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A New Numerical Method for Solving the Solute Transport Equation

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ABSTRACT

The solute transport equation can be solved numerically by approximating the water flow field by a network of stream tubes and using a Green's function solution within each stream tube. Compared to previous methods, this approach permits greater computational efficiency and easier representation of small discontinuities, and the results are easier to interpret physically. The method has been used to study hypothetical sites for disposal of high-level radioactive waste.

1. INTRODUCTION

Subsurface waste migration is a subject of intense current interest, particularly in view of the more stringent requirements now demanded by law for protection of our waters. The model described herein was developed to assist in the analysis of sites for the deep geologic disposal of radioactive waste. This model incorporates a new numerical method for solving the solute transport equation.

The approach results in a high degree of computational efficiency, and allows relatively straightforward representation of small discontinuities. This avoids the difficulties which are encountered in solving the time-dependent solute transport problem by finite-element or finite-difference methods. The model is constructed so as to facilitate handling various hydrogeological characteristics and making sensitivity studies.

Rather than divide the region of interest into a lattice of cells, the groundwater flow pattern is approximated by a network of one-dimensional flow paths, which need not be straight. These flow paths or stream tubes can then be analyzed using a simple one-dimensional model for the transport. The model incorporates the effects of mass transport, chemical interactions, hydrodynamic dispersion, and radioactive decay. As long as each stream tube is sufficiently long the one-dimensional equation can be solved using a simple Green's function technique. Each stream tube is considered in sequence and appropriate calculations are made at the junction of different streams.

2. WASTE TRANSPORT IN A STREAM TUBE

Our basic model of flow through porous media consists of a series of interconnected one-dimensional pipes. Mathematically the pipes represent stream tubes and the complexity of the fluid flow defines the number and positioning of the pipes. For our initial models, fairly simple flow fields were chosen. More complicated and more realistic flow can be determined by two- and three-dimensional hydrology programs.

Under certain constraints the three-dimensional equation for dispersive flow reduces to a simple one-dimensional flow equation in stream coordinates. Quite generally, transport of contaminants in porous media is described by a diffusivity equation (Scheidegger [1964]). Hence if the concentration of nuclide r in the interstitial water is $C_r(\underline{x},t)$ and the interstitial fluid velocity is $\underline{v}$ then

$$\frac{\partial C_r}{\partial t} = \frac{\partial}{\partial x_i} D_{ij} \frac{\partial}{\partial x_j} C_r - \frac{\partial}{\partial x_i}(v_i C_r) - \lambda_r C_r + \sum_{s \neq r} \lambda_s^r C_s - \frac{\partial m_r}{\partial t} - \lambda_r m_r + \sum_{s \neq r} \lambda_s^r m_r \tag{1}$$

Here D_{ij} is the coefficient of convective dispersion and m_r is the amount of ion r adsorbed on the solid per unit volume. The subscripts i and j refer to the three Cartesian coordinates, and sums are understood in accordance with the usual convention.

For nuclides which undergo radiactive decay, λ_r is the radioactive decay constant and λ_s^r is the production rate of nuclide r from decay of nuclide s. This formulation would also serve to model the first-order reaction kinetics for the chemical transformation of compounds (Cho, 1971), where λ_r and λ_s^r would be reaction rate constants.

The following assumptions are now made:

- Flow is slow enough so that instantaneous ion exchange equilibrium results, and the exchange adsorption isotherm is linear

  That is, $m_r = \frac{\rho}{\varepsilon} K_d C_r$, where ρ is the bulk density, ε is the effective porosity, and K_d is the distribution coefficient

- The release rate R(t) of the contaminant is independent of species within any decay (or chemical reaction) chain.

- The retardation factor, B_r, is the same for all species in the same decay chain

  $(B_r = 1 + \frac{\rho}{\varepsilon} K_d)$

The first assumption allows us to write (Grove [1970]):

$$B_r \frac{\partial C_r}{\partial t} = \frac{\partial}{\partial x_i} D_{ij} \frac{\partial}{\partial x_j} C_r - \frac{\partial}{\partial x_i}(v_i C_r) - B_r \lambda_r C_r + \sum_{s \neq r} B_s \lambda_s^r C_s \quad (2)$$

Let $I_r(t)$ be the total amount of species r at time t in the contaminant. Then using the rest of the assumptions and defining $c_r = C_r/I_r$, we have for species in the same decay chain:

$$B \frac{\partial c_r}{\partial t} = \frac{\partial}{\partial x_i} D_{ij} \frac{\partial}{\partial x_j} c_r - \frac{\partial}{\partial x_i}(v_i c_r) + B \sum_s \lambda_s^r \frac{I_s}{I_r}(c_s - c_r) \quad (3)$$

Equation 3 is derived by substituting $c_r I_r$ for C_r in Eq. 2 and using the decay equation for I_r:

$$\frac{\partial I_r}{\partial t} = -\lambda_r I_r + \sum_s \lambda_s^r I_s \quad (4)$$

By the uniqueness theorem for differential equations, $c_r = c_s$, and the subscripts may be replaced with a single subscript denoting a group of species which have the same retardation factor. In the remainder of this chapter, we will omit the subscript. This simplification allows us to eliminate from our model a detailed analysis of nuclide decay chains. The assumption that the retardation factor is the same for all parent-daughter combinations is reasonable in light of our imprecise knowledge of these parameters.

The simplified three-dimensional transport equation can now be written as

$$B\frac{\partial c}{\partial t} = \frac{\partial}{\partial x_i} D_{ij} \frac{\partial}{\partial x_j} (v_i c) \tag{5}$$

We now assume that D_{ij} reduces to a linear function of the flow velocity in the one-dimensional case. Because of Onsager's relation, D_{ij} will be symmetric and have the form

$$D_{ij} = (\alpha-2\mu)\, \delta_{ij}\, V + \frac{2\mu v_i v_j}{V} \tag{6}$$

Equation 5 becomes

$$B\,\frac{\partial c}{\partial t} + \frac{\partial}{\partial x_i} (v_i c) = (\alpha-2\mu)\, \frac{\partial}{\partial x_i}\, V\, \frac{\partial c}{\partial x_i} + 2\mu\, \frac{\partial}{\partial x_i}\, \frac{v_i v_j}{V}\, \frac{\partial c}{\partial x_j} \tag{7}$$

For steady incompressible flow $\nabla\cdot\underline{v} = 0$ and this simplifies to

$$B\,\frac{\partial c}{\partial t} + \underline{v}\cdot\nabla c = (\alpha-2\mu)\ \nabla\cdot(V\nabla c) + 2\mu(\underline{v}\cdot\nabla)\ \frac{1}{V}\ (\underline{v}\cdot\nabla)c \tag{8}$$

We now rewrite this equation in coordinates determined by the velocity field. The natural coordinates (S,N,T) are those where S represents the distance along a streamline in the direction of stream flow. The equation reduces to

$$B\,\frac{\partial c}{\partial t} + V\,\frac{\partial c}{\partial S} = \alpha V\,\frac{\partial^2 c}{\partial S^2} + \frac{(\alpha-2\mu)}{h_1h_3}\,\frac{\partial}{\partial N}\,(h_1h_3V)\,\frac{\partial c}{\partial N} + \frac{(\alpha-2\mu)}{h_1h_2}\,\frac{\partial}{\partial T}\,(h_1h_2V)\,\frac{\partial c}{\partial T} \tag{9}$$

Here h_1, h_2, and h_3 are the scale factors. The similarity of this equation to the familiar result for $\underline{v} = V\hat{i}$ and V constant is clear. We note that in this formulation S, N and T represent distances along coordinate axes. Hence the order of differentiation cannot in general be interchanged. Introducing coordinates $(S^*,\ N^*,\ T^*)$ where $dS = h_1dS^*$, $dN = h_2dN^*$ and $dT = h_3dT^*$ and integrating over the cross-sectional area of the stream tube we have

$$B \frac{\partial}{\partial t} \iint_A c h_1 h_2 h_3 dN^* dT^* + \frac{\partial}{\partial S^*} \iint_A V c h_2 h_3 dN^* dT^* = \alpha \frac{\partial}{\partial S^*} \iint_A \frac{V}{h_1} \frac{\partial c}{\partial S^*} h_2 h_3 dN^* dT^* \qquad (10)$$

The area of the stream tube, the mean velocity and two mean concentrations can now be defined as

$$\begin{aligned} W &= \iint_A h_2 h_3 dN^* dT^* \\ \tilde{V} &= \iint_A V h_2 h_3 dN^* dT^* / W \\ \tilde{c} &= \iint_A V c h_2 h_3 dN^* dT^* / W \cdot \tilde{V} \\ \langle c \rangle &= \iint_A c h_2 h_3 dN^* dT^* / W \end{aligned} \qquad (11)$$

If h_1 is, to a first order approximation, independent of N^* and T^* across the pipe then it may be taken outside of the integrals and we have approximately

$$B \frac{\partial \langle c \rangle}{\partial t} + \tilde{V} \frac{\partial \tilde{c}}{\partial S} = \alpha \tilde{V} \frac{\partial^2 \tilde{c}}{\partial S^2} \quad (12)$$

The constraint on h_1 is equivalent to requiring the integral of the curvature across the stream to be much less than one. If the two average concentrations are approximately equal, we have regained the usual equation for one-dimensional transport.

There are several situations for which the averaged equation may not be a good representation of transport. Figure 1 shows a situation where a stream tube branches into two parts. Since the concentration is averaged over the width of the stream the initial concentration in each branch is the same. In order to represent situations in which the mixing is not expected to be complete at a given node, mixing factors can be included in the program.

Figure 2 illustrates a possible situation which is not handled. Here the majority of the contaminant flows laterally in a layer which only partially leads to the biosphere. The upper faster-moving layer leads directly to a river or lake. Contaminant can migrate upwards from the lower layer due to dispersion and enter the biosphere. Since in this case our stream tube consists only of the lower layer, the amount of waste entering the river is underestimated. Furthermore, since the upper layer is moving faster than the lower layer, contaminant will reach the river more quickly.

3. METHOD OF NUMERICAL SOLUTION

The previous section indicated that within each stream tube nuclide transport can be represented approximately by the equation

$$\frac{\partial c}{\partial t} + \frac{V}{B}\frac{\partial c}{\partial x} = \frac{\alpha V}{B}\frac{\partial^2 c}{\partial x^2} \tag{13}$$

where the velocity and concentration are averaged across the stream tube. The solution of Eq. 13 can be written as

$$c(x,t) = \frac{V}{BQ} \int_0^t R(\tau)\, G'(x,t-\tau)\, d\tau \tag{14}$$

where $R(\tau)$ is the rate at which waste enters the pipe and the Green's function, or impulse response, is

$$G'(x,t) = \frac{1}{\sqrt{4\ \pi\ \alpha\ \frac{V}{B}\ t}}\ \exp\left\{-\frac{\left(x - \frac{V}{B}\ t\right)^2}{4\ \alpha\ \frac{V}{B}\ t}\right\} \tag{15}$$

In order to solve the problem on the computer, this continuous-time solution must be modified to be compatible with discrete time intervals. The means by which this is done is illustrated graphically in Fig. 3. This procedure will be developed mathematically in the remainder of this section.

We first define

$$F(\ell) = \int_{T(\ell)}^{T(\ell+1)} Q\ c(z,t)\ dt \tag{16}$$

where Q is the volumetric water flow rate. Equation 16 represents approximately the amount of waste leaving the pipe at $x = z$ during the time period $(T(\ell),\ T(\ell+1))$. Substitution of Eq. 14 into Eq. 16 and reversal of the order of integration yields the following:

$$F(\ell) = \frac{V}{B}\int_{0}^{T(\ell)}\int_{T(\ell)}^{T(\ell+1)} R(\tau)G(z,t-\tau)\,dt\,d\tau + \frac{V}{B}\int_{T(\ell)}^{T(\ell+1)}\int_{\tau}^{T(\ell+1)} R(\tau)G(z,t-\tau)\,dt\,d\tau \tag{17}$$

We now move $R(\tau)$ outside the t integral and integrate the Green's function over a single time interval. The result is

$$\frac{V}{B}\int_{T(\ell)}^{T(\ell+1)} G(z,t-\tau)\,dt = \frac{1}{2}\,e^{\frac{z}{2\alpha}}\left[g(z,\,T(\ell+1)-\tau) - g(z,\,T(\ell)-\tau)\right] \tag{18a}$$

$$\frac{V}{B}\int_{\tau}^{T(\ell+1)} G(z,t-\tau)\,dt = \frac{1}{2}\,e^{\frac{z}{2\alpha}}\,g(z,\,T(\ell+1)-\tau) \tag{18b}$$

where

$$g(z,t) = \operatorname{erfc}\left\{\frac{z-\frac{V}{B}t}{\sqrt{\frac{4\alpha Vt}{B}}}\right\} e^{-\frac{z}{2\alpha}} - \operatorname{erfc}\left\{\frac{z+\frac{V}{B}t}{\sqrt{\frac{4\alpha Vt}{B}}}\right\} e^{\frac{z}{2\alpha}} \tag{19}$$

These integrals give the fraction of an impulse of waste entering the steam tube, or pipe, at time τ which would leave the pipe during the time interval $(T(\ell),\ T(\ell+1))$.

The quantity $F(\ell)$ from Eq. 17 can now be used as the input to the next pipe in the flow network. A new set of integrated Green's functions will then tell us the fraction of waste leaving the second pipe. This procedure, as generalized to more complicated networks, is described by

$$F(\ell,m) = \sum_{m'<m} \sum_{\ell'<\ell} F(\ell',m')\ \Theta(m',m)\ \psi(\ell',\ell,m',m) \qquad (20)$$

In this formula, one begins with $F(\ell',m')$, the amount of waste flowing from the entrance of a pipe (node m') during interval ℓ'. ψ, the quantity defined in Eq. 18, is the fraction of the waste entering the pipe during interval ℓ' which reaches the end of the pipe (node m) during interval ℓ. These quantities are multiplied by Θ, the fraction of the waste leaving m' which enters a pipe leading directly from m' to m. (If there is no such pipe, Θ is zero.) At points where flow paths branch, Θ is usually proportional to the amount of water entering each stream tube.

4. APPLICATION

This model has been used to study the potential for disposal of high-level radioactive waste in deep mined cavities. A generic site for disposal in shale was defined for purposes of analysis. Wastes can escape from the disposal cavity to an overlying sandstone aquifer through both the overlying rock layers and through fracture zones created by construction of the mine. The aquifer discharges to a river ten miles away. The flow paths considered are illustrated in Fig. 4; the assumptions made were derived from Golder Associates [1977a] and Holdsworth et al. [1977] and are listed in Table 1.

The results for a typical set of parameters are illustrated in Fig. 5. Releases of radioactivity to surface waters have been converted to doses to human beings by accepted modeling techniques (Koplik [1978]). The figure shows, as a function of time, the dose to an individual who eats only food from the potentially contaminated watershed and uses the same area for recreational purposes. The scale of the abscissa is normalized to show the release of waste from a single MWe-yr of power production and must be multiplied by the total amount of waste stored in the repository. The three peaks in the figure represent, from left to right, iodine and technetium escaping through fracture zones, the same nuclides escaping through undisturbed rock, and other fission products escaping through fracture zones. These results are a computation of risk given a particular scenario and are not by themselves a calculation of the actual risk from disposal of nuclear waste.

Even when a specific site for waste disposal has been chosen, there will be uncertainty in estimates of risk due to such factors as measurement error and failure of models to account for small-scale variability of geologic parameters. The efficiency of the stream tube model makes it possible to estimate the uncertainty in risk estimates due to parameter uncertainties through a "monte carlo" calculation. In this approach, a large number of different sets of parameter values describing the repository are generated so as to correspond to probability distributions representing the uncertainties in the parameters. For each set of parameter values, a hydrologic calculation is conducted and the dose to man is computed. The range of the computed doses will then give an indication of the uncertainty in risk estimates due to uncertainties in hydrologic parameters.

A preliminary study of uncertainty was conducted on the case illustrated in Fig. 4. It was assumed that the parameter values listed in Table 1 were measured in the field at some site. Uncertainties in those measurements were then estimated. These estimates are described in Berman et al. [1978]. Fifty sets of randomly generated parameter values were input to the model.

Figure 6 shows the distribution of peak individual doses which was obtained. This distribution is very broad, spreading over several orders of magnitude.

The computer model has also been used to study situations with other flow paths, such as poorly sealed boreholes, pervious backfill, and fractures in the undisturbed rock, in addition to those illustrated in Fig. 4. Studies have been carried out as well for a bedded salt repository. Possible geologic events, such as faulting and dissolution phenomena, have been studied by using the model in a probabilistic mode (Ross, Koplik and Crawford [1978]). A report describing these investigations in detail has been prepared (Berman et al. [1978]).

A principal conclusion from these studies is that the time delays involved in flow through aquifers from the repository site to a surface discharge play an important role in repository performance. As de Marsily et al. [1977] and Maini and Hocking

[1977] have observed, it is difficult to guarantee the complete hydrologic isolation of a disposal cavity.

5. SUMMARY

We have developed a new numerical approach to solving the solute transport equation. This method is based on dividing the flow field into a network of stream tubes and using a Green's function solution within each segment of the network. This method has the following principal advantages:

- It permits representation of small discontinuities without causing computational problems
- Large-scale regional flows can be incorporated in the same model as local phenomena
- Efficient computational routines reduce cost and make possible repeated model executions to study uncertainties and sensitivities
- Because the steam tube network can correspond closely to the stratigraphy, physical interpretation of the results is facilitated

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TABLE 1

ASSUMED PARAMETER VALUES

PATHWAY	LENGTH (m)	CROSS SECTION (m^2)	EFFECTIVE POROSITY	PERMEABILITY (cm/sec)
1st Segment of Tunnel Fracture Zone	1200	316	0.1	0.1
2nd Segment of Tunnel Fracture Zone	440	18.96	0.1	0.1
Fracture Zone Around Shaft in Depository Layer	100	10	10^{-3}	10^{-4}
Fracture Zone Around Shaft in Shale Barrier Layer	200	5	10^{-3}	10^{-4}
Depository Layer	100	5×10^6	0.05	10^{-9}
Barrier Layer	200	5×10^6	0.05	10^{-7}
Aquifer	1.6×10^4	4×10^5	0.1	10^{-4}

PARAMETER	VALUE
Dissolution Rate of the Waste (yr^{-1})	10^{-4}
Time for Resaturation of Depository (yr)	100
Excess Hydraulic Head of Lower Aquifer (m)	60
Horizontal Hydraulic Gradient of Upper Aquifer	0.005
Dispersion Constant (m)	50
Retardation Factor for ^{129}I and ^{99}Tc	1
Retardation Factor for Other Fission Products	100
Retardation Factor for Actinides and Their Daughters	10^4

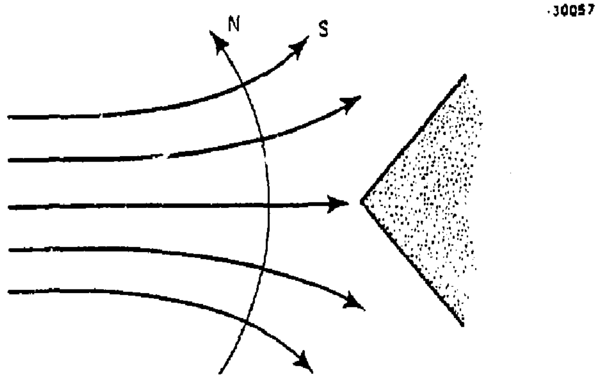

Figure 1 Branching of a Stream Tube

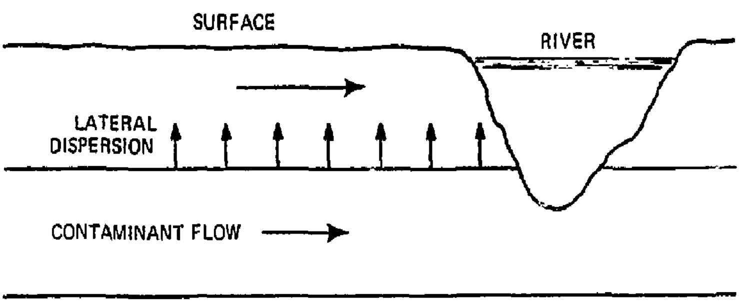

Figure 2 Dispersive Flow Into a River

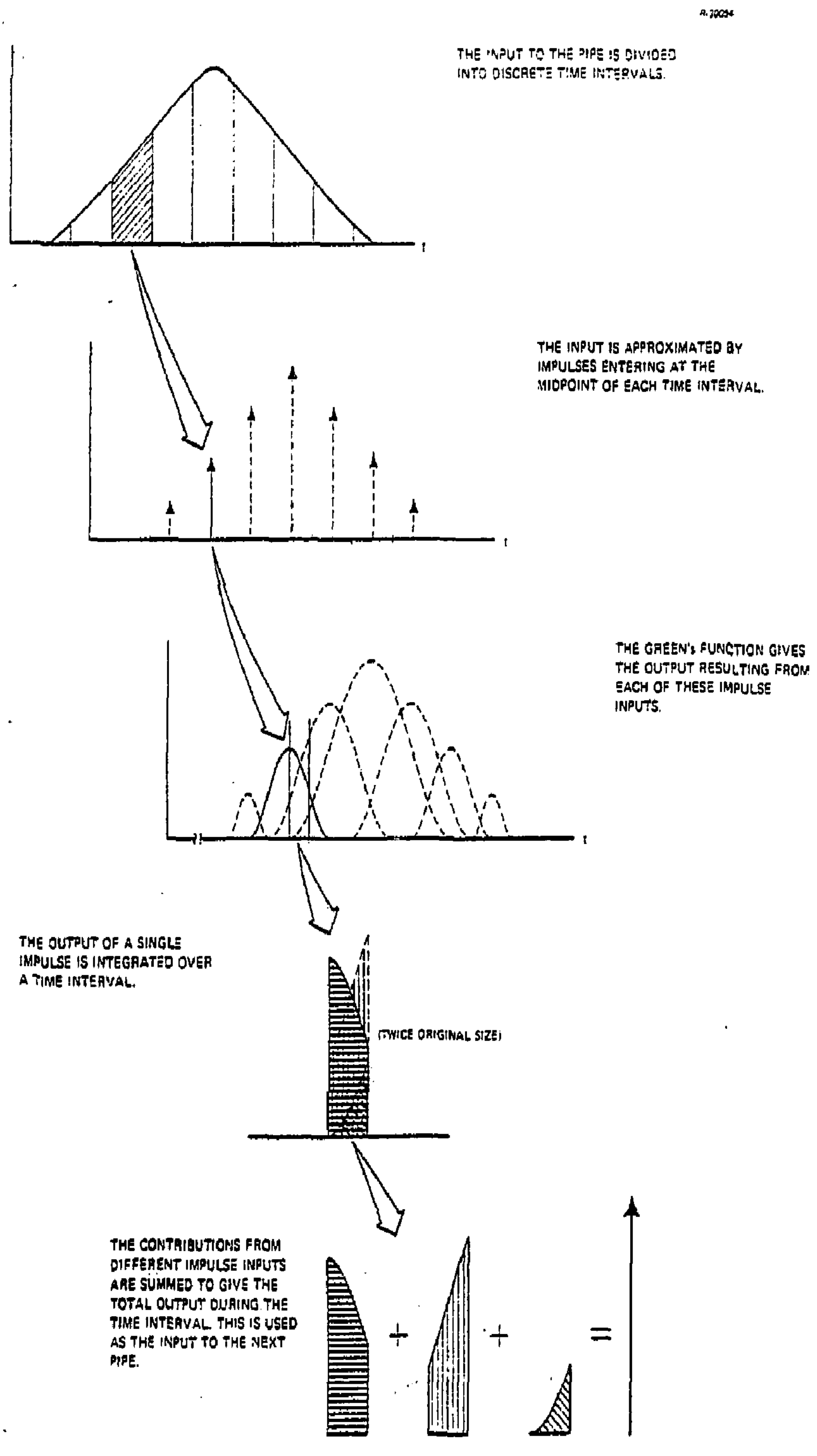

Figure 3 Procedure for Calculating the Output of a Stream Tube Segment

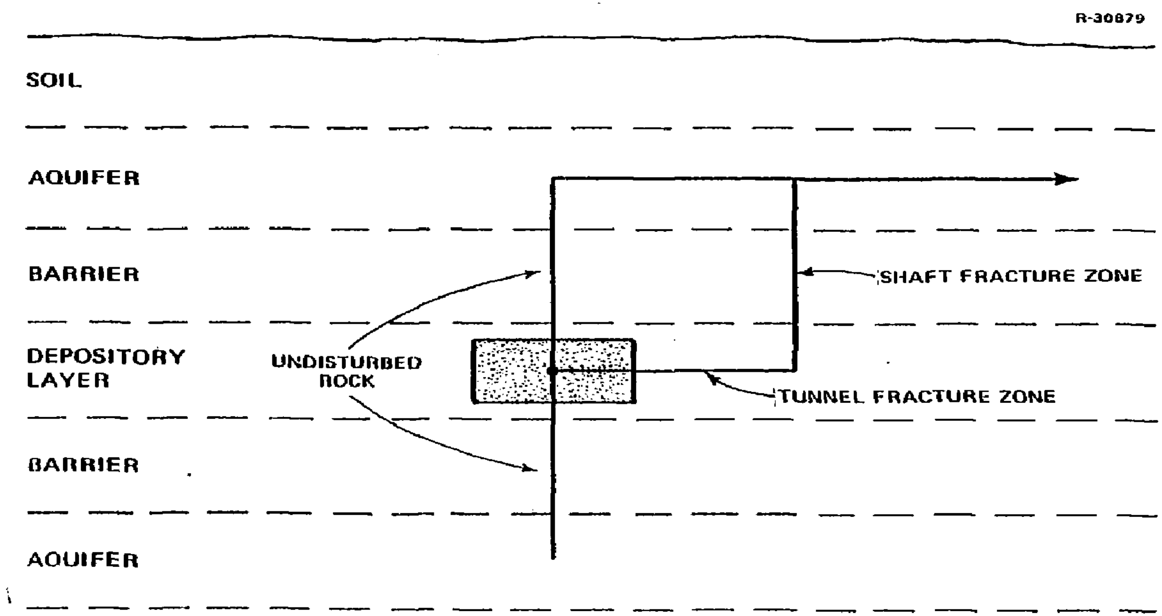

Figure 4   Flow Paths for Repository Discussed in Text

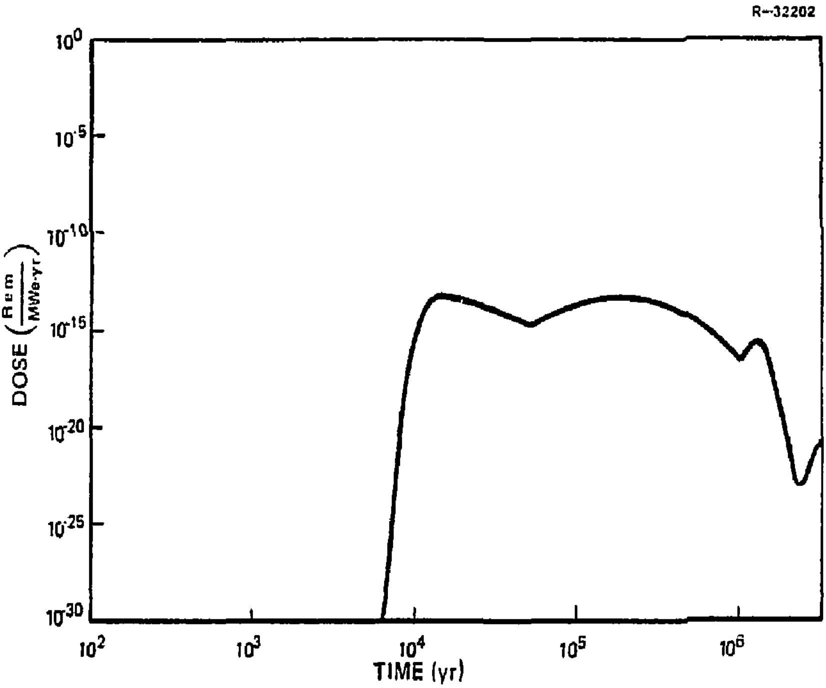

Figure 5 Whole-Body Radiation Dose over 50 Years to an Individual Living Near the Repository Shown in Fig. 4, Assuming Parameter Values Given in Table 1.

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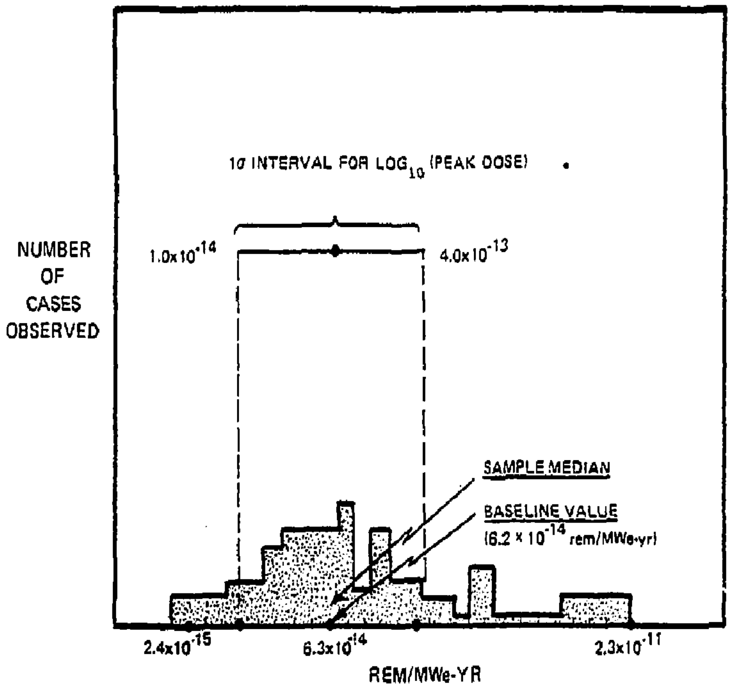

Figure 6 Distribution of 50 Peak Doses Due to Measurement Uncertainty