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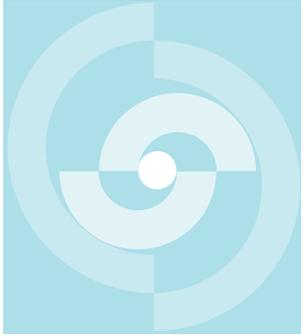
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TECHNICAL REPORT 83-06

A METHOD FOR MODELLING THE TRANS-
PORT OF NUCLIDES IN FISSURED ROCK
WITH DIFFUSION INTO THE SOLID MATRIX

R. J. HOPKIRK
D. J. GILBY

DECEMBER 1984

POLYDYNAMICS LTD, ZÜRICH

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A B S T R A C T

A technique is described, whereby diffusion of a chain of radionuclides in the microfissures or pores of the matrix material adjacent to water-bearing fractures may be calculated. The method enables the process to be calculated on all scales:

- in the individual blocks penetrated by a single fracture
- in an assembly of characteristic block sizes in a representative volume in a model on a coarser scale

The technique involves an analytical solution to the diffusion - decay - sorption equation in a block of simplified geometry. This is embedded into a numerical (finite difference) computer code which also handles the other mechanisms relevant to the transport of radionuclides in the geosphere.

The resulting program is verified against purely analytical solutions for matrix diffusion coupled with convective/dispersive transport of a single nuclide by means of benchmark calculations. Finally, the effect of matrix diffusion within blocks of varying sizes is demonstrated.

Z U S A M M E N F A S S U N G

In diesem Bericht wird eine Methode beschrieben, welche erlaubt, die Diffusion der einer Zerfallskette angehörenden Nuklide durch die Mikrorisse oder Mikroporosität des an den Haupttransportweg angrenzenden Feststoffs zu berechnen. Dabei ist es möglich, diesen Vorgang in verschiedenen Situationen zu untersuchen, wie zum Beispiel:

- in jedem einzelnen, an einen einzigen, wasserführenden Spalt angrenzenden Felsblock,
- in statistisch beschriebenen Konglomeraten von Blöcken verschiedener Grössen, wie sie in den repräsentativen Volumen eines größeren Modells enthalten sind.

Die Methode verwendet eine analytische Lösung für die Diffusions-Zerfalls-Sorptions-Gleichung in Körpern mit vereinfachter Geometrie. Diese Lösung wird in ein Computerprogramm eingebettet, welches auf numerischem Verfahren (Methode der finiten Differenzen) basiert, und für die Berechnung sämtlicher Migrationsmechanismen ausgelegt ist.

Das komplette Paket wird mit anderen, rein analytischen Lösungen des einfacheren, gekoppelten Problems (Advektion/Dispersion) mit Matrixdiffusion für ein einziges Nuklid verglichen. Schliesslich wird die Auswirkung der Matrixdiffusion in Blöcken verschiedener Grössen gezeigt.

R E S U M E

Le rapport présente une méthode propre à calculer la diffusion d'une chaîne de radionucléides dans les microfissures et les pores d'une matrice rocheuse adjacente à des fractures dans lesquelles s'effectue le transport de l'eau. Cette méthode permet de calculer le processus aux niveaux suivantes:

- dans des blocs individuels pénétrés par une fracture donnée
- dans un groupe de blocs caractéristiques à l'intérieur d'un volume représentatif dans le cadre d'un modèle à échelle plus grossière.

La technique mise en oeuvre utilise une solution analytique de l'équation de diffusion - décroissance - sorption à l'intérieur d'un bloc pour une géométrie simplifiée. Cette solution analytique est incorporée dans un programme numérique (aux différences finies) tenant aussi compte des autres mécanismes influençant le transport des radionucléides dans la géosphère.

Le programme ainsi développé a été vérifié par comparaison avec des solutions entièrement analytiques pour le cas de la diffusion dans la matrice couplée avec un transport convectif et dispersif d'un seul nucléide. On a pu mettre en évidence l'effet de la diffusion dans la matrice à l'intérieur de blocs de dimensions différentes.

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1. INTRODUCTION

Water can move through a fissured solid both via the pores in the solid matrix and via the fissures. When the solid is a crystalline rock, such as granite, it requires only a very minor crack to offer a hydraulic resistance many orders of magnitude less than that of the surrounding matrix.

Under these circumstances, convective water movement may be considered as being exclusively in the fissures. However, the rock matrix, while not exactly permeable, is slightly porous due to the presence of micro-fissures. Evidence is accumulating [1, 2, 3, 4, 5, 6 and 7] which indicates that diffusive transport in the solid matrix could be a process of some importance in the migration of trace concentrations of radionuclides. Whereas it is suspected [9] that variations may occur in the altered rock adjacent to water-bearing fissures, this effect has not yet been investigated in detail. The experimental work [8, 9], however, clearly demonstrates that the diffusive process should be taken into account in both coated and freshly formed fissures.

This report describes a method derived for predicting the effects of nuclide diffusion within the rock matrix. The method makes use of a closed analytical solution embedded into a numerical (finite difference) calculation.

2. SURVEY OF THE PROBLEM

2.1 The Fractured Medium

A discussion of the physical problem is called for before the method itself is presented. The principle feature of the transport problem is the difficulty in defining the flow paths. It has been shown [10, 13] that the fissures in a fractured rock mass may not always be treated as an equivalent anisotropic permeability. A superior model could be made in such cases by considering a network of discrete fissures, although to be able to do this each fissure should first be specified.

Present methods of fissure detection do not yield such detailed information, so at this stage the only reasonable way of deciding on those flow paths is via the use of statistics [see 10, 11, 12 and 13] combined with such observations as are possible.

Just as the available flowpaths are difficult to define, so also are the exact shapes of the blocks of rock between the fissures. It can further be seen that even when, by some statistical means the fissure systems are artificially generated, the inter-crack solid regions will tend, as in nature, to be of irregular shape and size.

The purpose of nuclide migration modelling is to predict the transport of nuclides with groundwater through the geosphere to the biosphere. The detailed geometrical modelling of each individual block of rock along the way would involve a prohibitive effort. However, the level of knowledge of the system which leads to the statistical determination of fissure systems can also lead to the determination of typical block dimensions. This is the information required for the method proposed here.

2.2 Implications for the Modeller

If it is possible to estimate the typical dimensions of the blocks in a given region of fractured rock, it will be possible also to estimate the maximum distances along which diffusion can take place in the solid, as well as the surface areas exposed to the water flowing in the fissure.

The basic computer code (TROUGH) to be used for the transport calculations in fissured or porous permeable media solves the relevant equations expressed in finite difference form. Exchange of a nuclide between the flowing water and the stagnant, micropore water in the solid matrix depends on the concentration gradient within the matrix normal to and at the fracture interface.

Effective diffusivities of nuclides are expected to be much lower in the micropores of the matrix than in the macroscopic flow channels (see [6]). This leads to very steep concentration gradients in the matrix. To model such gradients accurately by a numerical method would demand an unacceptably large number of spatial cells.

The proposed method of attack is to make use of an analytical solution to the diffusion within the rock matrix. A particular version of this method involves treating all blocks as being of simple shapes (slabs or spheres), the sizes and quantities of which may be adjusted to fit the characterisations of diffusion distance and surface-to-volume ratio of solid in each part of the field (figure 3-1 at the end of chapter 3).

It is for this version, wherein due to the simple shapes a one-dimensional diffusion solution may be used, that the derivation is presented.

Implied in the use of such a model are two assumptions. These are that diffusion is only significant in the direction normal to the local fissures and that an essentially symmetrical distribution of the diffused nuclides across the solid blocks may be considered.

At each position in the flow domain (this means each spatial cell in the TROUGH finite difference code) the blocks may then be allotted characteristic sizes and the nuclide concentration in the flowing water around them may be taken as locally uniform.

3. DIFFUSION IN THE SOLID BLOCKS

3.1 Formulation of the Equation

The one-dimensional problem in a slab or sphere which is treated here is characterised by the following features:

- nuclide diffusion in the water-saturated micropores of a solid block;
- the boundary concentration may vary randomly with time, but is identical at a given time at all points of the boundary of a given block;
- there exists instantaneous equilibrium between the concentration of a nuclide in the micropore water and that which is sorbed on to the walls of the microporosity;
- the nuclides undergo creation and annihilation in a decay chain.

If now:

C	is the concentration of a nuclide per unit volume of liquid or solid [atoms m ⁻³]
D _p	the diffusion coefficient in the pores or microfissures, taking account of their tortuosity and constrictivity [m ² yr ⁻¹]
λ	the decay constant [yr ⁻¹]
t	time [yr]
ξ	distance in the direction of diffusion [m]
ε	micro-porosity of the solid [-]

with the subscripts and superscripts:

*	indicates a property of the parent nuclide
l	in the liquid
s	in the solid

the diffusion equation for transport into or out of an infinite slab may be written:

$$\frac{\partial}{\partial t} \left(\epsilon C_1 + (1-\epsilon) C_s \right) = \frac{\partial}{\partial \xi} \left(\epsilon D_p \frac{\partial C_1}{\partial \xi} \right) + \lambda^* \left(\epsilon C_1^* + (1-\epsilon) C_s^* \right) - \lambda \left(\epsilon C_1 + (1-\epsilon) C_s \right) + S \quad (3.1)$$

where S is a general source term

The assumption of instantaneous equilibrium in the sorption process allows the use of the equilibrium distribution coefficient K_d to eliminate the nuclide concentration in the solid.

It will be observed that C_s occurs in identical combination with C_1 in the time dependent and decay sink terms in equation 3.1. Reformulating the expression:

$$\left(\epsilon C_1 + (1-\epsilon) C_s \right)$$

it becomes

$$\left(1 + \frac{1-\epsilon}{\epsilon} \frac{C_s}{C_1} \right) \epsilon C_1$$

Now, since the concentrations are expressed in terms of volume, the distribution coefficient is written

$$K_d = \frac{C_s}{\rho_s C_1}$$

where ρ_s is the solid density,

so that by eliminating C_s , an expression results which is defined as the Retention Factor:

$$R = \left\{ 1 + \frac{(1-\varepsilon)}{\varepsilon} \rho_s K_d \right\}$$

Under the assumption that ε and R are independent of position, equation 3.1 may be simplified now to become:

$$\frac{\partial C_1}{\partial t} = \frac{\partial}{\partial \xi} \left(D \frac{\partial C_1}{\partial \xi} \right) + \mu - \lambda C_1$$

where:

$$D = \frac{D_p}{R} \text{ (known as the apparent diffusivity)}$$

$$\mu = \lambda^* \frac{R^*}{R} C_1^* + \frac{S}{\varepsilon R}$$

It must be noted that the coefficient μ represents the creation of new nuclides by decay of the parent, plus any direct source of new nuclides. Because of the distribution of parent concentration μ will in general be a function of ξ .

With the additional assumption that D_p and hence D is constant along a diffusion path, we now drop the suffix 1 and the equation may be rewritten as:

$$\frac{\partial^2 C}{\partial \xi^2} - \frac{1}{D} \frac{\partial C}{\partial t} = \frac{\lambda}{D} C - \frac{\mu}{D} \quad (3.2)$$

It is this equation which is to be solved for the slab form of the solid. In spherical polar coordinates equation 3.2 takes a somewhat different form:

$$\frac{\partial^2 C}{\partial \xi^2} + \frac{2}{\xi} \frac{\partial C}{\partial \xi} - \frac{1}{D} \frac{\partial C}{\partial t} = \frac{\lambda}{D} C - \frac{\mu}{D} \quad (3.3)$$

However, introducing the substitution

$$C' = \xi C$$

the spherical form of the equation reduces to:

$$\frac{\partial^2 C'}{\partial \xi^2} - \frac{1}{D} \frac{\partial C'}{\partial t} = \frac{\lambda}{D} C' - \frac{\mu \xi}{D} \quad (3.4)$$

which, apart from a minor difference in the right hand side expression, is identical with the slab form.

The boundary conditions for the slab case are the time varying nuclide concentration in the neighbouring macroscopic flow channel, expressed as $\phi(t)$, and a zero diffusive flux across the centre of the slab, or zero C' at the centre of the sphere. We also have the initial condition that C is everywhere zero at time zero. Expressing these boundary conditions mathematically, and noting that ξ is measured from the mid-point of the solid:

$$\left. \begin{array}{ll} C = \phi(t) & \text{at } \xi = \pm 1 \\ C = 0 & \text{at } t = 0 \end{array} \right\} \quad (3.5a)$$

The symmetrical nature of the problem implies further that:

$$\frac{\partial C}{\partial \xi} = 0 \quad \text{at } \xi = 0 \quad (3.5b)$$

for a slab of thickness $2l$. For a sphere of radius l the conditions become:

$$C' = \phi(t) \quad \text{at} \quad \xi = 1$$

$$C' = 0 \quad \text{at} \quad t = 0$$

However, in this case the boundary condition at the centre of the sphere for the modified variable C' is:

$$C' = 0 \quad \text{at} \quad \xi = 0$$

The situation is thus mathematically identical to that of a slab whose thickness is equal to the radius of the sphere and which is exposed to a zero concentration on one face and a time-varying concentration on the other. The present report will describe in detail the development of the solution for the symmetrically loaded slab.

3.2 Preparation for Solution

It must be remembered that a solution method is sought for use within the framework of a time-dependent numerical solution. That is to say that equation 3.2 must be solved for each characteristic block size within every control volume (finite-difference cell)

- at each new step forwards in time, and
- for each nuclide.

The function $\phi(t)$, representing the time-varying surface boundary condition, is unknown and is certainly not a simple expression. It is convenient for the calculation to normalise the surface concentration to zero at the start of each time step and to approximate $\phi(t)$ as a linear ramp function during the step, i.e. the true surface concentration time history is approximated by a piecewise linear function. The implications of the normalisation are that all concentrations in the solid are expressed relative to the surface concentration at the beginning of the time step, and that the time scale in the analytical expression is a local one measured from the beginning of the finite difference time step.

A consequence of the local time scale is that at local time = 0 there will be some distribution of concentration within the micro-fissured solid, that prevailing at the end of the previous time step.

The normalisation to the relative concentration (i.e. to the difference between local and initial surface levels) causes no problems in the diffusive term, nor in the creation term. However, the rate of annihilation due to radioactive decay is proportional to the absolute and not the relative concentration, which may be negative.

Let the concentration at the surface of the solid block be C_f (where suffix f implies "fissure"). At the start of a time step ($t=0$) we have:

$$C_f = C_f^0$$

and at time t, following a ramp function as mentioned above:

$$C_f^t = C_f^0 + \alpha t \quad (3.6)$$

The equation 3.2, being re-expressed in terms of the relative concentration c (where $c = C - C_f^0$) instead of C, now becomes:

$$\frac{\partial^2 c}{\partial \xi^2} - \frac{1}{D} \frac{\partial c}{\partial t} = \frac{\lambda}{D} (c + C_f^0) - \frac{\mu}{D}$$

Rearranging the right-hand side of this gives:

$$\frac{\partial^2 c}{\partial \xi^2} - \frac{1}{D} \frac{\partial c}{\partial t} = \frac{\lambda c}{D} - \frac{\mu'}{D} \quad (3.7)$$

where the effective creation rate is now

$$\mu' = \mu - \lambda C_f^0$$

Following the approach used by Carslaw and Jaeger [14] a transformation of variables is now made in order to allow use of easily defined boundary and initial conditions.

Provisionally, let $u = ce^{\lambda t}$ so that equation 3.7 becomes:

$$\frac{\partial^2 u}{\partial \xi^2} - \frac{1}{D} \frac{\partial u}{\partial t} = - \frac{\mu'}{D} e^{\lambda t} \tag{3.8}$$

and now let $v+w = u$, so that:

$$\left. \begin{aligned} \frac{\partial^2 v}{\partial \xi^2} - \frac{1}{D} \frac{\partial v}{\partial t} &= - \frac{\mu'}{D} e^{\lambda t} & (a) \\ \text{and} & & \\ \frac{\partial^2 w}{\partial \xi^2} - \frac{1}{D} \frac{\partial w}{\partial t} &= 0 & (b) \end{aligned} \right\} \tag{3.9}$$

The physical boundary and initial conditions for the case of a parallel-sided slab of thickness $2l$, as given in equation 3.5, can be reformulated in terms of the new variables v and w as follows:

$$\left. \begin{aligned} v = e^{\lambda t} \alpha t & \quad \text{at } \xi = \pm 1 & (a) \\ v = 0 & \quad \text{at } t = 0 & \\ w = f(\xi) & \quad \text{at } t = 0 & (b) \\ w = 0 & \quad \text{at } \xi = \pm 1 & \end{aligned} \right\} \tag{3.10}$$

It has been found that the spatial distribution of relative concentration $c(\xi)$ in the solid at the start of a time step ($t=0$) can be closely approximated by a function:

$$f(\xi) = F(\xi) - C_f^0 \quad \text{at } t = 0 \text{ (see (3.10))}$$

where:

$$F(\xi) = a_0 + \sum_{k=1}^m a_k \cosh\left(\frac{hk\xi}{l}\right) + \sum_{k=1}^{m'} b_k \cos\left(\frac{\pi k\xi}{l}\right) \quad (3.11)$$

The symmetrical nature of this function permits a good fit to the essentially symmetrical slab problem. In practice the fit is optimised by appropriate selection of the parameters m , m' and h .

Also μ' can be expressed in terms of the averaged local absolute concentration $\overline{C^*}(\xi)$ of parent nuclide during the timestep. Thus

$$\mu' = \mu_0 + \sum_{k=1}^m \mu_k \cosh\left(\frac{hk\xi}{l}\right) + \sum_{k=1}^{m'} \eta_k \cos\left(\frac{\pi k\xi}{l}\right)$$

where:

$$\mu_k = \lambda^* \overline{a_k^*} \text{ and } \eta_k = \lambda^* \overline{b_k^*}$$

are respectively the averages of $\lambda^* a_k^*$ and $\lambda^* b_k^*$ during the timestep, except in the case of the first coefficient, where

$$\mu_0 = \left(\lambda^* \overline{a_0^*} - \lambda C_f^0 + \frac{S}{\epsilon R} \right)$$

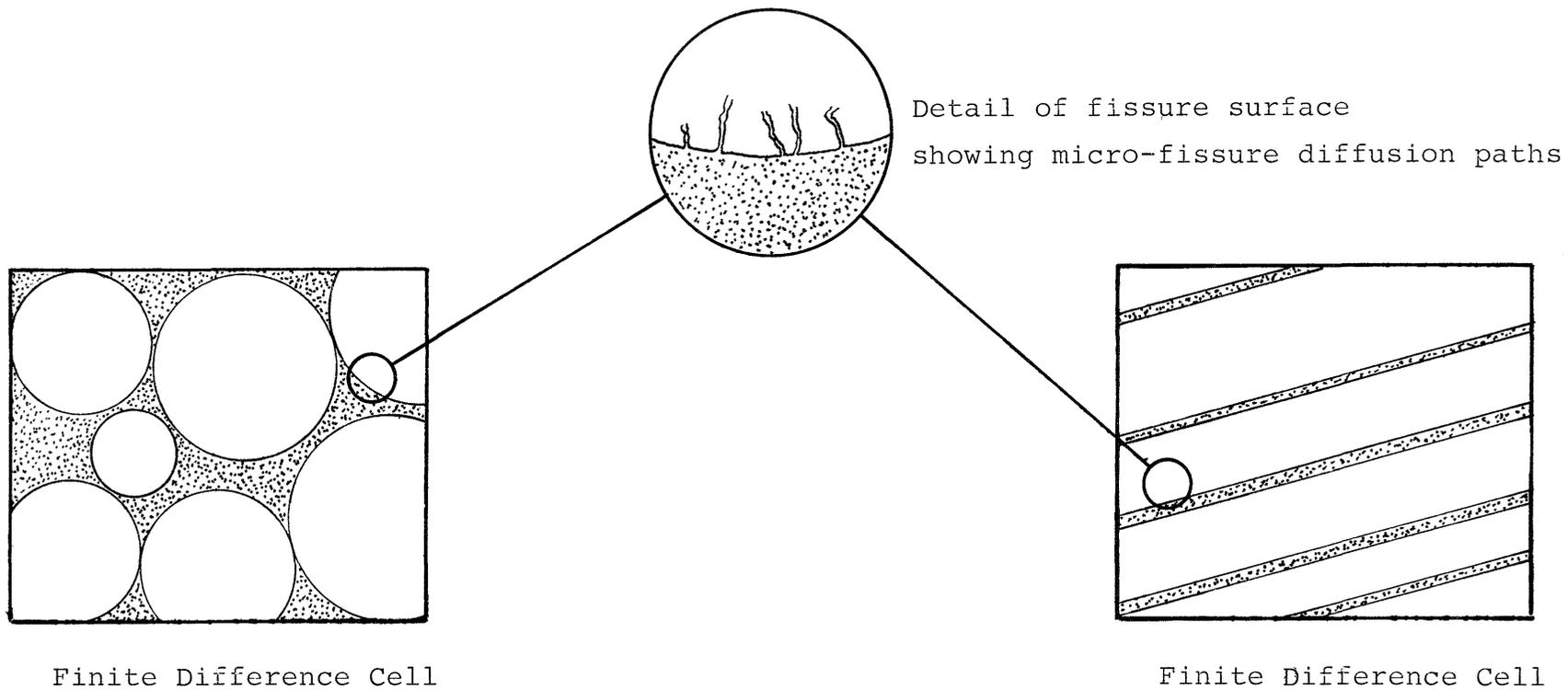


Fig. 3.1: Lumped parameter representation of solid blocks in a finite difference cell. Surface/volume ratio and diffusion path lengths may be adjusted by changing the number and characteristic dimensions of spheres or slabs.

4. ANALYTICAL SOLUTION

Two equations, 3.9(a) and 3.9(b) require solution. We start here with the former, for which a particular integral is developed by using Laplace Transforms:

$$\frac{\partial^2 \bar{v}}{\partial \xi^2} - \frac{p\bar{v}}{D} = - \frac{\mu'}{D(p-\lambda)} \quad (4.1)$$

for which:

$$\bar{v} = \frac{1}{(p-\lambda)} \left(\frac{\mu_0}{p} + \sum_{k=1}^m \frac{\mu_k}{p-\nu_k} \cosh\left(\frac{hk\xi}{l}\right) + \sum_{k=1}^{m'} \frac{\eta_k}{p-\psi_k} \cos\left(\frac{\pi k\xi}{2l}\right) \right)$$

where:

$$\nu_k = \left(\frac{hk}{l}\right)^2 D$$

$$\psi_k = \left(\frac{\pi k}{2l}\right)^2 D$$

This can be inverted to give:

$$v_{PI}(\xi, t) = \frac{\mu_0}{\lambda} (e^{\lambda t} - 1) + \sum_{k=1}^m \frac{\mu_k (e^{\lambda t} - e^{\nu_k t})}{\lambda - \nu_k} \cosh\left(\frac{hk\xi}{l}\right) + \sum_{k=1}^{m'} \frac{\eta_k (e^{\lambda t} - e^{-\psi_k t})}{\lambda + \psi_k} \cos\left(\frac{\pi k\xi}{2l}\right) \quad (4.2)$$

The Complementary Function for (3.9(a)) is the solution to:

$$\frac{\partial^2 v}{\partial \xi^2} - \frac{1}{D} \frac{\partial v}{\partial t} = 0 \quad (4.3)$$

Because of the non-zero value of the particular integral at $\xi = \pm 1$ the boundary condition for this equation is:

$$v = \psi(t) = \alpha t e^{\lambda t} - v_{PI}(\pm 1, t) \quad \text{at } \xi = \pm 1$$

and at $t = 0$:

$$v = 0$$

According to Carslaw and Jaeger [14], page 104 the solution to (4.3) is:

$$v_{CF} = \frac{\pi D}{l^2} \sum_{n=0}^{\infty} (-1)^n (2n+1) e^{-(2n+1)^2 \pi^2 D t / 4l^2} \cos \frac{(2n+1)\pi \xi}{2l} \cdot \int_0^t e^{(2n+1)^2 \pi^2 D \tau / 4l^2} \psi(\tau) d\tau \quad (4.4)$$

It is convenient at this stage to introduce certain abbreviations:

$$d = \frac{\pi D}{l^2}$$

$$\phi_n = \frac{(2n+1)\pi}{2}$$

$$r_n = \frac{(2n+1)^2 \pi^2 D}{4l^2} = \frac{\phi_n^2 d}{\pi}$$

$$\kappa_k = v_k - \lambda$$

$$\chi_k = \psi_k + \lambda$$

$$\beta_k = \frac{\mu_k \cosh(rk)}{\kappa_k} \quad (\text{where } r = \frac{h}{l})$$

$$\gamma_k = \frac{\eta_k \cos(\frac{\pi}{2} k)}{\chi_k}$$

$$\Sigma_1 \text{ signifying } \sum_{k=1}^m$$

$$\Sigma_2 \text{ signifying } \sum_{k=1}^{m'}$$

$$\Sigma_3 \text{ signifying } \sum_{n=0}^{\infty}$$

so that upon integration and simplification one is left with:

$$v_c = \left\{ at + \Sigma_1 (1 - e^{-\kappa_k t}) \beta_k + \Sigma_2 (1 - e^{-\chi_k t}) \gamma_k + a d \Sigma_3 (-1)^n {}_n I_A \cos(\phi_n \xi) + d \Sigma_3 (-1)^n {}_n I_B \cos(\phi_n \xi) \right\} e^{\lambda t} \quad (4.5)$$

where ${}_n I_A$ and ${}_n I_B$ are functions of time only, namely:

$${}_n I_A = \frac{(2n+1)}{r_n (r_n + \lambda)} \left[\frac{\lambda}{(r_n + \lambda)} - (1 + \lambda t) + \frac{r_n e^{-(r_n + \lambda)t}}{(r_n + \lambda)} \right] \quad (4.6)$$

$$\begin{aligned}
 n I_B = & \frac{(2n+1)}{r_n} \left[\frac{\mu_0}{\lambda} (1 - e^{-\lambda t}) + \sum_1 \frac{\nu_k}{r_n + \nu_k} \left(e^{\kappa_k t} - e^{-(r_n + \lambda)t} \right) \right] \beta_k \\
 & + \sum_2 \frac{\psi_k}{r_n - \psi_k} \left(e^{-\chi_k t} - e^{-(r_n + \lambda)t} \right) \gamma_k \\
 & - \frac{\lambda}{(r_n + \lambda)} \left(1 - e^{-(r_n + \lambda)t} \right) \left[\frac{\mu_0}{\lambda} + \sum_1 \beta_k + \sum_2 \gamma_k \right]
 \end{aligned} \tag{4.7}$$

The addition of the particular integral (4.2) to the complementary function (4.5) results in the following expression for v:

$$\begin{aligned}
 v e^{-\lambda t} = & at + \theta + \alpha d \sum_{n=0}^{\infty} (-1)^n n I_A \cos(\phi_n \xi) \\
 & + d \sum_{n=0}^{\infty} (-1)^n n I_B \cos(\phi_n \xi)
 \end{aligned} \tag{4.8}$$

where:

$$\begin{aligned}
 \theta = & \sum_1 \frac{(1 - e^{\kappa_k t})}{\kappa_k} \left[\mu_k [\cosh(hk) - \cosh(\frac{hk\xi}{l})] \right] \\
 & + \sum_2 \frac{(1 - e^{-\chi_k t})}{\chi_k} \left[\eta_k [\cos(\frac{\pi k}{2}) - \cos(\frac{\pi k \xi}{2l})] \right]
 \end{aligned} \tag{4.9}$$

Turning now to equation 3.9(b), it will be found that Carslaw and Jaeger [14] also present a solution for this:

$$w = \frac{2}{l} \sum_{n=0}^{\infty} e^{-r_n t} \cos(\phi_n \xi) \int_0^1 [f(\zeta) - C_f^0] \cos(\phi_n \zeta) d\zeta \tag{4.10}$$

where $f(\zeta)$ is the initial concentration distribution, defined in (3.11). This yields an expression for w in the same form as (4.8) gives for v .

$$we^{-\lambda t} = 2 \sum_{n=0}^{\infty} (-1)^n {}_n I_C \cos(\phi_n \xi) \quad (4.11)$$

where:

$${}_n I_C = \left[\frac{a_0 - C_f^0}{\phi_n} + \phi_n \Sigma_1 \frac{a_k \cosh(hk)}{(hk)^2 + \phi_n^2} + \phi_n \Sigma_2' \frac{b_k \cos(\frac{\pi}{2} k)}{(\frac{\pi k}{2})^2 - \phi_n^2} + (-1)^n \frac{b_{2n+1}}{2} \right] e^{-(r_n + \lambda)t} \quad (4.12)$$

$$\Sigma_2' = \Sigma_2 \text{ for } k \neq 2n+1$$

Retransformation of the variables leads to an expression for the concentration distribution in the solid block

$$C = (v+w)e^{-\lambda t} + C_f^0$$

or:

$$C(\xi, t) = C_f^0 + \alpha t + \theta + \sum_{n=0}^{\infty} \left(\alpha d {}_n I_A + d {}_n I_B + 2 {}_n I_C \right) (-1)^n \cos(\phi_n \xi) \quad (4.13)$$

This expression is now fitted to a series of the type given in (3.11) in preparation for the next timestep, care being taken to ensure mass conservation (see Chapter 6).

5. COMBINING ANALYTICAL AND NUMERICAL SOLUTIONS

For the TROUGH codes, transport equations are expressed in the conservative form. The equation for a concentration C_f of a nuclide in the flowing groundwater always contains the following types of terms (using tensor notation):

$$\frac{\partial}{\partial t} (PC_f) + \frac{\partial}{\partial x_i} (Q_i C_f) = \frac{\partial}{\partial x_i} \left(R_{ij} \frac{\partial C_f}{\partial x_j} \right) + \Sigma S \quad (5.1)$$

time dependent convective diffusive sources

where P , Q , R are the appropriate coefficients. These are not specified more closely as they differ for a continuum or a discrete fissure flow.

The source terms in this equation can be due to:

- injection
- leaching
- creation or annihilation by decay
- direct surface sorption
- dissolution or precipitation
- production or removal by chemical reaction
- diffusion into or out of the solid

In the present context it is the last of these which is of interest and may be defined at any instant of time (t) as:

$$S(t) = \text{solid/liquid flux} = - \sigma \epsilon D_p \frac{\partial C}{\partial \xi} \Big|_{\xi=\pm 1} \quad (5.2)$$

where σ is the specific area of the interface, usually expressed in $[m^2 m^{-3}]$

However, in the discretised form employed for numerical calculation the source term S is an average source/sink rate during a time step. A true

analytical average of $S(t)$ is employed here.

In the implicit difference scheme used for solving the equations of transport and motion in TROUGH, the spatial domain is divided into rectangular cells with scalar properties defined at their centres and component velocities on their faces, as shown in figure 5-1 at the end of this chapter.

The coefficients linking the scalar values in the neighbouring cells and with the previous (old) timestep being defined, the conservation equation for cell P is written in the canonical form:

$$\Sigma(A_i)C_f^P = \Sigma(A_i C_f^i) + S_i + S_d C_f^P \quad (5.3)$$

where $i = E, W, N, S, B, F, O$ ("O" \equiv old value) in a 3-D continuum model.

The source term is split into two components forming a linear expression in C_f^P . This technique assists in achieving a stable solution [15] where the source strength is strongly variable-dependent.

The finite difference scheme assumes linear gradients locally in space and time, so the analytical solution developed for the solid concentration in the previous chapter, demanding a linear ramp function in time for the boundary concentration, can be immediately adapted to it.

$$S = - \frac{\sigma \epsilon D_p}{\Delta t} \int_0^{\Delta t} \left\{ \sum_{n=0}^{\infty} [\alpha d_n I_A + d_n I_B + \frac{2}{1} d_n I_C] \phi_n + \frac{\partial \theta}{\partial \xi} \Big|_{\xi=1} \right\} dt \quad (5.4)$$

Noting that for a time step Δt ,

$$\alpha = (C_f^P - C_f^O) / \Delta t,$$

this may now be divided up into the linearised source form:

$$S = S_i + S_d C_f^P$$

so that in the cell P

$$\left. \begin{aligned} S_i &= - \sigma \epsilon D_p \left[\sum_{n=0}^{\infty} \left(- \frac{C_f^O}{\Delta t} d_{n A}^{I'} + d_{n B}^{I'} + \frac{2}{l} d_{n C}^{I'} \right) \phi_n + \left(\frac{\partial \theta}{\partial \xi} \Big|_{\xi=1} \right)' \right] \\ S_d &= - \sigma \epsilon D_p d \sum_{n=0}^{\infty} \frac{1}{\Delta t} d_{n A}^{I'} \phi_n \end{aligned} \right\} \quad (5.5)$$

where the primed terms represent the time averages of the corresponding terms in (5.4).

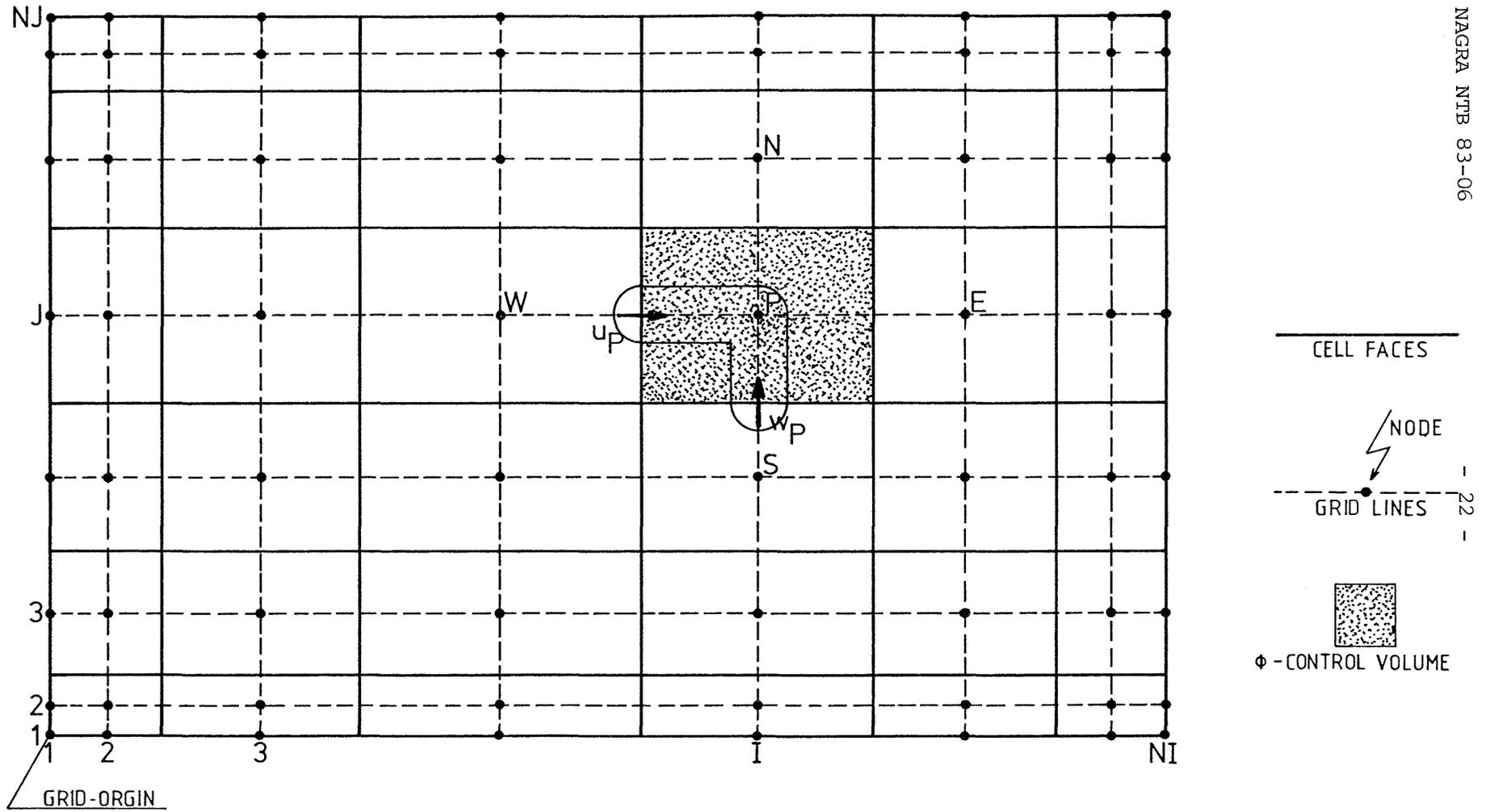


Fig. 5.1: Finite difference grid showing cell numbering scheme.

6. FITTING THE INITIAL DISTRIBUTION FUNCTION

At the end of a finite difference time step it is necessary to fit a function of the form 3.11 to the concentration distribution described by the analytical solution 4.13. This fitting must be carried out for each nuclide and must take account of the following considerations:

- a) The fitted function forms the initial condition for the analytical solution in the next time step. Therefore, if accuracy is to be maintained through a sequence of time steps, the function must reliably model the concentration distribution at the end of the current step.
- b) In order that the overall mass consistency of the full transport model is maintained the fitted function must lead to a correct value of the total quantity of each nuclide held in the micro-pores.

The general form of the fitting function was selected as being suitable for the type of distribution patterns to be expected in a slow diffusion time varying surface concentration problem. Using a conventional least squares method to fit such a function to a number of discrete points on a concentration distribution, i.e. specific solutions of 4.13, will therefore generally meet the condition imposed by (a) above.

A standard least squares fitting will not, however, necessarily meet the condition of mass consistency expressed in (b). Since it is of primary importance to ensure overall mass consistency at every stage in a calculation of nuclide transport possibly extending over many thousands of years it was therefore considered necessary to introduce a modification to the conventional least-squares fitting technique so as to guarantee this consistency when determining the parameters a_k and b_k of 3.11.

To develop expressions for the fitting parameters we first define a total fitting square error as follows:

$$E^2 = \sum_i \left[C_i - a_0 - \sum_1 a_k \cosh\left(\frac{hk\xi_i}{1}\right) - \sum_2 b_k \cos\left(\frac{\pi k\xi_i}{2l}\right) \right]^2 \quad (6.1)$$

where the subscript i refers to the data points, concentrations C_i at prescribed points ξ_i , to which the expression is to be fitted. Following the usual procedure we equate the differential of this expression with respect to each of the fitting parameters in turn to zero and obtain a set of equations in a_k and b_k . These equations can be summarised in matrix form:

$$[S] [a] = [y] \quad (6.2)$$

[S] being a symmetric square matrix of order $(m + m' + 1)$ with elements defined as follows:

for j and $k \leq m + 1$

$$S_{j,k} = \sum_i \cosh\left(\frac{(j-1)h\xi_i}{1}\right) \cdot \cosh\left(\frac{(k-1)h\xi_i}{1}\right)$$

for j and $k > m + 1$, and $j' = j - m - 1$, $k' = k - m - 1$

$$S_{j,k} = \sum_i \cos\left(\frac{j'\pi\xi_i}{2l}\right) \cdot \cos\left(\frac{k'\pi\xi_i}{2l}\right)$$

for $j \leq m + 1$ and $k > m + 1$

$$S_{j,k} = \sum_i \cosh\left(\frac{(j-1)h\xi_i}{1}\right) \cdot \cos\left(\frac{k'\pi\xi_i}{2l}\right)$$

The vector [a] has elements: m' , a_0 , a_1 , a_2 a_m , b_1 b_m , and the elements of the vector [y] are defined by:

$$Y_i = \sum_i \left(\cosh \left(\frac{(j-1)h\xi_i}{l} \right) \cdot C_i \right) \quad j \leq m + 1$$

$$Y_j = \sum_i \left(\cos \left(\frac{j' \pi \xi_i}{2l} \right) \cdot C_i \right) \quad j > m + 1$$

These equations are sufficient to define all the fitting parameters provided there are more data points than parameters.

The total quantity of nuclide held in the micropores is given by:

$$N = \int_0^l C(\xi) d\xi \quad (6.3)$$

Using first the analytical expression and then the still-to-be-fitted function for the new concentration distribution $C(\xi)$ under the integral we derive two expressions for N as follows:

from 4.13:

$$N = (C_f^0 + \alpha t)l + \int_0^l \theta d\xi + \sum_{n=0}^{\infty} \frac{1}{\phi_n} \left(\alpha d_n I_A + d_n I_B + 2 d_n I_C \right) \quad (6.4)$$

and from 3.11 using the coefficients which still have to be determined:

$$N = a_0 l + \sum_1 \left(\frac{a_k l}{hk} \right) \sinh \left(\frac{hk}{l} \right) + \sum_2 \left(\frac{2b_k l}{\pi k} \right) \sin \left(\frac{\pi k}{2l} \right) \quad (6.5)$$

Equating these we obtain an extra equation to add to the set 6.2, this extra condition being that the total quantity of nuclides is correct.

Since there are now more equations than unknown a_k and b_k parameters and since it is not desirable to introduce additional terms to the fitted

function, it is necessary to discard one of the least squares fitting conditions.

After some experimentation it was found that the least disturbance to the quality of the fit, as quantified by the square error E^2 , resulted when the equation omitted was that resulting from the differentiation with respect to a_1 .

The consequence of this step is that the value of E^2 is no longer a minimum with respect to the a_1 parameter. However, since this least squares condition still applies to the other parameters, and the space integral of the concentration distribution is exactly correct the loss of this constraint does not significantly reduce the quality of the fit.

7. VERIFICATION OF THE METHOD

In order to increase the level of understanding of the processes involved in radionuclide migration, to examine the effects of different modelling techniques and to compare performances of different computer codes, an international comparison project, INTRACOIN, was set up in 1981.

The first level of INTRACOIN [17] provided a set of simple benchmark test cases, one of which has been selected as a test problem for verification of the method described in this report. Results for case 5 calculated with TROUGH 1-D are compared with those produced from an analytical solution to the problem. The analytical solution is that one produced at the Royal Institute of Technology in Stockholm and described by Rasmuson & Neretnieks [2, 16]. The results of the calculations with NUCDIF, which incorporates this solution, have been reported in [18].

Case no. 5 concerns the migration of an initial inventory of 3 nuclides, expressed in arbitrary units of activity, migrating along a pathway consisting of parallel fissures separated by slabs: The three nuclides form a decay chain. Water flows fast through the fissures with a high hydrodynamic dispersion coefficient. The definition of the case is as follows:

Inventory:

Nuclide (k)	Half life ($t_{1/2}$ (k))	Initial inventory ($I_k(0)$)
^{245}Cm	8.500×10^3	0.700
^{237}Np	2.140×10^6	1.000
^{233}U	1.592×10^5	0.004

The inventory is expressed in arbitrary activity units, normalised to the initial level of the longest-lived member of the decay chain (^{237}Np)

Time to leach (T) at a constant mass removal rate:	10^5 yr
Length of 1-dimensional migration path (L):	500 m
Water flux velocity (v):	500 m/yr
Water flux (\dot{q}):	$1 \text{ m}^3/\text{yr}$
Hydrodynamic dispersion coefficient (D_1):	$2.5 \times 10^4 \text{ m}^2/\text{yr}$
Fissure aperture (2b):	10^{-4} m
Fissure spacing (s):	5 m
Porosity of the rock matrix (ϵ):	5×10^{-3}
Bulk density of the rock matrix (ρ):	$2.7 \times 10^3 \text{ kg/m}^3$
Effective diffusivity in the rock matrix (ϵD_p):	$10^{-12} \text{ m}^2/\text{s}$

Volumetric Equilibrium Distribution Coefficients

Nuclide (k)	$K_v(k) [\text{m}^3/\text{m}^3]$
^{245}Cm	570
^{237}Np	80
^{233}U	30

The upstream boundary condition treated is a boundary concentration, defined by the leaching time into the groundwater:

$$C_k(0, t) = \frac{I_k(t)}{\dot{q} T} [H(t) - H(t-T)]$$

where the Heavyside function $H(\tau) = 0$ for $\tau < 0$ and is unity for $\tau > 0$

The time history of the nuclide concentrations at a point 500 m downstream from the prescribed concentration boundary has been calculated using TROUGH with the embedded analytical solution of matrix diffusion. The results are plotted on Figure 7.1 for the two dominant nuclides,

together with the corresponding results from Magnusson et al. [18]. The agreement between the two methods is extremely good over the whole time history.

The TROUGH calculation indicates a slightly lower and earlier peak concentration. This suggests a small amount of numerical dispersion or truncation error in the analytical solution.

However, the discrepancies are so small as to be barely distinguishable on the plots. Over most of the time history the difference between the two sets of results is less than 5 %.

INTRACOIN Level 3 [19] was concerned specifically with advective dispersion of a single nuclide in a fractured medium. The medium consists of a set of cubic blocks of rock separated by narrow planar fissures through which the nuclide carrying groundwater moves. Twelve cases were defined, each being a specific combination of the possible retardation mechanisms:

- Sorption on fracture surface
- Hydrodynamic dispersion
- Matrix diffusion
- Limited nuclide solubility

The parameter values for the physical model are given below:

Parameter Values for the Central Case of INTRACOIN, Level 3

Parameter	Symbol	Value
Water flux in the rock	Q	$0.2 \times 10^{-3} \text{ m}^3/\text{m}^2, \text{ yr}$
Travel distance	Z_0	100 m
Flow porosity (1)	ϵ_f	$10^{-4} \text{ m}^3/\text{m}^3$
Matrix porosity	ϵ_m	$5 \times 10^{-3} \text{ m}^3/\text{m}^3$
Block size (cubes)	S	5 m
Half life ^{237}Np	T 1/2	2.1×10^6 years
Sorption distribution coefficient	K_d	$5 \text{ m}^3/\text{kg}$
Density of the rock	ρ	$2'700 \text{ kg}/\text{m}^3$
Nuclide inventory (^{237}Np only)	m_i	1.5 Ci (2.114 kg)
Leach time	T	10^5 years
Dispersion length in the fissures(2)	α	20 m
Diffusivity in the matrix	D_p	$3.15 \times 10^{-4} \text{ m}^2/\text{yr}$

$$(1) \quad \epsilon_f = 3 \frac{2b}{S}, \quad \text{where } 2b \text{ is the width of the fractures}$$

$$(2) \quad D_L = \frac{\alpha Q}{\epsilon_f}, \quad \text{where } D_L \text{ is the lateral dispersion coefficient}$$

The case of interest in the present context is variant 8 in which hydrodynamic dispersion, sorption and matrix diffusion are all featured.

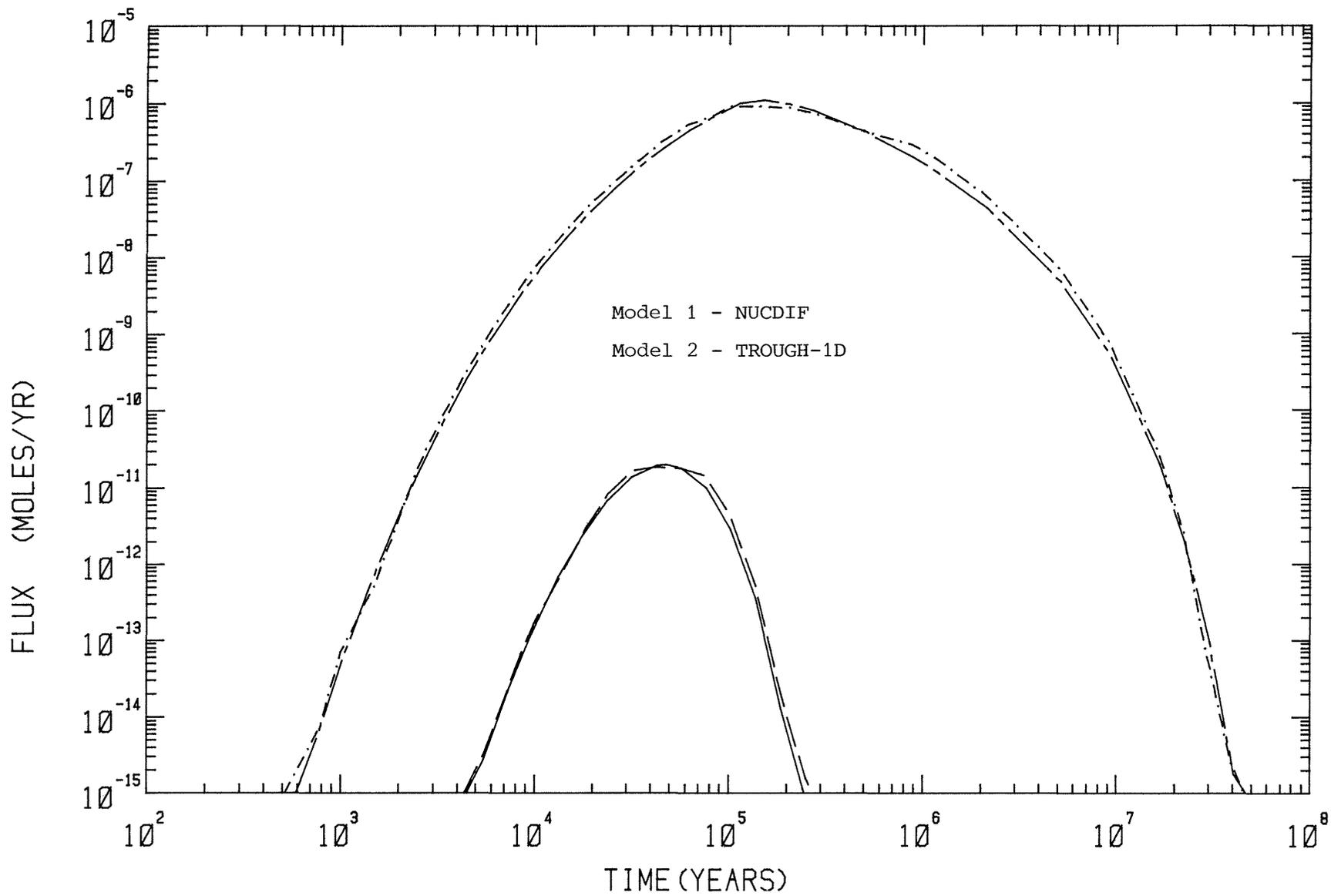
Once again concentration outflow time histories were calculated and have been compared with results derived from analytical models. The following table shows the maximum calculated activity flux and the time of the maximum given by TROUGH, RANCHM [20] and NAMID [21].

Comparison of Predictions of Maximum Discharge Rate and the Time of the Maximum from Different Models

	TROUGH	RANCHM D	NAMID
Maximum Flux [Ci/yr]	2.77×10^{-13}	2.89×10^{-13}	2.97×10^{-13}
Time of Maximum [yr]	9.38×10^6	9.50×10^6	9.50×10^6

It can be seen that, as before, the TROUGH code gives a prediction of peak concentration that is a little lower and earlier than that predicted by the analytical codes.

Finer discretisation of the numerical model both in space and time, will yield closer correlation between it and analytical codes, but at the expense of computation time.



CM-245 1
NP-237 1
CM-245 2
NP-237 2

NAGRA NTB 83-06

Figure 7.1:

COMPARISON OF FLUX TIME HISTORIES FROM MODELS 1 & 2

8. THE EFFECTS OF MATRIX DIFFUSION

In this chapter we present results of a series of calculations carried out to demonstrate the way in which matrix diffusion could influence the transport of radionuclides.

The basic demonstration problem concerns the transport of a 3-member nuclide decay chain in groundwater flowing through a fissured rock. The radionuclides are assumed to be leaching from an initial inventory over a period of 10'000 years into groundwater moving at a velocity of 10 m/yr in the fissures. Dispersion and convection take place within the moving groundwater and nuclides are exchanged between a mobile component and immobile components, either being adsorbed onto the fissure surface or diffusing perpendicularly to the main flow in the stationary groundwater trapped in the micro-fissures.

The effective porosity of the fissured rock, i.e. the fraction of the total cross-section of 100 m^2 taken up by flowing fissures, is assumed to be 0.001, i.e. the groundwater flux is $1 \text{ m}^3/\text{yr}$. Within the flowing fissures the relatively high velocity gives rise to a strong hydrodynamic dispersion with a coefficient of $500 \text{ m}^2/\text{yr}$. The consequence of this is that the concentration in the moving groundwater changes very little along the flow path.

Three different cases are considered as follows:

- a) No matrix diffusion. Equilibrium exists at all times between the nuclide concentration in the groundwater and the immobile nuclides sorbed onto the fissure surfaces.
- b) Matrix diffusion with a characteristic slab half-thickness of 0.1 m.
- c) Matrix diffusion with two characteristic slab thicknesses, 0.01 m and 0.1 m.

The area of the interface between the rock matrix and the flowing fissure is chosen, such that the total volume of rock into which nuclides may diffuse is the same in cases b) and c). Furthermore, in case c) the total rock volume is divided equally between the two characteristic slab thicknesses. Thus, we have:

Length of Flow Path Over Which Matrix Diffusion Occurs

	<u>0.1 m Slab</u>	<u>0.01 m Slab</u>
Case a)	0 - 24 m	-
Case b)	0 - 12 m	12 - 132 m

With these conditions, and the physical parameters presented in Table 8.1 the concentration time history for each nuclide at a point 150 m downstream from the leaching point was calculated for each of the three cases. The results are presented in Figures 8.1 - 8.3 and illustrate quite clearly the effects of the matrix diffusion.

The concentrations of activity of the three nuclides at the monitoring point generally rise during the leaching period and fall thereafter in each case.

When there is no matrix diffusion (Figure 8.1) the relatively abundant Cm-245 and Np-237 reach a maximum concentration during the leach period. A small drop from the maximum can be seen for the fast decaying Cm-245.

The U-233 concentration rises slowly towards a peak at the end of the leach period, the rise being fed by both the initial inventory and the decay of the Np-237. After the end of the leach period the concentration falls steadily as the sorbed nuclides reenter solution. The concentration of the strongly sorbed Cm-245 falls relatively slowly, despite its much faster decay rate, while the U-233 remains in equilibrium with its parent Np-237.

When matrix diffusion occurs the concentration time histories take on a somewhat different form (Figures 8.2 and 8.3).

During the leach period the rate of rise of concentration is slowed as nuclides diffuse strongly into the matrix. Peak concentrations, reached only at the end of the leach period, are of course greatly reduced.

Nuclides diffuse slowly back into the flowing water after the leach period extending the time during which the monitored concentration remains at a significant level. The dominant feature in the Cm-245 release profile is the decay rate which is fast in comparison with the rate at which curium nuclides diffuse out of the matrix.

When the matrix diffusion occurs in two equal volume zones (Figure 8.3), the smaller thickness slabs rapidly become saturated with nuclides. After the leach period these smaller slabs also release their nuclides more quickly. Consequently the major differences between cases b) and c) occur at the beginning of each of the transient processes. There is also some small difference in the peak concentrations achieved, those in case c) being lower as a result of the greater volume of rock matrix containing relatively high concentrations of nuclides.

Table 8.1:

Parameter	Symbol	Unit	Nuclide 1 Cm-245	Nuclide 2 Np-237	Nuclide 3 U-233
Initial inventory	I	[-]	0.7	1.0	0.004
Half-life	t 1/2	[yr]	8'500	2'100'000	160'000
Decay constant	λ	[1/yr]	8.16×10^{-5}	3.24×10^{-7}	4.35×10^{-6}
Retention Factor	R	[-]	500	100	60
Apparent diffusivity in micro-fissures	D	[m ² /yr]	5.5×10^{-6}	3.9×10^{-5}	1.1×10^{-4}

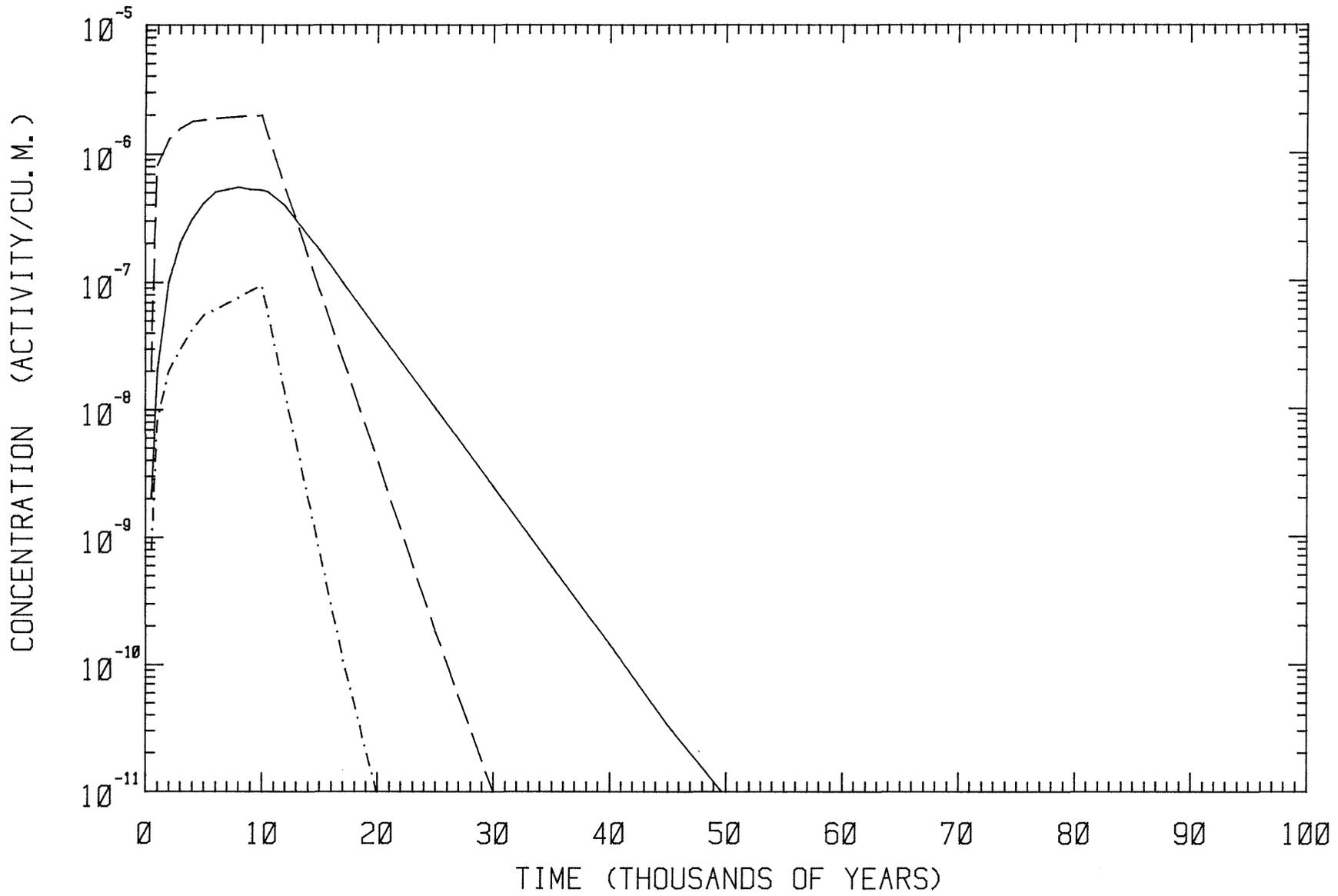


Figure 8.1: Nuclide concentration time histories - no solid diffusion, equilibrium sorption

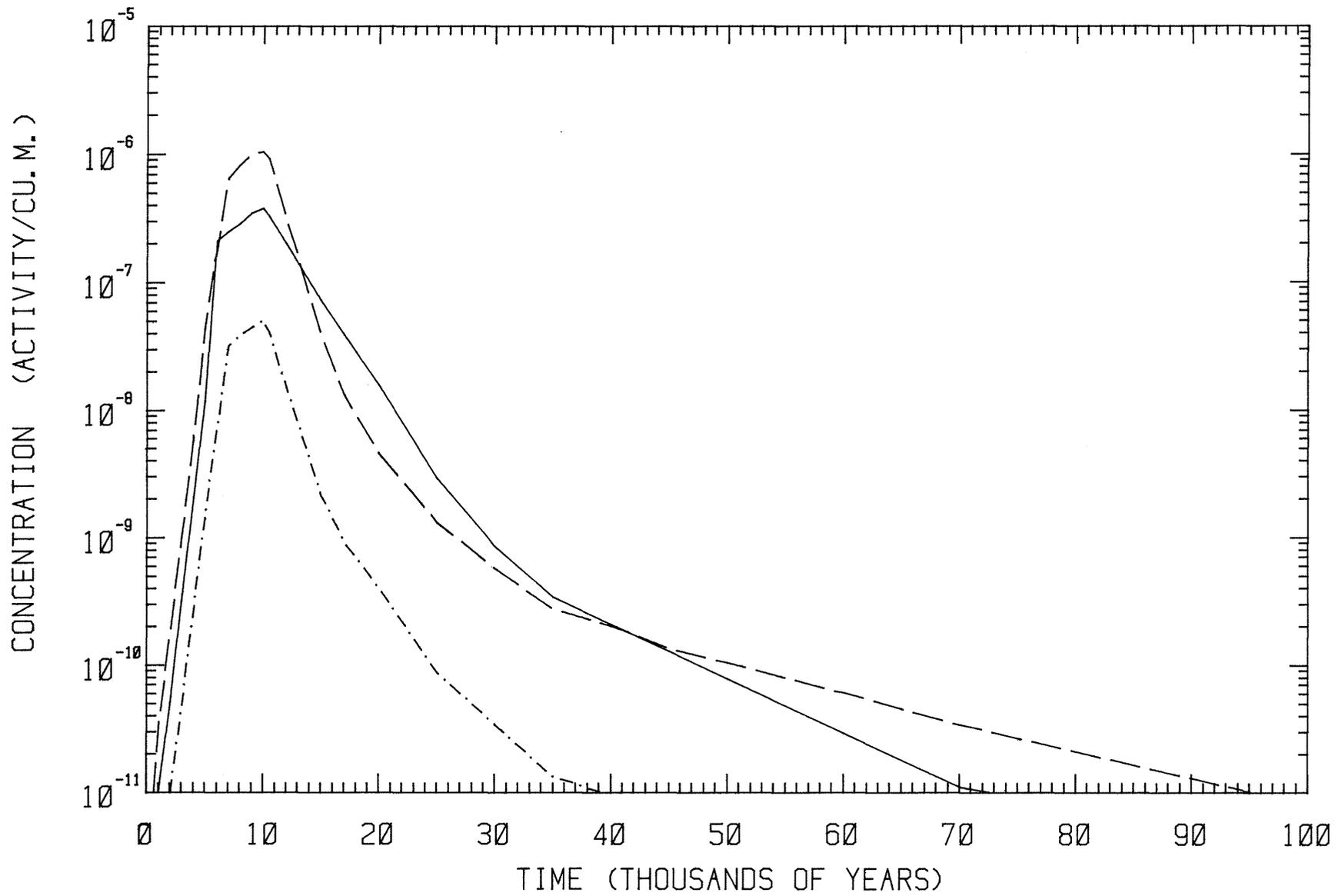


Figure 8.2: Nuclide concentration time histories - with solid diffusion

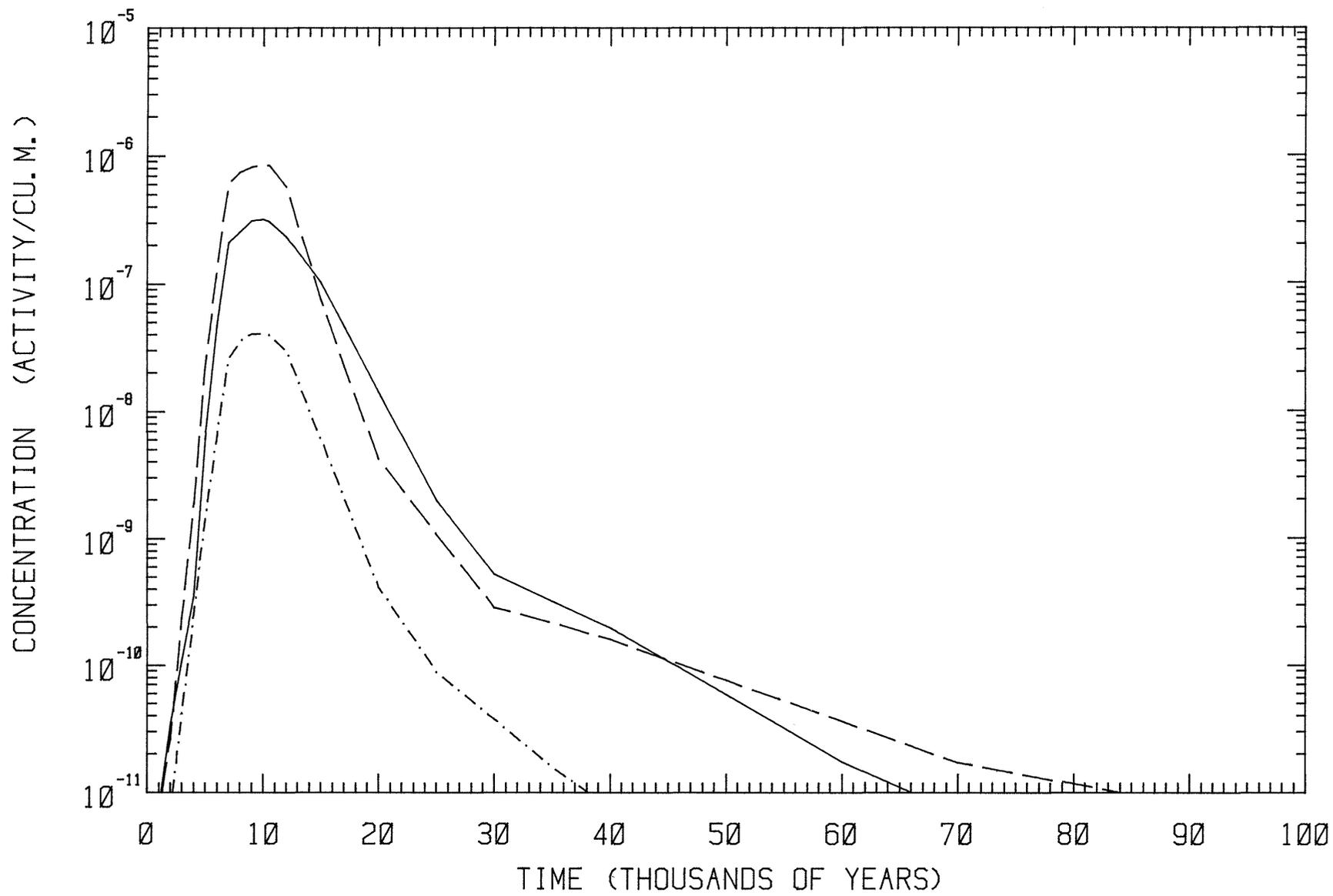


Figure 8.3: Nuclide concentration time histories - solid diffusion with two characteristic block sizes

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10. NOMENCLATURE LIST

a,b,d,h	Constants
C	Concentration of radionuclides in the micro-fissures [atoms per m ³ of space]
c	Concentration of radionuclides in the micro-fissures relative to interface concentration
D	Apparent diffusivity of microfissure system [m ² /yr]
D _p	Diffusion coefficient in the pore or micro-fissures [m ² yr ⁻¹]
I	A collection of terms following an integration
K _d	Equilibrium concentration ratio (N _s /ρN _l) [m ³ /kg]
l	Depth of micro-fissure system [m]
l	(subscript) referring to liquid
m,n	Summation indices
o	(superscript) referring to "old" values, i.e. values at the end of the previous time step in the numerical calculation
R	Retention factor $(1 + \frac{1-\epsilon}{\epsilon} \rho_s K_d)$ [-]
r _n	series term $[(2n+1)^2 \pi^2 D/4l^2]$
S	General source term [atoms/(yr m ³ of space)]
s	(subscript) referring to solid
S _d	Coefficient of dependent component of source term
S _i	Independent component of source term
t	Time [yr]
α	Rate of change of concentration [atoms/(m ³ yr)]
ε	Porosity [-]
λ	Decay constant [1/yr]
μ	Creation rate of nuclides by decay of parent [atoms/(yr m ³ of solid)]
μ _k , η _k	Coefficients in the distribution function of μ
ξ	1-dimensional space coordinate in the microfissure system (0 ≤ ξ ≤ 1) [m]
ρ	Density [kg/m ³]
σ	Specific surface area per m ³ of space [m ² /m ³]

ϕ Concentration of radionuclides in liquid [atoms/kg of liquid)
 ϕ_n Series term $[(2n+1)\pi/2l]$