

CONVECTIVE-DISPERSIVE TRANSPORT OF SOLUTES INVOLVED IN SEQUENTIAL FIRST-ORDER DECAY REACTIONS

M. TH. VAN GENUCHTEN

U.S. Salinity Laboratory, 4500 Glenwood Drive, Riverside, California 92501, U.S.A.

(Received 24 July 1983; revised 8 February 1984; received for publication 26 November 1984)

Abstract—Problems of solute transport involving sequential first-order decay reactions frequently occur in soil systems. Examples are the migration of radionuclides, in which the chain members form a first-order decay reaction, and the simultaneous movement of various interacting nitrogen species. This study presents analytical solutions that describe the simultaneous convective-dispersive transport of up to four species involved in such a consecutive chain reaction. Evaluation of the analytical solutions is not straightforward but requires, among other things, the calculation of complex complementary error functions. A FORTRAN IV computer program (CHAIN) that can be used to evaluate the analytical solutions is described. Application of this program to problems of solute transport is illustrated with two examples, one dealing with radionuclide transport and one with nitrification.

Key Words: Convective-dispersive solute transport, First-order decay reactions, Nitrogen transport, Radionuclide transport, Transport modeling.

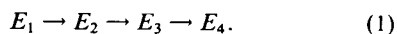
INTRODUCTION

Problems of solute transport involving sequential first-order decay reactions frequently occur in soil and groundwater systems. One often quoted example is that of the migration of various radionuclides (Lester, Jansen, and Burkholder, 1975; Rogers, 1978; Gureghian and Jansen, 1983). Other examples are concerned with the simultaneous movement of interacting nitrogen species (Cho, 1971; Misra, Nielson, and Biggar, 1974a, b; Wagenet, Biggar, and Nielson, 1976), organic phosphates (Castro and Rolston, 1977), or pesticides (Bromilow and Leistra, 1980). Previous attempts to derive analytical solutions have been limited either to three chain members, using relatively simple boundary conditions (Cho, 1971; Wagenet, Biggar, and Nielson, 1976; Harada and others, 1980), or to transport conditions, where dispersion phenomena could be neglected (Higashi and Pigford, 1980; Harada and others, 1980). This paper presents several analytical solutions that are applicable to sequential first-order decay reactions with up to four chain members. The boundary conditions are formulated in such a way that different sets of input source functions can be included in the formulation. The solutions are evaluated with a FORTRAN IV computer program termed CHAIN.

THEORETICAL

Governing transport equations

This study considers the transport of four species (E_i , $i = 1, 4$) involved in a consecutive first-order decay chain of the form



The following set of coupled differential equations describes the one-dimensional convective-dispersive transport of the four chain members under transient flow conditions:

$$\frac{\partial}{\partial t}(\theta c_i + \rho s_i) = \frac{\partial}{\partial x} \left(\theta D \frac{\partial c_i}{\partial x} - q c_i \right) - \mu_{w,i} \theta c_i - \mu_{s,i} \rho s_i, \quad (2a)$$

$$\frac{\partial}{\partial t}(\theta c_i + \rho s_i) = \frac{\partial}{\partial x} \left(\theta D \frac{\partial c_i}{\partial x} - q c_i \right) + \mu_{w,i-1} \theta c_{i-1} + \mu_{s,i-1} \rho s_{i-1} - \mu_{w,i} \theta c_i - \mu_{s,i} \rho s_i \quad (i = 2, 3, 4), \quad (2b)$$

where c is the solution concentration (ML^{-3}), s the adsorbed concentration (MM^{-1}), θ the volumetric water content ($\text{L}^3 \text{L}^{-3}$), q the volumetric flux (LT^{-1}), D the dispersion coefficient ($\text{L}^2 \text{T}^{-1}$), ρ the porous medium bulk density (ML^{-3}), x distance (L), and t time (T); the subscript i delineates the i th chain member. The coefficients $\mu_{w,i}$ and $\mu_{s,i}$ are rate constants for first-order decay (T^{-1}) in the liquid and solid phases of the soil, respectively. These two rate coefficients probably will have different values for both chemical and microbiological degradation. However, for radioactive decay we may assume that they are identical:

$$\mu_{w,i} = \mu_{s,i} (= \mu_i). \quad (3)$$

Two assumptions are introduced. First, it is assumed that the soil system is homogeneous and that θ and q are constant in time and space (steady-state flow). Second, we assume that adsorbed concentrations (s_i) can be related to solution concentrations (c_i) by linear (or linearized) and reversible isotherms of the form

$$s_i = k_i c_i \quad (i = 1, 4), \quad (4)$$

where the k_i are empirical distribution coefficients ($M^{-1}L^3$). Using equations (3) and (4) and the assumptions of steady-state flow and linear equilibrium transport, equations (2a, b) reduce to

$$R_1 \frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial x^2} - v \frac{\partial c_1}{\partial x} - \mu_1 R_1 c_1, \quad (5a)$$

$$R_i \frac{\partial c_i}{\partial t} = D \frac{\partial^2 c_i}{\partial x^2} - v \frac{\partial c_i}{\partial x} + \mu_{i-1} R_{i-1} c_{i-1} - \mu_i R_i c_i \quad (i = 2, 3, 4), \quad (5b)$$

where $v (=q/\theta)$ is the average pore-water velocity and the retardation factors R_i are given by

$$R_i = 1 + \frac{\rho k_i}{\theta}. \quad (6)$$

Note that equations (5a, b) apply only for the limiting situation when the decay constants $\mu_{w,i}$ and $\mu_{s,i}$ are identical [equation (3)]. If degradation is limited only to the liquid phase ($\mu_{s,i} = 0$), the retardation factors should be dropped from the decay terms in equations (5a, b). Other situations for which $\mu_{s,i} \neq \mu_{w,i}$ can be considered easily by redefining appropriately the general decay coefficient μ_i .

Initial and boundary conditions

In this study we restrict ourselves to analytical solutions that are applicable to semi-infinite systems ($0 \leq x < \infty$) using the boundary conditions

$$\frac{\partial c_i}{\partial x}(\infty, t) = 0, \quad t \geq 0 \quad (i = 1, 4) \quad (7)$$

and to systems that are initially free of solutes:

$$c_i(x, 0) = 0, \quad x \geq 0. \quad (8)$$

The input boundary conditions at $x = 0$ are described by either first-type (or concentration-type) conditions of the form

$$c_i(0, t) = \begin{cases} f_i(t), & 0 < t \leq t_0, \\ 0, & t > t_0, \end{cases} \quad (9)$$

or third-type (or flux-type) conditions of the form

$$\left(-D \frac{\partial c_i}{\partial x} + v c_i \right) \Big|_{x=0} = \begin{cases} v f_i(t), & 0 < t \leq t_0, \\ 0, & t > t_0, \end{cases} \quad (10)$$

where f_i ($i = 1, 4$) is given by

$$f_1(t) = B_1 e^{-\lambda_1 t}, \quad (11a)$$

$$f_2(t) = B_2 e^{-\lambda_1 t} + B_3 e^{-\lambda_2 t}, \quad (11b)$$

$$f_3(t) = B_4 e^{-\lambda_1 t} + B_5 e^{-\lambda_2 t} + B_6 e^{-\lambda_3 t}, \quad (11c)$$

$$f_4(t) = B_7 e^{-\lambda_1 t} + B_8 e^{-\lambda_2 t} + B_9 e^{-\lambda_3 t} + B_{10} e^{-\lambda_4 t}, \quad (11d)$$

in which the coefficients B_j ($j = 1, 10$) and λ_i ($i = 1, 4$) are all constants. The multiple terms in equations (11a-d) are a consequence of decay reactions in the waste site (e.g., a nuclear waste repository) and also account for a finite rate of release of each chain member from the waste site into the environment. For one particular release mechanism, the constants B_j are related to each other through the Bateman equations (Bateman, 1910). This situation is described briefly.

Consider a waste site that initially for each chain member contains an amount M_i^0 per unit cross-sectional area perpendicular to the direction of flow. It is assumed that the release rate from the repository into the environment for each member is proportional to the amount remaining in the repository. The total amount of each chain member in the waste site as a function of time, $M_i(t)$, then can be calculated by solving the set of equations (Higashi and Pigford, 1980):

$$\frac{dM_1}{dt} = -\mu_1 M_1 - \gamma_1 M_1 \quad (12a)$$

$$\frac{dM_i}{dt} = \mu_{i-1} M_{i-1} - \mu_i M_i - \gamma_i M_i \quad (i = 2, 3, 4), \quad (12b)$$

where the proportionality constant γ_i for each member determines the release rate from the repository. Hence, the first term on the right-hand side of equation (12a) defines the rate of decay of the parent member, and the second term defines the release rate into the environment. Solving equations (12a, b), subject to the initial condition

$$M_i(t) = M_i^0 \quad (t = 0) \quad (13)$$

gives

$$M_1(t) = A_1 e^{-\lambda_1 t}, \quad (14a)$$

$$M_2(t) = A_2 e^{-\lambda_1 t} + A_3 e^{-\lambda_2 t}, \quad (14b)$$

$$M_3(t) = A_4 e^{-\lambda_1 t} + A_5 e^{-\lambda_2 t} + A_6 e^{-\lambda_3 t}, \quad (14c)$$

$$M_4(t) = A_7 e^{-\lambda_1 t} + A_8 e^{-\lambda_2 t} + A_9 e^{-\lambda_3 t} + A_{10} e^{-\lambda_4 t}, \quad (14d)$$

where

$$\lambda_i = \mu_i + \gamma_i; \quad (15)$$

$$A_1 = M_1^0, \quad A_2 = \frac{\mu_1 M_1^0}{\lambda_2 - \lambda_1}, \quad A_3 = M_2^0 - A_2,$$

$$A_4 = \frac{\mu_2 A_2}{\lambda_3 - \lambda_1}, \quad A_5 = \frac{\mu_2 A_3}{\lambda_3 - \lambda_2}, \quad A_6 = M_3^0 - A_4 - A_5, \quad (16)$$

$$A_7 = \frac{\mu_3 A_4}{\lambda_4 - \lambda_1}, \quad A_8 = \frac{\mu_3 A_5}{\lambda_4 - \lambda_2}, \quad A_9 = \frac{\mu_3 A_6}{\lambda_4 - \lambda_3},$$

$$A_{10} = M_4^0 - A_7 - A_8 - A_9.$$

Equations (14)–(16) are known as the Bateman equations.

Continuity of the solute flux ($q_{s,i}$) for each species across the boundary of the waste site requires

$$q_{s,i} = \gamma_i M_i = \left(-\theta D \frac{\partial c_i}{\partial x} + \theta v c_i \right) \Big|_{x=0} \quad (i = 1, 4), \quad (17)$$

where, as before, the amount of material of each chain member in the waste repository (M_i) is defined per unit cross-sectional area perpendicular to the direction of flow. Combining equations (14) and (17) and dividing by the volumetric water content (θ) leads directly to equations (10) and (11), provided that the coefficients B_j are defined as

$$\begin{aligned} B_1 &= \frac{\gamma_1 A_1}{q}, & B_2 &= \frac{\gamma_2 A_2}{q}, & B_3 &= \frac{\gamma_2 A_3}{q}, \\ B_4 &= \frac{\gamma_3 A_4}{q}, & B_5 &= \frac{\gamma_3 A_5}{q}, & B_6 &= \frac{\gamma_3 A_6}{q}, \\ B_7 &= \frac{\gamma_4 A_7}{q}, & B_8 &= \frac{\gamma_4 A_8}{q}, & B_9 &= \frac{\gamma_4 A_9}{q}, \\ B_{10} &= \frac{\gamma_4 A_{10}}{q}. \end{aligned} \quad (18)$$

Equations (10) and (11) apply to the most general situation of preferential release from the repository; that is, all γ_i 's can have different values. Other and more simple boundary conditions can be formulated by forcing appropriate constants (λ_i , B_j) in equations (11) to be zero.

Analytical solutions

Standard Laplace transform techniques were used for the derivation of the analytical solutions presented here (van Genuchten, 1981a; van Genuchten and Alves, 1982). Details of the derivation are omitted.

The solution of equations (5), (7), and (8) for the flux-type boundary condition [equations (10) and (11)] is

$$\begin{aligned} c_i^*(x, t) &= \begin{cases} c_i^*(x, t), & 0 < t \leq t_0, \\ c_i^*(x, t) - \exp(-\lambda_i t_0) c_i^*(x, t - t_0), & t > t_0, \end{cases} \\ &\quad (19) \end{aligned}$$

where $c_i^*(x, t)$ is given in Appendix 1. The analytical solution for the concentration-type boundary condition [equations (9) and (11)] is identical to this solution, except that the term F_{ijk} in Appendix 1 must be replaced by

$$\begin{aligned} F_{ijk} &= \exp(-a_{ijk} t) \left\{ \frac{1}{2} \exp \left[\frac{(v-w)x}{2D} \right] \operatorname{erfc} \left[\frac{R_i x - wt}{2(DR_i t)^{1/2}} \right] \right. \\ &\quad \left. + \frac{1}{2} \exp \left[\frac{(v+w)x}{2D} \right] \operatorname{erfc} \left[\frac{R_i x + wt}{2(DR_i t)^{1/2}} \right] \right\}. \quad (20) \end{aligned}$$

PROGRAM CHAIN

A FORTRAN IV computer program termed CHAIN was written to evaluate the two analytical solutions derived here; the program is listed in Appendix 2. Much of the notation used in CHAIN is identical to the notation of the theoretical section. The most significant program variables are defined in Appendix 3, and Appendix 4 gives instructions for setting up the input data file.

The program consists of a main program (MAIN), two functions (F and EXF), and one subroutine (CEXF). Most of the calculations are carried out in MAIN, including input and output instructions and the calculation of various constants. The function F evaluates the variables F_{ijk} or S_{ij} according to (see also Appendix 1)

$$F(I, JJ) = \begin{cases} F_{ij0} & \text{if } JJ \leq 0 \quad (I = i, JJ = -j), \\ S_{ij} & \text{if } JJ > 0 \quad (I = i, JJ = j). \end{cases} \quad (21)$$

The function EXF(A , B) defines the product of the exponential function ($\exp$) and the complementary error function (erfc) as follows:

$$\operatorname{EXF}(A, B) = \exp(A) \operatorname{erfc}(B), \quad (22)$$

where

$$\operatorname{erfc}(B) = \frac{2}{\sqrt{\pi}} \int_B^\infty \exp(-\tau^2) d\tau. \quad (23)$$

Two different approximations are used for EXF(A , B). For $0 \leq B \leq 2.5$ [see also equation (7.1.26) of Gautschi, 1964]:

$$\begin{aligned} \operatorname{EXF}(A, B) &\simeq \exp(A - B^2)(a_1 \tau \\ &\quad + a_2 \tau^2 + a_3 \tau^3 + a_4 \tau^4 + a_5 \tau^5), \quad (24) \end{aligned}$$

where

$$\tau = \frac{1}{1 + 0.3275911B}; \quad (25)$$

$$\begin{aligned} a_1 &= 0.254829592, & a_2 &= -0.284496736, \\ a_3 &= 1.421413741, & a_4 &= -1.453152027, \\ a_5 &= 1.061405429; \end{aligned} \quad (26)$$

and, for $B > 2.5$ [see also equation (7.1.14) of Gautschi, 1964]:

$$\operatorname{EXF}(A, B) \simeq \frac{1}{\sqrt{\pi}} \frac{\exp(A - B^2)}{(B + 0.5/(B + 1/(B + 1.5/(B + 2/(B + 2.5/(B + 1))))))}. \quad (27)$$

For negative values of B , the following additional relation is needed:

$$\text{EXF}(A, B) = 2 \exp(A) - \text{EXF}(A, -B). \quad (28)$$

The function $\text{EXF}(A, B)$ cannot be used for small or large values of its arguments A, B . The function returns zero for the following two conditions:

$$\begin{cases} |A| > M, \\ B \leq 0, \end{cases} \quad \text{or} \quad \begin{cases} |A - B^2| > M, \\ B > 0, \end{cases} \quad (M = 170). \quad (29)$$

Subroutine $\text{CEXF}(A, B, Z, U, V)$ is used to evaluate the complementary error function for complex arguments:

$$U + iV = \exp(Z) \operatorname{erfc}(A + iB). \quad (30)$$

Complex erfc functions occur in the formulation whenever w^2 (see last equation of Appendix 1) becomes negative.

For $(A + B) > 6$ with $A \geq 0$ and $B \geq 0$, equation (30) is evaluated as follows [see also equations (7.13, 7.14, 7.4.13, and 7.1.14) of Gautschi, 1970]:

$$U = \frac{1}{\pi} \exp(Z + B^2 - A^2) [T \cos(2AB) - W \sin(2AB)], \quad (31)$$

$$V = \frac{1}{\pi} \exp(Z + B^2 - A^2) [-T \sin(2AB) - W \cos(2AB)], \quad (32)$$

$$T = \int_{-\infty}^{\infty} \frac{A \exp(-\tau^2)}{(B + \tau)^2 + A^2} d\tau, \quad (33)$$

$$W = \int_{-\infty}^{\infty} \frac{(B + \tau) \exp(-\tau^2)}{(B + \tau)^2 + A^2} d\tau, \quad (34)$$

where T and W are evaluated with a Hermitian quadrature scheme of the form

$$\int_{-\infty}^{\infty} \exp(-\tau^2) g(\tau) d\tau = \sum_{i=1}^n w_i g(\tau_i). \quad (35)$$

Values for the weighting factors w_i and the associated abscissas τ_i were obtained from Table 25.10 of Davis and Polonsky (1964). The program uses 20 quadrature points ($n = 20$).

For $0 < A + B < 6$ with $A > 0, B \geq 0$, equation (30) is evaluated with equation (7.1.29) of Gautschi (1970). Specifically, the following set of equations is used:

$$U = \exp(Z) \operatorname{erfc}(A) - \exp(Z - A^2) \times \left[\frac{1 - C}{2\pi A} + \sum_{n=1}^{\infty} \alpha(2A - b_1 C + b_2 S) \right], \quad (36)$$

$$V = \exp(Z - A^2) \left[\frac{S}{2\pi A} + \sum_{n=1}^{\infty} \alpha(b_1 S + b_2 C) \right], \quad (37)$$

where

$$\alpha = \frac{2 \exp(-n^2/4)}{\pi(n^2 + 4A^2)}; \quad (38a)$$

$$C = \cos(2AB), \quad S = \sin(2AB); \quad (38b)$$

$$b_1 = 2A \cosh(nB), \quad b_2 = n \sinh(nB). \quad (38c)$$

This method gives an accuracy of at least six significant digits if the number of terms in the series expansions of equations (36) and (37) equals or exceeds m , where m is the integer closest to $(12 + 2B)$.

The two methods for evaluating the complex erfc -function hold for nonnegative values of A and B . Given equation (30) for positive A and B , the two methods can be extended to negative values by making use of the relationships

$$2 \exp(Z) - U - iV = \exp(Z) \operatorname{erfc}(-A - iB), \quad (39a)$$

$$2 \exp(Z) - U + iV = \exp(Z) \operatorname{erfc}(-A + iB), \quad (39b)$$

$$U - iV = \exp(Z) \operatorname{erfc}(A - iB). \quad (39c)$$

Initially, serious round-off errors were observed when the approximations for erfc were used directly in the equations for F_{ijk} , especially for the solution of the flux-type boundary condition (Appendix 1). Such round-off errors become immediately apparent when the expressions for F_{ijk} are evaluated for relatively large values of R, x , and/or t . Fortunately, round-off errors could be limited greatly by first substituting the appropriate erfc approximations into the F_{ijk} terms and subsequently combining and simplifying various terms in the resulting series expansions. Although straightforward in principle, this manipulation and simplification of the various terms in F_{ijk} required extensive bookkeeping. Details are omitted here; if needed, they can be retrieved from the function $F(I, JJ)$ in Appendix 2.

Finally, the program also contains the statement

$$\text{CALL ERRSET}(208, 0, -1, 1), \quad (40)$$

which suppresses all underflow error messages during execution. If this subroutine is not available, underflow problems in most situations can be avoided by using $M = 100$ in equation (29), rather than the present value of 170 (these changes are made easily at selected places in the program).

EXAMPLES

Two examples are presented here to illustrate typical results that can be obtained with the program. Appendix 5 lists the input data for the examples; the associated output file is given in Appendix 6. The first example applies to the three-species nitrification chain



and is the same as described earlier by Cho (1971),

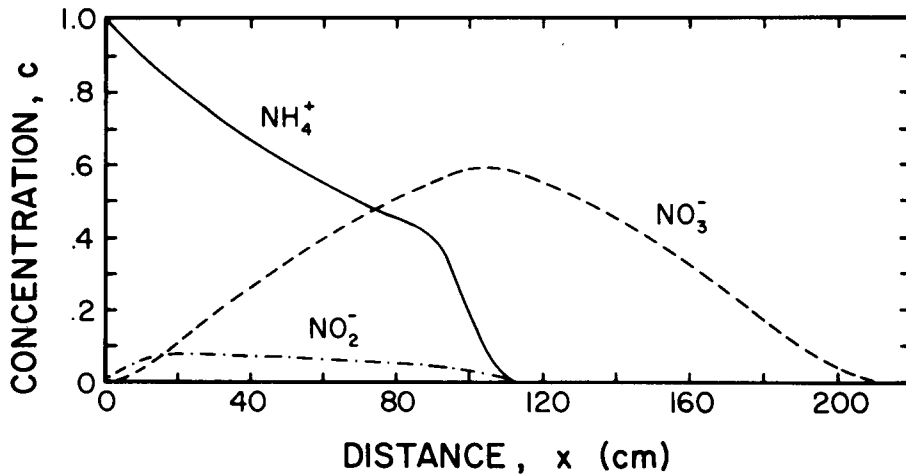


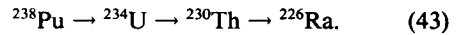
Figure 1. Calculated distributions for nitrogen decay chain $\text{NH}_4^+ \rightarrow \text{NO}_2^- \rightarrow \text{NO}_3^-$ at $t = 200$ h.

except that the constant concentration boundary condition for Cho's solution is replaced here by a constant flux condition, that is, by equations (10) and (11) with $B_1 = 1$, $B_j = 0$ ($j = 2, 10$), and $\lambda_1 = 0$. Other parameter values for the example are

$$\begin{aligned} v &= 1 \text{ cm h}^{-1}, \quad D = 0.18 \text{ cm}^2 \text{ h}^{-1}, \\ R_1 &= 2 \quad R_2 = R_3 = 1, \\ \mu_1 R_1 &= 0.01 \text{ h}^{-1}, \quad \mu_2 = 0.1 \text{ h}^{-1}, \quad \mu_3 = 0. \end{aligned} \quad (42)$$

Calculated concentration distributions for the three species at $t = 200$ h are shown in Figure 1. The distributions are nearly identical to those given by Cho (1971), thus showing that the effect of a different boundary condition is small for the parameter values of this example.

The second example is concerned with the radionuclide decay chain



This decay chain was recently used by Higashi and Pigford (1980) as an example in predictive modeling of subsurface radionuclide transport. Of the four species in equation (43), ^{226}Ra presumably is the more crucial chain member, partly because of the high biological hazard associated with this species, and partly because the relatively low R value of ^{226}Ra leads to a higher mobility in the system than is the situation with the other chain members. Values of the various input parameters, listed in Appendices 5 (input file) and 6 (output file), are the same as those used in the original study by Higashi and Pigford

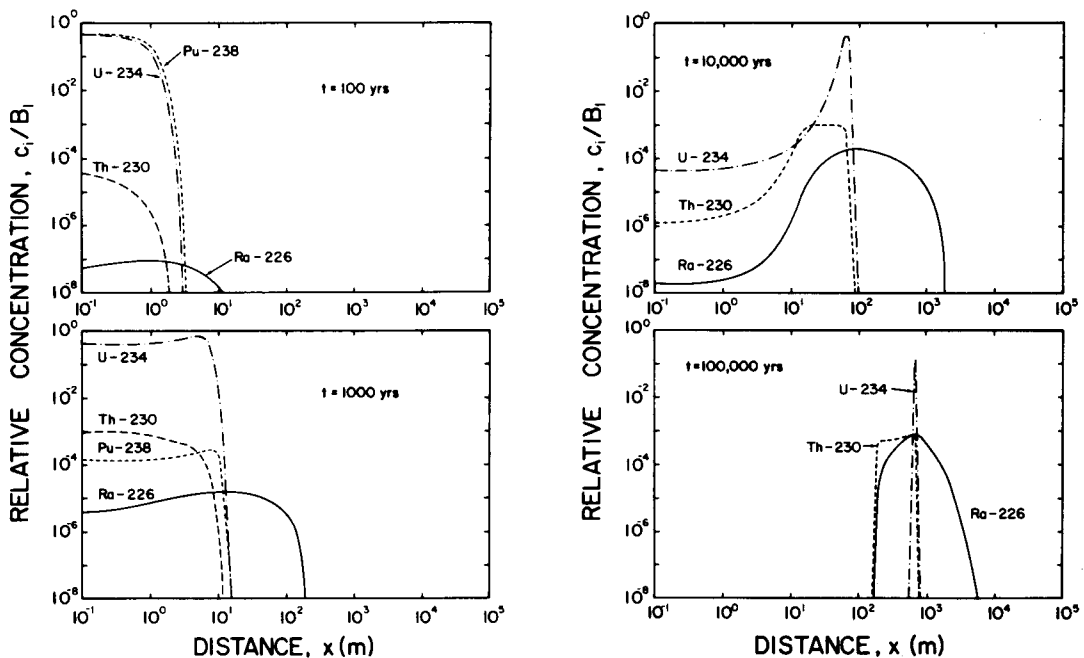


Figure 2. Calculated distributions for radionuclide decay chain $^{238}\text{Pu} \rightarrow ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ at four different times.

(1980). An arbitrary value of $10m^2/\text{year}$ was assigned to the dispersion coefficient D (Higashi and Pigford neglected dispersion phenomena in their analysis). The reader is referred to the original paper of Higashi and Pigford (1980) for a more detailed discussion of the physical problem.

Figure 2 shows relative concentration distribution at four different times; concentrations are expressed as fractions of the input concentration (B_1) of ^{238}Pu at $x = 0$ [see equation (18)]. A careful comparison of the plotted results with figure 3 of Higashi and Pigford (1980) shows that the shapes of the distributions are nearly identical. Hence, the effects of dispersion are relatively small in this example, although it should be realized that these effects are not clearly visible in graphic form because of the logarithmic scale of the figures.

CONCLUSION

This study presents a FORTRAN IV computer program (CHAIN) that can be used to evaluate the convective-dispersive transport of up to four chain members that are involved in consecutive first-order decay reactions. The analytical solutions for this problem, especially those for the fourth chain member, are complicated and require special programming care if serious round-off errors are to be avoided. Examples concerned with nitrogen and radionuclide transport illustrate the type of problems that can be considered with the program. Compared with numerical transport models, the analytical formulation presented here may prove to be useful in several situations. First, the program can be used to obtain initial and approximate estimates for solute transport for a number of simplified field situations (notably for steady-state flow in one dimension). For example, the long-term vertical transport of chemicals through the unsaturated zone may be described accurately with appropriate analytical solutions (Wierenga, 1977). Second, analytical solutions of the type given here are important tools for estimating transport parameters from observed laboratory or field data, notably with respect to dispersion coefficients, retardation factors, or decay coefficients (see Misra, Nielsen, and Biggar, 1974b, and van Genuchten, 1981b, for examples). Finally, the analytical model may be used for verification of appropriate numerical multi-ion chain solutions.

REFERENCES

- Bateman, H., 1910, The solution of a system of differential equations occurring in the theory of radioactive transformations: *Proc. Cambridge Philos. Soc.*, v. 15, pt V, p. 423-427.
- Bromilow, R. H., and Leistra, M., 1980, Measured and simulated behaviour of aldicarb and its oxidation products in fallow soils: *Pestic. Sci.*, v. 11, no. 4, p. 389-395.
- Castro, C. L., and Rolston, D. E., 1977, Organic phosphate transport and hydrolysis in soil: theoretical and experimental evaluation: *Soil Sci. Soc. America Jour.*, v. 41, no. 6, p. 1085-1092.
- Cho, C. M., 1971, Convective transport of ammonium with nitrification in soil, *Can. Jour. Soil Sci.*, v. 51, no. 3, p. 339-350.
- Davis, P. J., and Polonsky, I., 1964, Numerical interpolation, differentiation and integration, in Abramowitz, M., and Stegun, I. A., eds., *Handbook of mathematical functions*: U.S. Government Printing Office, Washington D.C., p. 875-924.
- Gautschi, W., 1964, Error function and Fresnel integrals, in Abramowitz, M., and Stegun, I. A., eds., *Handbook of mathematical functions*: U.S. Government Printing Office, Washington D.C., p. 295-329.
- Gureghian, A. B., and Jansen, G., 1983, LAYFLO: A one-dimensional semianalytical model for the migration of a three-member decay chain in a multilayered geologic medium: Tech. Rep. ONWI-466, Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, Ohio, 83 p.
- Harada, M., Chambre, P. L., Foglia, M., Higashi, K., Iwamoto, F., Leung, D., Pigford, T. H., and Ting, D., 1980, Migration of radionuclides through sorbing media, analytical solutions—I: Rep. no. LBL-10500 (UC-11), Lawrence Berkeley Laboratory, Univ. California, Berkeley, 245 p.
- Higashi, K., and Pigford, T. H., 1980, Analytical models for migration of radionuclides in geologic sorbing media: *Jour. Nucl. Sci. and Techn.*, v. 17, no. 9, p. 700-709.
- Lester, D. H., Jansen, G., and Burkholder, H. C., 1975, Migration of radionuclide chains through an adsorbing medium, in Adsorption and ion exchange: *Am. Inst. Chem. Eng., Symp. Series no. 152*, v. 71, p. 202-213.
- Misra, C., Nielsen, D. R., and Biggar, J. W., 1974a, Nitrogen transformations in soil during leaching: I. Theoretical considerations: *Soil Sci. Soc. America Proc.*, v. 38, no. 2, p. 289-293.
- Misra, C., Nielsen, D. R., and Biggar, J. W., 1974b, Nitrogen transformations in soil during leaching: II. Steady state nitrification and nitrate reduction: *Soil Sci. Soc. America Proc.*, v. 38, no. 2, p. 294-299.
- Rogers, V. C., 1978, Migration of radionuclide chains in groundwater: *Nucl. Techn.*, v. 40, no. 3, p. 315-320.
- van Genuchten, M. Th., 1981a, Analytical solutions for chemical transport with simultaneous adsorption, zero-order production and first-order decay: *Jour. Hydrol.*, v. 49, no. 3-4, p. 213-233.
- van Genuchten, M. Th., 1981b, Non-equilibrium transport parameters from miscible displacement experiments: *Research Rep. no. 119*, U.S. Salinity Laboratory, Riverside, California, 88 p.
- van Genuchten, M. Th., and Alves, W. J., 1982, Analytical solutions of the one-dimensional convective-dispersive solute transport equation: *Tech. Bull.*, no. 1661, U.S. Dep. of Agriculture, 151 p.
- Wagenet, R. J., Biggar, J. W., and Nielsen, D. R., 1976, Analytical solutions of miscible displacement equations describing the sequential microbiological transformations of urea, ammonium and nitrate: *Research Rep. no. 6001*, Dep. of Water Science and Engineering, Univ. California, Davis, 53 p.
- Wierenga, P. J., 1977, Solute distribution profiles computed with steady-state and transient water movement models, *Soil Sci. Soc. America Jour.*, v. 41, no. 6, p. 1050-1055.

APPENDIX 1

Expressions for $c_i^*(x, t)$ in equation (19) for the analytical solution of equations (5), (7), (8), (10) and (11)

$$c_1^* = B_1 F_{110},$$

$$c_2^* = B_2 F_{210} + B_3 F_{220} + \frac{\mu_1 R_1 B_1 (S_{12} - F_{210} + F_{110})}{R_{12} \lambda_1 - \mu_{12}},$$

$$c_3^* = B_4 F_{310} + B_5 F_{320} + B_6 F_{330} + \frac{\mu_2 R_2 B_2 (S_{23} - F_{310} + F_{210})}{R_{23} \lambda_1 - \mu_{23}} \\ + \frac{\mu_2 R_2 B_3 (S_{23} - F_{320} + F_{220})}{R_{23} \lambda_2 - \mu_{23}} + \frac{A_1 B_1 R_{12} (F_{210} - F_{110} - S_{12})}{R_{12} \lambda_1 - \mu_{12}} \\ + \frac{A_1 B_1 R_{13} (S_{13} - F_{310} + F_{110})}{R_{13} \lambda_1 - \mu_{13}} + \frac{A_1 B_1 R_{23} (F_{310} - F_{210} - S_{23})}{R_{23} \lambda_1 - \mu_{23}},$$

$$c_4^* = B_7 F_{410} + B_8 F_{420} + B_9 F_{430} + B_{10} F_{440} + \frac{\mu_3 R_3 B_4 (S_{34} - F_{410} + F_{310})}{R_{34} \lambda_1 - \mu_{34}} \\ + \frac{\mu_3 R_3 B_5 (S_{34} - F_{420} + F_{320})}{R_{34} \lambda_2 - \mu_{34}} + \frac{\mu_3 R_3 B_6 (S_{34} - F_{430} + F_{330})}{R_{34} \lambda_3 - \mu_{34}} \\ + \frac{A_2 B_2 R_{34} (F_{410} - F_{310} - S_{34})}{R_{34} \lambda_1 - \mu_{34}} + \frac{A_2 B_2 R_{23} (F_{310} - F_{210} - S_{23})}{R_{23} \lambda_1 - \mu_{23}} \\ + \frac{A_2 B_2 R_{24} (S_{24} - F_{410} + F_{210})}{R_{24} \lambda_1 - \mu_{24}} + \frac{A_2 B_3 R_{34} (F_{420} - F_{320} - S_{34})}{R_{34} \lambda_2 - \mu_{34}} \\ + \frac{A_2 B_3 R_{23} (F_{320} - F_{220} - S_{23})}{R_{23} \lambda_2 - \mu_{23}} + \frac{A_2 B_3 R_{24} (S_{24} - F_{420} + F_{220})}{R_{24} \lambda_2 - \mu_{24}} \\ + \frac{A_3 B_1 R_{14}^2 (S_{14} - F_{410} + F_{110})}{G_{134} G_{124} (R_{14} \lambda_1 - \mu_{14})} + \frac{A_3 B_1 R_{24}^2 (F_{410} - F_{210} - S_{24})}{G_{124} G_{234} (R_{24} \lambda_1 - \mu_{24})} \\ + \frac{A_3 B_1 R_{34}^2 (S_{34} - F_{410} + F_{310})}{G_{134} G_{234} (R_{34} \lambda_1 - \mu_{34})} + \frac{A_3 B_1 R_{13}^2 (F_{310} - F_{110} - S_{13})}{G_{123} G_{134} (R_{13} \lambda_1 - \mu_{13})} \\ + \frac{A_3 B_1 R_{23}^2 (S_{23} - F_{310} + F_{210})}{G_{123} G_{234} (R_{23} \lambda_1 - \mu_{23})} + \frac{A_3 B_1 R_{12}^2 (S_{12} - F_{210} + F_{110})}{G_{123} G_{124} (R_{12} \lambda_1 - \mu_{12})};$$

$$A_1 = \frac{\mu_1 \mu_2 R_1 R_2}{G_{123}}, \quad A_2 = \frac{\mu_2 \mu_3 R_2 R_3}{G_{234}}, \quad A_3 = \mu_1 \mu_2 \mu_3 R_1 R_2 R_3;$$

$$G_{ijk} = \mu_i R_i R_{kj} + \mu_j R_j R_{ik} + \mu_k R_k R_{ji}; \quad S_{ij} = F_{jji} - F_{iji};$$

$$R_{ij} = R_i - R_j; \quad \mu_{ij} = \mu_i R_i - \mu_j R_j;$$

$$F_{ijk} = \exp(-a_{ijk}t) \left\{ \frac{v}{v+w} \exp\left[\frac{(v-w)x}{2D}\right] \operatorname{erfc}\left[\frac{R_i x - w t}{2(DR_i t)^{1/2}}\right] + \frac{v}{v-w} \exp\left[\frac{(v+w)x}{2D}\right] \operatorname{erfc}\left[\frac{R_i x + w t}{2(DR_i t)^{1/2}}\right] \right\} \\ + \frac{2v^2}{w^2 - v^2} \exp\left(\frac{vx}{D} - \mu_i t\right) \operatorname{erfc}\left[\frac{R_i x + vt}{2(DR_i t)^{1/2}}\right] \quad (\mu_i \neq a_{ijk}),$$

$$F_{ijk} = \exp(-a_{ijk}t) \left\{ \frac{1}{2} \operatorname{erfc}\left[\frac{R_i x - vt}{2(DR_i t)^{1/2}}\right] + \left(\frac{v^2 t}{\pi DR_i}\right)^{1/2} \exp\left[-\frac{(R_i x - vt)^2}{4DR_i t}\right] \right. \\ \left. - \frac{1}{2} \left(1 + \frac{vx}{D} + \frac{v^2 t}{DR_i}\right) \exp\left(\frac{vx}{D}\right) \operatorname{erfc}\left[\frac{R_i x + vt}{2(DR_i t)^{1/2}}\right] \right\} \quad (\mu_i = a_{ijk});$$

$$a_{ijk} = \begin{cases} \lambda_j, & k = 0, \\ \frac{\mu_{ij}}{R_{ij}}, & k > 0; \end{cases}$$

$$w = [v^2 + 4DR_i(\mu_i - a_{ijk})]^{1/2}.$$

APPENDIX 2

Listing of program CHAIN

```

001 C *****
002 C *
003 C *      MULTI-ION SOLUTE TRANSPORT      CHAIN
004 C *
005 C *      FOUR SPECIES
006 C *      PREFERENTIAL RELEASE
007 C *
008 C *      MAY,1982
009 C *
010 C *****
011 C
012 C      IMPLICIT REAL*8 (A-H,O-Z)
013 C      COMMON R(4),DONE(4),RLAM(4),D,V,X,T,KSURF
014 C      DIMENSION ZERO(4),GAMMA(4),TITLE(20),A(10),B(10)
015 C      CALL ERRSET(208,0,-1,1)
016 C
017 C      ----- READ NUMBER OF CASES CONSIDERED -----
018 C      READ(5,1000) NC
019 C      DO 14 NCASE=1,NC
020 C      READ(5,1001) TITLE
021 C
022 C      ----- READ INPUT PARAMETERS
023 C      READ(5,1000) NS,NR,NB,KSURF,KDB,KPR,V,D,WC,TP
024 C      READ(5,1002) (R(I),I=1,4),(DONE(I),I=1,4)
025 C      READ(5,1002) (ZERO(I),I=1,4),(GAMMA(I),I=1,4)
026 C      READ(5,1002) XI,DX,XM,TI,DT,TM
027 C
028 C      -----
029 C      DO 1 I=1,4
030 C      ZERO(I)=NB*ZERO(I)
031 C      1 RLAM(I)=NB*DONE(I)+GAMMA(I)
032 C      IF(NB.EQ.0) GO TO 3
033 C
034 C      ----- CALCULATE BATEMAN EQUATIONS IF NB=1 -----
035 C      Q=V*WC
036 C      A(1)=ZERO(1)
037 C      A(2)=DONE(1)*A(1)/(RLAM(2)-RLAM(1))
038 C      A(3)=ZERO(2)-A(2)
039 C      A(4)=DONE(2)*A(2)/(RLAM(3)-RLAM(1))
040 C      A(5)=DONE(2)*A(3)/(RLAM(3)-RLAM(2))
041 C      A(6)=ZERO(3)-A(4)-A(5)
042 C      A(7)=DONE(3)*A(4)/(RLAM(4)-RLAM(1))
043 C      A(8)=DONE(3)*A(5)/(RLAM(4)-RLAM(2))
044 C      A(9)=DONE(3)*A(6)/(RLAM(4)-RLAM(3))
045 C      A(10)=ZERO(4)-A(7)-A(8)-A(9)
046 C      K=0
047 C      DO 2 I=1,4
048 C      DO 2 J=1,I
049 C      K=K+1
050 C      2 B(K)=A(K)*GAMMA(I)/Q
051 C      GO TO 4
052 C      3 READ(5,1002) (B(I),I=1,10)
053 C
054 C      ----- WRITE INPUT PARAMETERS -----
055 C      4 IF(NS.LT.0) GO TO 14
056 C      WRITE(6,1003)
057 C      IF(KSURF.EQ.0) WRITE(6,1004)
058 C      IF(KSURF.NE.0) WRITE(6,1005)
059 C      WRITE(6,1006) TITLE
060 C      WRITE(6,1007) NS,V,D,NR,WC,TP,NB
061 C      WRITE(6,1008) (I,ZERO(I),R(I),DONE(I),GAMMA(I),RLAM(I),I=1,NS)
062 C      WRITE(6,1009) (I,B(I),I=1,10)
063 C
064 C      ----- MULTIPLY DONE WITH R IF NR=1 -----
065 C      IF(NR.EQ.0) GO TO 6
066 C      DO 5 I=1,4
067 C      5 DONE(I)=R(I)*DONE(I)
068 C
069 C      ----- CALCULATE VARIOUS CONSTANTS -----
070 C      6 D12=DONE(1)-DONE(2)
071 C      D13=DONE(1)-DONE(3)
072 C      D23=DONE(2)-DONE(3)
073 C      D14=DONE(1)-DONE(4)
074 C      D24=DONE(2)-DONE(4)

```



```

075      D34=DONE(3)-DONE(4)
076      R12=R(1)-R(2)
077      R13=R(1)-R(3)
078      R23=R(2)-R(3)
079      R14=R(1)-R(4)
080      R24=R(2)-R(4)
081      R34=R(3)-R(4)
082      B123=-DONE(1)*R23+DONE(2)*R13-DONE(3)*R12
083      B124=-DONE(1)*R24+DONE(2)*R14-DONE(4)*R12
084      B134=-DONE(1)*R34+DONE(3)*R14-DONE(4)*R13
085      B234=-DONE(2)*R34+DONE(3)*R24-DONE(4)*R23
086      DD=DONE(1)*DONE(2)
087      DP=DONE(2)*DONE(3)
088      DDD=DD*DONE(3)
089      C21=DONE(1)*B(1)/(D12-R12*RLAM(1))
090      C31=DONE(2)*B(2)/(D23-R23*RLAM(1))
091      C32=DONE(2)*B(3)/(D23-R23*RLAM(2))
092      C331=DD*B(1)*R12/(B123*(R12*RLAM(1)-D12))
093      C332=DD*B(1)*R13/(B123*(R13*RLAM(1)-D13))
094      C333=DD*B(1)*R23/(B123*(R23*RLAM(1)-D23))
095      C41=DONE(3)*B(4)/(D34-R34*RLAM(1))
096      C42=DONE(3)*B(5)/(D34-R34*RLAM(2))
097      C43=DONE(3)*B(6)/(D34-R34*RLAM(3))
098      C441=DP*B(2)*R34/(B234*(R34*RLAM(1)-D34))
099      C442=DP*B(2)*R23/(B234*(R23*RLAM(1)-D23))
100      C443=DP*B(2)*R24/(B234*(R24*RLAM(1)-D24))
101      C451=DP*B(3)*R34/(B234*(R34*RLAM(2)-D34))
102      C452=DP*B(3)*R23/(B234*(R23*RLAM(2)-D23))
103      C453=DP*B(3)*R24/(B234*(R24*RLAM(2)-D24))
104      C461=DDD*B(1)*R14**2/(B134*B124*(R14*RLAM(1)-D14))
105      C462=DDD*B(1)*R24**2/(B124*B234*(R24*RLAM(1)-D24))
106      C463=DDD*B(1)*R34**2/(B134*B234*(R34*RLAM(1)-D34))
107      C464=DDD*B(1)*R13**2/(B123*B134*(R13*RLAM(1)-D13))
108      C465=DDD*B(1)*R23**2/(B123*B234*(R23*RLAM(1)-D23))
109      C466=DDD*B(1)*R12**2/(B123*B124*(R12*RLAM(1)-D12))
110  C
111  C      ----- PRINT COEFFICIENTS IF KDB.GT.0 -----
112      IF(KDB.GT.0) WRITE(6,1010) C21,C31,C32,C331,C332,C333,C41,C42,C43
113      1,C441,C442,C443,C451,C452,C453,C461,C462,C463,C464,C465,C466
114      F12=0.
115      F13=0.
116      F23=0.
117      F32=0.
118      F14=0.
119      F24=0.
120      F42=0.
121      F34=0.
122      F43=0.
123      F44=0.
124      E=0.
125      P12=DABS(1000.*R12/R(1))
126      P13=DABS(1000.*R13/R(1))
127      P23=DABS(1000.*R23/R(2))
128      P14=DABS(1000.*R14/R(1))
129      P24=DABS(1000.*R24/R(2))
130      P34=DABS(1000.*R34/R(3))
131      IF(DX.EQ.0.) DX=1.0
132      IF(DT.EQ.0.) DT=1.0
133      IMAX=(XM+DX-XI)/DX
134      JMAX=(TM+DT-TI)/DT
135  C
136  C      ----- DYNAMIC PART OF PROGRAM -----
137      DO 12 JJ=1,JMAX
138      IF(IMAX.GE.JJ) WRITE(6,1011)
139      TIME=TI+(JJ-1)*DT
140      DO 12 II=1,IMAX
141      X=XI+(II-1)*DX
142      VVO=0.0
143      IF(X.EQ.0.) GO TO 7
144      VVO=V*TIME/X
145      7 DO 10 KK=1,2
146      C1=0.0
147      C2=0.0
148      C3=0.0
149      C4=0.0
150      E1=1.0
151      E2=1.0
152      E3=1.0
153      E4=1.0

```

```

154      T=TIME+(1-KK)*TP
155      IF(T.LE.0.) GO TO 10
156      IF(KK.EQ.1) GO TO 8
157      E1=EXF(-RLAM(1)*TP,E)
158      E2=EXF(-RLAM(2)*TP,E)
159      E3=EXF(-RLAM(3)*TP,E)
160      E4=EXF(-RLAM(4)*TP,E)
161      8 F110=F(1,0)*E1
162      C
163      C      ----- FIRST TRACER -----
164      C1=B(1)*F110
165      IF(NS.EQ.1) GO TO 9
166      C
167      C      ----- SECOND TRACER -----
168      F220=F(2,0)*E2
169      F210=F(2,-1)*E1
170      IF(P12.GT.1.) F12=F(1,2)*E1
171      C2=B(2)*F210+B(3)*F220+C21*(F210-F110-F12)
172      IF(NS.EQ.2) GO TO 9
173      C
174      C      ----- THIRD TRACER -----
175      F310=F(3,-1)*E1
176      F320=F(3,-2)*E2
177      F330=F(3,0)*E3
178      IF(P13.GT.1.) F13=F(1,3)*E1
179      IF(P23.GT.1.) F23=F(2,3)
180      F32=F23*E2
181      F23=F23*E1
182      IF(KDB.EQ.2) WRITE(6,1012) F110,F210,F220,F12,F310,F320,F330,F13,F
183      123,F32
184      C3=B(4)*F310+B(5)*F320+B(6)*F330+C31*(F310-F210-F23)+C32*(F320-F22
185      10-F32)+C331*(F210-F110-F12)+C332*(F13-F310+F110)+C333*(F310-F210-F
186      223)
187      IF(NS.EQ.3) GO TO 9
188      C
189      C      ----- FOURTH TRACER -----
190      F410=F(4,-1)*E1
191      F420=F(4,-2)*E2
192      F430=F(4,-3)*E3
193      F440=F(4,0)*E4
194      IF(P14.GT.1.) F14=F(1,4)*E1
195      IF(P24.GT.1.) F24=F(2,4)
196      IF(P34.GT.1.) F34=F(3,4)
197      F42=F24*E2
198      F24=F24*E1
199      F43=F34*E3
200      F44=F34*E2
201      F34=F34*E1
202      IF(KDB.EQ.2) WRITE(6,1013) F410,F420,F430,F440,F14,F24,F42,F34,F4
203      1,F44
204      C4=B(7)*F410+B(8)*F420+B(9)*F430+B(10)*F440+C41*(F410-F310-F34)+C4
205      12*(F420-F320-F44)+C43*(F430-F330-F43)+C441*(F410-F310-F34)+C442*(F
206      2310-F210-F23)+C443*(F24-F410+F210)+C451*(F420-F320-F44)+C452*(F320
207      3-F220-F32)+C453*(F42-F420+F220)+C461*(F110-F410+F14)+C462*(F410-F2
208      410-F24)+C463*(F34-F410+F310)+C464*(F310-F110-F13)+C465*(F23-F310+F
209      5210)+C466*(F12-F210+F110)
210      9 IF(KK.EQ.2) GO TO 10
211      CONC1=C1
212      CONC2=C2
213      CONC3=C3
214      CONC4=C4
215      10 CONTINUE
216      CONC1=CONC1-C1
217      CONC2=CONC2-C2
218      CONC3=CONC3-C3
219      CONC4=CONC4-C4
220      IF(KPR.EQ.0) WRITE(6,1014) X,TIME,VVO,CONC1,CONC2,CONC3,CONC4
221      12 IF(KPR.EQ.1) WRITE(6,1015) X,TIME,VVO,CONC1,CONC2,CONC3,CONC4
222      14 CONTINUE
223      C
224      C      -----
225      1000 FORMAT(6I5,5F10.0)
226      1001 FORMAT(20A4)
227      1002 FORMAT(8F10.0)
228      1003 FORMAT(1H1,10X,82(1H*)/11X,1H*,80X,1H*/11X,1H*,9X,'MULTI-ION SOLUT
229      1E TRANSPORT',30X,'CHAIN',10X,1H*/11X,1H*,80X,1H*/11X,1H*,9X,'FOUR
230      2SPECIES',59X,1H*/11X,1H*,9X,'PREFERENTIAL RELEASE',51X,1H*/11X,1H*
231      3,80X,1H*)

```

```

232 1004 FORMAT(11X,1H*,9X,'FIRST-TYPE BOUNDARY CONDITION',42X,1H*)
233 1005 FORMAT(11X,1H*,9X,'THIRD-TYPE BOUNDARY CONDITION',42X,1H*)
234 1006 FORMAT(11X,1H*,80X,1H*/11X,1H*,20A4,1H*/11X,1H*,80X,1H*/11X,82(1H*
235 1))
236 1007 FORMAT(/11X,'INPUT PARAMETERS'/11X,16(1H=)/11X,'NS =' ,I5,10X,'V
237 1=' ,F12.4,10X,'D =' ,F12.4/11X,'NR =' ,I5,10X,'WC =' ,F11.4,10X,'TP ='
238 2,F11.4/11X,'NB =' ,I5)
239 1008 FORMAT(/9X,'TRACER',7X,'ZERO',13X,'R',11X,'DONE',11X,'GAMMA',11X
240 1,'RLAM'/(11X,I2,D16.5,F14.3,3F15.8))
241 1009 FORMAT(/11X,'B(I)-COEFFICIENTS'/11X,17(1H=)/11X,4('I',9X,'B(I)'
242 1,9X)/(10X,4(I2,D15.6,6X)))
243 1010 FORMAT(/9X,'C21=' ,D13.5,6X,'C31=' ,D13.5,6X,'C32=' ,D13.5/9X,
244 1'C331=' ,D12.5,6X,'C332=' ,D12.5,6X,'C333=' ,D12.5/9X,'C41=' ,
245 2D13.5,6X,'C42=' ,D13.5,6X,'C43=' ,D13.5/9X,'C441=' ,D12.5,6X,
246 3'C442=' ,D12.5,6X,'C443=' ,D12.5/9X,'C451=' ,D12.5,6X,'C452=' ,
247 4D12.5,6X,'C453=' ,D12.5/9X,'C461=' ,D12.5,6X,'C462=' ,D12.5,6X,
248 5'C463=' ,D12.5/9X,'C464=' ,D12.5,6X,'C465=' ,D12.5,6X,'C466=' ,
249 6D12.5)
250 1011 FORMAT(/8X,'DISTANCE',6X,'TIME',7X,'PORE VOLUME',4X,19(1H-),'CON
251 1CENTRATION',19(1H-)/11X,'(X)',8X,'(T)',9X,'(VVO)',11X,'(1)',11X,'(
252 22)',11X,'(3)',11X,'(4)')
253 1012 FORMAT(/9X,'F110=' ,D12.5,4X,'F210=' ,D12.5,4X,'F220=' ,D12.5/9X,
254 1'F12=' ,D13.5,4X,'F310=' ,D12.5,4X,'F320=' ,D12.5/9X,'F330=' ,D12.
255 25,4X,'F13=' ,D13.5,4X,'F23=' ,D13.5/9X,'F32=' ,D13.5)
256 1013 FORMAT(/9X,'F410=' ,D12.5,4X,'F420=' ,D12.5,4X,'F430=' ,D12.5/9X,
257 1'F440=' ,D12.5,4X,'F14=' ,D13.5,4X,'F24=' ,D13.5/9X,'F42=' ,D13.5,
258 24X,'F34=' ,D13.5,4X,'F43=' ,D13.5/9X,'F44=' ,D13.5)
259 1014 FORMAT(2X,3F13.3,2X,4F14.5)
260 1015 FORMAT(2X,3F13.3,2X,4D14.5)
261 STOP
262 END

```

```

263 FUNCTION F(I,JJ)
264 C
265 C PURPOSE: TO CALCULATE FIJK-TERMS
266 C
267 IMPLICIT REAL*8 (A-H,O-Z)
268 COMMON R(4),DONE(4),RLAM(4),D,V,X,T,KSURF
269 C
270 C -----
271 J=IABS(JJ)
272 IF(JJ.LT.0) A=RLAM(J)
273 IF(JJ.EQ.0) A=RLAM(I)
274 IF(JJ.GT.0) A=(DONE(I)-DONE(J))/(R(I)-R(J))
275 U2=V*V+4.*D*(DONE(I)-A*R(I))
276 S1=2.*DSQRT(D*R(I)*T)
277 U=DSQRT(DABS(U2))
278 VT=V*T
279 UT=U*T
280 E=0.0
281 C
282 C ----- CASE FOR POSITIVE VALUES OF U2 -----
283 IF(U2.LT.0.) GO TO 28
284 AM=0.5*(V-U)*X/D-A*T
285 BM1=(R(I)*X-UT)/S1
286 BP1=(R(I)*X+UT)/S1
287 AP=0.5*(U+V)*X/D-A*T
288 IF(KSURF.EQ.0) GO TO 20
289 C
290 C ----- CASE FOR THIRD-TYPE BOUNDARY CONDITION -----
291 CP1=(R(I)*X+VT)/S1
292 ALP=.3275911
293 CM1=(R(I)*X-VT)/S1
294 DUV=DABS(1.-U/V)
295 IF((BP1.GT.4.).AND.(CP1.GT.4.)) GO TO 4
296 IF((BP1.LT.2.5).AND.(CP1.LT.2.5)) GO TO 2
297 IF(DUV.GT.0.1) GO TO 1
298 IF(CP1.GT.3.) GO TO 4
299 GO TO 2
300 1 E1=2./((U/V)**2-1.)
301 G1=V*X/D-DONE(I)*T/R(I)
302 F1=V/(U-V)*EXF(AP,BP1)-E1*EXF(G1,CP1)
303 GO TO 6
304 2 D1=-CM1**2-DONE(I)*T/R(I)
305 BETA=ALP*V*DSQRT(T/(D*R(I)))
306 T1=1./(1.+ALP*BP1)
307 T2=1./(1.+ALP*CP1)

```

```

308 C1=T1*T2*(1.+ALP*CM1)
309 C2=T1*(C1-BETA*T2**2)
310 C3=T1*(C2-BETA*T2**3)
311 C4=T1*(C3-BETA*T2**4)
312 C5=T1*(C4-BETA*T2**5)
313 F1=V*(.2548296*C1-.2844967*C2+1.421414*C3-1.453152*C4+1.061405*C5)
314 1*EXF(D1,E)/(V+U)
315 GO TO 6
316 4 B=DONE(I)*T/R(I)+CM1**2
317 H=1./(BP1*(R(I)*X+VT))
318 WT=H*VT
319 HT=H*UT
320 B1=WT+2.*HT
321 B2=WT*B1+2.*HT**2
322 B3=WT*B2+2.*HT**3
323 B4=WT*B3+2.*HT**4
324 B5=WT*B4+2.*HT**5
325 B6=WT*B5+2.*HT**6
326 B7=WT*B6+2.*HT**7
327 B8=WT*B7+2.*HT**8
328 Z1=H*R(I)*X
329 Z2=Z1**2
330 Z3=Z1*Z2
331 Z4=Z1*Z3
332 Z5=Z1*Z4
333 Z6=Z1*Z5
334 Z7=Z1*Z6
335 Z8=Z1*Z7
336 Z9=Z1*Z8
337 A1=Z1-WT
338 A2=Z3-WT*(B2+3.*(B1*Z1+Z2))
339 A3=Z5-WT*(B4+5.*(B3*Z1+Z4)+10.*(B2*Z2+B1*Z3))
340 A4=Z7-WT*(B6+7.*(B5*Z1+Z6)+21.*(B4*Z2+B1*Z5)+35.*(B3*Z3+B2*Z4))
341 A5=Z9-WT*(B8+9.*(B7*Z1+Z8)+36.*(B6*Z2+B1*Z7)+84.*(B5*Z3+B2*Z6)+126
342 1.*(B4*Z4+B3*Z5))
343 F1=.5641896*V*EXF(-B,E)*(A1-.5*A2+.75*A3-1.875*A4+6.5625*A5)/(U+V)
344 6 IF(JJ.GT.0) GO TO 8
345 F=V/(V+U)*EXF(AM,BM1)-F1
346 RETURN
347 8 S2=2.*DSQRT(D*R(J)*T)
348 BM2=(R(J)*X-UT)/S2
349 BP2=(R(J)*X+UT)/S2
350 CP2=(R(J)*X+VT)/S2
351 CM2=(R(J)*X-VT)/S2
352 IF((BP2.GT.4.).AND.(CP2.GT.4.)) GO TO 14
353 IF((BP2.LT.2.5).AND.(CP2.LT.2.5)) GO TO 10
354 IF(DUV.GT.0.1) GO TO 9
355 IF(CP2.GT.3.) GO TO 14
356 GO TO 10
357 9 E2=2./((U/V)**2-1.)
358 G2=V*X/D-DONE(J)*T/R(J)
359 AP=0.5*(V+U)*X/D-A*T
360 F2=V/(U-V)*EXF(AP,BP2)-E2*EXF(G2,CP2)
361 GO TO 18
362 10 D2=-CM2**2-DONE(J)*T/R(J)
363 BETA=ALP*V*DSQRT(T/(D*R(J)))
364 T1=1./(1.+ALP*BP2)
365 T2=1./(1.+ALP*CP2)
366 C1=T1*T2*(1.+ALP*CM2)
367 C2=T1*(C1-BETA*T2**2)
368 C3=T1*(C2-BETA*T2**3)
369 C4=T1*(C3-BETA*T2**4)
370 C5=T1*(C4-BETA*T2**5)
371 F2=V*(.2548296*C1-.2844967*C2+1.421414*C3-1.453152*C4+1.061405*C5)
372 1*EXF(D2,E)/(V+U)
373 GO TO 16
374 14 B=DONE(J)*T/R(J)+CM2**2
375 H=1./(BP2*(R(J)*X+VT))
376 WT=H*VT
377 HT=H*UT
378 B1=WT+2.*HT
379 B2=WT*B1+2.*HT**2
380 B3=WT*B2+2.*HT**3
381 B4=WT*B3+2.*HT**4
382 B5=WT*B4+2.*HT**5
383 B6=WT*B5+2.*HT**6
384 B7=WT*B6+2.*HT**7
385 B8=WT*B7+2.*HT**8

```

```

386      Z1=H*(J)*X
387      Z2=Z1**2
388      Z3=Z1*Z2
389      Z4=Z1*Z3
390      Z5=Z1*Z4
391      Z6=Z1*Z5
392      Z7=Z1*Z6
393      Z8=Z1*Z7
394      Z9=Z1*Z8
395      A1=Z1-WT
396      A2=Z3-WT*(B2+3.*(B1*Z1+Z2))
397      A3=Z5-WT*(B4+5.*(B3*Z1+Z4)+10.*(B2*Z2+B1*Z3))
398      A4=Z7-WT*(B6+7.*(B5*Z1+Z6)+21.*(B4*Z2+B1*Z5)+35.*(B3*Z3+B2*Z4))
399      A5=Z9-WT*(B8+9.*(B7*Z1+Z8)+36.*(B6*Z2+B1*Z7)+84.*(B5*Z3+B2*Z6)+126
400      1.*(B4*Z4+B3*Z5))
401      F2=.5641896*V*EXF(-B,E)*(A1-.5*A2+.75*A3-1.875*A4+6.5625*A5)/(U+V)
402  16 IF((BM1.GT.0.).OR.(BM2.GT.0.)) GO TO 18
403      Q=-BM2
404      BM2=-BM1
405      BM1=Q
406  18 F=V/(V+U)*(EXF(AM,BM2)-EXF(AM,BM1))+F1-F2
407      RETURN
408  C
409  C ----- CASE FOR FIRST-TYPE BOUNDARY CONDITION -----
410  20 IF(JJ.GT.0) GO TO 22
411      F=0.5*(EXF(AM,BM1)+EXF(AP,BP1))
412      RETURN
413  22 S2=2.*DSQRT(D*R(J)*T)
414      BM2=(R(J)*X-UT)/S2
415      BP2=(R(J)*X+UT)/S2
416      IF((BM1.GT.0.).OR.(BM2.GT.0.)) GO TO 24
417      Q=-BM2
418      BM2=-BM1
419      BM1=Q
420  24 F=0.5*(EXF(AM,BM2)-EXF(AM,BM1)+EXF(AP,BP2)-EXF(AP,BP1))
421      RETURN
422  C
423  C ----- CASE FOR NEGATIVE VALUES OF U2 -----
424  28 AM=0.5*V*X/D-A*T
425      ARG=0.5*U*X/D
426      CS=DCOS(ARG)
427      SN=DSIN(ARG)
428      A1=R(I)*X/S1
429      A2=UT/S1
430      CALL CEXF(A1,A2,AM,X1,Y1)
431      IF(KSURF.EQ.0) GO TO 32
432      CP1=(R(I)*X+VT)/S1
433      CM1=V*X/D-DONE(I)*T/R(I)
434      C1=2.*V/(V*V-U2)
435      C2=0.5*V*V/(D*(DONE(I)-A*R(I)))
436      IF(JJ.GT.0) GO TO 30
437      F=C1*(CS*(X1*V-Y1*U)-SN*(X1*U+Y1*V))+C2*EXF(CM1,CP1)
438      RETURN
439  30 CONTINUE
440      S2=2.*DSQRT(D*R(J)*T)
441      CM2=V*X/D-DONE(J)*T/R(J)
442      C3=0.5*V*V/(D*(DONE(J)-A*R(J)))
443      CP2=(R(J)*X+VT)/S2
444      A1=R(J)*X/S2
445      A2=UT/S2
446      CALL CEXF(A1,A2,AM,X2,Y2)
447      F=C1*(CS*(V*(X2-X1)-U*(Y2-Y1))-SN*(U*(X2-X1)+V*(Y2-Y1)))+C3*EXF(CM
448      12,CP2)-C2*EXF(CM1,CP1)
449      RETURN
450  32 IF(JJ.GT.0) GO TO 34
451      F=CS*X1-SN*Y1
452      RETURN
453  34 S2=2.*DSQRT(D*R(J)*T)
454      A1=R(J)*X/S2
455      A2=UT/S2
456      CALL CEXF(A1,A2,AM,X2,Y2)
457      F=CS*(X2-X1)-SN*(Y2-Y1)
458      RETURN
459      END

460      FUNCTION EXF(A,B)
461  C

```

```

462 C   PURPOSE: TO CALCULATE EXP(A) ERFC(B)
463 C
464     IMPLICIT REAL*8 (A-H,O-Z)
465     EXF=0.0
466     IF((DABS(A).GT.170.).AND.(B.LE.0.)) RETURN
467     IF(B.NE.0.0) GO TO 1
468     EXF=DEXP(A)
469     RETURN
470 1 C=A-B*B
471   IF((DABS(C).GT.170.).AND.(B.GT.0.)) RETURN
472   IF(C.LT.-170.) GO TO 4
473   X=DABS(B)
474   IF(X.GT.2.5) GO TO 2
475   T=1./(1.+3275911*X)
476   Y=T*(.2548296-T*(.2844967-T*(1.421414-T*(1.453152-1.061405*T))))
477   GO TO 3
478 2 Y=.5641896/(X+.5/(X+1./(X+1.5/(X+2./(X+2.5/(X+3./(X+1.))))))
479 3 EXF=Y*DEXP(C)
480 4 IF(B.LT.0.0) EXF=2.*DEXP(A)-EXF
481   RETURN
482   END

483     SUBROUTINE CEXF(A,B,Z,U,V)
484 C
485 C   COMPLEX ERFC-FUNCTION: U+IV=EXP(Z)ERFC(A+IB)
486 C
487     IMPLICIT REAL*8 (A-H,O-Z)
488     DIMENSION W(10),H(10)
489     DATA W/.4622437,.2866755,.1090172,.02481052,.3243773D-2,
490     1.2283386D-3,.7802556D-5,.1086069D-6,.4399341D-9,.2229394D-12/,
491     2H/.2453407,.7374737,1.234076,1.738538,2.254974,
492     32.788806,3.347855,3.944764,4.603682,5.387481/
493 C
494 C   -----
495     X=DABS(A)
496     Y=DABS(B)
497     U=0.0
498     V=0.0
499     E=0.0
500     XYZ=Y*Y+Z-X*X
501     IF(DABS(XYZ).GT.170.)RETURN
502     COS=DCOS(2.*X*Y)
503     SIN=DSIN(2.*X*Y)
504     IF((X+Y).LT.6.) GO TO 2
505     T=0.0
506     DO 1 K=1,10
507       T=T+W(K)*((X/((Y-H(K))**2+X*X)))+(X/((Y+H(K))**2+X*X))
508 1 V=V+W(K)*((Y-H(K))/((Y-H(K))**2+X*X)+(Y+H(K))/((Y+H(K))**2+X*X))
509     U=.3183099*DEXP(XYZ)*(T*COS-V*SIN)
510     V=.3183099*DEXP(XYZ)*(-T*SIN-V*COS)
511     IF(X.EQ.0.)U=DEXP(DMIN1(Z,1.7D2))
512     GO TO 8
513 2 IF(X.GT.2.5) GO TO 3
514   T=1./(1.+3275911*X)
515   U=T*(.2548296-T*(.2844967-T*(1.421414-T*(1.453152-1.06145*T))))
516   GO TO 4
517 3 U=.5641896/(X+.5/(X+1./(X+1.5/(X+2./(X+2.5/(X+3./(X+1.))))))
518 4 IF(Y.EQ.0.) GO TO 7
519   IF(X.EQ.0.) V=-.3183099*Y
520   IF(X.EQ.0.) GO TO 5
521   U=U-.1591549*(1.D0-COS)/X
522   V=V-.1591549*SIN/X
523 5 NT=12.+2.*Y
524   DO 6 I=1,NT
525     P=I
526     ARG=P*Y
527     F1=X*(DEXP(ARG)+DEXP(-ARG))
528     F2=0.5*P*(DEXP(ARG)-DEXP(-ARG))
529     EX=.6366198*DEXP(-0.25*P*P)/(4.*X*X+P*P)
530     U=U-EX*(2.*X-F1*COS+F2*SIN)
531 6 V=V-EX*(F1*SIN+F2*COS)
532     V=V*DEXP(Z-X*X)
533 7 U=U*DEXP(Z-X*X)
534 8 IF(B.LT.0.) V=-V
535   IF(A.LT.0.) U=2.*EXF(Z,E)-U
536   RETURN
537   END

```

APPENDIX 3

Definition of the most significant variables

<u>Variable</u>	<u>Definition</u>
A(10)	Vector containing the values of the coefficients $A_1 - A_{10}$.
B(10)	Vector containing the values of the coefficients $B_1 - B_{10}$.
C1,...,C4	Dummy variables for the concentration of the four species.
CONC1,...,CONC4	Calculated concentrations of the four chain members.
D	Dispersion coefficient.
DONE(4)	Values of μ_i for the four chain members.
DT	t-increment for printout.
DX	x-increment for printout.
F12	Value of the term S_{12} ; more general:
FIJ	Value of the term S_{ij} .
F110	Value of the term F_{110} ; more general:
FIJK	Value of the term F_{ijk} .
GAMMA(4)	Values of γ_i for the four chain members.
KDB	Debugging code. If $KDB > 0$, various coefficients will be printed during execution. In addition, values of all terms F_{ijk} and S_{ij} will be printed during execution when $KDB = 2$.
KPR	Format code for printout. If $KPR = 0$, concentrations are printed in regular F14.5 format; if $KPR = 1$, concentrations are printed in the exponential D14.5 format.
KSURF	Code specifying the input boundary condition. If $KSURF = 0$, the solution for a first-type boundary condition will be given; if $KSURF \neq 0$, the solution for a third-type boundary conditions is given.
NB	Boundary code that specifies if the Bateman equations must be used for the input boundary conditions. If $NB = 1$, the parameters A_i and B_i are calculated in the program. If $NB = 0$, the Bateman equations are not used; values of the parameters B_i are then read in.
NC	Number of examples to be executed.
NR	Code specifying the form of the decay terms. If $NR = 0$, the retardation factors R_i are dropped from the decay terms in the transport equations; if $NR = 1$, the decay terms are not altered.
NS	Number of chain members considered ($NS \leq 4$).
Q	Volumetric flux density.
R(4)	Values of R_i for the four chain members.
RLAM(4)	Values of λ_i for the four chain members.
T	Dummy variable for time.
TI	Smallest t-value for which concentrations are calculated.
TIME	Time, t.
TITLE(20)	Title (input label).
TM	Maximum value of t for which concentrations are calculated.
TP	Pulse time, t_0 .
V	Average pore-water velocity, v.
VVO	Local value of the amount of pore volumes ($VVO = vt/x$).
WC	Volumetric water content, θ (dummy variable if $NB = 0$).

X	Distance, x.
XI	Smallest value of x for which concentrations are calculated.
XM	Maximum value of x for which concentrations are calculated.
ZERO(4)	Values of M_1^0 for the four chain members; not needed if NB=0.

APPENDIX 4

Input data instructions

Line	Variables	Format	Comments
1	NC	I5	Number of problems to be considered. The remaining lines are read in for each case (hence, they are repeated NC times).
2	TITLE	20(A4)	Title.
3	NS,NR,NB,KSURF, KDB,KPR,V,D,WC, TP	6I5,4F10.0	Value for WC is not needed if NB = 0.
4	(ZERO(I) (GAMMA(I), I=1,4)	8F10.0	Use dummy values for ZERO(I) = M_1^0 if NB = 0.
5	XI,DX,XM TI,DT,TM	8F10.0	These parameters define the x and t-values for which concentrations are calculated.
6,7	(B(I),I=1,10)	8F10.0	These two lines are needed only if NB = 0.

APPENDIX 5

Input data for the two examples considered in this study

2	EXAMPLE 1: NITRIFICATION (CHO,1971)								
3	0	0	1	0	0	1.0	0.18	1.00	200.0
	2.00	1.00	1.00	5.0	0.01	0.10	0.0	0.95	
	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	0.0	5.0	200.0	200.0	0.0	200.0			
	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	0.0	0.0							
	EXAMPLE 2: RADIONUCLIDE TRANSPORT								
4	1	1	1	0	1	100.00	10.0	0.4	10000.
	10000.0	14000.0	50000.0	500.0	.0079	0.0000028	0.0000087	0.00043	
	50000.	0.0	.0	0.0	.001	.001	.001	.001	
	0.0	5.0	250.0	1000.0	9000.0	10000.0			

APPENDIX 6

Output file for the data of Appendix 5

```

*****
*
*      MULTI-ION SOLUTE TRANSPORT                      CHAIN
*
*      FOUR SPECIES
*      PREFERENTIAL RELEASE
*
*      THIRD-TYPE BOUNDARY CONDITION
*
*      EXAMPLE 1: NITRIFICATION (CHO,1971)
*
*****

```


INPUT PARAMETERS

=====

NS =	3	V =	1.0000	D =	0.1800
NR =	0	WC =	1.0000	TP =	200.0000
NB =	0				

TRACER	ZERO	R	DONE	GAMMA	RLAM
1	0.00000D+00	2.000	0.01000000	0.00000000	0.00000000
2	0.00000D+00	1.000	0.10000000	0.00000000	0.00000000
3	0.00000D+00	1.000	0.00000000	0.00000000	0.00000000

B(I)-COEFFICIENTS

=====

I	B(I)	I	B(I)	I	B(I)	I	B(I)
1	0.100000D+01	2	0.000000D+00	3	0.000000D+00	4	0.000000D+00
5	0.000000D+00	6	0.000000D+00	7	0.000000D+00	8	0.000000D+00
9	0.000000D+00	10	0.000000D+00				

DISTANCE (X)	TIME (T)	PORE VOLUME (VVO)	-----CONCENTRATION-----			
			(1)	(2)	(3)	(4)
0.000	200.000	0.000	0.99821	0.00173	0.00006	0.00000
5.000	200.000	40.000	0.94961	0.03871	0.01168	0.00000
10.000	200.000	20.000	0.90338	0.05951	0.03712	0.00000
15.000	200.000	13.333	0.85940	0.07048	0.07012	0.00000
20.000	200.000	10.000	0.81756	0.07554	0.10690	0.00000
25.000	200.000	8.000	0.77775	0.07706	0.14519	0.00000
30.000	200.000	6.667	0.73989	0.07648	0.18363	0.00000
35.000	200.000	5.714	0.70387	0.07470	0.22143	0.00000
40.000	200.000	5.000	0.66960	0.07226	0.25815	0.00000
45.000	200.000	4.444	0.63700	0.06947	0.29354	0.00000
50.000	200.000	4.000	0.60599	0.06653	0.32748	0.00000
55.000	200.000	3.636	0.57648	0.06356	0.35995	0.00000
60.000	200.000	3.333	0.54842	0.06063	0.39095	0.00000
65.000	200.000	3.077	0.52172	0.05778	0.42050	0.00000
70.000	200.000	2.857	0.49632	0.05503	0.44865	0.00000
75.000	200.000	2.667	0.47215	0.05239	0.47545	0.00000
80.000	200.000	2.500	0.44901	0.04986	0.50097	0.00000
85.000	200.000	2.353	0.42508	0.04738	0.52522	0.00000
90.000	200.000	2.222	0.38943	0.04453	0.54804	0.00000
95.000	200.000	2.105	0.31490	0.03979	0.56824	0.00000
100.000	200.000	2.000	0.19272	0.03122	0.58260	0.00000
105.000	200.000	1.905	0.07679	0.01995	0.58714	0.00000
110.000	200.000	1.818	0.01794	0.01024	0.58083	0.00000
115.000	200.000	1.739	0.00231	0.00449	0.56635	0.00000
120.000	200.000	1.667	0.00016	0.00183	0.54705	0.00000
125.000	200.000	1.600	0.00001	0.00073	0.52498	0.00000
130.000	200.000	1.538	0.00000	0.00029	0.50106	0.00000
135.000	200.000	1.481	0.00000	0.00012	0.47562	0.00000
140.000	200.000	1.429	0.00000	0.00005	0.44876	0.00000
145.000	200.000	1.379	0.00000	0.00002	0.42047	0.00000
150.000	200.000	1.333	0.00000	0.00001	0.39072	0.00000
155.000	200.000	1.290	0.00000	0.00000	0.35943	0.00000
160.000	200.000	1.250	0.00000	0.00000	0.32652	0.00000
165.000	200.000	1.212	0.00000	0.00000	0.29193	0.00000
170.000	200.000	1.176	0.00000	0.00000	0.25557	0.00000
175.000	200.000	1.143	0.00000	0.00000	0.21736	0.00000
180.000	200.000	1.111	0.00000	0.00000	0.17741	0.00000
185.000	200.000	1.081	0.00000	0.00000	0.13628	0.00000
190.000	200.000	1.053	0.00000	0.00000	0.09581	0.00000
195.000	200.000	1.026	0.00000	0.00000	0.05947	0.00000
200.000	200.000	1.000	0.00000	0.00000	0.03134	0.00000

```

*****
*
*      MULTI-ION SOLUTE TRANSPORT                      CHAIN
*
*      FOUR SPECIES
*      PREFERENTIAL RELEASE
*
*      THIRD-TYPE BOUNDARY CONDITION
*
*      EXAMPLE 2: RADIONUCLIDE TRANSPORT
*
*****

```

INPUT PARAMETERS

=====

```

NS =    4          V =   100.0000          D =    10.0000
NR =    1          WC =    0.4000          TP = 10000.0000
NB =    1

```

TRACER	ZERO	R	DONE	GAMMA	RLAM
1	0.50000D+05	10000.000	0.00790000	0.00100000	0.00890000
2	0.00000D+00	14000.000	0.00000280	0.00100000	0.00100280
3	0.00000D+00	50000.000	0.00000870	0.00100000	0.00100870
4	0.00000D+00	500.000	0.00043000	0.00100000	0.00143000

B(I)-COEFFICIENTS

=====

I	B(I)	I	B(I)	I	B(I)	I	B(I)
1	0.125000D+01	2	-0.125044D+01	3	0.125044D+01	4	0.443684D-03
5	0.593431D+00	6	-0.593874D+00	7	-0.516740D-06	8	0.120853D-01
9	-0.122637D-01	10	0.178925D-03				

DISTANCE (X)	TIME (T)	PORE VOLUME (VVO)	-----CONCENTRATION-----			
			(1)	(2)	(3)	(4)
0.000	1000.000	0.000	0.17223D-03	0.46515D+00	0.11223D-02	0.43196D-05
5.000	1000.000	20000.000	0.28532D-03	0.86173D+00	0.32843D-03	0.18007D-04
10.000	1000.000	10000.000	0.20949D-03	0.23503D-01	0.15595D-05	0.19105D-04
15.000	1000.000	6666.667	0.87774D-07	0.13240D-06	0.38435D-11	0.18089D-04
20.000	1000.000	5000.000	0.31171D-15	0.11647D-15	0.15561D-20	0.17095D-04
25.000	1000.000	4000.000	0.52002D-29	0.95033D-30	0.72587D-35	0.16128D-04
30.000	1000.000	3333.333	0.36043D-48	0.40354D-49	0.20127D-54	0.15188D-04
35.000	1000.000	2857.143	0.99318D-73	0.76193D-74	0.00000D+00	0.14274D-04
40.000	1000.000	2500.000	0.00000D+00	0.00000D+00	0.00000D+00	0.13386D-04
45.000	1000.000	2222.222	0.00000D+00	0.00000D+00	0.00000D+00	0.12525D-04
50.000	1000.000	2000.000	0.00000D+00	0.00000D+00	0.00000D+00	0.11690D-04
55.000	1000.000	1818.182	0.00000D+00	0.00000D+00	0.00000D+00	0.10883D-04
60.000	1000.000	1666.667	0.00000D+00	0.00000D+00	0.00000D+00	0.10102D-04
65.000	1000.000	1538.462	0.00000D+00	0.00000D+00	0.00000D+00	0.93487D-05
70.000	1000.000	1428.571	0.00000D+00	0.00000D+00	0.00000D+00	0.86226D-05
75.000	1000.000	1333.333	0.00000D+00	0.00000D+00	0.00000D+00	0.79242D-05
80.000	1000.000	1250.000	0.00000D+00	0.00000D+00	0.00000D+00	0.72536D-05
85.000	1000.000	1176.471	0.00000D+00	0.00000D+00	0.00000D+00	0.66110D-05
90.000	1000.000	1111.111	0.00000D+00	0.00000D+00	0.00000D+00	0.59969D-05
95.000	1000.000	1052.632	0.00000D+00	0.00000D+00	0.00000D+00	0.54114D-05
100.000	1000.000	1000.000	0.00000D+00	0.00000D+00	0.00000D+00	0.48548D-05
105.000	1000.000	952.381	0.00000D+00	0.00000D+00	0.00000D+00	0.43276D-05
110.000	1000.000	909.091	0.00000D+00	0.00000D+00	0.00000D+00	0.38300D-05
115.000	1000.000	869.565	0.00000D+00	0.00000D+00	0.00000D+00	0.33624D-05
120.000	1000.000	833.333	0.00000D+00	0.00000D+00	0.00000D+00	0.29251D-05
125.000	1000.000	800.000	0.00000D+00	0.00000D+00	0.00000D+00	0.25185D-05
130.000	1000.000	769.231	0.00000D+00	0.00000D+00	0.00000D+00	0.21429D-05
135.000	1000.000	740.741	0.00000D+00	0.00000D+00	0.00000D+00	0.17985D-05
140.000	1000.000	714.286	0.00000D+00	0.00000D+00	0.00000D+00	0.14857D-05
145.000	1000.000	689.655	0.00000D+00	0.00000D+00	0.00000D+00	0.12045D-05
150.000	1000.000	666.667	0.00000D+00	0.00000D+00	0.00000D+00	0.95504D-06
155.000	1000.000	645.161	0.00000D+00	0.00000D+00	0.00000D+00	0.73722D-06
160.000	1000.000	625.000	0.00000D+00	0.00000D+00	0.00000D+00	0.55073D-06
165.000	1000.000	606.061	0.00000D+00	0.00000D+00	0.00000D+00	0.39497D-06
170.000	1000.000	588.235	0.00000D+00	0.00000D+00	0.00000D+00	0.26898D-06

175.000	1000.000	571.429	0.00000D+00	0.00000D+00	0.00000D+00	0.17130D-06
180.000	1000.000	555.556	0.00000D+00	0.00000D+00	0.00000D+00	0.99831D-07
185.000	1000.000	540.541	0.00000D+00	0.00000D+00	0.00000D+00	0.51629D-07
190.000	1000.000	526.316	0.00000D+00	0.00000D+00	0.00000D+00	0.22713D-07
195.000	1000.000	512.821	0.00000D+00	0.00000D+00	0.00000D+00	0.80500D-08
200.000	1000.000	500.000	0.00000D+00	0.00000D+00	0.00000D+00	0.21611D-08
205.000	1000.000	487.805	0.00000D+00	0.00000D+00	0.00000D+00	0.41075D-09
210.000	1000.000	476.190	0.00000D+00	0.00000D+00	0.00000D+00	0.52264D-10
215.000	1000.000	465.116	0.00000D+00	0.00000D+00	0.00000D+00	0.42319D-11
220.000	1000.000	454.545	0.00000D+00	0.00000D+00	0.00000D+00	0.21017D-12
225.000	1000.000	444.444	0.00000D+00	0.00000D+00	0.00000D+00	0.62278D-14
230.000	1000.000	434.783	0.00000D+00	0.00000D+00	0.00000D+00	0.10719D-15
235.000	1000.000	425.532	0.00000D+00	0.00000D+00	0.00000D+00	0.10562D-17
240.000	1000.000	416.667	0.00000D+00	0.00000D+00	0.00000D+00	0.58848D-20
245.000	1000.000	408.163	0.00000D+00	0.00000D+00	0.00000D+00	0.18361D-22
250.000	1000.000	400.000	0.00000D+00	0.00000D+00	0.00000D+00	0.31844D-25

DISTANCE (X)	TIME (T)	PORE VOLUME (VVO)	-----CONCENTRATION-----			
			(1)	(2)	(3)	(4)
0.000	10000.000	0.000	0.28126D-38	0.55998D-04	0.15580D-05	0.23970D-07
5.000	10000.000	200000.000	0.46609D-38	0.11391D-03	0.16208D-04	0.16912D-06
10.000	10000.000	100000.000	0.77239D-38	0.23171D-03	0.14107D-03	0.15111D-05
15.000	10000.000	66666.667	0.12800D-37	0.47133D-03	0.79188D-03	0.10577D-04
20.000	10000.000	50000.000	0.21211D-37	0.95877D-03	0.12984D-02	0.35898D-04
25.000	10000.000	40000.000	0.35150D-37	0.19503D-02	0.12595D-02	0.63125D-04
30.000	10000.000	33333.333	0.58249D-37	0.39672D-02	0.12686D-02	0.90103D-04
35.000	10000.000	28571.429	0.96528D-37	0.80700D-02	0.12715D-02	0.11690D-03
40.000	10000.000	25000.000	0.15996D-36	0.16416D-01	0.12697D-02	0.14343D-03
45.000	10000.000	22222.222	0.26508D-36	0.33392D-01	0.12585D-02	0.16954D-03
50.000	10000.000	20000.000	0.43928D-36	0.67925D-01	0.12280D-02	0.19491D-03
55.000	10000.000	18181.818	0.72796D-36	0.13815D+00	0.11584D-02	0.21892D-03
60.000	10000.000	16666.667	0.12063D-35	0.27885D+00	0.10095D-02	0.24029D-03
65.000	10000.000	15384.615	0.19991D-35	0.49784D+00	0.71635D-03	0.25653D-03
70.000	10000.000	14285.714	0.33128D-35	0.50542D+00	0.30805D-03	0.26436D-03
75.000	10000.000	13333.333	0.54898D-35	0.16927D+00	0.53143D-04	0.26424D-03
80.000	10000.000	12500.000	0.90973D-35	0.13664D-01	0.26364D-05	0.26109D-03
85.000	10000.000	11764.706	0.15048D-34	0.23341D-03	0.31209D-07	0.25760D-03
90.000	10000.000	11111.111	0.24057D-34	0.80838D-06	0.81080D-10	0.25414D-03
95.000	10000.000	10526.316	0.30950D-34	0.57275D-09	0.45632D-13	0.25073D-03
100.000	10000.000	10000.000	0.22346D-34	0.89203D-13	0.59308D-17	0.24736D-03
105.000	10000.000	9523.810	0.66130D-35	0.37366D-17	0.21940D-21	0.24404D-03
110.000	10000.000	9090.909	0.67537D-36	0.65890D-22	0.36646D-26	0.24075D-03
115.000	10000.000	8695.652	0.21927D-37	0.86333D-27	0.47609D-31	0.23751D-03
120.000	10000.000	8333.333	0.21735D-39	0.10942D-31	0.60324D-36	0.23432D-03
125.000	10000.000	8000.000	0.64370D-42	0.13856D-36	0.76390D-41	0.23116D-03
130.000	10000.000	7692.308	0.56249D-45	0.17537D-41	0.96611D-46	0.22804D-03
135.000	10000.000	7407.407	0.14390D-48	0.21856D-46	0.11916D-50	0.22497D-03
140.000	10000.000	7142.857	0.10722D-52	0.23819D-51	0.12357D-55	0.22193D-03
145.000	10000.000	6896.552	0.23189D-57	0.16519D-56	0.77441D-61	0.21893D-03
150.000	10000.000	6666.667	0.14520D-62	0.51493D-62	0.21249D-66	0.21598D-03
155.000	10000.000	6451.613	0.26271D-68	0.58278D-68	0.21085D-72	0.21306D-03
160.000	10000.000	6250.000	0.13717D-74	0.21571D-74	0.00000D+00	0.21018D-03
165.000	10000.000	6060.606	0.00000D+00	0.00000D+00	0.00000D+00	0.20733D-03
170.000	10000.000	5882.353	0.00000D+00	0.00000D+00	0.00000D+00	0.20453D-03
175.000	10000.000	5714.286	0.00000D+00	0.00000D+00	0.00000D+00	0.20176D-03
180.000	10000.000	5555.556	0.00000D+00	0.00000D+00	0.00000D+00	0.19902D-03
185.000	10000.000	5405.405	0.00000D+00	0.00000D+00	0.00000D+00	0.19632D-03
190.000	10000.000	5263.158	0.00000D+00	0.00000D+00	0.00000D+00	0.19366D-03
195.000	10000.000	5128.205	0.00000D+00	0.00000D+00	0.00000D+00	0.19103D-03
200.000	10000.000	5000.000	0.00000D+00	0.00000D+00	0.00000D+00	0.18843D-03
205.000	10000.000	4878.049	0.00000D+00	0.00000D+00	0.00000D+00	0.18587D-03
210.000	10000.000	4761.905	0.00000D+00	0.00000D+00	0.00000D+00	0.18334D-03
215.000	10000.000	4651.163	0.00000D+00	0.00000D+00	0.00000D+00	0.18085D-03
220.000	10000.000	4545.455	0.00000D+00	0.00000D+00	0.00000D+00	0.17839D-03
225.000	10000.000	4444.444	0.00000D+00	0.00000D+00	0.00000D+00	0.17596D-03
230.000	10000.000	4347.826	0.00000D+00	0.00000D+00	0.00000D+00	0.17356D-03
235.000	10000.000	4255.319	0.00000D+00	0.00000D+00	0.00000D+00	0.17119D-03
240.000	10000.000	4166.667	0.00000D+00	0.00000D+00	0.00000D+00	0.16885D-03
245.000	10000.000	4081.633	0.00000D+00	0.00000D+00	0.00000D+00	0.16655D-03
250.000	10000.000	4000.000	0.00000D+00	0.00000D+00	0.00000D+00	0.16427D-03