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THE SAVANNAH RIVER LABORATORY DOSTOMAN CODE -
A COMPARTMENTAL PATHWAYS COMPUTER MODEL OF CONTAMINANT TRANSPORT

by

C. M. King, E. L. Wilhite, R. W. Root, Jr.,
D. J. Fauth, K. R. Routt, R. H. Emslie, and R. R. Beckmeyer

E. I. du Pont de Nemours and Company
Savannah River Laboratory
Aiken, SC 29808

and

R. A. Fjeld, G. A. Hutto, and J. A. Vandeven

Clemson University
Dept. of Environmental Systems Engineering
Clemson, SC

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C. M. King, E. L. Wilhite, R. W. Root, Jr., D. J. Fauth, K. R. Rott, R. H. Emslie, and R. R. Beckmeyer
E. I. du Pont de Nemours and Company
Savannah River Laboratory
Aiken, SC 29808

and

R. A. Fjeld, G. A. Hutto, and J. A. Vandeven
Clemson University
Dept. of Environmental Systems Engineering
Clemson, SC

ABSTRACT

The Savannah River Laboratory DOSTOMAN code has been used since 1978 for environmental pathway analysis of potential migration of radionuclides and hazardous chemicals. The DOSTOMAN work will be reviewed including a summary of historical use of compartmental models, the mathematical basis for the DOSTOMAN code, examples of exact analytical solutions for simple matrices, methods for numerical solution of complex matrices, and mathematical validation/calibration of the SRL code. The review includes the methodology for application to nuclear and hazardous chemical waste disposal, examples of use of the model in contaminant transport and pathway analysis, a user's guide for computer implementation, peer review of the code, and use of DOSTOMAN at other Department of Energy sites.

INTRODUCTION

Assessing the impact on man of radioactive and hazardous chemical waste disposal is an important problem in environmental science and engineering. An essential part of impact assessment is prediction of the long term transport of material in the environment. However, environmental transport is difficult to predict with precision, especially for time periods extending far into the future. This is because of the complex nature of the environment and environmental transport. Environmental systems are highly

heterogeneous and subject to change and environmental transport is governed by a variety of physical, chemical, and biological processes that are difficult to quantify. In spite of these difficulties and uncertainties, transport estimates must be made in order to assess the hazard associated with existing disposal sites or to evaluate the suitability of new ones. For example, federal regulations¹⁻² governing the licensing of radioactive waste disposal sites require estimates of long term transport. For low-level waste, predictions are required to 500 years; for high-level waste they are required to 10,000 years. It is reasonable to expect analogous requirements to be established eventually for hazardous chemical waste sites.

One method of predicting transport through complex environmental systems is by compartmental modeling. Developed and used extensively in biological tracer applications,³⁻⁶ the compartmental method has only recently been applied to environmental transport problems.⁷⁻⁹

Although not very elegant, compartmental modeling is an extremely practical method for making transport predictions. It is a semiempirical technique in which complex environmental transport pathways are approximated as a series of discrete, interconnected, homogeneous compartments. An environmental compartment is conceptually analogous to the continuous-flow, stirred-tank reactor (CFSTR) used in chemical engineering reactor modeling. Material accumulation in a CFSTR is dependent on influent flow concentration and various gains and losses within the reactor vessel. An environmental compartment is essentially a CFSTR in which material inputs, material outputs, and reactions are approximated as first-order processes and thus are quantified by first-order rate constants. The rate constants are given the name transfer coefficients and are based either on field data, laboratory data, or theory.

The time rate of change of material inventory in a given compartment is given by a first-order differential equation. A complete compartmental model consists of a series of simultaneous, linear, first-order differential equations. Solution of the set of equations yields compartment inventories as a function of time. Generally, closed-form analytical solutions are possible only for simple systems. For example, systems consisting of compartments in series with unidirectional transport are described by a set of equations identical to those for a radioactive decay chain. Solution of the set of equations yields the Bateman equations.¹⁰ Systems with only a few compartments and bidirectional transfer, such as those encountered in many biological applications, can be solved analytically using Laplace transforms.¹¹ A four compartment system which includes the transport of radioactive daughters of a transuranic nuclide was solved by the eigenvalue technique.⁸ For systems containing more than three compartments with either multiple inputs to any given compartment or bidirectional transport, the analytical techniques mentioned above are generally not practical and numerical methods are usually required.¹² One such large system is a 70 compartment model used to estimate long term dose to man due to shallow land burial of radioactive wastes at the Savannah River Plant.⁷

THE COMPARTMENTAL MODEL: HISTORICAL REVIEW

Compartmental modeling began in the field of mathematical biology. In the 1930's, isotopic tracers were used to identify metabolic pathways in mammalian systems. As the use of tracers proliferated in the 1940's, experiments were designed to be more quantitative in analyzing biological processes. This led to the use of simple compartmental models in analyzing experimental data. Terminology was also established during this time period and the term "compartment" was first used by Sheppard.¹⁴ A compartment was defined as having "homogeneous contents that are separated by real boundaries or," for radiotracer purposes, "can be generalized so that a substance such as a chemical element can be considered to be in a different state of chemical combination." In the radiotracer literature there is some confusion in the distinction between compartments and metabolic pools. They are treated as the same in some references;⁴⁻⁶ however, pools can be distinguished as a mixture of compounds that are lumped together due to their kinetic equivalence in the synthesis of biological macromolecules. An example is the lumping together of amino acids for protein synthesis. This distinction, however, only represents a more precise biological terminology and does not have any consequence on the theoretical derivation of compartmental modeling.

The use of compartmental models has extended beyond the realm of the biological radiotracer applications. Of specific interest here are applications involving metabolic and environmental transport of radionuclides. The International Commission on Radiological Protection (ICRP) uses compartmental models to quantify the transport of radionuclides to various body organs due to inhalation (lung model) or ingestion (gastrointestinal model).¹⁵ The ICRP combines transport predictions with radiation dose calculations to establish limits for intakes of radionuclides by nuclear workers. Environmental transport applications include migration and distribution of radionuclides in lakes;¹⁶ the movement of radionuclides in agricultural systems;¹⁸ the transport of radioactive iodine, strontium, and cesium in the forage-cow-milk pathway;¹⁷ and the global cycling of long-lived radionuclides such as ^3H , ^{14}C , ^{85}Kr , and ^{131}I .¹⁶ The 70 compartment model used at the Savannah River Plant is based on the forage-cow-milk model developed at Oak Ridge National Laboratory¹⁷. It is unique, however, due to its incorporation of both environmental and metabolic compartments in a single model and its resulting large size. The model is used to estimate radiation dose to man due to low-level burial operations at the site.

THEORETICAL BASIS

Environmental compartmental models in use today are based largely on intuition and empiricism. However, a theoretical foundation for the method can be established from the general mass transport equation¹⁸ for an incompressible fluid. The full derivation is presented in a Clemson University Department of Environmental Systems Engineering Thesis.¹⁹

The equation governing radionuclide movement accounts for the four factors determining the radionuclide inventory in a compartment: 1) transfer in from other compartments, 2) transfer out to other compartments, 3) source or sink terms, and 4) radioactive decay. These factors are all included in the following generalized linear differential equation:

$$\frac{dQ_n}{dt} = \sum_{m=1}^N \lambda_{n,m} Q_m - \sum_{m=1}^N \lambda_{m,n} Q_n - \lambda_R Q_n \pm S_n \quad (1)$$

where Q_n represents the quantity of nuclide in compartment n (curies), Q_m represents the quantity of nuclide in compartment m (curies), $\lambda_{n,m}$ is the transfer coefficient for the transport of radionuclide from compartment m to compartment n (year^{-1}), $\lambda_{m,n}$ is the transfer coefficient for the transport of a radionuclide from compartment n to compartment m (year^{-1}). λ_R is the decay constant for the radionuclide (year^{-1}), S_n is a source or sink term in compartment n (curies/year), and N is the maximum number of compartments under consideration. Equation (1) constitutes a finite difference relationship describing the change in radionuclide inventory in compartment n over time.

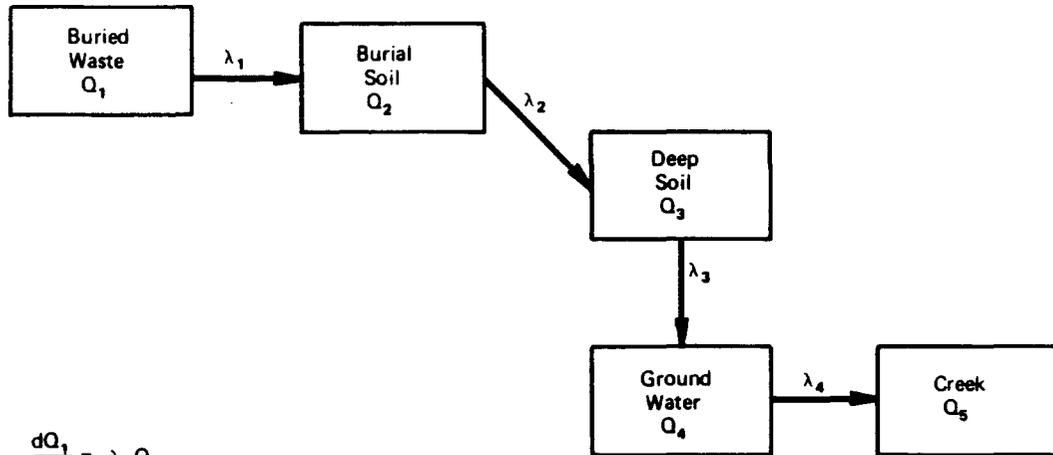
Thus, the first term to the right of the equal sign in the equation is the sum of all input rates to compartment n , the second term is the sum of all loss rates from compartment n , the third term is the loss from compartment n due to radioactive decay, and the fourth term is the gain or loss in compartment n due to sources or sinks, respectively.

Example of an Exact Analytical Solution

Simple compartmental models consisting of less than four compartments or involving unidirectional transport between compartments can generally be solved by standard analytical techniques such as substitution or Laplace transforms. However these straightforward approaches are either not applicable or impractical for large, complex systems; and numerical methods, such as finite difference, or alternate analytical methods, such as the matrix analytical technique¹⁹, are necessary. A closed-form analytical solutions for one simple system is presented in Figure 1. These solutions are used to verify the mathematical accuracy of the finite difference and matrix analytical solutions. This system is analogous to the successive decay of the members of a radioactive series. The solution to the set of differential equations which describe successive decay is the general Bateman equation¹⁰. The solution is obtained in a straightforward manner by successive substitution. This involves solving the first differential equation, substituting the result into the second equation, solving the second equation and continuing the process.

Mathematical Validation

To lend credence to these calculations, computer codes used to simulate environmental transport should be mathematically validated against suitable, site-specific radionuclide data bases. By derivation of an exact analytical solution of a set of linear differential equations which simulate radionuclide transport from buried waste, through the unsaturated zone, to the groundwater table, we have been able to demonstrate that the set of equations simulating transport are mathematically valid, with the error introduced by numerical solution techniques at <0.2%.



$$\frac{dQ_1}{dt} = -\lambda_1 Q_1$$

$$\frac{dQ_2}{dt} = \lambda_1 Q_1 - \lambda_2 Q_2$$

$$\frac{dQ_3}{dt} = \lambda_2 Q_2 - \lambda_3 Q_3$$

$$\frac{dQ_4}{dt} = \lambda_3 Q_3 - \lambda_4 Q_4$$

Initial Conditions: $Q_1(0) = Q_2(0)$
 $Q_2(0) = Q_3(0) = Q_4(0) = 0$

Solutions

$$Q_1(t) = Q_1(0)e^{-\lambda_1 t}$$

$$Q_2(t) = \frac{Q_1(0)\lambda_1}{(\lambda_2 - \lambda_1)} [e^{-(\lambda_1)t} - e^{-(\lambda_2)t}]$$

$$Q_3(t) = \frac{Q_1(0)\lambda_1\lambda_2 e^{-(\lambda_1)t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} - \frac{Q_1(0)\lambda_1\lambda_2 e^{-(\lambda_2)t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} + \frac{Q_1(0)\lambda_1\lambda_2 e^{-(\lambda_3)t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}$$

$$Q_4(t) = \frac{Q_1(0)\lambda_1\lambda_2\lambda_3 e^{-(\lambda_1)t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)} - \frac{Q_1(0)\lambda_1\lambda_2\lambda_3 e^{-(\lambda_2)t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)}$$

$$+ \frac{Q_1(0)\lambda_1\lambda_2\lambda_3 e^{-(\lambda_3)t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)} + \frac{Q_1(0)\lambda_1\lambda_2\lambda_3 e^{-(\lambda_4)t}}{(\lambda_1 - \lambda_4)(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)}$$

FIGURE 1. Exact Analytical Solution to a 5-Compartment Unidirectional Model

For a simplified description of mass transport of radionuclides into the environment, an exact analytical solution can be derived, as shown in Figure 1. Even in this form, the exact solution is rather complex. The exact solution is analogous to the Bateman equations for successive radioactive decay of a nuclide.¹⁰ The exact solution can now be compared with the numerical solution techniques - used for solution of the more complex problem - based upon Gauss-Jordan elimination/finite element/matrix inversion methods. Results are illustrated in Figure 2. Several environmental compartments are shown since the mass transfer coefficients (λ_n 's) will vary considerably as a function of environmental compartment and radionuclide environmental properties. The results well illustrate that the numerical approximation methods employed introduce minor error (<0.2%) into the environmental impact analysis. The numerical approximation methods are mathematically valid.

METHODOLOGY FOR NUCLEAR AND HAZARDOUS CHEMICAL WASTE DISPOSAL

This mathematical model has been developed to provide estimates of long-term dose to man from buried waste. The model consists of compartments which represent different portions of the environment, including vegetation, herbivores, atmosphere, ground water, surface water, and man. Movement of radionuclides between compartments is controlled by transfer coefficients, which specify the fraction of radionuclide entering or leaving a compartment during a specified time period. Time controls account for the time lag in movement induced by such factors as the delayed presence of man. Sources and sinks independent of the natural radionuclide movement are provided.

Flow Diagram

The computer program²⁰ consists of a main program (module DOSTOMAN) and seven subroutines (Figure 3).

Module DOSTOMAN performs the main control function by coordinating the input and output of data and the calculations of compartment radionuclide inventories. The entire module is run for each time step specified and subroutines are called in clockwise beginning with subroutine INPUT.

Module DOSTOMAN is divided into two basic sections:

- 1) calculation of radionuclide inventories in compartments assuming transfer coefficients are continuous exponential or Gaussian functions with time or discontinuous step functions and
- 2) calculation of inventories assuming that some component of a transfer coefficient changes either additively or geometrically during the period being simulated.

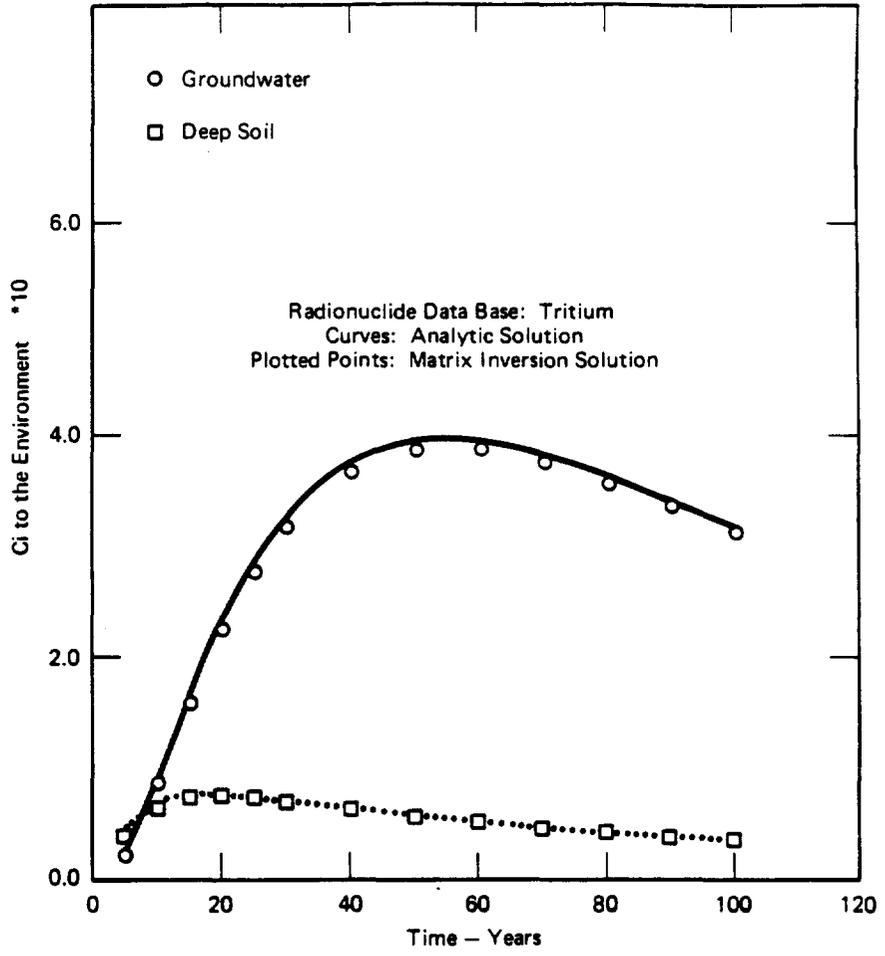


FIGURE 2. Model Mathematical Validation Exact Analytical Solution Verses Matrix Inversion

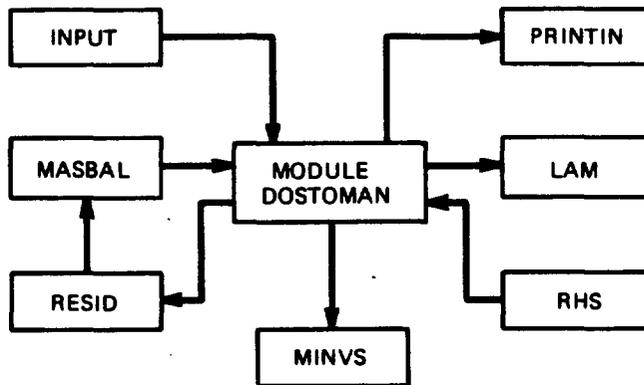


FIGURE 3. Flow Diagram for the DOSTOMAN Computer Program

Subroutine INPUT reads in the initial data from seven records, including: radioactive decay constant, number of compartments, number with sources or sinks, time step, values and locations of sources or sinks, initial compartment radionuclide inventory, transfer coefficient components, time functions to control the application of the transfer coefficients, and factors to perturb components of transfer coefficients, if applicable. Other input records give plotting and editing specifications. The only calculations performed in this subroutine involve the transfer coefficients, which are of the form $A \cdot V^X$. The individual components A, V, and X are read at this time and the transfer coefficients are calculated. If requested, all input data is printed by subroutine PRINTIN.

In the main program, all transfer coefficients, sources, and sinks are corrected for time dependence, if any, and the coefficient matrix is set up. Subroutine LAM is then called to calculate the terms on the main diagonal in the A matrix. This involves summing the λ 's from $m = 1$ to $m = N$ and subtracting $1/\Delta t$ and λ_R . After this calculation, coefficients of the A matrix are ready for the solution of the simultaneous equation.

Subroutine RHS is then called to set up the right-hand side of the simultaneous equations (equation 1). This involves making each term (B_n) in the B matrix equal to $-Q_n^0/\Delta t$ where $n = 1..N$. If compartment n contains a source or sink, the time dependence of the source or sink is accounted for and the B_n values are adjusted accordingly.

Upon returning to the main program, the matrices A, X, and B are ready for solution by matrix inversion. The solution is accomplished by calling subroutine MINVS, which calculates the determinant and the inverse of the A matrix (the λ values) for the matrix problem $AX = B$ and finds the solution vector X. Upon return from MINVS, matrix A contains the inverse matrix; therefore, the original matrix A is destroyed. Matrix B then contains the solution vector X (in this case, the Q_n). The solution is accomplished by the standard Gauss-Jordan elimination method. Details of this method can be found in matrix algebra texts. The Q_n values are then printed and any requested plots are made.

At this point in the program, time is incremented by either a specified or a calculated time step and the model is rerun to calculate Q_n values for a new time. This is done until the specified simulation time is achieved or the number of time steps is consumed.

An initial inventory is provided to the system by the Q_n values. If no sources or sinks are added to or removed from the system during the run, this initial amount of radionuclide must be maintained throughout the run, adjusted, of course, for radioactive decay. Subroutine MASBAL calculates the state of mass balance by summing all of the Q_n values at a particular time and comparing the total to the sum of initial Q_n values. Ideally, the deviation from mass balance should be zero; again, round-off and truncation errors may contribute to a small deviation.

Two means of evaluating the numerical stability of the code are included in the program. Subroutine RESID determines the difference (residual) between the calculated values for the right- and left-hand sides of the set of simultaneous equations solved by matrix inversion in subroutine MINVS. The subroutine RHS uses the values calculated at a particular time; therefore, the difference between the two sides of each equation is a measure of how accurate the solution is. Ideally, the residual should be zero; some small residual can be accounted for by round-off and truncation errors in the calculations and is usually insignificant.

The time required to run a simulation with the model will depend greatly on the characteristics of the computer facilities available to the user. On the IBM 360/195 at the Savannah River Laboratory, a simulation involving 200 time steps, 200 transfer coefficients, and 69 separate compartments requires about six minutes of central processing unit time. About 500 K bytes of core are required for such a simulation. Only one radionuclide can be considered in each run: the radioactive decay constant and many of the compartment inventories and transfer coefficients will be specific to that radionuclide.

EXAMPLES OF USE OF THE MODEL IN TRANSPORT AND PATHWAY ANALYSES

The DOSTOMAN code developed by SRL solves a mass balance equation based on a set of simultaneous linear differential equations that simulate radionuclide transport. The code extends the calculations to a number of pathways by scenario analysis. Realistic scenarios such as hydrologic transport and hypothetical scenarios such as future land occupation are used to estimate environmental impacts, usually stated as dose commitments. These in turn are used to evaluate site performance, radionuclide disposal limits, improved concepts for land disposal of waste, and decommissioning alternatives. The code relies on site-specific input data such as radionuclide inventory, chemical form, release rate, K_d 's, and geohydrologic parameters.

During the past year, modeling studies with the DOSTOMAN code included identification of factors for reduction of dose commitments, evaluations of disposal limits for transuranic (TRU) waste, and analysis of the projected behavior of the mobile radionuclides (tritium, Tc-99, and I-129).

Factors for Reduction of Exposure

Model analyses for a variety of radionuclides have shown that the two most significant pathways for potential exposure from radioactive waste burial grounds are:

- hydrologic transport of radionuclides such as tritium, Tc-99, and I-129 that have low soil adherence

- biotic transport via plant uptake of radionuclides such as Sr-90, Cs-137, Pu-238, and Pu-239 that have high soil adherence

Several factors that will minimize these routes have been identified by sensitivity analyses with the model. These factors are: radionuclide inventory control; moisture infiltration barriers, such as low permeability matrices or site caps; site vegetation control or overburden; and deeper burial, which requires a deeper water table. The model can place a decremental environmental impact effect on each factor. These factors have been incorporated into new concepts for improved shallow land burial operations and for greater confinement disposal.

TRU Waste Disposal Limits

The effect of a 100 nCi/g demarcation value for land disposal of TRU radionuclides was evaluated using site-specific transport modeling and pathway analysis. Since 1968, the demarcation value has been 10 nCi/g. However, prior to 1968 all TRU waste at SRP (including that >100 nCi/g) was routinely buried. The consequences of this practice have been carefully monitored, and the monitoring data supply input to the transport model for evaluating potential TRU disposal limits. The incremental environmental impact of the higher demarcation value (100 nCi/g) depends on the scenario chosen, but in all cases studied was insignificant.

Transportable Radionuclides

Tritium behavior in the closed 31-hectare burial ground at SRP has been modeled. The calculations predict that tritium in groundwater will increase about 10% in coming years and then decrease mostly by radioactive decay. The rate of outcrop to surface streams is calculated to be less than 500 Ci/yr, which has a negligible effect at the site boundary. Scenario calculations based on 100-year institutional control followed by land occupation of the site predict that the hydrologic pathway will continue to dominate. However, because of decay of the buried tritium and long migration paths, the environmental effect will be minimal.

Tc-99, primarily present as the pertechnetate anion, has been shown to have low soil adherence. Model projections on movement of Tc-99 to the water table are in good agreement with experimental observations. Concentrations of Tc-99 in the groundwater at the SRP burial ground are predicted to remain small and well below standards for surface streams. Scenario calculations predict that the water pathway will contribute 70% and vegetative uptake 30% to the environmental effect. The vegetative pathway is significant because plant uptake factors for Tc-99 are reported to be large.

I-129 migration was modeled using field data, the estimated inventory of I-129 as silver iodide, and near-zero soil adherence. Under these conditions, the flux of I-129 to the water table is calculated to be about

10⁻⁵/yr, with groundwater concentration of about 10⁻² pCi/L, well below environmental standards for I-129. The calculation was also performed as a function of soil permeability to demonstrate the effect of a clay cap that reduces rainwater infiltration. In this case, the flux is about 10⁻⁸/yr, and the groundwater concentration is about 10⁻⁵ pCi/L. Finite soil adherence of I-129, as found in laboratory tests, would be expected to decrease calculated I-129 fluxes and concentrations by at least an order of magnitude.

Biotic Transport Calculations

Vegetative uptake of Sr-90 is the primary pathway leading to exposure because the calculation assumes that roots will penetrate the buried waste. The code predicts that vegetative uptake of Sr-90 can be controlled by increasing the distance from the surface to the waste. This could be accomplished by adding 3-5 m of overburden above the waste, which is a decommissioning alternative, or by deeper burial, which is an option for new disposals.

Exposures from Tc-99 are predicted to be minor, in part because of the small inventory of Tc-99 in the buried waste. Technetium, in the form of pertechnetate anion, is a case where the mobile species has little or no soil adherence. The calculated value of Tc-99 concentration in groundwater is in good agreement with field measurements at the burial ground.

Cs-137 transport and exposures are calculated to be less significant than those of Sr-90 because vegetative uptake is less for cesium.

A sensitivity analysis of the model projections was completed, using the Sr-90 data base. Key input parameters, the dosimetry data base, the scenarios, and various burial ground management options all were varied to determine the most sensitive features of the calculation. This exercise demonstrated the importance of vegetative uptake of Sr-90 and showed that this pathway could be eliminated by increased depth from surface to waste.

USER'S GUIDE FOR THE COMPARTMENTAL PATHWAYS MODEL

A User's Guide²¹ which documents the development and computer implementation of the Savannah River Laboratory compartmental pathways computer code used to simulate radionuclide transport was published in 1981. The User's Guide provides all the necessary information for the prospective user to input the required data, execute the computer program, and display results.

PEER REVIEW AND IMPLEMENTATION OF THE MODEL AT DEPARTMENT OF ENERGY SITES

In 1982, personnel from EG&G Idaho and the National Low Level Waste Management Program conducted a peer review of available codes for pathway

analysis and environmental impact for evaluation of radionuclide land disposal operations. Pathway analysis codes from DOE sites were specifically studied. Included were codes from:

- SRL (DOSTOMAN)
- Battelle PNL (ARRRG, FOOD, KRONIC, SUBDOSA, DACRIN, PABLM, MAXI)
- Los Alamos National Lab (BIOTRAN, CREAMS)
- Idaho National Engineering Lab (BURYIT)
- Nuclear Regulatory Commission (GRWATER, INTRUDE)
- Environmental Protection Agency (PRESTO, AMRAW).

The DOSTOMAN Code was evaluated to be flexible, straight forward, easily understood, and sufficiently well documented to be used at other sites. In addition, its dynamic or time-dependent nature was considered advantageous as opposed to many assessment models which presume equilibrium or no net flow of contaminants.

In 1983, the DOSTOMAN computer code and available details on methodology, user's guide, matrix inversion, and transfer coefficient calculations were transmitted to EG&G/Idaho. Similar information has also been transmitted to the Argonne National Laboratory and the Environmental Protection Agency.

In 1984, the SRL code was applied to the development of Threshold Guidance Limits for the potential management of waste below regulatory concern (so called "de minimis" waste) by EG&G personnel (DOE/LLW-40T/Draft, "Development of Threshold Guidance", January 1985).

TECHNICAL MANUAL ON THE SRL DOSTOMAN CODE

In October 1985, the Savannah River Laboratory will publish a TECHNICAL MANUAL on the SRL Code for widespread distribution to DOE sites. The Code is currently being used for transport analysis and human health risk assessment for hazardous chemical and mixed waste sites.²²

ACKNOWLEDMENT

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