

THE SUPERPOSITION SOLUTION OF THE TRANSPORT OF A  
RADIONUCLIDE CHAIN THROUGH A SORBING MEDIUM

M. Foglia, F. Iwamoto, M. Harada,  
P. L. Chambre, and T. H. Pigford

November 1979

RECEIVED  
LAWRENCE  
BERKELEY LABORATORY

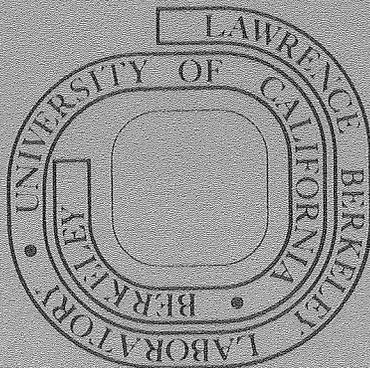
FEB 25 1980

LIBRARY AND  
DOCUMENTS SECTION

Prepared for the U.S. Department of Energy  
under Contract W-7405-ENG-48

**TWO-WEEK LOAN COPY**

*This is a Library Circulating Copy  
which may be borrowed for two weeks.  
For a personal retention copy, call  
Tech. Info. Division, Ext. 6782.*



LBL-10346 C. 2

## DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

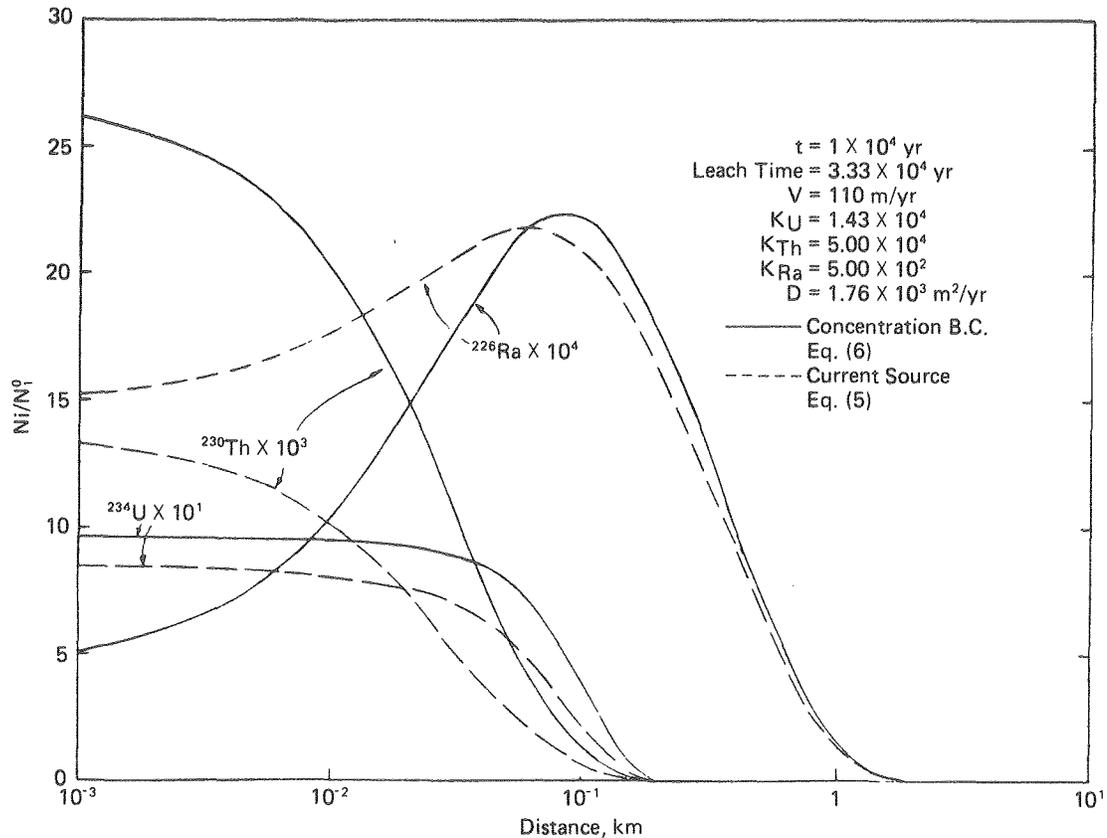


Fig. 1. Comparison of concentration profiles for different source boundary conditions (pure  $^{234}\text{U}$  source at  $t = 0$ ).

$W^0/F$ , where  $T$  is the leach time. The source term is then

$$F_i(t) = \frac{N_i(t)W^0}{T}, \quad (5)$$

where  $N_i(t)$  is the time-dependent amount of nuclide  $i$  per unit amount of waste material, given by the Bateman equation. In the special case of no diffusion or dispersion, the source term must equal the convection flow at  $z = 0$ , which yields the usual approximation of a concentration boundary condition

$$N_i(0, t) = \frac{F_i(t)}{v}. \quad (6)$$

The analytical solutions for the concentration boundary condition have been presented elsewhere.

The source boundary condition of Eq. (5) then results in the analytical solutions for a step release in Ref. 1, but with the function  $E(i, j; k)$  defined as

$$E(i, j; k) = \exp(z/2\kappa - \beta_{ij}t) \int_0^{\infty} \frac{2}{\sqrt{\pi}} \times \exp\{-[\gamma y^2 + (z/4\kappa y)^2]\} dy \\ = \frac{\exp(-\beta_{ij}t + z/2\kappa)}{2\sqrt{\gamma}} \left\{ \exp(-Z\sqrt{\gamma}/2\kappa) \operatorname{erfc} \left[ \frac{Z - v_k t \sqrt{\gamma}}{(4\kappa v_k t)^{1/2}} \right] \right. \\ \left. - \exp(Z\sqrt{\gamma}/2\kappa) \operatorname{erfc} \left[ \frac{Z + v_k t \sqrt{\gamma}}{(4\kappa v_k t)^{1/2}} \right] \right\} \\ (\gamma > 0). \quad (7)$$

The band-release solution is then obtained by applying the superposition method of Foglia et al.<sup>2</sup>

Figure 1 is an illustration showing the differences of the concentration profiles for the two different boundary conditions, calculated for the decay chain  $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ , and with assumed parameters listed on the figure. The concentration profiles for the two boundary conditions are appreciably different in the regions near the repository. At greater penetrations into the geologic medium, the errors due to the use of the concentration boundary condition become less important. The two solutions become identical in the case of no dispersion.

1. M. FOGLIA, F. IWAMOTO, M. HARADA, P. L. CHAMBRÉ, and T. H. PIGFORD, "The Superposition Solution of the Transport of a Radionuclide Chain Through a Sorbing Medium," UCB-NE-3348 (June 1979).
2. M. FOGLIA, T. H. PIGFORD, and P. L. CHAMBRÉ, "The Superposition Equation for the Band Release of Decaying Radionuclides Through Sorbing Media," UCB-NE-3935 (Mar. 1979).

## 6. The Superposition Solution of the Transport of a Radionuclide Chain Through a Sorbing Medium, M. Foglia, F. Iwamoto, M. Harada, P. L. Chambré, T. H. Pigford (UC-Berkeley)

The prediction of the space-time-dependent concentrations of radionuclides undergoing hydrogeologic transport is of primary concern in the evaluation of geologic disposal of



radioactive wastes. Analytical and numerical solutions have previously been published.<sup>1,2</sup> Here we present a new superposition technique<sup>3</sup> for developing an analytical solution, we demonstrate the application of this technique to the migration of a three-member decay chain, and we apply the resulting solution to demonstrate the effects of dispersion on the space-time-dependent concentration of three-member decay chain  $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ .

The one-dimensional transport equation for the aqueous concentration  $N_i(z, t)$  of radionuclide  $i$  in groundwater moving at velocity  $v$ , assuming local sorption equilibrium, is

$$K_i \partial N_i / \partial t - D \partial^2 N_i / \partial z^2 + v \partial N_i / \partial z + \lambda_i K_i N_i = \lambda_{i-1} K_{i-1} N_{i-1} \\ \lambda_0 = 0, \quad i = 1, 2, \dots, \quad 0 < z < \infty, \quad t > 0, \quad (1)$$

where  $K_i^{-1}$  is the equilibrium ratio at the amount of nuclide  $i$  in the water to the amount in soil plus water.

The initial condition and the infinite-medium boundary condition are

$$N_i(z, 0) = 0, \quad N_i(\infty, t) = 0; \quad t > 0. \quad (2)$$

The remaining boundary conditions must be specified by the source term at the repository. This is frequently specified as the time-dependent concentration in water at the repository (at  $z = 0$ ), which neglects the effect of diffusional transport at the source.<sup>4</sup> For a band release, such that all of the waste material has dissolved after a leach time  $T$ , the source boundary condition is then

$$N_i(0, t) = B_i(t)H(t) - B_i(t)H(t - T), \quad (3)$$

where  $H(t)$  denotes the Heaviside step function and  $B_i(t)$  is given by the Bateman equation

$$B_i(t) = \sum_{j=1}^i b_{ji} \exp(-\lambda_j t), \\ b_{ji} = \sum_{m=1}^j N_m^0 \prod_{\ell=m}^i (\lambda_\ell / \lambda_i) \bigg/ \prod_{\substack{\ell=m \\ (\ell \neq i)}}^i (\lambda_\ell - \lambda_j). \quad (4)$$

Assuming a constant rate of solution of the waste matrix material over the time period  $T$ , the initial concentration  $N_i^0$  (at  $z = 0, t = 0$ ) is given by

$$N_i^0 = M_i^0 / QT, \quad (5)$$

where  $M_i^0$  is the initial amount of nuclide  $i$  per unit amount of waste, and  $Q$  is the volume flow rate of water per unit initial amount of waste.

The solution can be developed in terms of step-release (solutions  $N_i^S$ ), such that

$$N_i^S(0, t) = B_i(t)H(t). \quad (6)$$

The results are:

$$N_1^S(z, t) = b_{1,1} E(1, 0; 1)$$

$$N_2^S(z, t) = \sum_{j=1}^2 b_{j,2} E(j, 0; 2) + \frac{b_{1,1} \lambda_1}{v_1 (\lambda_{12} - \lambda_1 \Gamma_{12})} [E(1, 0; 2) - E(1, 0; 1) + E(1, 2; 1) - E(1, 2; 2)]$$

$$N_3^S(z, t) = \sum_{j=1}^3 b_{j,3} E(j, 0; 3) + \frac{\lambda_2}{v_2} \sum_{j=1}^2 \frac{b_{j,2}}{(\lambda_{23} - \lambda_j \Gamma_{23})} [E(j, 0; 3) - E(j, 0; 2) + E(2, 3; 2) - E(2, 3; 3)]$$

$$+ \frac{\lambda_1 \lambda_2 b_{1,1}}{v_1 v_2} \sum_{j=1}^3 \left[ \frac{E(1, 0; j)}{(\lambda_{kj} - \lambda_1 \Gamma_{kj})(\lambda_{lj} - \lambda_1 \Gamma_{lj})} + \frac{\Gamma_{kj} E(k, j; j)}{(\lambda_{kj} - \lambda_1 \Gamma_{kj})(\Gamma_{lj} \lambda_{kj} - \Gamma_{kj} \lambda_{lj})} + \frac{\Gamma_{lj} E(l, j; j)}{(\lambda_{lj} - \lambda_1 \Gamma_{lj})(\Gamma_{kj} \lambda_{lj} - \Gamma_{lj} \lambda_{kj})} \right]$$

$$(k \neq \ell, \quad k \neq j, \quad \ell \neq j; \quad \ell, k = 1, 2, 3), \quad (7)$$

where

$$\Lambda_{kj} = \frac{\lambda_k}{v_k} - \frac{\lambda_j}{v_j}, \quad \Gamma_{kj} = v_k^{-1} - v_j^{-1}$$

$$E(i, j; k) = \exp(z/2\kappa - \beta_{ij}t) \int_0^\infty \frac{2}{z(4\kappa v_k t)^{1/2} \sqrt{\pi}} \\ \times \exp\{-[y^2 + (z/4\kappa y)^2 \gamma]\} dy \\ = \frac{1}{2} \exp(-\beta_{ij}t + z/2\kappa) \\ \times \left\{ \exp(z\sqrt{\gamma}/2\kappa) \operatorname{erfc} \left[ \frac{z + v_k t \sqrt{\gamma}}{(4\kappa v_k t)^{1/2}} \right] \right. \\ \left. + \exp(-z\sqrt{\gamma}/2\kappa) \operatorname{erfc} \left[ \frac{z - v_k t \sqrt{\gamma}}{(4\kappa v_k t)^{1/2}} \right] \right\} \quad (\gamma > 0) \\ \beta_{ij} = \begin{cases} \lambda_i & , j = 0 \\ \Lambda_{ij}/\Gamma_{ij} & , j \neq 0 \end{cases}, \quad \gamma = 1 + 4\kappa(\lambda_k - \beta_{ij})/v_k \\ \kappa = D/v, \quad v_i = v/K_i. \quad (8)$$

The band-release solution can then be calculated by application of the superposition theorem developed by Foglia et al.<sup>3</sup>

$$N_i^B(z, t) = N_i^S(z, t, b_{ji}) - N_i^S[z, t - T; b_{ji} \exp(-\lambda_j T)], \quad (9)$$

where  $N_i^S(z, t; b_{ji})$  denotes the step-release solution with the argument  $b_{ji}$  in Eq. (4).

Figure 1 shows the instantaneous spatial distribution for the  $^{234}\text{U}$ - $^{230}\text{Th}$ - $^{226}\text{Ra}$  decay chain, calculated for assumed parameters listed in the figure. The curves for the lower value of the dispersion coefficient are essentially the same as for zero dispersion. These calculations illustrate that the effect of dispersion on the concentration profiles of  $^{234}\text{U}$  and  $^{230}\text{Th}$  is relatively large, whereas this degree of dispersion has relatively little effect on the concentration profile of  $^{226}\text{Ra}$ . Even though its concentration is relatively small,  $^{226}\text{Ra}$  is one of the more hazardous radionuclides to be considered in predictive modeling. Our more detailed analyses suggest that, for a range of system parameters, reasonably accurate estimates of the peak concentrations of  $^{226}\text{Ra}$  can be estimated by the simpler dispersion-free equations which have been presented elsewhere.<sup>5</sup>

1. D. H. LESTER, G. JANSEN, and H. C. BURKHOLDER, *AICHE Symp. Ser.*, **71**, 202 (1975).
2. A. D. LITTLE, Intera Environmental Consultants, "Development of Radioactive Waste Migration Model," for Sandia Labs. (Sep. 1977).

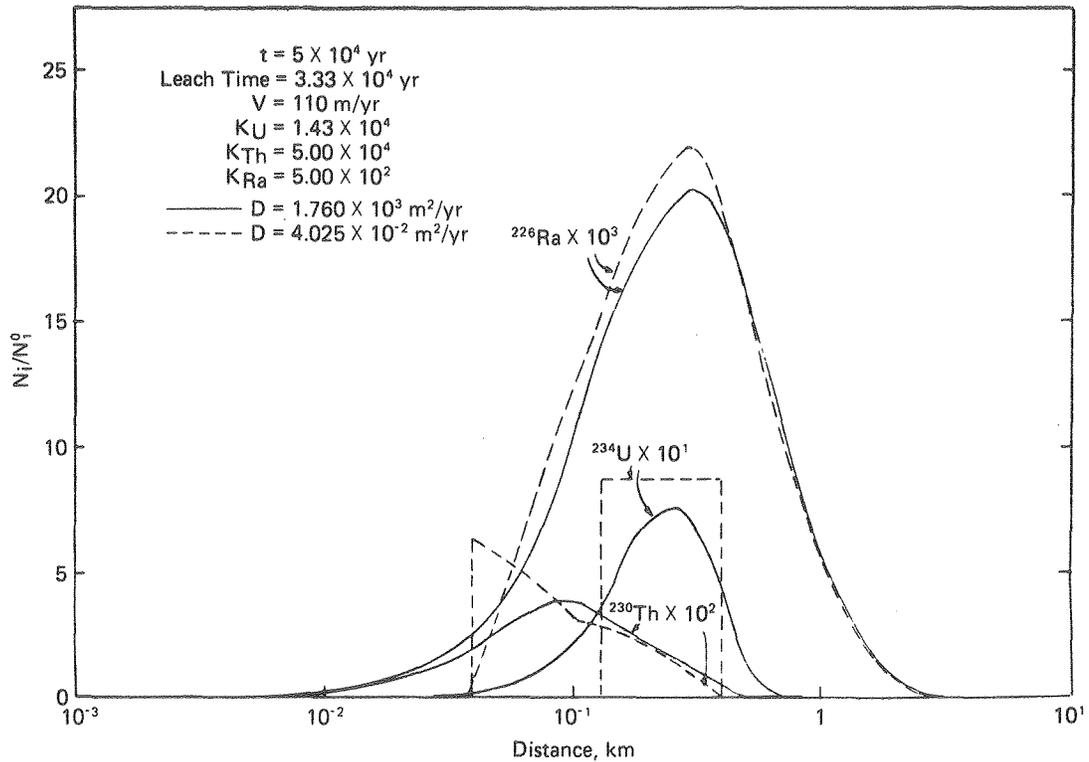
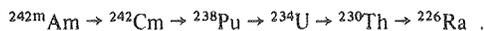


Fig. 1. Migration pattern of  $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$  decay chain (pure  $^{234}\text{U}$  source at  $t = 0$ ).

3. M. FOGLIA, T. H. PIGFORD, and P. L. CHAMBRÉ, "The Superposition Equation for the Band Release of Decaying Radionuclides Through Sorbing Media," UCB-NE-3935 (Mar. 1979).
4. M. HARADA, F. IWAMOTO, and T. H. PIGFORD, "Effect of Source Boundary Conditions in Predicting the Migration of Radionuclides Through Geologic Media," UCB-NE-3349 (June 1979).
5. K. HIGASHI and T. H. PIGFORD, "General Solutions for the Migration of Radionuclides Through Sorbing Media Without Dispersion," UCB-NE-3351 (Mar. 1979).

### 7. Migration Behavior of the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ Decay Chain, K. Higashi, \* M. Harada, F. Iwamoto, T. H. Pigford (UC-Berkeley)

One of the important nuclide chains to be considered in the predictive modeling of the hydrogeological transport of radionuclides in high-level wastes is:



The most important species is usually  $^{226}\text{Ra}$ , because of its relatively high biological hazard and its relatively high mobility in geologic media. The important time scales for the appearance of  $^{226}\text{Ra}$  are on the order of tens of thousands of years, as controlled by the long life of its precursor  $^{230}\text{Th}$ . Therefore, because of the relatively short lives of the first three members of the above chain, the analysis of  $^{226}\text{Ra}$  transport can be reduced to the analysis of the last three members of the above chain, with the assumption that the first three members have already decayed to form the

long-lived  $^{234}\text{U}$ . Analytical studies of the transport of this decay chain have been reported elsewhere.<sup>1-3</sup> In the present paper we analyze the loci of maximum concentrations of  $^{226}\text{Ra}$  and we analyze the effect of dispersion on the concentration profile of  $^{226}\text{Ra}$ .

The radionuclide transport equation to be solved is the one-dimensional (1-D) equation which assumes constant water velocity and local sorption equilibrium.<sup>1-3</sup> Figure 1 shows the isopleths of radium concentration, calculated for the dispersion-free case<sup>3</sup> with assumed leaching of the waste matrix material at a constant rate over a leach time  $T$ . Other assumed parameters are indicated on the figure. The isopleths indicate that the peak time-dependent radium concentration at any given distance from the source increases with source distance until a maximum concentration is reached, after which it decreases monotonically with distance. This is a result of (a) the slow growth with time of  $^{226}\text{Ra}$  in the waste material, and (b) the reconcentration effect due to the distribution of sorbed  $^{230}\text{Th}$  through the media, as has been discussed elsewhere.<sup>3,4</sup>

When thorium sorption ( $K_2$ ) is greater than uranium sorption ( $K_1$ ), the location of the maximum of the radium concentration at any time  $t$  is near the front of the uranium chromatographic band, as shown by the line  $z = K_1 t/v$  in Fig. 1.

Figure 2 shows the effects of dispersion and of the overall radium sorption constant on the concentration profile of radium, at a given instant in time and as a function of distance from the source. The lower value of the dispersion coefficient gives results equivalent to a dispersion-free transport analysis. The curves indicate that dispersion has relatively little effect on the concentration profile of radium in this time period, unless the radium sorption is much greater

\*Present address: Kyoto University, Japan.