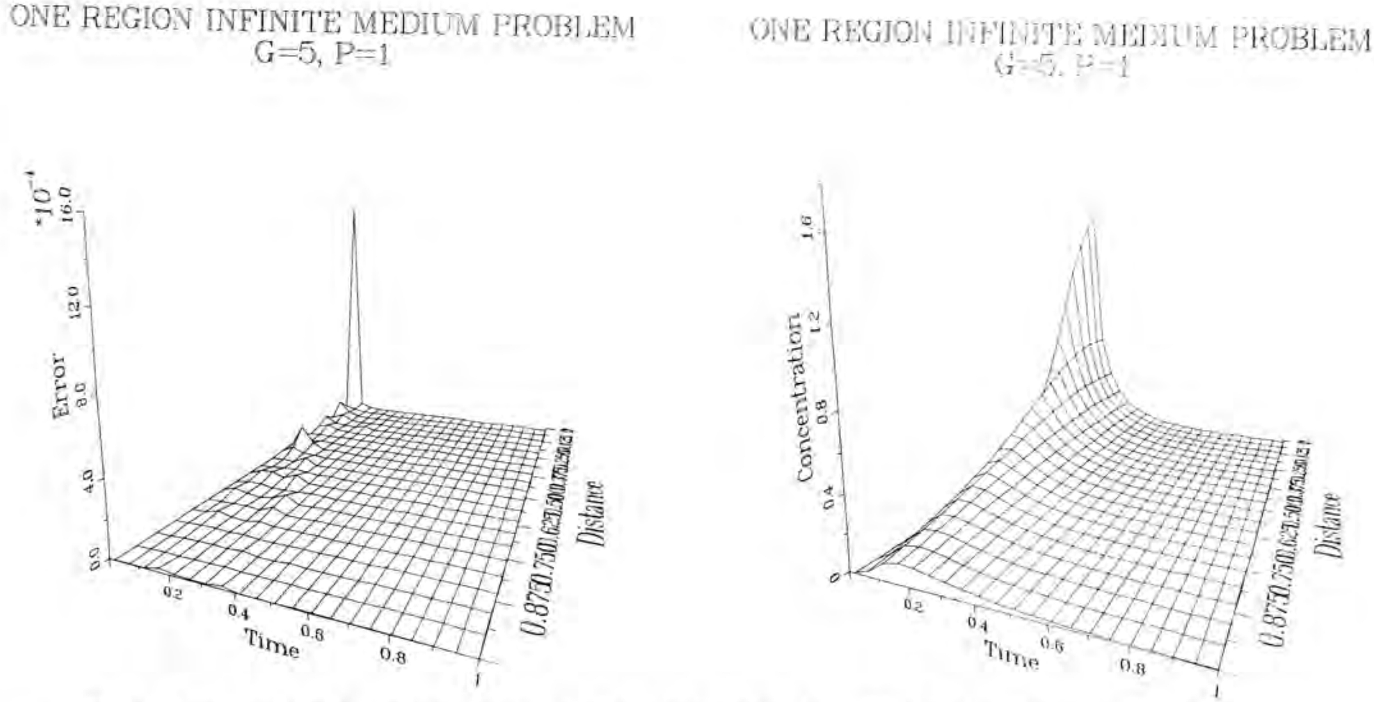

Fig. 1. Error Analysis for the One Region Infinite Medium Problem. (a) Absolute Error. (b) Concentration.

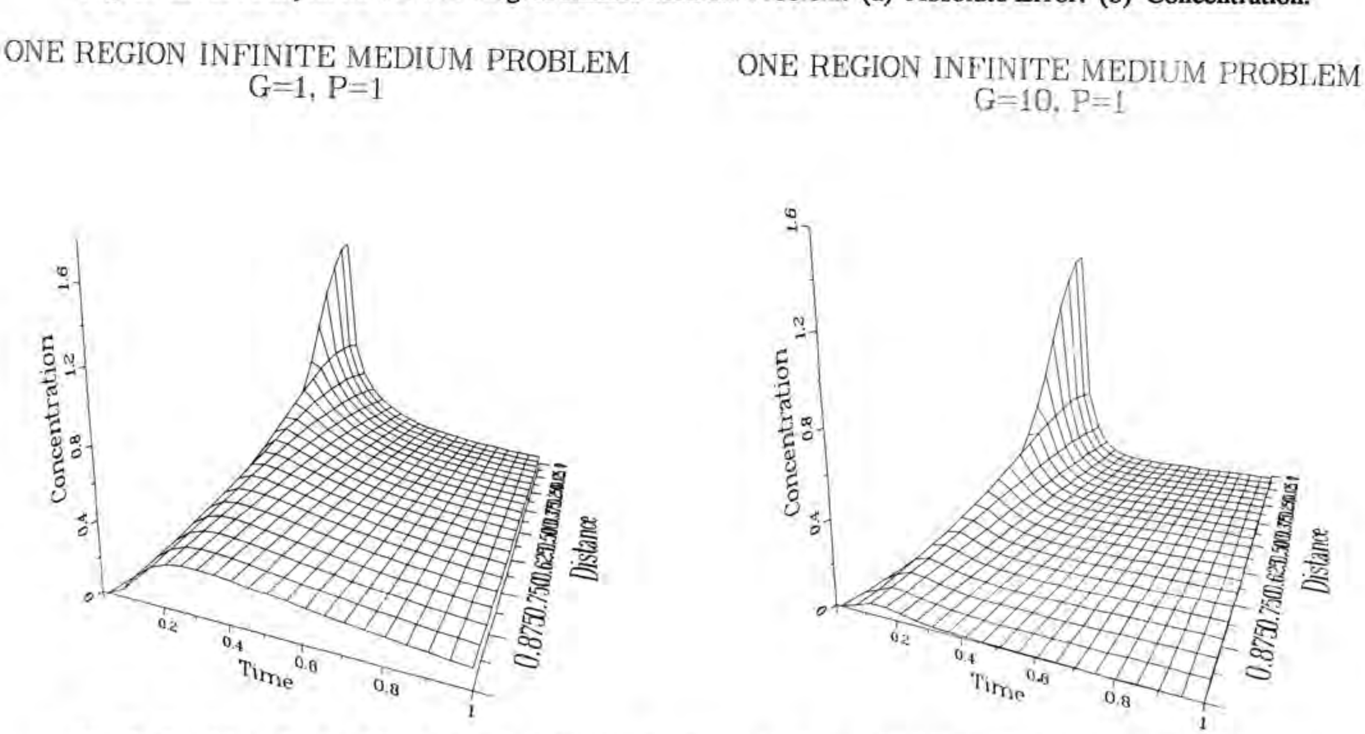

Fig. 2. Sensitivity Study of the Retardation/Decay Number. (a) Results for G = 1. (b) Results for G = 10.

tolerance by virtue of the proximity of γ_j and γ_{j-1}. If the contour is increased away from γ according to

$$\gamma_0 = 1.5\gamma$$

$$\gamma_j = \gamma + 1.5\,(\gamma_{j-1} - \gamma) \quad , \; j = 1, 2, \ldots J \; . \qquad \text{(Eq.13)}$$

The multiplying factor 1.5 was chosen from experience. The primary reason for including the contour iteration is to provide confidence in the result since if contour convergence is not achieved then most probably the numerical inversion is incorrect. If, however, contour convergence is achieved without any other error indication, then the user can be confident in the result. The addition of the contour iteration increases the required computational time but is well worth the expense if incorrect results can be avoided.

RESULTS AND DISCUSSION

Of the geometrical configurations examined, only the infinite homogeneous medium has a simple analytical solution. The initial condition used in this case was an infinite plane source pulsed at time zero. The solution to this problem is

$$N(x,t) = \frac{1}{2\sqrt{\pi t/p}} \exp\left[- \frac{(x-t)^2}{4t/p} - Gt \right] . \qquad \text{(Eq.14)}$$

Equation 14 was used as a benchmark for evaluating the Laplace transform inversion previously described. Representative deviations between the numerical Laplace inverse method and the analytical solution are presented in Fig. 1 along with the concentrations calculated from the inversion. The absolute errors were several orders of magnitude below the values of the concentration until the concentrations fell essentially to zero. Agreement with the analytical values can be improved by retaining more terms in the inversion series, however obtaining this increased accuracy would require more computational time.

The retardation/decay number, G, is a measure of the magnitude of radioactive decay and retardation of movement. As G increases so does the decay rate relative to the isotope displacement. More of the isotope now decays before moving a given distance. One would therefore expect to see the concentration fall off more rapidly with distance. This can be seen in the results of the infinite medium

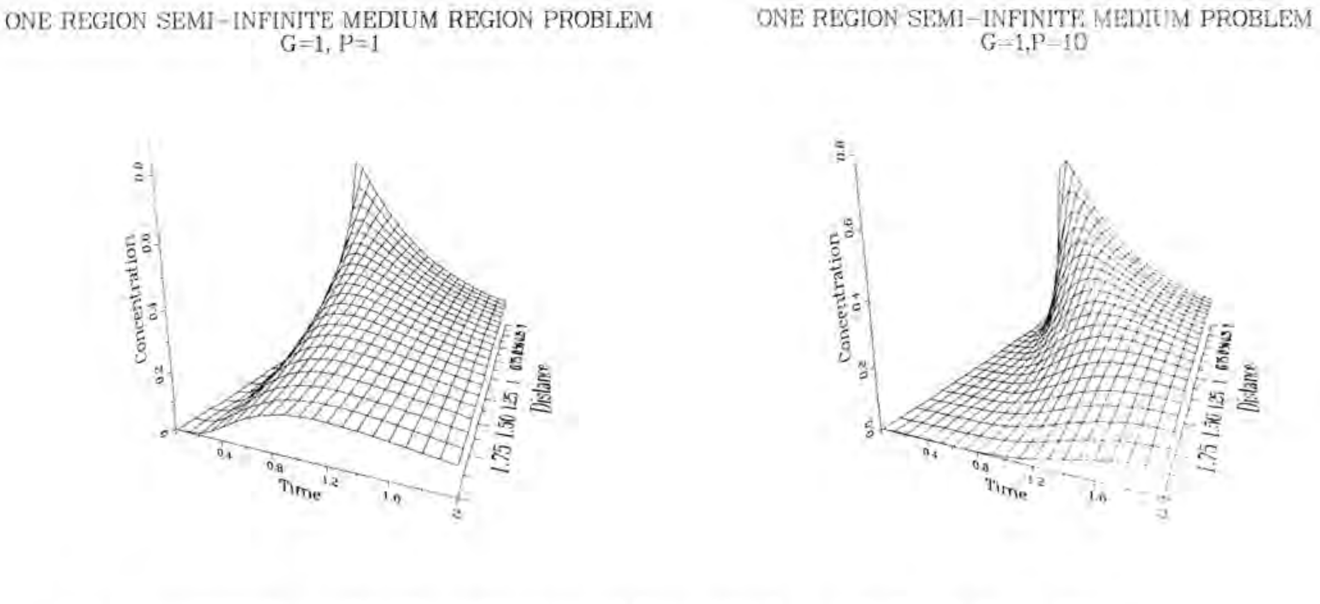

Fig. 3. Sensitivity Study of the Peclet Number. (a) Results for P = 1. (B) Results for P = 10.

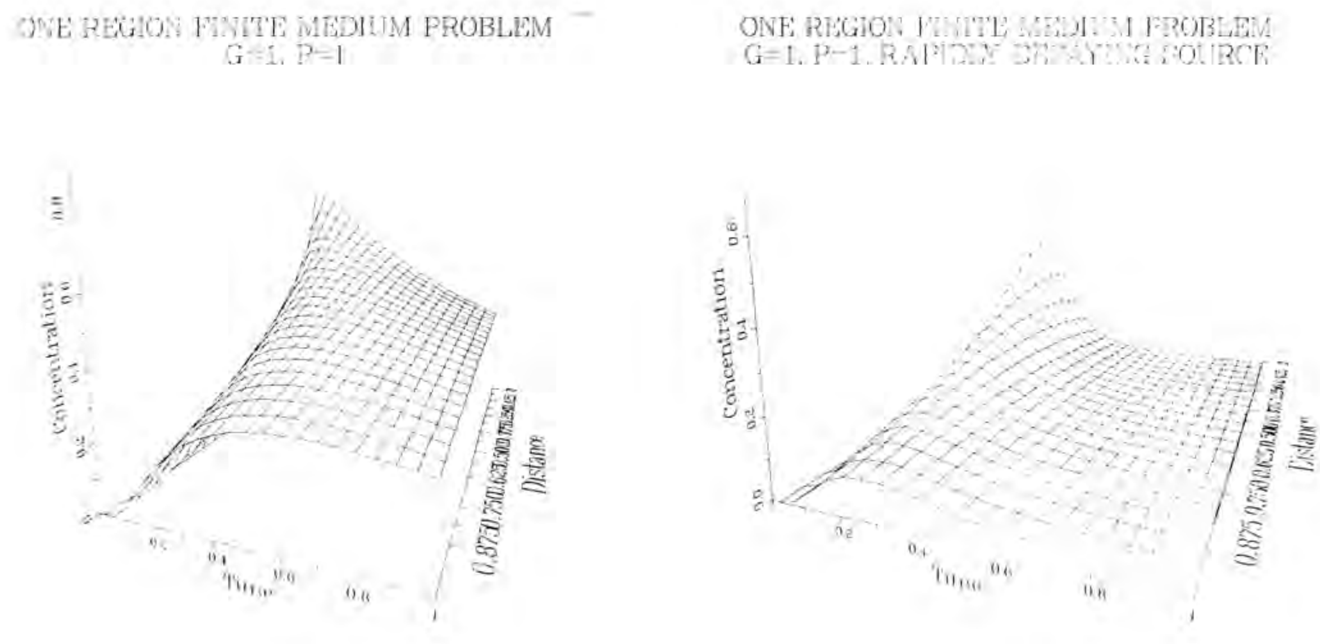

Fig. 4. Sensitivity Study of the Source Decay Rate. (a) Moderate Decay Rate. (b) Rapid Decay Rate.

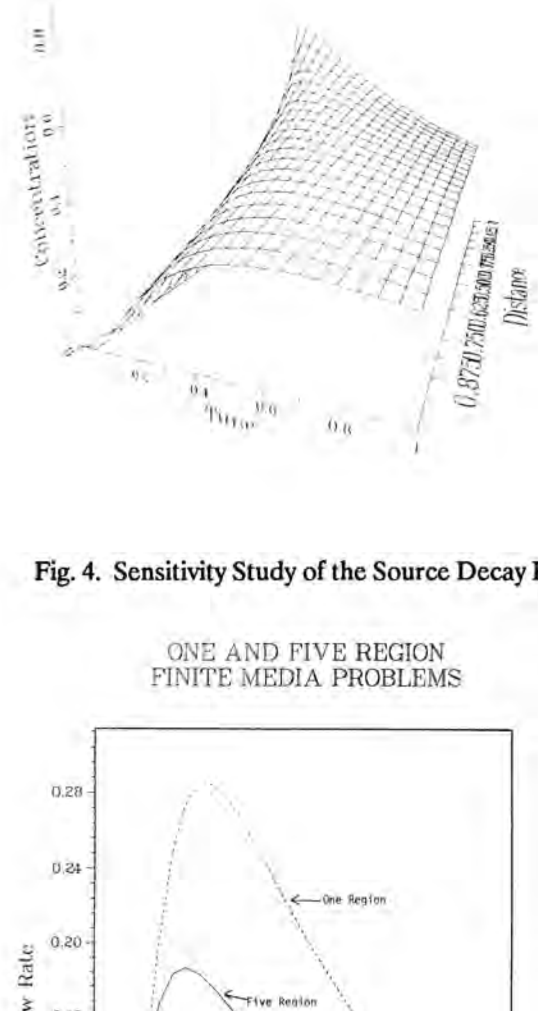

Fig. 5. Comparison of a One Region and a Five Region Finite Medium Analysis.

analysis shown in Fig. 2.

The Peclet number, p, indicates the importance of advection with regard to diffusion. Through advection the isotope moves along its flow path. Diffusion, on the other hand, spreads out the isotope relative to this path. In a medium with a higher p, the isotope migrates a given distance with a higher concentration. Thus, the concentration profile stays more peaked in time as the isotope travels through the medium. This phenomenon can be seen in Fig. 3 which is an example of results obtained from the semi-infinite medium cases with a exponentially time decaying source at $x=0$.

While a pulsed source was used in the infinite medium configuration, a finite source decaying exponentially in time was applied in all other cases. For higher decay rates, the concentration of the isotope falls off more rapidly with time. One would also expect to see a sharper drop in the concentration as a function of distance since a larger fraction of the isotope is released at the earlier times. The results for a finite medium of length 1, given in Fig. 4, is an example of this behavior.

In the interest of brevity only one example of each sensitivity study has been presented. In gathering this data the infinite, semi-infinite, and finite medium cases were all systematically examined by varying p and G. Each of these variables was assigned values of 1, 5, and 10. In addition, select cases for the semi-infinite and finite media were evaluated with a more rapidly decaying source. In each of the studies made, the results were similar to those given above. All trends were exactly as described.

Finally, a multiregion finite medium configuration with uniform p and G within a region was evaluated. This result is given in Fig. 5. Table 1 gives the regional values of D, p, and G, for a far-field zone composed of five regions. In addition the volume weighted average of these parameters are also presented. In Figure 5, R_I, the out flow rate to the accessible environment boundary (x=1),

$$R_I = \left[p_I \, N_I(x,t) - \frac{\partial N_I}{\partial x} \right|_{x=1} \right] \quad \text{(Eq. 15)}$$

is displayed. Region I is the last region before the accessible environment (at x=1). Also present in Figure 5 is the outflow assuming that the far-field region is one homogenous medium. From this comparison, the homogenous calculation provides a conservative result.

TABLE I

Parameters for Multiregion Analysis

Region	Diffusion	Peclet Number	Retardaion/Decay
1	5.8011	9.5051	7.8637
2	2.9762	4.5370	0.062619
3	2.7574	3.0565	6.8910
4	3.8266	1.3290	8.3186
5	5.8298	0.98625	2.7655
Average	4.7240	3.4932	4.8950

CONCLUSION

The numerical Laplace transform inversion provides a quick, inexpensive method for determining the release rate of an isotope to the accessible environment at a given time without requiring calculation of concentrations at previous times. The technique is also applicable to multimedia problems.

Work is currently underway to make the analysis quicker and improve the reliability of the multimedia applications.

ACKNOWLEDGEMENTS

The authors would like to thank Randal Baker for supplying the three-dimensional plotting program used to display and evaluate the results.

REFERENCES

1. D. P. Hodgkinson, et al, Ann. Nucl. Energy, 11, 111 (1984).
2. J. C. Walton, et al, Waste Management Conference, 1988.
3. B. Davies and B. Martin, J. Comp. Phys., 33, 1 (1979).
4. H. Dubner and J. Abate, J. ACM, 15, 115 (1968).
5. H. Hurwitz, and P. Zweifel, M.T.A.C., 10, 170 (1956).
6. A. Miller, Fortran Program for Scientist and Engineers, Sybex, Berkley, Ca., 1982.
7. W. Press, et al, Numerical Recipes, Cambridge Univ. Press, 1986.