# PREDICTION OF THE MIGRATION OF SEVERAL RADIONUCLIDES IN OCEAN SEDIMENT WITH THE COMPUTER CODE IONMIG: A PRELIMINARY REPORT



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# PREDICTION OF THE MIGRATION OF SEVERAL RADIONUCLIDES IN OCEAN SEDIMENT WITH THE COMPUTER CODE IONMIG: A PRELIMINARY REPORT*

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## ABSTRACT

A computer code, IONMIG, which is used to calculate the far-field transport of radionuclides through ocean sediment by diffusion and convection is described. The code uses a two-dimensional, axisymmetric, explicit finite difference formulation. Preliminary results for several species (Cs, Pu, I, Tc) are given.



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## INTRODUCTION

As part of a study to determine the feasibility of radioactive waste disposal in seabed sediment, a radionuclide migration code, IONMIG, is being developed. This code is a two-dimensional planar or axisymmetric code which solves the transport equation including convection, axial and transverse dispersion, molecular diffusion, concentration dependent adsorption and radioactive decay. Assumptions used in its formulation are that the presence of the radionuclides does not change the fluid properties or the behavior of other nuclides and that absorbtive processes are reversible and describable in terms of an empirically determined equilibrium constant. Near-field details are not treated and species are injected as volumetric source terms. A brief description of the code is given in the CODE DESCRIPTION section of this report.

IONMIG differs from another Sandia nuclide migration code, SWIFT,[2] which was developed with Intera Corporation to analyze transport through hydrologic formations, in that it is much simpler to use (SWIFT input requirements are about 30 times greater than IONMIG's). IONMIG can use concentration dependent adsorption coefficients and is part of a modular solution system (IONMIG interfaces with the incompressible fluid thermo code, MARIAH, and the plot code, SEAPLT).

The purpose of this report is to describe some preliminary results on several species which are representative of the elements of concern in a waste canister inventory. These are $^{137}Cs$, $^{99}Tc$, $^{129}I$, and $^{239}Pu$.

## CODE DESCRIPTION

### Theory

The equations describing the migration of contaminant ions in a porous saturated bed are of the form:

$$\frac{\partial (C_i \varepsilon K_i)}{\partial t} + \nabla \cdot (C_i \vec{v}) = -\sum_{k=1}^{N} (\lambda_{ik} K_i \varepsilon C_i) + \sum_{k=1}^{N} \lambda_{ki} K_k \varepsilon C_k + S_i \quad , \qquad (1)$$

where,

$C_i$ = the ion species concentration ($kg/m^3$)

$\varepsilon$ = the porosity of the medium

$K_i$ = the species equilibrium coefficient for species i = $(1 + \frac{1 - \varepsilon}{\varepsilon} \rho_{soil} K_{d_i})$ where $K_d$ is the equilibrium distribution coefficient

$\lambda_{ik}$ = the radioactive decay rate from species i to species k

$S_i$ = a source term for continuous or step function addition of ions corresponding to the leach rate

$\vec{v}$ = the total ion velocity which is assumed to be of the form:

$$u - \frac{D_x}{C_i} \frac{\partial C_i}{\partial x} \vec{i} + v - \frac{D_y}{C_i} \frac{\partial C_i}{\partial y} \vec{j}$$

$u$ = convective velocity in the x-direction

$v$ = convective velocity in the y-direction

$D_{x,y}$ = diffusion dispersion coefficient in the x,y-direction

$D_x = \alpha_L |u| + \alpha_T |v| + D_o$

$\alpha_L, \alpha_T$ = longitudinal and transverse dispersion coefficients

$D_o$ = molecular diffusion coefficient

to a level in which they will disappear in several global time steps. This permits the inclusion of transition species in the chain without going to excessively small time steps, but of course, the instantaneous concentrations of such short lived species will be greatly overestimated and should not be considered as part of the useable output of the code. Any element for which $\lambda\Delta t > 3$ is treated as such a transition species.

In order to retain the flexibility of use provided by the options of axisymmetric geometry and variable mesh calculations, conservation differencing, which becomes rather unwieldy for those options, was not used. It is possible, therefore, that discretization and truncation errors may introduce some source error into the calculations. To partially correct for that possibility, a separate and more accurate time integration of the global quantities is performed and compared with the mesh point summations. An option to correct the mesh point values at each time step is included in the code.

Geometry and Boundary Conditions

The computational domain is a rectangular region, with boundary conditions as indicated in Figure 1, which represents a cross section of an axisymmetric region with x as the radial coordinate. The concentrations and sources are initially specified and the code computes subsequent distributions. If an external convective velocity field is not provided, an internally generated conservative convective cell field is assumed with its peak velocity specified as input data (peak may be zero).

The rectangular region shown in Figure 2 represents a cross section of the axisymmetric computational domain, as in Figure 1, with the dimensions used in the calculations and the canister location indicated. The streamlines shown in Figure 2 are those calculated with the finite element code, MARIAH, at 100 years after burial. The canister is assumed to be 3 m long and 0.3 m in diameter and to contain 10 year old reactor waste. It is buried with its center 30 metres below the ocean-sediment interface.

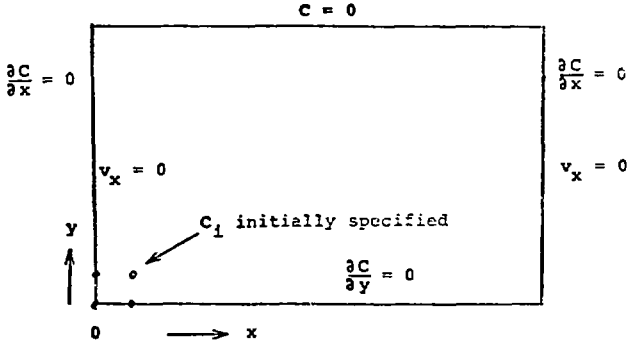


FIGURE 1. Geometry and Boundary Conditions Assumed in IONMIG.

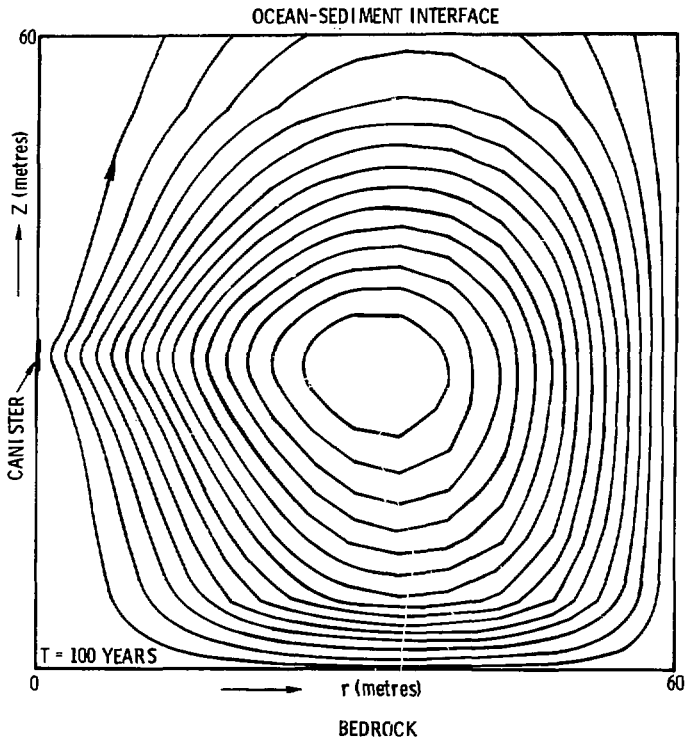


FIGURE 2. GEOMETRY AND STREAMLINES IN THE SEDIMENT COMPUTATIONAL REGION.

Input Parameters

The heat generated by the canister is assumed to be 1.5 kW at burial and to be decaying according to a schedule given in the ORIGEN code.[3] The temperature history near the canister surface is shown in Figure 3. In a few hundred years, the temperature is within a few degrees of the ambient (1.5°C). The same is true for all points in the region.

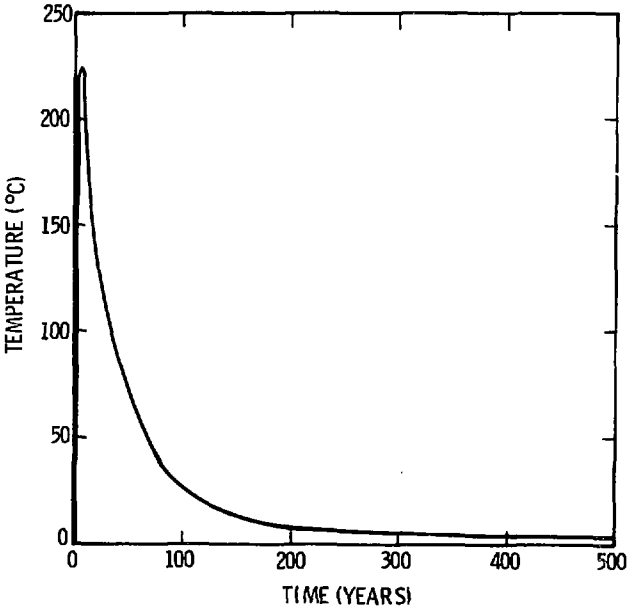


FIGURE 3. TEMPERATURE HISTORY ADJACENT TO A WASTE CANISTER 30 METRES BELOW THE SEDIMENT SURFACE.

The velocity variation with time at its maximum point adjacent to the canister is shown in Figure 4. It is seen that this velocity is very small, and after 100 years, convective transport is negligible compared to molecular diffusion.

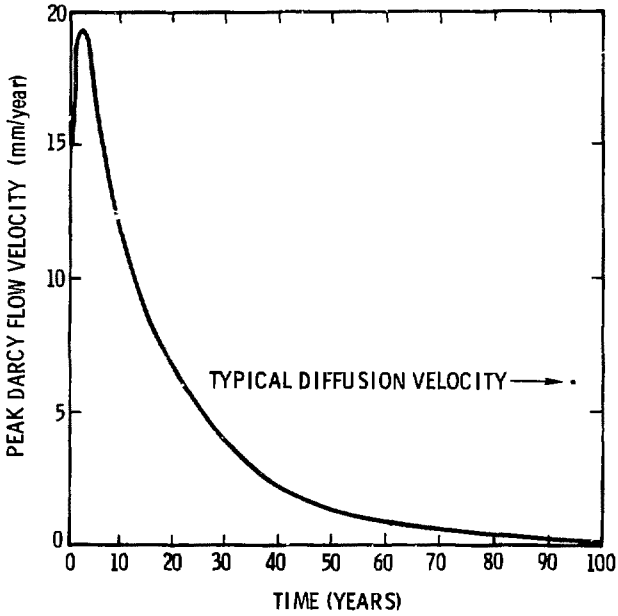


FIGURE 4. FLOW VELOCITY ADJACENT TO A WASTE CANISTER 30 METRES BELOW THE SEDIMENT SURFACE.

For all elements, except iodine, it is assumed that the molecular diffusion coefficient is 0.01 $m^2$/year (3.0 E - 10 $m^2$/s), longitudinal dispersivity factor is 6.1 m,* the transverse dispersivity factor is 0.61 m, the porosity of the sediment is uniform and equal to 0.75. For iodine, the molecular diffusion coefficient is 0.018 $m^2$/year. Values for the equilibrium partition coefficient, $K_d$, were obtained by fitting a limited amount of laboratory data, and were approximated as follows:

Plutonium: $$K_d = \frac{100}{1 + 3E8C} + 0.01$$

Cesium: $$K_d = \frac{10}{1 + 200000C} + 0.1$$

Iodine and Technetium: $$K_d = \frac{0.0001}{1 + 10000C}$$

where C is the fluid ionic concentration in kg/$m^3$ ($K_d$ values are in $m^3$/kg; to obtain ml/g multiply by 1000). Valence states of Cs and Pu are assumed to be +1 and +4, respectively.

The fluid properties used by the code MARIAH to calculate the convective velocities vary with temperature and plots of these quantities are shown in Figure 5. A nominal fluid density of 1000 kg/$m^3$ was used. Additional sediment property values used in the MARIAH calculations are:

Density = 2950 kg/$m^3$

Specific Heat = 880 J/kg$^{o}$C

Thermal Conductivity = 1.92 exp(-6.376 E-6 T) W/m$^{o}$C

Porosity = 0.8

Horizontal Permeability = 5 x $10^{-16}$ $m^2$

Vertical Permeability = 5 x $10^{-17}$ $m^2$

*This conservative overestimate of dispersivity factor is used because good data is not currently available, and the low velocities calculated make dispersion very small compared to molecular diffusion so that errors in this factor do not affect the results.

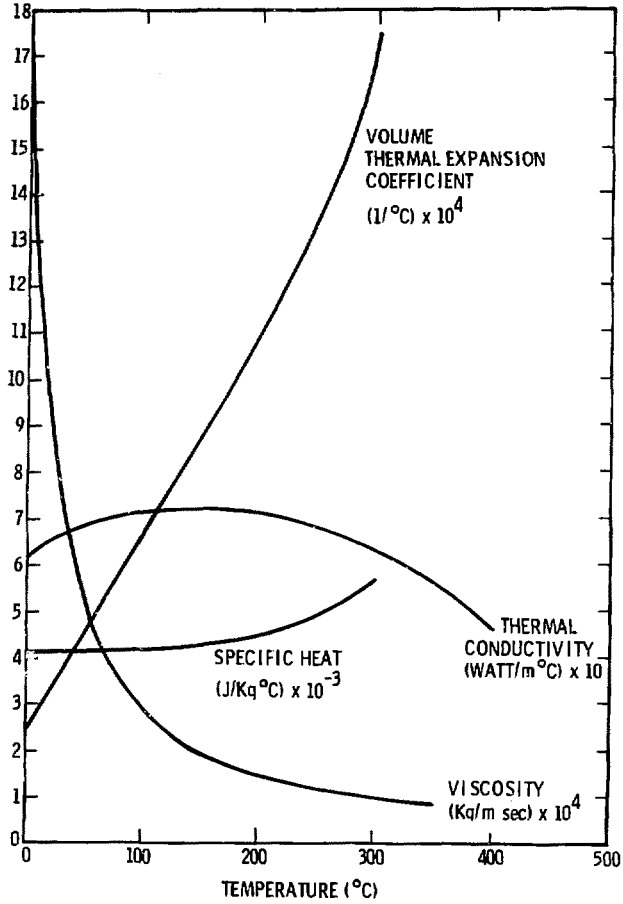


FIGURE 5. VARIATION OF FLUID PROPERTIES WITH TEMPERATURE AT 600 BARS.

The release in each case is assumed to be of the whole canister inventory at zero time. At the bedrock and the outer periphery of the computational domain, a zero flux condition is assumed so that all nuclides released from the canister (which do not decay) will eventually diffuse to the surface.

Although the results of only four nuclide migration calculations are discussed, in order to obtain those results parent and daughter nuclides were considered simultaneously. Table 1 lists all the nuclides associated with each calculation and their assumed canister inventory value at zero time.

TABLE 1

INITIAL CANISTER INVENTORIES OF ISOTOPES USED IN THE CALCULATIONS

| Radionuclide | Mass in Canister (kg) |
|---|---|
| $^{243}Am$ | 0.121 |
| $^{239}Np$ | 1.0 E-7 |
| $^{243}Cm$ | 6.92 E-5 |
| $^{239}Pu$ | 3.58 E-2 |
| $^{235}U$ | 1.0 E-5 |
| $^{129}I$ | 0.305 |
| $^{129}Xe$ | 1.178 E-4 |
| $^{99}Tc$ | 1.13 |
| $^{99}Ru$ | 4.32 E-5 |
| $^{137}Cs$ | 1.33 |
| $^{137}Ba$ | 2.01 E-7 |

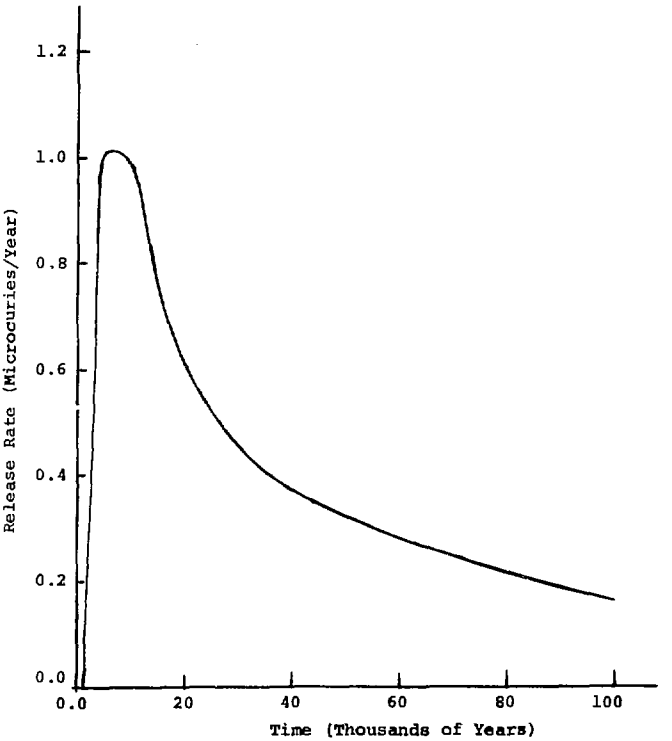


FIGURE 7. Release Rate of $^{129}I$ from the Sediment Surface Covering a Single Canister at 30 m Depth.

Figure 8 shows a similar release rate profile for $^{99}Tc$. The peak value of 180 μCi/year is much higher than that for $^{129}I$ because both the initial canister inventory and the specific activity of $^{99}Tc$, which beta decays to $^{99}Ru$ with a half-life of 213,000 years, are higher.

The significance of such release rates in terms of human or ecological hazards can only be determined by a comprehensive analysis of the water column transport mechanisms and marine bio system concentration mechanisms. Some perspective on the predicted release rates can be obtained, however, by a comparison with other oceanic radioactive sources. A single storm in the mid-sixties could wash out more than $10^6$ μCi of beta activity from residual atmospheric sources.[4] Total oceanic atmospheric deposition of $^{90}Sr$ alone in 1978 exceeded $8 \times 10^{10}$ μCi.[5] Such rates are very much higher than the worst calculated predictions (even for thousands of canisters). In addition to atmospheric deposition, natural radioactive substances are constantly diffusing from the pelagic clay sediment. For the axisymmetric case for which the IONMIG calculations were made, the per canister area is based on a canister spacing of 120 m. This results in an average per canister release of $8.9 \times 10^{-5}$ μCi/year-$m^2$ for $^{129}I$ and $1.6 \times 10^{-2}$ μCi/year-$m^2$ for $^{99}Tc$. These fluxes compare to natural radium and radon fluxes of $3.5\text{-}8.8 \times 10^{-4}$ μCi/year-$m^2$ for $^{226}Ra$ and 0.26-0.88 μCi/year-$m^2$ for $^{222}Rn$.[6,7] Based on this comparison, one may hypothesize that the immediate exposure effects of the released $^{129}I$ and $^{99}Tc$ on benthic organisms would be negligible in as much as they evolved in a much more intense field. Confirmation of this hypothesis awaits completion of ongoing research and experimentation on biological concentration, water column transport mechanisms, and development of pathways-to-man models.

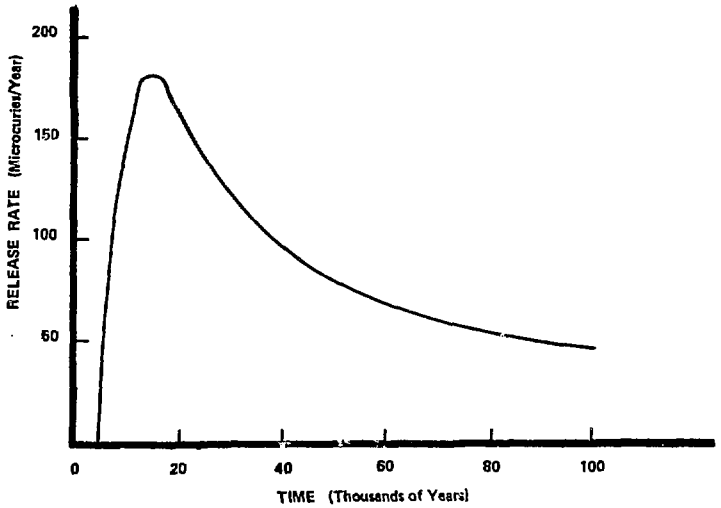


FIGURE 8. Release Rate of $^{99}Tc$ from the Sediment Surface Covering a Single Canister at 30 m Depth.

## CONCLUSIONS

The development of a computer code to be used as a tool in assessing the transport of waste nuclides through ocean sediment is proceeding satisfactorily. Preliminary calculations of the migration of Cs, Pu, I, and Tc from a point 30 metres below the sediment surface have been made. Elements with short half-lives (less than 500 years) such as $^{137}Cs$ decay to negligible values before reaching the surface. Actinides many of which have large values $K_d$ (above 10,000 gm/ml) move very slowly and also do not reach the surface until beyond 1,000,000 years. Long-lived fission products with low adsorption coefficients, such as $^{129}I$ and $^{99}Tc$, are the most difficult to contain. For those nuclides, it must be shown that the low release rate and low specific activity reduce the hazards from them to an acceptable level, or additional engineered barriers must be provided.

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