MIGRATION OF RADIONUCLIDES THROUGH SORBING MEDIA ANALYTICAL SOLUTION - I

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MIGRATION OF RADIONUCLIDES THROUGH SORBING MEDIA ANALYTICAL SOLUTION - I

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1. INTRODUCTION

This report presents analytical solutions for the transport of radionuclides through porous sorbing media. These solutions are developed to serve as analytical standards for benchmark checking of numerical solutions and to use as predictive techniques.

The basic transport equation is derived and is simplified for the case of one-dimensional transport through isotropic porous media, with local chemical equilibria between radionuclides in water solution and sorbed by the solid. Analytical solutions in the present report are limited to this simplified model and are also limited to an infinite plane source of dissolving radionuclides.

Possible modes of release of radionuclides from the waste solid into the ground water are examined. Release-mode equations and boundary conditions are developed for the cases of (a) a constant overall rate of dissolution of the waste matrix and its contained radionuclides and (b) a constant fractional rate of dissolution of each radionuclide species. Each release mode can be written in terms of a concentration boundary condition foreach radionuclide at the point of dissolution, which is applicable if transport by liquid diffusion at the waste location can be neglected, or, more generally, as a volumetric source term to be used directly in the transport equation.

A finite dissolution rate of the waste material results in a "band release", wherein a chromatographic band of the released radionuclides propagates outward through the geologic medium. In Chapter 3 a superposition theorem is developed such that the relatively complicated equations for band release can be constructed from the simpler solutions for a "step release", which assumes that the waste material dissolves at a finite rate over an infinite period of time.

In Chapter 4 recursive and general solutions of the transport equation, with and without dispersion, are presented. Solutions are presented for individual members in a three-member decay chain and also for any member in a decay chain involving an arbitrary number of members.

In Chapter 5 the recursive and general solutions are applied to the various release modes, including the limiting cases of impulse release and step release, for an infinite geologic medium surrounding the radioactive waste. The solutions are demonstrated for the three-member chain

$$^{234}\mathrm{U} \rightarrow {}^{230}\mathrm{Th} \rightarrow {}^{226}\mathrm{Ra}$$

which is of practical importance in the analysis of possible long-term environmental releases from high-level waste repositories. Effects of the alternative source boundary conditions are demonstrated. The properties of the maximum concentration of Ra-226, its increase with distance from the repository, and its relative insensitivity to the dissolution rate, are illustrated.

The analytical solutions are also demonstrated for the radionuclide chain

$$^{237}\mathrm{Np} \rightarrow {}^{233}\mathrm{Pa} \rightarrow {}^{233}\mathrm{U} \rightarrow {}^{229}\mathrm{Th} \rightarrow {}^{225}\mathrm{Ra}$$

which is also important in analyses of long-term environmental effects.

In Chapter 6 the general solutions are applied to multilayered geologic media and explicitly for a two-media system consisting of a medium of finite thickness adjacent to the radioactive waste, sandwiched between infinite half spaces of a second medium. Explicit solutions are developed for a three-member decay chain without dispersion. A recursive formula is developed for the system with dispersion.

In the Appendix is presented a simple computer program written to demonstrate the migration features of radionuclides in a three-member chain, with and without dispersion.

The analyses presented herein were developed in part under financial support from the University of California and were extended under financial support from the Office of Waste Isolation (ONWI) of the U. S. Department of Energy. Work under ONWI support performed during the period April 1 through August 31, 1979 is included in this report. The ONWI Project Officer is Dr. H. C. Burkholder.

More detailed demonstration of the use of these equations, as well as analytical solutions for non-equilibrium sorption and multidimensional cases, will be the subject of future reports.

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2. THE RADIONUCLIDE TRANSPORT EQUATION

2.1 Introduction

The accurate prediction of the probable pattern of contaminant migration is of primary concern in the evaluation of geological disposal of high-level radioactive waste. This chapter is concerned with the formulation of the basic differential equation for the transport of radionuclides in a geological medium saturated with groundwater. Section 2.2 contains a discussion of the transport equation for porous media. In Section 2.3 a linearized formulation is discussed.

2.2 Transport Equation for Radionuclides in Porous Media

In this section the equations for transport of radionuclides through porous media will be developed with the aid of the averaging concept for a heterogeneous water-solid medium, as proposed by Whitakker (W1) and Slattery (S2).

2.2.1 General transport equation in porous media

The geological medium is considered to be a composite material which consists of fluid and solid phases. The phenomena of nuclide migration through a geological medium are complicated microscopically because the porous medium is not an ordered structure. We can formulate the transport equation governing the nuclide migration in a geological medium according to a statistical approach (W1,S2) which smooths out the properties in complicated composite material by averaging over a small volume domain in this medium.

Let us consider a composite medium as shown in Figure 2.1, in which arbitrary positions are measured by the reference coordinate. The volume of an arbitrary domain D is designated by V. The domain is fixed in time and in space. The center of the domain occupies the point O', the position of which is designed as $\mathbb{X}$ on the reference coordinate. The position of arbitrary point in the domain D is designed as $\mathbf{x}$, the relative position vector from the point O'.

We can select the domain D of a volume element which is small enough compared to the characteristic length of the composite medium and large enough compared to the microscopic feature of the composite medium, e.g., the inter-particle pore size.[a1]

Consider a dynamical property, Ψ, which is generally described as

$$\psi = \psi(\mathbb{X}, \mathbf{x}, t) \tag{2.1}$$

[a1] In this system, there are two kinds of pores. One is a pore of the inter-soil-solid particle and the other an intra-fine pore in the solid phase. The latter should be included in the solid phase.

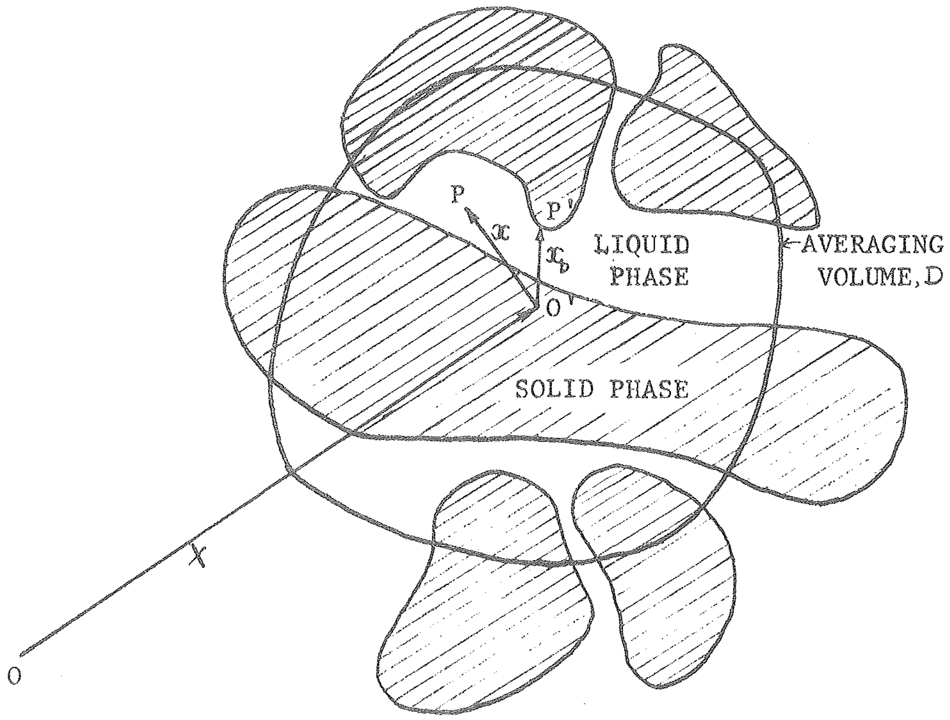

Fig. 2.1 - A schema of averaging volume domain comprising the solid phase and fluid phase

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The transport equation which governs the time-dependent and spatially-dependent properties in the composite medium system can be obtained by the following three steps:

1. Smooth the property in the domain with the aid of volume averaging:

$$\hat{\psi} = \frac{1}{V}\int_V \psi(\mathbb{X}, x, t)\, dx \qquad (2.2)$$

2. Assign the smoothed value to the property at the center of the domain. The property is thus reduced to

$$\hat{\psi} = \hat{\psi}(\mathbb{X}, t) \qquad (2.3)$$

   $\hat{\psi}$ is to be considered as a point function with respect to the position, $\mathbb{X}$.

3. Derive the transport equation for the point function, $\hat{\psi}$.

The values of the dynamical property are different for solid and fluid phases, s and f, in the domain.

$$\psi(\mathbb{X}, x, t) = \begin{cases} \psi_f(\mathbb{X}, x, t), & x \in x_f \\ \\ \psi_s(\mathbb{X}, x, t), & x \in x_s \end{cases} \qquad (2.4)$$

where x_α denotes the position in the phase α. We can define a function as

$$H(x \in x_\alpha) = \begin{cases} 1 , & x \in x_\alpha \\ 0 , & x \notin x_\alpha \end{cases} \tag{2.5}$$

From the above definition, the Ψ values in each phase and in the entire domain can be defined as

$$\Psi_\alpha(X, x, t) \equiv \psi_\alpha H(x \in x_\alpha) \tag{2.6a}$$

$$\Psi(X, x, t) \equiv \psi_s H(x \in x_s) + \psi_f H(x \in x_f) \tag{2.6b}$$

The volume average for (2.6) is defined as

$$\hat{\psi}_\alpha \equiv \frac{1}{V}\int_V \Psi_\alpha \, dx = \frac{1}{V}\int_{V_\alpha} \psi_\alpha \, dx = \varepsilon_\alpha \tilde{\psi}_\alpha \tag{2.7a}$$

$$\hat{\psi} \equiv \frac{1}{V}\int_V \Psi \, dx = \hat{\psi}_f + \hat{\psi}_s \tag{2.7b}$$

where

$$\tilde{\psi}_\alpha = \frac{1}{V_\alpha}\int_{V_\alpha} \psi_\alpha \, dx \tag{2.8}$$

$$\varepsilon_\alpha = V_\alpha / V = \text{volume fraction of phase } \alpha. \tag{2.9}$$

The average, $\tilde{\Psi}_\alpha$, is an intrinsic value for the α-phase. V_α is the volume of α-phase in the domain and is generally a function of time.

Next we will find the averaging rules for the time derivative and the position derivative with respect to $\mathbf{X}$, which usually appear in the transport equation for a homogenous medium. Differentiating (2.7a) with respect to time under constant volume, V, we obtain

$$\frac{\partial \hat{\psi}_\alpha}{\partial t} = \frac{1}{V}\int_V \frac{\partial \psi_\alpha}{\partial t} H(x \in x_\alpha)\, dx + \frac{1}{V}\int_V \psi_\alpha \frac{\partial H(x \in x_\alpha)}{\partial t}\, dx \tag{2.10}$$

We can use the following equation

$$\begin{aligned}\frac{\partial H(x \in x_\alpha)}{\partial t} &= \frac{\partial}{\partial x_b} H(x \in x_\alpha) \cdot \frac{dx_b}{dt} \\ &= \delta(x_b - x)\,\mathbf{w} \cdot \mathbf{n}_\alpha\end{aligned} \tag{2.11}$$

where $\mathbf{x}_b$ denotes the position on the interface between the solid and fluid phases, $\mathbf{n}_\alpha$ is the unit normal vector pointing out of the interface from the α-phase, and $\mathbf{w}$ is the velocity of the interface. $\delta(x)$ is the Dirac delta function. The second term of (2.10) is rewritten with the aid of (2.11).

$$\begin{aligned}\frac{1}{V}\int_V \psi_\alpha \frac{\partial H(\mathbf{x} \in \mathbf{x}_\alpha)}{\partial t}\, d\mathbf{x} &= \frac{1}{V}\int_V \psi_\alpha\, \delta(\mathbf{x}_b - \mathbf{x})\, \mathbf{w} \cdot \mathbf{n}_\alpha\, d\mathbf{x} \\ &= \frac{1}{V}\int_A \psi_\alpha(\mathbf{x}_b, t)\, \mathbf{w} \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b\end{aligned} \tag{2.12}$$

where A is the interfacial area within the domain.

Introducing (2.12) into (2.10),

$$\frac{\partial \hat{\psi}_\alpha}{\partial t} = \widehat{\frac{\partial \psi_\alpha}{\partial t}} + \frac{1}{V}\int_A \psi_\alpha(\mathbf{x}_b, t)\, \mathbf{w} \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b \tag{2.13}$$

Writing (2.13) for each phase and adding, we obtain

$$\frac{\partial \hat{\psi}}{\partial t} = \widehat{\frac{\partial(\psi_s + \psi_f)}{\partial t}} + \frac{1}{V}\int_A [\psi_s(\mathbf{x}_b, t) - \psi_f(\mathbf{x}_b, t)]\, \mathbf{w} \cdot \mathbf{n}_s\, d\mathbf{x}_b \tag{2.14}$$

(2.13) and (2.14) are the averaging rules for the time derivative.

We can derive the averaging rule for the position derivative from a procedure similar to the above. The dynamical property Ψ can be written as

$$\psi(\mathbf{X}, x, t) = \psi(\mathbf{X} + x, t) \tag{2.15}$$

because Ψ is a point function with respect to the position vector. Ψ is rewritten with use of the definition function (2.5):

$$\Psi_\alpha(\mathbf{X}, x, t) = \psi_\alpha(\mathbf{X} + x, t)\, H[(\mathbf{X} + x) \in (\mathbf{X} + x)_\alpha]$$
$$\alpha = s \text{ or } f$$
$$\Psi(\mathbf{X}, x, t) = \Psi_f(\mathbf{X} + x, t) + \Psi_s(\mathbf{X} + x, t) \tag{2.16}$$

The value of Ψ_α integrated over the fixed domain is now differentiated with respect to the position vector $\mathbf{X}$:

$$\begin{aligned}
&\nabla_{\mathbf{X}} \int_V \psi_\alpha H[(\mathbf{X} + x) \in (\mathbf{X} + x)_\alpha]\, dx \\
&= \int_V \nabla_{\mathbf{X}} [\psi_\alpha H\{(x + \mathbf{X}) \in (x + \mathbf{X})_\alpha\}]\, dx \\
&= \int_V \nabla_x [\psi_\alpha H\{(x + \mathbf{X}) \in (x + \mathbf{X})_\alpha\}]\, dx \\
&= \int_V (\nabla_x \psi_\alpha) H\{(x + \mathbf{X}) \in (x + \mathbf{X})_\alpha\}]\, dx + \int_V \psi_\alpha \nabla_x H\{(x + \mathbf{X}) \in (x + \mathbf{X})_\alpha\}\, dx \\
&= \int_V (\nabla_x \psi_\alpha) H\{(x + \mathbf{X}) \in (x + \mathbf{X})_\alpha\}\, dx - \int_V \psi_\alpha \delta(x_b - x) n_\alpha\, dx \\
&= \int_{V_\alpha} \nabla_x \psi_\alpha\, dx - \int_A \psi_\alpha(x_b, t) n_\alpha\, dx_b
\end{aligned} \tag{2.17}$$

Dividing both hand-sides by V, the volume average can be obtained as

$$\nabla_x \hat{\psi}_\alpha = \widehat{\nabla_x \psi_\alpha} - \frac{1}{V}\int_A \psi_\alpha(x_b, t)\, n_\alpha\, dx_b \tag{2.18}$$

For Ψ,

$$\nabla_x \hat{\psi} = \widehat{\nabla_x \psi} - \frac{1}{V}\int_A \{\psi_s(x_b, t) - \psi_f(x_b, t)\}\, n_s\, dx_b \tag{2.19}$$

These two equations (S1) relate the derivative of the averaged value to the average of the derivative of Ψ. Henceforth, $\Psi_f(x_b, t)$ is simply designated as $\Psi_f(x_b)$.

As a first contribution to the transport equation, the material derivative of the dynamical property is given by

$$\frac{d}{dt}\int_V \psi_\alpha H(x \in x_\alpha)\, dx = \frac{d}{dt}\int_{V_\alpha(t)} \psi_\alpha\, dx$$

$$= \int_{V_\alpha} \frac{\partial \psi_\alpha}{\partial t}\, dx + \int_{S_\alpha} \psi_\alpha v_\alpha \cdot n_\alpha\, dS + \int_A \psi_\alpha(x_b)\, v_\alpha(x_b) \cdot n_\alpha\, dx_b \tag{2.20}$$

where S_α is the exit or entrance surface area enclosing the phase α in the domain. $v_\alpha(x_b)$ is the velocity for α-phase at the interface. From (2.20) and (2.13), we can obtain the general transport equation.

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$$\frac{d}{dt}\int_V \psi_\alpha H(\mathbf{x}\in \mathbf{x}_\alpha)\,d\mathbf{x} = \frac{\partial}{\partial t}\int_V \psi_\alpha H(\mathbf{x}\in \mathbf{x}_\alpha)\,d\mathbf{x} + \int_A \psi_\alpha(\mathbf{x}_b)\left[\mathbf{v}_\alpha(\mathbf{x}_b)-\mathbf{w}\right]\cdot \mathbf{n}_\alpha\,d\mathbf{x}_b + \int_{S_\alpha} \psi_\alpha \mathbf{v}_\alpha \cdot \mathbf{n}_\alpha\,dS$$

= birth rate of Ψ_α in the α-phase (2.21)

The left hand side of this equation is equal to the birth rate of Ψ in α-phase of the domain. From the Gauss integral theorem, this equation can be rewritten as

$$\frac{\partial}{\partial t}\int_V \psi_\alpha H(\mathbf{x}\in \mathbf{x}_\alpha)\,d\mathbf{x} + \int_{V_\alpha} \nabla_{\mathbf{x}}\cdot(\psi_\alpha \mathbf{v}_\alpha)\,d\mathbf{x} - \int_A \psi_\alpha(\mathbf{x}_b)\,\mathbf{w}\cdot \mathbf{n}_\alpha\,d\mathbf{x}_b$$

= birth rate of Ψ_α in α-phase (2.22)

For the whole value of Ψ within the domain

$$\frac{d}{dt}\int_V \Psi\,d\mathbf{x} = \frac{\partial}{\partial t}\int_V \Psi\,d\mathbf{x} + \int_V \nabla_{\mathbf{x}}\cdot(\psi_s \mathbf{v}_s + \psi_f \mathbf{v}_f)\,d\mathbf{x} - \int_A \{\psi_s(\mathbf{x}_b) - \psi_f(\mathbf{x}_b)\}\,\mathbf{w}\cdot \mathbf{n}_s\,d\mathbf{x}_b$$

= birth rate of Ψ in the whole space
of the domain (2.23)

For a mixture of m chemical species, consider m continuous bodies, each of which is visualized by the region it occupies in the physical space. The bodies are allowed to occupy a common portion of the physical space. In this case, the material derivative of the dynamical property for the k-th chemical species is written by

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$$\frac{d}{dt}\int_V \psi_{k\alpha} H(\mathbf{x} \in \mathcal{X}_\alpha)\, d\mathbf{x} = \frac{d}{dt}\int_{V_\alpha} \psi_{k\alpha}\, d\mathbf{x}$$

$$= \int_{V_\alpha} \frac{\partial \psi_{k\alpha}}{\partial t}\, d\mathbf{x} + \int_{S_\alpha} \psi_{k\alpha} \mathbf{v}_{k\alpha} \cdot \mathbf{n}_\alpha\, dS + \int_A \psi_{k\alpha} \mathbf{v}_{k\alpha}(\mathbf{x}_b) \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b \tag{2.24}$$

where subscript k denotes the value for k-th species. Introduction of (2.13) into (2.24) gives the transport equation for the k-th species

$$\frac{\partial}{\partial t}\int_{V_\alpha} \psi_{k\alpha}\, d\mathbf{x} + \int_{V_\alpha} \nabla_{\mathbf{x}} \cdot (\psi_{k\alpha} \mathbf{v}_{k\alpha})\, d\mathbf{x} - \int_A \psi_{k\alpha}\, \mathbf{w} \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b$$

$$= \text{birth rate of k-th species in } \alpha\text{-phase} \equiv R_{k\alpha} \tag{2.25}$$

(2.22) and (2.25) can be rewritten with help of (2.18).

$$\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{\psi}_\alpha) + \nabla_{\mathbf{x}} \cdot (\widetilde{\psi_\alpha \mathbf{v}_\alpha}\, \varepsilon_\alpha) + \frac{1}{V}\int_A \psi_\alpha(\mathbf{x}_\alpha)[\mathbf{v}_\alpha(\mathbf{x}_b) - \mathbf{w}] \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b$$

$$= \text{birth rate of } \Psi_\alpha \text{ in the } \alpha\text{-phase} \tag{2.26}$$

$$\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{\psi}_{k\alpha}) + \nabla_{\mathbf{x}} \cdot (\widetilde{\psi_{k\alpha} \mathbf{v}_{k\alpha}}\, \varepsilon_\alpha) + \frac{1}{V}\int_A \psi_{k\alpha}(\mathbf{x}_b)[\mathbf{v}_{k\alpha}(\mathbf{x}_b) - \mathbf{w}] \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b$$

$$= \text{birth rate of } \Psi_{k\alpha} \text{ in the } \alpha\text{-phase} \tag{2.27}$$

In the above derivation we have used the relation:

$$\frac{1}{V}\int_V \psi_\alpha H(\mathbf{x} \in \mathcal{X}_\alpha)\, d\mathbf{x} = \frac{1}{V}\int_{V_\alpha} \psi_\alpha\, d\mathbf{x} = \varepsilon_\alpha \tilde{\psi}_\alpha \tag{2.28}$$

(2.27) can be rewritten by introducing the relative flux $\mathring{\mathbf{j}}_{k\alpha}$ of the property Ψ of k-th species within the α-phase, which moves at a velocity $(\mathbf{v}_{k\alpha} - \mathbf{v}_\alpha)$ relative to the α-phase, i.e.,

$$\mathring{\mathbf{j}}_{k\alpha} = \psi_{k\alpha}(\mathbf{v}_{k\alpha} - \mathbf{v}_\alpha) \tag{2.29}$$

If the solid phase is stationary, $\mathring{j}_{k\alpha}$ measured at the solid-fluid interface is the net flux across the interface. Substituting (2.29) into (2.27) results in

$$\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{\psi}_{k\alpha}) + \nabla_X \cdot (\varepsilon_\alpha \widetilde{\psi_{k\alpha} v_\alpha})$$

$$+ \frac{1}{V}\int_A [\, \mathring{j}_{k\alpha}(x_b) + \psi_{k\alpha}(x_b)\{v_\alpha(x_b) - w\}] \cdot n_\alpha \, dx_b$$

$$= -\nabla_X \cdot (\varepsilon_\alpha \tilde{\mathring{j}}_{k\alpha}) + \text{birth rate of } \Psi_{k\alpha} \text{ in } \alpha\text{-phase} \qquad (2.30)$$

(2.26) and (2.30) are the general transport equations for a single body and for multi-bodies of a mixture, respectively.

These equations can also be obtained by integrating the single-phase transport equation over the volume domain and by using the averaging rules, (2.13) and (2.18).

2.2.2. The Equation of continuity

In case Ψ is selected as the density of the α-phase, the birth rate of Ψ becomes zero, because radioactive decay contributes negligibly to a mass change. Then, from (2.26),

$$\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{\rho}_\alpha) + \nabla_X \cdot (\varepsilon_\alpha \widetilde{\rho_\alpha v_\alpha}) + \frac{1}{V}\int_A \rho_\alpha (v_\alpha - w) \cdot n_\alpha \, dx_b = 0 \qquad (2.31)$$

This equation is the mass-conservation equation for the α-phase.

The last term denotes the rate of change of the α-phase to another phase. The sum of (2.31) with respect to both solid and fluid phases yields

$$\frac{\partial \hat{\rho}}{\partial t} + \nabla_X \cdot (\widehat{\rho v}) = 0 \tag{2.32}$$

where

$$\hat{\rho} = \varepsilon_s \hat{\rho}_s + \varepsilon_f \hat{\rho}_f \ , \ \widehat{\rho v} = \varepsilon_s \widehat{\rho_s v_s} + \varepsilon_f \widehat{\rho_f v_f}$$

and we have used the continuity equation at the interface:

$$\int_A \rho_f [v_f(x_b) - w] \cdot n_f \, dx_b + \int_A \rho_s [v_s(x_b) - w] \cdot n_s \, dx_b = 0$$

Next, we will derive the continuity equation of the k-th chemical species in the mixture. In this case, the general transport equation (2.30) for the mixture should be used. Selecting the molar concentration C_{k_α} as Ψ_{k_α}, equation (2.30) can be written as

$$\begin{aligned} &\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{C}_{k\alpha}) + \nabla_X \cdot (\varepsilon_\alpha \widetilde{C_{k\alpha} v_\alpha}) + \nabla_X \cdot (\varepsilon_\alpha \tilde{\mathring{j}}_{k\alpha}) \\ &+ \frac{1}{V} \int_A [\mathring{j}_{k\alpha}(x_b) + C_{k\alpha}(x_b) \{ v_\alpha(x_b) - w \} \cdot n_\alpha \, dx_b \\ &= \hat{R}_{k\alpha} \end{aligned} \tag{2.33}$$

where

$$\hat{R}_{k\alpha} = \frac{1}{V} \int_{V_\alpha} R_{k\alpha} \, dx$$

and $R_{k\alpha}$ is the birth rate of the k-th species in the α-phase.

The second term in (2.33) represents convective transport, the third term represents diffusive transport, and the fourth term represents the net flow of the k-th species across the solid-fluid interface. Summing over k, (2.33) is reduced to (2.31), which is the continuity equation for total mass in the α-phase.

We denote the deviations of $C_{k\alpha}$ and $\mathbf{v}_\alpha$ from their averaged values in the domain as $\mathring{C}_{k\alpha}$ and $\mathring{\mathbf{v}}_\alpha$, respectively. Then,

$$C_{k\alpha} = \tilde{C}_{k\alpha} + \mathring{C}_{k\alpha}, \quad \mathbf{v}_\alpha = \tilde{\mathbf{v}}_\alpha + \mathring{\mathbf{v}}_\alpha \tag{2.34}$$

Each deviation is reduced to zero if averaged:

$$\int_{V_\alpha} \mathring{C}_{k\alpha}\, d\mathbf{x} = \int_{V_\alpha} \mathring{\mathbf{v}}_\alpha\, d\mathbf{x} = 0 \tag{2.35}$$

Using (2.34) and (2.35),

$$\nabla_X \cdot (\varepsilon_\alpha \widetilde{C_{k\alpha}\mathbf{v}_\alpha}) = \nabla_X \cdot (\varepsilon_\alpha \tilde{C}_{k\alpha}\tilde{\mathbf{v}}_\alpha) + \nabla_X \cdot (\varepsilon_\alpha \widetilde{\mathring{\mathbf{v}}_\alpha \mathring{C}_{k\alpha}}) \tag{2.36}$$

Introduction of (2.36) into (2.33) yields

$$\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{C}_{k\alpha}) + \nabla_X \cdot (\varepsilon_\alpha \tilde{C}_{k\alpha}\tilde{\mathbf{v}}_\alpha) + \nabla_X \cdot [\varepsilon_\alpha \tilde{\mathbf{j}}_{k\alpha} + \varepsilon_\alpha \widetilde{\mathring{C}_{k\alpha}\mathring{\mathbf{v}}_\alpha}]$$
$$+ \frac{1}{V}\int_A [\mathbf{j}_{k\alpha}(\mathbf{x}_b) + C_{k\alpha}(\mathbf{x}_b)\{\mathbf{v}_\alpha(\mathbf{x}_b) - \mathbf{w}\}] \cdot \mathbf{n}_\alpha\, d\mathbf{x}_b$$
$$= \frac{1}{V}\int_{V_\alpha} R_{k\alpha}\, d\mathbf{x} \tag{2.37}$$

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Here the mass flux $\mathring{j}_{k\alpha}$ is the net molecular flux for the k-th chemical species relative to the motion of the α-phase. In this work, the general constitutive relations governing $\mathring{j}_{k\alpha}$ will not be treated. Instead, we assume Fick's law of diffusion:

$$\mathring{j}_{k\alpha} = - D_{k\alpha} \nabla_{x} C_{k\alpha} \tag{2.38}$$

With the help of (2.38), (2.37) is reduced to

$$\begin{aligned} & \frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{C}_{k\alpha}) + \nabla_{\mathbf{x}} \cdot (\varepsilon_\alpha \tilde{C}_{k\alpha} \tilde{v}_\alpha) + \nabla_{\mathbf{x}} \cdot (\varepsilon_\alpha \widetilde{\mathring{v}_\alpha \mathring{C}_{k\alpha}}) \\ & - \nabla_{\mathbf{x}} \cdot \left[D_{k\alpha} \nabla_{\mathbf{x}} (\varepsilon_\alpha \tilde{C}_{k\alpha}) \right] - \nabla_{\mathbf{x}} \left[\frac{1}{V} \int_A D_{k\alpha} C_{k\alpha} n_\alpha \, d\mathbf{x}_b \right] \\ & + \frac{1}{V} \int_A \left[\mathring{j}_{k\alpha}(\mathbf{x}_b) + C_{k\alpha}(\mathbf{x}_b) \{ v_\alpha(\mathbf{x}_b) - w \} \right] \cdot n_\alpha \, d\mathbf{x}_b \\ & = \varepsilon_\alpha \tilde{R}_{k\alpha} \end{aligned} \tag{2.39}$$

where the spatial change of $D_{k\alpha}$ in the averaging domain was neglected. The fifth term in the left hand side of (2.39) means a kind of the "tortuosity" effect (S2,G1) which yields the reduction of diffusion flow.

We are primarily concerned here with the migration of radionuclides, which may exist in the ground water in various

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chemical forms. Let us consider a decay chain such that the i-th nuclide species within a given phase exists in m different chemical forms, each form k containing S_i^k atoms of nuclide i per molecule.

The birth rate of the i-th nuclide takes the form

$$\sum_{k=1}^{m} S_i^k \tilde{R}_{k\alpha} = \lambda_{i-1} \sum_{k=1}^{m} S_{i-1}^k \tilde{C}_{k\alpha} - \lambda_i \sum_{k=1}^{m} S_i^k \tilde{C}_{k\alpha} + \tilde{f}_{i\alpha}(\mathbb{X}, t) \qquad (2.40)$$

where $\tilde{f}_{i\alpha}(\mathbb{X}, t)$ is the source term of the i-th nuclide in the α-phase at the position $\mathbb{X}$ and at time t. If we multiply (2.39) by S_i^k and add the resultant equation with respect to k, it follows that

$$\begin{aligned}
&\frac{\partial}{\partial t}(\varepsilon_\alpha \tilde{N}_{i\alpha}) + \nabla_{\mathbb{X}} \cdot (\varepsilon_\alpha \tilde{\mathbf{v}}_\alpha \tilde{N}_{i\alpha}) \\
&+ \nabla_{\mathbb{X}} \cdot \Big\{ \varepsilon_\alpha \widetilde{\mathring{\mathbf{v}}_\alpha \mathring{N}_{i\alpha}} - \sum_{k=1}^{m} S_i^k D_{k\alpha} \nabla_{\mathbb{X}} (\varepsilon_\alpha \tilde{C}_{k\alpha}) \Big\} \\
&- \nabla_{\mathbb{X}} \cdot \Big\{ \frac{1}{V} \int_A \sum_{k=1}^{m} S_i^k D_{k\alpha} C_{k\alpha} \mathbf{n}_\alpha \, d\mathbf{x}_b \Big\} \\
&+ \frac{1}{V} \int_A \Big[\sum_{k=1}^{m} S_i^k \mathbf{j}_{k\alpha}(\mathbf{x}_b) + N_{i\alpha}(\mathbf{x}_b) \{ \mathbf{v}_\alpha(\mathbf{x}_b) - \mathbf{w} \} \Big] \cdot \mathbf{n}_\alpha \, d\mathbf{x}_b \\
&= (\lambda_{i-1} \tilde{N}_{i-1,\alpha} - \lambda_i \tilde{N}_{i,\alpha}) \varepsilon_\alpha + \varepsilon_\alpha \tilde{f}_{i\alpha}(\mathbb{X}, t)
\end{aligned}$$

$$(2.41)$$

where $\tilde{N}_{i\alpha}$ is the atomic concentration of the i-th nuclide,

$$\tilde{N}_{i\alpha} = \sum_{k=1}^{m} S_i^k \tilde{C}_{k\alpha}$$

and λ_i is the decay constant for i-th nuclide. This equation is the balance for i-th nuclide and is a general transport equation governing the migration of nuclide through a geological medium, to be further developed in the following section.

2.3 Governing Equations for Radionuclide Migration

In this section we simplify the transport equation (2.41), and we develop the governing equation for the nuclide migration process which will be the subject of analyses described in the later chapters.

Because of the complexity of the geological media, we postulate that the radionuclides are transported by ground water moving in a deterministic flow path with a flow velocity given a priori. In this case the transport equations for chemical species and radionuclides are given by (2.39) and (2.41).

2.3.1 Simplifying assumptions

We postulate the following assumptions:

1. The fluctuation terms for the convective transport

in the water phase, $\varepsilon_f \widetilde{\mathring{v}_f \mathring{C}_{kf}}$ and $\varepsilon_f \widetilde{\mathring{v}_f \mathring{N}_i}$, are described as a stochastic random process. This assumption is formulated as

$$\varepsilon_f \widetilde{\mathring{v}_f \mathring{C}_{kf}} = -\mathbb{D}_{df} \cdot \nabla_X (\varepsilon_f \widetilde{C}_{kf}), \quad \varepsilon_f \widetilde{\mathring{v}_f \mathring{N}_{if}} = -\mathbb{D}_{df} \cdot \nabla_X (\varepsilon_f \widetilde{N}_{if}) \qquad (2.42)$$

where $\mathbb{D}_{df}$ is the dispersion tensor.

The convective term within the solid phase is assumed to play no role in the transport of radionuclides.

2. The molecular diffusion coefficients in the water phase are assumed to have a value D_{mf} for each chemical species. This assumption leads to

$$\frac{1}{V} \nabla_X \int_{V_f} \sum_k D_{kf} C_{kf} S_i^k \, dX = \nabla_X (\varepsilon_f D_{mf} \widetilde{N}_{if}) \qquad (2.43)$$

This assumption is reasonable, because the molecular diffusion coefficients of several chemical species are of the same order and because transport by molecular diffusion process is less important than transport by dispersive diffusion, as treated in assumption 1.

The molecular diffusion in the solid phase is also assumed to be independent of chemical species, so that

$$D_{k,s} = D_s \qquad (2.44)$$

3. The tortuosity term is neglected. Because this term usually reduces the flow due to the molecular diffusion, this assumption is conservative for nuclide migration. Scheidegger (S1) has proposed a dispersion tensor in which both convective fluctuation and tortuosity terms are included.

4. The motion of the interface is negligible, and the mass-averaged velocities of the two phases across the interface are assumed to be zero.

$$w = v_f(x_b) = v_s(x_b) = 0 \qquad (2.45)$$

This assumption is reasonable when there is no phase change between the water and the solid phases.

5. The bulk motion of the solid phase is neglected.

$$v_s = 0 \qquad (2.46)$$

The transport equations for chemical species and for radionuclies are then given by

$$\frac{\partial}{\partial t}(\varepsilon_f \tilde{C}_{kf}) + \nabla_X \cdot (\varepsilon_f \tilde{C}_{kf} \tilde{v}_f)$$

$$- \nabla_X \cdot \left[\mathbb{D}_f \cdot \nabla_X (\varepsilon_f \tilde{C}_{kf}) \right] + \frac{1}{V} \int_A \mathbf{j}_k(x_b) \cdot n_f \, dx$$

$$= \varepsilon_f \tilde{R}_{kf}$$

(2.47a)

$$\frac{\partial}{\partial t}(\varepsilon_s \tilde{C}_{ks}) - \nabla_X \cdot \left[D_s \nabla_X (\varepsilon_s \tilde{C}_{ks}) \right]$$

$$- \frac{1}{V} \int_A \mathbf{j}_k(x_b) \cdot n_f \, dx_b = \varepsilon_s \tilde{R}_{ks}$$

(2.47b)

$$\frac{\partial}{\partial t}(\varepsilon_f \tilde{N}_{if}) + \nabla_X \cdot (\varepsilon_f \tilde{v}_f \tilde{N}_{if})$$

$$- \nabla_X \cdot \left[\mathbb{D}_f \cdot \nabla_X (\varepsilon_f \tilde{N}_{if}) \right] + \frac{1}{V} \int_A \sum_k S_i^k \mathbf{j}_k(x_b) \cdot n_f \, dx_b$$

$$= \varepsilon_f \left[\lambda_{i-1} \tilde{N}_{i-1,f} - \lambda_i \tilde{N}_{i,f} \right] + \varepsilon_f \tilde{f}_{if}(X, t)$$

(2.48a)

$$\frac{\partial}{\partial t}(\varepsilon_s \tilde{N}_{is}) - \nabla_X \cdot \left[D_s \nabla_X (\varepsilon_s \tilde{N}_{is}) \right]$$

$$- \frac{1}{V} \int_A \sum_k S_i^k \mathbf{j}_k(x_b) \cdot n_f \, dx_b$$

$$= \varepsilon_s \left(\lambda_{i-1} \tilde{N}_{i-1,s} - \lambda_i \tilde{N}_{i,s} \right) + \varepsilon_s \tilde{f}_{is}(X, t)$$

(2.48b)

where

$$\mathbb{D}_f = \mathbb{D}_{df} + D_{mf}\mathbb{1}$$

and $\mathbb{1}$ is the unit tensor.

2.3.2 Transport with local chemical-sorptive equilibria

At this stage, we encounter a problem concerned with the rate processes involving in the migration of radionuclides through geological media, e.g., sorption and chemical reaction processes.

A geologic medium suitable for radioactive waste isolation must have a water velocity small enough to isolate the nuclide from biosphere. With such slow transport, one might expect that local thermochemical equilibrium will exist within each phase and between water and solid phases. To examine the migration features under the limiting conditions that chemical and sorption processes are locally equilibrated, we further assume that

6. At each position $\mathbb{X}$ the sorption equilibrium is described as a linear relation:

$$\tilde{C}_{ks} = k_{D,k}\,\tilde{C}_{kf} \qquad (2.49)$$

where $k_{D,k}$ is a distribution coefficient for the k-th chemical species.

7. Different chemical forms of a given chemical element are in equilibrium within a given phase α, so that

$$\tilde{C}_{k\alpha} k^{t}_{R,k\alpha} = \tilde{C}_{t\alpha} \qquad (2.50)$$

where $\tilde{C}_{t\alpha}$ is the concentration of the t-th chemical species involving the nuclide i and $k^{t}_{R,k\alpha}$ denotes an equilibrium constant.

From (2.49) and (2.50), the overall sorption equilibrium is expressed in terms of nuclide concentrations $\tilde{N}_{if}$ and $\tilde{N}_{is}$.

$$\begin{aligned}\tilde{N}_{is} &= \left[\sum_{k} \left(k_{D,k} S_i^{k} / k^{t}_{R,ks} \right) / \sum_{k} \left(S_i^{k} / k^{t}_{R,ks} \right) \right] \tilde{N}_{if} \\ &\equiv K_{D,i} \tilde{N}_{if} \end{aligned} \qquad (2.51)$$

where $K_{D,i}$ is the overall distribution coefficient for the i-th nuclide, i.e.,

$$K_{d,i} = \frac{\text{concentration of nuclide i in the solid}}{\text{concentration of nuclide i in the liquid}}$$

The concentrations $\tilde{N}_i$ are expressed in consistent units (e.g., moles/liter), so that K_{di} is dimensionless.

Summing up (2.48) with respect to phase and using (2.51) the transport equation governing the i-th nuclide migration is given by the single equation:

$$\begin{aligned}&\frac{\partial}{\partial t}(\varepsilon_f K_i \tilde{N}_{if}) + \nabla_X \cdot \left[\left\{ \varepsilon_f \mathbb{V}_f - \mathbb{D}_f \cdot \nabla_X \varepsilon_f - D_s \nabla_X (K_{D,i}\, \varepsilon_s) \right\} \tilde{N}_{if} \right] \\ &- \nabla_X \cdot \left[(\mathbb{D}_f \varepsilon_f + D_s K_{D,i} \varepsilon_s \mathbb{1}) \cdot \nabla_X \tilde{N}_{if} \right] \\ &= \varepsilon_f K_{i-1} \lambda_{i-1} \tilde{N}_{i-1,f} - \varepsilon_f K_i \lambda_i \tilde{N}_{if} \\ &+ \varepsilon_f \tilde{f}_{if}(X,t) + \varepsilon_s \tilde{F}_{is}(X,t)\end{aligned} \qquad (2.52)$$

where

$$K_i = 1 + \varepsilon_s K_{D,i} / \varepsilon_f \qquad (2.53)$$

2.3.3 One-Dimensional transport with local chemical-sorptive equilibria

For simplicity, we also assume:

8. The deterministic water pathway is one-dimensional and the nuclide flow out of the pathway due to transverse diffusion can be neglected. The geologic medium is isotropic.

(2.52) is simplified by this assumption to

$$\begin{aligned}&\frac{\partial}{\partial t}(\varepsilon_f K_i \tilde{N}_{if}) + \frac{\partial}{\partial z}(\varepsilon_f v_f A_i \tilde{N}_{if}) \\ &\qquad - \frac{\partial}{\partial z}\left(D_f \varepsilon_f B_i \frac{\partial}{\partial z} \tilde{N}_{if} \right) \\ &= \varepsilon_f K_{i-1} \lambda_{i-1} \tilde{N}_{i-1,f} - \varepsilon_f K_i \lambda_i \tilde{N}_{if} \\ &\quad + \varepsilon_f \tilde{f}_{if} + \varepsilon_s \tilde{f}_{is} \end{aligned} \tag{2.54}$$

where

$$A_i = 1 - \frac{D_f(\partial \varepsilon_f/\partial z) + D_s(\partial K_{D,i} \varepsilon_s/\partial z)}{v_f \varepsilon_f}$$

$$B_i = 1 + \frac{D_s \varepsilon_s K_{D,i}}{D_f \varepsilon_f} \tag{2.55}$$

and z is the distance in the direction of water flow.

D_f is the dispersion coefficient in fluid phase, and v_f is the velocity of the water phase in z-direction. If diffusion within solid phase plays no role in the transport of the nuclide, then B_i becomes unity.

When ε_f, K_{di}, K_i and v_f are constant in space and time the above equation is reduced to

$$\begin{aligned}&\frac{\partial \tilde{N}_{if}}{\partial t} + v_f \frac{\partial \tilde{N}_{if}}{\partial z} - D_i \frac{\partial^2 \tilde{N}_{if}}{\partial z^2} \\ &= K_{i-1} \lambda_{i-1} \tilde{N}_{i-1,f} - K_i \lambda_i \tilde{N}_{if} + \tilde{f}_{if} + \tilde{f}_{is} \frac{\varepsilon_s}{\varepsilon_f}\end{aligned} \tag{2.56}$$

where $D_i = D_f B_i$.

Equations (2.54) and (2.56) are the fundamental equations to be solved in the succeeding chapters of this report.

2.3.4 One-dimensional transport without local chemical equilibrium

For future studies it is helpful to formulate the governing equations of the nuclide migration in a more general form involving the effects of non-equilibrium sorption and chemical reaction processes.

First, we consider the problem evaluating the mass flux across the interface $\mathring{j}_k(x_b)$. The mass transfer between two phases arises from sorption processes and information concerning the sorption mechanism is necessary for a strict formulation of this flux across the interface. Sorption occurs via three steps; diffusion within the water phase, diffusion through fine pores in the solid phase, and adsorption or ion-exchange. We can postulate some limiting cases, e.g., a process controlled by diffusion or a process controlled by the rate of the reactions.

In the case of diffusion controlling, we can formulate $\dot{j}_k(x_b)$ by the following equation:

$$\dot{j}_k(x_b) = k_{M,k}\left(\tilde{C}_{kf} - \frac{1}{K_{D,k}}\tilde{C}_{ks}\right)n_f \qquad (2.57)$$

where $k_{M,k}$ is the overall mass-transfer coefficient for the k-th species which includes the effects of the diffusional resistances in both phases. This equation is not strictly correct but holds approximately if sorption process is in a near-equilibrium state.

When mass transfer is controlled by reaction rates, the mass flux across the interface is obtained from an assumed first-order reaction rate with respect to the k-th species.

$$\dot{j}_k(x_b) = k_{A,k}\left[\tilde{C}_{kf}\tilde{C}_{vs} - \frac{1}{K_A}\tilde{C}_{mf}\tilde{C}_{ks}\right]\frac{V_s}{A}n_f \qquad (2.58)$$

where $k_{A,k}$ is the reaction-rate constant. In the above equation, $\tilde{C}_{vs}$ and $\tilde{C}_{mf}$ are some characteristic concentrations. For an ion-exchange reaction process,

$\tilde{C}_{vs}$ = the concentration of the ion-exchangeable species, except for the k-th species, in the solid phase

$\tilde{C}_{mf}$ = the concentration of the ion-exchangeable species, except for the k-th species, in the liquid phase (2.59)

For an adsorption process, we can select

$\tilde{C}_{vs}$ = the vacancy site concentration in the solid phase

$\tilde{C}_{mf} = 1$

(2.60)

There may exist competing sorptive species in each phase, the concentrations of which may be greater than that of the radionuclide. It is, therefore, reasonable to assume that the $\tilde{C}_{vs}$ and the $\tilde{C}_{mf}$ are constant, independent of the concentration of k-th species. In this case, (2.58) is formally reduced to (2.57).

The integral terms in (2.47) and (2.48) are rewritten for the mass-transfer rate of (2.57) as

$$\frac{1}{V}\int_A \mathring{j}_k(x_b)\cdot n_f \, dx_b = k_{M,k}\, a\left(\tilde{C}_{kf} - \frac{1}{k_{D,k}}\tilde{C}_{kS}\right) \tag{2.61}$$

$$\frac{1}{V}\sum_k \int_A \mathring{j}_k(x_b)\cdot n_f S_i^k \, dx_b = \sum_k k_{M,k}\, a\left(\tilde{C}_{k,f} - \frac{1}{k_{D,k}}\tilde{C}_{k,s}\right) S_i^k \tag{2.62}$$

where a is the interfacial area per unit volume of the geologic medium. In general, the values of $k_{M,k}$ and $k_{D,k}$ are dependent upon the chemical form. It is possible that a given nuclide may exist in more than one chemical form, even within a given phase. In such cases (2.62) cannot be evaluated in terms of the concentration of nuclide i alone. The prediction of the radionuclide migration then requires the simultaneous solution of the transport equations for chemical species. If the radionuclide is in a single chemical form within a given phase, (2.62) can be simplified as

$$\frac{1}{V}\int_A \sum_k \mathring{j}_k(x_b)\cdot n_f S_i^k \, dx_b = k_{M,i}\, a\left(\tilde{N}_{if} - \frac{1}{k_{D,i}}\tilde{N}_{iS}\right) \qquad (2.63)$$

In this case, the simultaneous transport equations for the nuclide (2.48) can be solved directly with help of (2.63)

Next we consider the possibility that a radionuclide may exist in more than one chemical form within a phase. As explained previously, the sorbing process is strongly influenced by the chemical form of the nuclide. If a certain chemical species is difficult to sorb compared with the other chemical species, the chemical species which sorbs less easily moves more rapidly through the geological medium. Some experimental results suggest the importance of different chemical forms upon the migration Pu and Am nuclide (F3). This problem is in principle evaluated by solving the simultaneous transport equations for the different chemical forms of a nuclide with appropriate chemical-reaction rates and mass-transfer rates as discussed previously.

Another problem concerned with chemical reaction is the mineralization reaction, i.e., the nuclide may react irreversibly with some component in the soil. The assumption of reversible sorption, i.e., ignoring irreversible mineralization, may always be conservative, but the effect of mineralization upon transport requires elucidation (B5).

This mineralization process can also be considered as the problem concerned with different chemical forms of a radionuclide in the solid phase. We consider two chemical forms, one is desorbable and the other is impossible to desorb. In this case, the transport equations for the solid phase can be described from (2.47):

$$\begin{aligned} \frac{\partial}{\partial t}(\varepsilon_S \tilde{C}_{1S}) - \nabla_x \cdot [D_S \nabla_x (\varepsilon_S \tilde{C}_{1S})] - \frac{1}{V}\int_A \mathbf{j}_1(x_b) \cdot n_f \, dx_b \\ = \varepsilon_S \tilde{R}_{1S} \\ \frac{\partial}{\partial t}(\varepsilon_S \tilde{C}_{2S}) - \nabla_x \cdot [D_S \nabla_x (\varepsilon_S \tilde{C}_{2S})] = \varepsilon_S \tilde{R}_{2S} \end{aligned} \qquad (2.64)$$

where $\mathcal{C}_{1s}$ and $\mathcal{C}_{2s}$ are the concentrations of the chemical forms of the desorbable nuclide, i_1, and of the nuclide impossible to desorb, i_2. For the nuclide concentrations, the above equations can be rewritten as

$$\frac{\partial}{\partial t}(\varepsilon_s \tilde{N}_{i_1 s}) - \nabla_x \cdot [D_s \nabla_x (\varepsilon_s \tilde{N}_{i_1 s})]$$

$$- \frac{1}{V}\int_A S_i^1 j_1(\mathbf{x}_b) \cdot \mathbf{n}_f \, d\mathbf{x}_b = \varepsilon_s S_i^1 \tilde{R}_{1s}$$

$$\frac{\partial}{\partial t}(\varepsilon_s \tilde{N}_{i_2 s}) - \nabla_x \cdot [D_s \nabla_x \varepsilon_s \tilde{N}_{i_2 s}] = \varepsilon_s S_i^2 \tilde{R}_{2s}$$

(2.65)

where

$$S_i^1 \tilde{R}_{i_1 s} = S_{i_1}^{i-1} \lambda_{i-1} \tilde{N}_{i-1\,s} - \lambda_i \tilde{N}_{i_1 s} - k_i \tilde{N}_{i_1 s}$$

$$S_i^2 \tilde{R}_{i_2 s} = S_{i_2}^{i-1} \lambda_{i-1} \tilde{N}_{i-1\,s} - \lambda_i \tilde{N}_{i_2 s} + k_i \tilde{N}_{i_1 s}$$

(2.66)

The last term of the above equation, $k_i N_{i_1 s}$, denotes the rate of mineralization, where we have assumed that the rate can be expressed by a first order chemical reaction. $S_{i_1}^{i-1}$ is the fraction of the (i-1)-th nuclide that is transformed by decay to the desorbable chemical species of the i-th nuclide. In this case, (2.65) and (2.48a) are the fundamental equations to be solved, with the interfacial transfer rate given by:

$$\frac{1}{V}\int_A \sum_k \mathbf{j}_k(x_b)\cdot n_f S_i^k \, dx_b = k_{Mi}\, a\left(\tilde{N}_{if} - \frac{1}{k_{d,i}}\tilde{N}_{i,s}\right) \qquad (2.67)$$

Solution of these non-equilibria forms of the transport equation will be a subject of future studies.

2.3.5 Transport along interfaces

The fundamental premise in developing the transport equations in this chapter has been that the geological medium is composed only of solid and liquid phases. Transport of radionuclide along the interface between two phases has been neglected, although this might become important for strongly sorbing species and with very low water velocities. The treatment of inter facial transport is left for future studies.

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3. NUCLIDE RELEASE MODES

3.1 Introduction

The analyses herein provide for various possible modes by which radionuclides can be released at the repository (H5). These release modes affect the boundary conditions at the repository, and they thereby affect the migration features of the radionuclides through the geologic media. In Sections 3.2 mathematical models are developed for the various release modes assumed for this study. In Section 3.3 the boundary condition and source term for transport equation are discussed. In Section 3.4 we present a general superposition relation for constructing the "band release" solutions.

3.2 Nuclide Release Modes

3.2.1 Constant rate of dissolution of waste material

Consider a radioactive waste consisting of radionuclides dispersed through a solid matrix. It is assumed that:

(a) a constant amount of the total waste material dissolves into ground water per unit time,

(b) all of the waste material begins to dissolve at $t=0$. Dissolution is complete within a leach time T, and

(c) all radionuclides contained within the waste material go into solution when the waste matrix dissolves.

Assumption (b) results in a "band release" of radionuclides into the ground water.

For an initial amount M_T^0 of total waste material, the rate of dissolution $\dot{M}_T$ of the total material is

$$\dot{M}_T = \frac{M^0}{T} \qquad (0 \leq t \leq T) \qquad (3.1)$$

The rate of dissolution $M_i(t)$ of radionuclide <u>i</u> in the waste material is then

$$\dot{M}_i(t) = n_i(t)\dot{M}_T \qquad (3.2)$$

where $n_i(t)$ is the amount of nuclide <u>i</u> per unit amount of waste material.

If the radionuclides dissolve uniformly into water flowing at a volumetric flow rate Q past the waste material, and neglecting the diffusional transport of the dissolved radionuclides in the vicinity of the waste material, the concentration $N_i(t)$ of a dissolved radionuclide at this location $(z = 0)$ is

$$N_i(t) = \frac{\dot{M}_i(t)}{Q} \quad , \quad (0 \leq t \leq T) \qquad (3.3)$$

Or, using (3.1) and (3.2):

$$N_i(t) = \frac{n_i(t) M^o{}_T}{QT} \qquad (0 \leq t \leq T) \quad (3.4)$$

The waste concentration $n_1(t)$ of a mother nuclide ($\lambda_o = 0$) in a decay chain is related to its initial waste concentration n_1^o by

$$N_1(t) = N_1^o e^{-\lambda_1 t} \qquad (3.5)$$

For the decay daughter

$$n_2(t) = \frac{n_1^o \lambda_1}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + n_2^o e^{-\lambda_2 t} \qquad (3.6)$$

etc., which continues in the form of the Bateman equation for radioactive decay (B1). Therefore, the time-dependent concentration $N_i(t)$ in the liquid at the point of dissolution ($z = 0$) is also given by a Bateman equation. More formally,

$$N_i(t) = \begin{cases} 0 & , \quad t < 0 \\ B_i(t) & , \quad 0 < t < T \\ 0 & , \quad t > T \end{cases} \qquad (3.7)$$

The above equation can be rewritten as

$$N_i(t) = B_i(t)\left[h(t) - h(t+T)\right] \qquad (3.8)$$

where h(ξ) is the Heaviside step function

$$h(\xi) = \begin{cases} 0 & , \ \xi < 0 \\ 1 & , \ \xi > 0 \end{cases}$$

$B_i(t)$ can be obtained from the solution of the differtial equation

$$\frac{dB_i(t)}{dt} = \lambda_{i-1} B_{i-1} - \lambda_i B_i \, , \quad (i = 1, 2, \cdots , \lambda_0 = 0) \qquad (3.9)$$

with the initial condition

$$B_i(0) = N_i(0) \qquad (3.10)$$

$N_i(0)$ is the concentration in the water phase at the time zero when dissolution begins. The solution of Equation (3.10) is the Bateman equation

$$B_i(t) = \sum_{m=1}^{i} \left(\frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \sum_{n=m}^{i} \frac{e^{-\lambda_n t}}{\prod\limits_{\substack{l=m \\ l \neq n}}^{i} (\lambda_l - \lambda_n)} \equiv \sum_{j=1}^{i} B_{ij} \, e^{-\lambda_j t} \qquad (3.11)$$

where

$$B_{ij} = \sum_{m=1}^{j} N_m^0 \left(\frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \Big/ \prod_{\substack{l=m \\ l \neq j}}^{i} (\lambda_l - \lambda_j) \qquad (3.12)$$

the product term in the denominator of Equation (3.12) is defined as unity when m = j = i.

The $B_i(t)$ values for i = 1, 2 and 3 are written as

$$B_1(t) = N_1^0 e^{-\lambda_1 t} \qquad (3.13)$$

$$B_2(t) = N_2^0 e^{-\lambda_2 t} + \frac{N_1^0 \lambda_1}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) \qquad (3.14)$$

$$B_3(t) = N_3^0 e^{-\lambda_3 t} + \frac{N_2^0 \lambda_2}{\lambda_3 - \lambda_2}\left(e^{-\lambda_2 t} - e^{-\lambda_3 t}\right)$$

$$+ \lambda_1 \lambda_2 N_1^0 \left[\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right] \qquad (3.15)$$

When the band-release time T becomes very small and approaches zero, the release mode becomes an impulse release, such that

$$N_i(t) = N_i^0 T \delta(t) \qquad (3.16)$$

where $\delta(t)$ is the Dirac delta function.

When the band-release time T approaches infinity, the release mode becomes a step release, such that

$$N_i(t) = B_i(t) \hbar(t) \qquad (3.17)$$

On physical grounds, waste material may reasonably be expected to dissolve at an infinitesimal rate in the case of a step release, resulting in only differential concentrations of the dissolved radionuclides. However, it is useful to construct step-release solutions which satisfy the boundary condition of finite concentration in Equation (3.17). If these finite concentrations are those determined from the band-release dissolution model of Equation (3.4) with finite T, then the resulting relatively simple step-release solutions can be used in the superposition equation (Eq.(3.60) developed in Section 3.4 to construct the more complicated band-release solutions.

3.2.2 Nuclide-Dependent fractional rate of dissolution

Here we assume that the waste material and its contained radionuclides begin to dissolve into the water at $t = 0$, and we assume that each radionuclide leaches from the waste at a fractional rate k_i, which is a constant for a given radionuclide. The amount $M_i(t)$ of radionuclide <u>i</u> within the repository at time t is given by

$$\frac{dM_i}{dt} = -k_i M_i - \lambda_i M_i + \lambda_{i-1} M_{i-1} \qquad (3.18)$$

with the initial condition

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$$M_i(0) = M_i^0 \tag{3.19}$$

The solution of (3.18) and (3.19) is obtained as

$$M_i(t) = \sum_{m=1}^{i} \frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r M_m^0 \sum_{n=m}^{i} \frac{e^{-\Lambda_n t}}{\prod_{\substack{l=m \\ (l \neq n)}}^{i} (\Lambda_l - \Lambda_n)} \tag{3.20}$$

where

$$\Lambda_i \equiv \lambda_i + k_i \tag{3.21}$$

Assuming, as in Section 3,2.1, that the radionuclides dissolve uniformly into water flowing at a volumetric rate Q past the waste material, and neglecting the diffusional transport of the dissolved radionuclides in the vicinity of the waste material, the concentration $N_i(t)$ in the water phase at z = 0 is given by

$$N_i(t) = \sum_{m=1}^{i} \left(\frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \frac{k_i}{k_m} N_m^0 \sum_{n=m}^{i} \frac{e^{-\Lambda_n t}}{\prod_{\substack{l=m \\ (l \neq n)}}^{i} (\Lambda_l - \Lambda_n)} \tag{3.22}$$

or

$$N_i(t) = \sum_{j=1}^{i} B'_{ij} e^{-\Lambda_j t}, \qquad t > 0$$

where

$$B'_{ij} \equiv \sum_{m=1}^{j} \frac{\left(\frac{k_i}{k_m} N_m^0 \right) \prod_{r=m}^{i-1} \lambda_r}{\prod_{\substack{l=m \\ (l \neq j)}}^{i} (\Lambda_l - \Lambda_j)} \tag{3.23}$$

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N_m^o is the concentration of nuclide m in the water phase at $z = 0$, $t = 0$. For three members

$$N_1(t) = N_1^0 e^{-\Lambda_1 t} \tag{3.24}$$

$$N_2(t) = N_2^0 e^{-\Lambda_2 t} + \frac{k_2}{k_1} N_1^0 \frac{\lambda_1}{\Lambda_2 - \Lambda_1}\left(e^{-\Lambda_1 t} - e^{-\Lambda_2 t}\right) \tag{3.25}$$

$$\begin{aligned} N_3(t) = {} & N_3^0 e^{-\Lambda_3 t} + \frac{k_3}{k_2} N_2^0 \frac{\lambda_2}{\Lambda_3 - \Lambda_2}\left(e^{-\Lambda_2 t} - e^{-\Lambda_3 t}\right) \\ & + \frac{k_3}{k_1} N_1^0 \lambda_1 \lambda_2 \left\{ \frac{e^{-\Lambda_1 t}}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1)} + \frac{e^{-\Lambda_2 t}}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2)} + \frac{e^{-\Lambda_3 t}}{(\Lambda_1 - \Lambda_3)(\Lambda_2 - \Lambda_3)} \right\} \end{aligned} \tag{3.26}$$

Equations (3.22), (3.23), (3.24), (3.25) and (3.26) describe the "preferential release" mode.

If all species dissolve at the same fractional rate, then all have the same fractional rate constant k ($k_i = k$), and Equation (3.22) becomes

$$N_i(t) = \sum_{m=1}^{i} N_m^0 \left(\frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r\right) \sum_{n=m}^{i} \frac{e^{-(\lambda_n + k)t}}{\prod\limits_{\substack{l=m \\ (l \neq n)}}^{i} (\lambda_l - \lambda_n)} \tag{3.27}$$

where

$$N_m^0 = k M_m^0 Q \tag{3.28}$$

The release mode of Equations (3.27) and (3.28) is called the "exponential release mode".

The release modes of Section 3.2 have been illustrated for the case of no diffusional transport of the released radionuclide at the point of release, so that a concentration boundary condition at the source can be specified, as in Equations (3.4) and (3,28). In the next section we apply these release modes in the development of a generalized source term at the point of dissolution.

3.3 Boundary Conditions and Source Terms

The concentrations $N_i(t)$ obtained in Section 3.2 are the real concentrations $N_i(0,t)$ at the boundary only if transport by diffusion can be neglected at $z = 0$. However, these concentrations $N_i(t)$ can also yield a nuclide source term to be used directly in radionuclide transport equation, a useful approach if diffusional transport at the source is to be considered and/or if the transport equation is to be solved for spatially distributed sources.

The physical picture for the source condition is illustrated in Figure 3.1, in which the dispersion coefficient D_i is assumed to be zero in the up-stream zone and non-zero in the down-stream zone.

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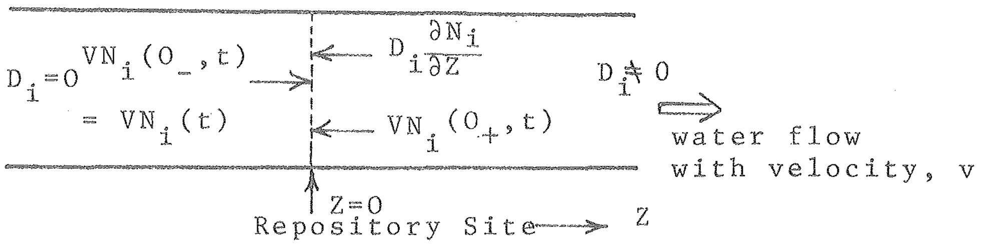

Figure 3.1 Boundary condition at the repository (z=0), diffusional transport in flow direction only.

In this case, the nuclide balance at the repository for one-dimensional transport, with flow at velocity v in the Z direction, yields

$$-D_i\left[\frac{\partial N_i(z,t)}{\partial Z}\right]_{Z=0_+} = v\left[N_i(t) - N_i(0_+,t)\right] \quad (3.29)$$

If the effect of dispersion at z = 0 is neglected, Equation (3.29) reduces to the simple equation.

$$N_i(0_+, t) = N_i(t) \quad (3.30)$$

This concentration boundary condition has been used in other published analyses of radionuclide migration.

A more fundamental approach, necessary when diffusional transport both upstream and downstream of the repository is to be considered, is to use the dissolution rate as a source term in the transport equation. The rate of dissolution $M_i^0(0,t)$ is given from the previous solutions by

$$M_i^0(t) = N_i(t)Q \qquad (3.31)$$

The equivalent distributed volumetric source term $f_i(z,t)$ to be used in the transport equation is then

$$f_i(z,t) = N_i(t)\frac{Q}{S}\,\delta(z) \qquad (3.32)$$

where S is the effective cross-sectional area for water flow associated with the dissolving waste, or

$$f_i(z,t) = N_i(t)v\,\delta(z) \qquad (3.33)$$

where v is the water velocity at the point of dissolution, as illustrated in Figure 3.2.

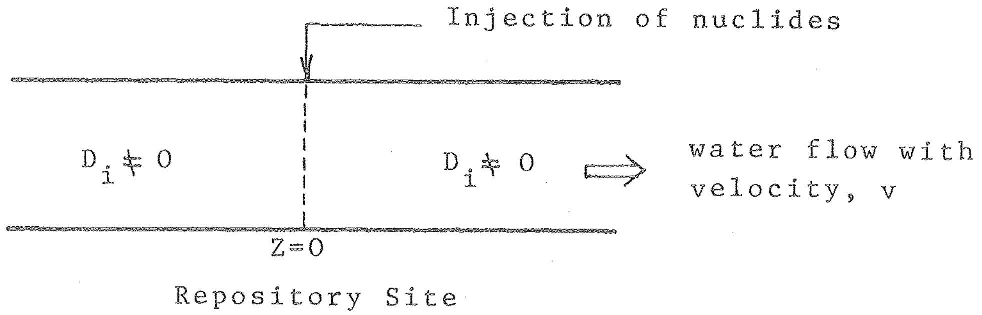

Figure 3.2 Dissolution as a volumetric source term.

3.4 General Superposition Equation for Band Release (F1)

In this section, we develop the general superposition relation for the band release of decaying radionuclides through sorbing media. As shown before, there are two standpoints for connecting the nuclide-release phenomena with the nuclide migration. One is to treat the release as a boundary condition at the repository, and the transport equation without source term is solved using this bounday condition. The other is to treat the release as a source term in the transport equation.

With the first version, the transport equation (2.54) is reduced to

$$L_i(N_i) = -K_{i-1}\lambda_{i-1}\epsilon N_{i-1} \quad , \quad i=1,2,\cdots\cdots, 0<z<\infty, t>0$$

$$L_i(N_i) \equiv \frac{\partial}{\partial z}\left(\epsilon D_i \frac{\partial N_i}{\partial z}\right) - \frac{\partial}{\partial z}(\epsilon v A_i N_i) - \frac{\partial}{\partial t}(\epsilon K_i N_i) - K_i\lambda_i\epsilon N_i \quad (3.34)$$

where $N_i(z,t)$ is the nuclide concentration in water, v is the water velocity, D_i the effective diffusion coefficient and K_i is the overall sorption coefficient. A_i is defined in (2.55). For simplicity, the subscript f has not been included in (3.34). The governing equation for "mother", i.e., the first member of the decay chain, is obtained by formally setting $\lambda_o=0$. The properties ϵ, D_i, v, A_i, B_i and K_i are considered generally to be functions of z.

The initial condition is

$$N_i(z,0) = 0 \quad , \quad i=1,2,\cdots\cdots, \quad 0<z<\infty \quad (3.35)$$

We further prescribe

$$N_i(z,t) = 0 \quad , \quad i=1,2,\cdots\cdots, 0<z<\infty, t<0 \quad (3.36)$$

In the following we will be concerned with two special forms of the solution to the above equations. These are due to a step release of the radionuclides at the boundary z=0 where they obey the conditions

$$N_i^{s}(0,t) = B_i(t)h(t), \quad N_i^{*s}(0,t-T) = B_i(t)h(t-T) \qquad (3.37)$$

for i=1,2,3,...n. With the help of (3.37) we wish to construct the solution for the band release $N_i^b(z,t)$ which satisfies the boundary conditions

$$N_i^{b}(0,t) = B_i(t)h(t) - B_i(t)h(t-T) \qquad (3.38)$$

Where T is the leach time for the band release. We will show that the desired solution is given by

$$N_i^{b}(z,t) = N_i^{s}(z,t) - N_i^{*s}(z,t-T), \quad i=1,2,\text{-------} \quad 0<z<\infty, \; t>0 \qquad (3.39)$$

where $N_i^s(z,t)$ is the solution for the step release. Equation (3.37) and $\overset{*}{N}{}_i^s(z,t)$ is the step release solution satisfying the coundary condition:

$$N_i^{*s}(0,t) = B_i^{*}(t)h(t) = B_i(t+T)h(t) \qquad (3.40)$$

The system of equation (3.34) is linear in the N_i functions and simultaneously subject to non-trivial boundary conditions of the form of (3.37) for all of the nuclides. It is convenient to construct a solution through a set of subsidiary equations in which only one boundary condition arises, one at a time. Thus, we consider for example $N_i^s(z,t)$ in $0<z<\infty$, where i is any number of the chain $i=1,2....,$ n with $\lambda_o=0$. The contribution $N_i^{s(1)}$ due to the finite initial amount of the first member of the chain is given by

$$L_i(N_i^{S(1)}) = -K_{i-1}\lambda_{i-1}N_{i-1}^{S(1)} \quad , \quad i \geq 1 \ , \ \lambda_0 = 0 \qquad (3.41a)$$

$$N_1^{S(1)}(0,t) = B_1(t)h(t) \ , \quad N_1^{S(1)}(z,t<0) = 0 \qquad (3.41b)$$

$$N_i^{S(1)}(0,t) = 0, (-\infty<t<\infty), \quad N_i^{S(1)}(z,t<0)=0, \ i\geq 2 \qquad (3.41c)$$

The contribution $N_i^{s(2)}$ due to the finite amount of the second member of the chain is given by

$$L_i(N_i^{S(2)}) = -K_{i-1}\lambda_{i-1}N_{i-1}^{S(2)} \quad , \quad \lambda_1 = 0 \ , \quad i \geq 2 \qquad (3.42a)$$

$$N_2^{S(2)}(0,t) = B_2(t)h(t) \ , \quad N_2^{S(2)}(z,t<0) = 0 \qquad (3.42b)$$

$$N_i^{S(2)}(0,t) = 0, (-\infty<t<\infty), \quad N_i^{S(2)}(z,t<0) = 0, \ i\geq 3 \qquad (3.42c)$$

and so forth. In each subsequent subsystem, the starting index is increased by unity to represent the member which is to be a mother for the remainder of the chain. On account of the linearity it follow at once that

$$N_i^s(z,t) = \sum_{j=1}^{i} N_i^{s(j)}(z,t) \qquad (3.43)$$

and that $N_i^s(z,t)$ satisfies Equations (3.34), (3.35), (3.43) (3.36) and (3.37). In exactly the same way one can construct the solution for the function $N_i^{*s}(z,t-T)$ by rewriting (3.41), (3.44) and (3,43) in terms of the argument (t-T) after replacing $B_i(t)$ by $B_i^*(t)$. The combination of these two solutions giving $N_i^b(z,t)$, as postdated in (3.39), satisfies the partial differential equation (3.34). It also satisfies the initial conditions because $N_i^s(z,t)$ and $N_i^{*s}(z,t-T)$ vanish for $t<0$ and $t<T$. The boundary conditions force a constraint between the function $B_i(t)$ and $B_i^*(t)$, which is described as

$$B_i^*(t) = B_i(t+T) \qquad (3.44)$$

The second version of the nuclide release mode as mentioned previously asserts that the transport equation (3.34) is rewritten as

$$L_i(N_i) = -\left(K_{i-1}\lambda_{i-1}\varepsilon N_{i-1} + \varepsilon f_i(z,t)\right)$$

$$= -g_i(z,t), \quad i = 1, 2, \cdots\cdots \qquad (3.45)$$

$$\mathbb{D} \; ; \quad -\infty < t < \infty \; , \; -\infty < z < \infty$$

where we postulate that nuclides are released into the water phase only.

At the boundary $\mathbb{C}$ of the defined domain $\mathbb{D}$, the following homogeneous condition is satisfied

$$N_i(z,t) = 0 \quad , \quad z,t \in \mathbb{C} \tag{3.46}$$

We introduce a function $G_i(z,t;\varepsilon,\tau)$ which satisfies

$$L_i\left[G_i(z,t;\xi,\tau)\right] = -\delta(z-\xi)\,\delta(t-\tau) \tag{3.47}$$

$$G_i(z,t;\xi,\tau) = 0 \quad , \quad z,t \in \mathbb{C} \tag{3.48}$$

If we can find the solution for $G_i(z,t;\xi,\tau)$, then $N_i(z,t)$ is given by

$$N_i(z,t) = \iint_{\mathbb{D}} G_i(z,t;\xi,\tau)\, g_i(\xi,\tau)\, d\xi\, d\tau \tag{3.49}$$

The above relation can be easily shown as follows: Multiplying both sides of (3.47) by $g_i(\xi,\tau)$ and integrating the resultant equation over the domain $\mathbb{D}$, one can obtain

$$\begin{aligned} &\iint_{\mathbb{D}} g_i(\xi,\tau) L_i\left[G_i(z,t;\xi,\tau)\right] d\xi\, d\tau \\ &= -\iint_{\mathbb{D}} g_i(\xi,\tau)\, \delta(z-\xi)\cdot\delta(t-\tau)\, d\xi\, d\tau \\ &= -\, g_i(z,t) \end{aligned} \tag{3.50}$$

The left hand side of (3.50) can be rewritten as

$$\begin{aligned}
&\iint_{\mathbb{D}} g_i(\xi,\tau)\, L_i\left[G_i(z,t\,;\xi,\tau)\right] d\xi\, d\tau \\
&= \iint_{\mathbb{D}} L_i\left[G_i(z,t\,;\xi,\tau)\cdot g_i(\xi,\tau)\right] d\xi\, d\tau \\
&= L_i\left[\iint_{\mathbb{D}} G_i(z,t\,;\xi,\tau)\, g_i(\xi,\tau)\, d\xi\, d\tau\right]
\end{aligned} \tag{3.51}$$

Combining (3.50) and (3.51) with (3.45) we can find that (3.49) is the solution for (3.45) and (3.46). Equation (3.49) satisfies (3.45) because $G_i(z,t;\xi,\tau)$ satisfies (3.48). Equation (3.49) can be rewritten as

$$\begin{aligned}
N_i(z,t) = &\iint_{\mathbb{D}} G_i(z,t\,;\xi,\tau)\, \varepsilon(\xi)\, f_i(\xi,\tau)\, d\xi\, d\tau \\
&+ \iint_{\mathbb{D}} G_i(z,t\,;\xi,\tau)\, K_{i-1}(\xi)\, \varepsilon(\xi)\, \lambda_{i-1} N_{i-1}(\xi,\tau)\, d\xi\, d\tau
\end{aligned} \tag{3.52}$$

Equation (3.54) is a recursive solution. By iterative procedure, we can find that $N_i(z,t)$ is given by the linear combination with respect to $f_i, f_{i-1}, \ldots, f_1$. Then, the solution of the band release in which f_i^b is expressed by the combination of two step release modes,

$$f_i^b(z,t) = f_i^s(z,t)\, h(t) - f_i^s(z,t)\, h(t-T) \tag{3.53}$$

is given by the linear combination of the solutions for these step releases:

$$N_i^b(z,t) = N_i^s(z,t) - N_i^{*s}(z,t-T) \tag{3.54}$$

$N_i^{*s}(z,t)$ is the solution for the step release,

$$f_i^s(z,t+T)h(t) = f_i^{*s}(z,t)\,h(t) \tag{3.55}$$

Equation (3.54) agrees with (3.39) which has been obtained on account of the concentration boundary condition. When $f_i^s(z,t)$ is expressed by

$$f_i^s(z,t) = \Phi_i(t)\delta(z)h(t)\,,\quad f^{*s}(z,t) = \Phi_i^*(t)\,\delta(z)h(t) \tag{3.56}$$

the Φ_i term can be described with the help of (3.55) as follows:

$$\Phi_i(t) = QB_i(t)/S\,,\quad \Phi_i^*(t) = QB_i^*(t)/S \tag{3.57}$$

Q is the volumetric flow rate of water and S is the effective cross sectional area of the water phase. Combining (3.57) with (3.55) we obtain

$$B_i(t+T) = B_i^*(t) \tag{3.58}$$

To illustrate how this condition is met, let us assume that the $B_i(t)$ are given by Bateman equations with known coefficients B_{ij}, which has been shown in 3.2. In this case, (3.44) can be expressed as

$$B_i^*(t) = B_i(t+T) = \sum_{j=1}^{i} B_{ij}\, e^{-\lambda_j (t+T)} = \sum_{j=1}^{i} B_{ij}^*\, e^{-\lambda_j t} \qquad (3.59)$$

where $B_{ij}^* = B_{ij} e^{-\lambda_j T}$

Then, if we described the solution of the step release,

$$N_i^S(o,t) = B_i(t)h(t)$$

or

$$f_i^S(z,t) = \Phi_i(t)\delta(Z)\delta(t)$$

as $N_i^S(z,t:B_{ij})$, the solution for the band release can be expressed by

$$N_i^b(z,t) = N_i^s(z,t;B_{ij}) - N_i^s(z,t-T;B_{ij}^*) \qquad (3.60)$$

By use of the superposition equation, the relatively simple solution for step releases can be used to construct the exact but more complicated solution for band release.

Lester, et al (L1) have proposed the superposition relation for band release

$$N_i^b(z,t) = N_i^s(z,t;B_{ij}) - N_i^s(z,t-T;B_{ij}) \qquad (3.61)$$

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Equation (3.61) is incorrect, because it does not take into account the difference between Bateman coefficients, B_{ij}^* and B_{ij}. It is, in fact, applicable only to chemical chromatographic bands wherein radioactive decay does not occur. As seen how Equation (3.59), only when $\lambda_i=0$ are the B_{ij}^* and B_{ij} equal.

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4. RECURSIVE AND GENERAL SOLUTIONS OF THE TRANSPORT EQUATION FOR NUCLIDE MIGRATION

4.1 Introduction

We have obtained the fundamental transport equation (2.54) which governs the migration of the nuclide through geological media. When we neglect a term of f_{is} in (2.54), the governing equation can be expressed as

$$\frac{\partial}{\partial t}(K_i \varepsilon N_i) + \frac{\partial}{\partial z}(A_i \varepsilon v N_i) - \frac{\partial}{\partial z}\left(\varepsilon D_i \frac{\partial N_i}{\partial z}\right)$$
$$= \varepsilon(K_{i-1}\lambda_{i-1}N_{i-1} - K_i\lambda_i N_i) + \varepsilon f_i(z,t) \quad , \quad \lambda_0 = 0 \qquad (4.1)$$

K_i is the overall sorption coefficient, ε is the porosity, v, the water velocity, D_i, the diffusion coefficient and A_i is given by (2.55). These values are functions of z and time in general.

When $K_i A_i, \varepsilon, v$ and D_i are independent of the distance z and time t the above equation can be simplified as

$$D_i \frac{\partial^2 N_i}{\partial z^2} - v\frac{\partial N_i}{\partial z} - K_i \frac{\partial N_i}{\partial t} - K_i\lambda_i N_i$$
$$= -K_{i-1}\lambda_{i-1}N_{i-1} - f_i(z,t) \quad , \quad \lambda_0 = 0 \qquad (4.2)$$

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In this chapter, we shall be concerned with solving the fundamental equation (4.2) with arbitrary release modes. Section 4.2 discusses the solution of the transport equation with a nuclide source. Section 4.3 treats the recursive solution for the transport equation without a source term, in which the release phenomena are treated as a boundary condition at z = 0. Section 4.4 is concerned with the case of no-dispersion. Recursive solutions of different forms and the general solution will be proposed for an i - member decay chain and arbitrary release modes. Section 4.5 discusses the condition for local secular equilibrium in a decay chain in which very rapidly decaying nuclides are included.

4.2 Solution of the Transport Equation with a Nuclide Source

4.2.1 Recursive solution of transport equation

(4.2) can be rewritten as:

$$\eta_i \frac{\partial^2 N_i}{\partial z^2} - \upsilon_i \frac{\partial N_i}{\partial z} - \frac{\partial N_i}{\partial t} - \lambda_i N_i = -\frac{K_{i-1}}{K_i} \lambda_{i-1} N_{i-1} - \frac{f_i(z,t)}{K_i} \qquad (4.3ab)$$

$$\equiv -F_i(z,t)$$

$$\eta_i \equiv D_i/K_i \quad , \quad \upsilon_i \equiv \upsilon/K_i$$

The defined domain in which (4.3) holds is

$$\mathbb{D}: \quad -\infty < z < \infty \ , \ -\infty < t < \infty \tag{4.4a}$$

and the concentrations $N_i(z,t)$ at the boundary of the domain, $\mathbb{C}$, are zero

$$N_i(z,t) = 0 \quad , \quad z, t \in \mathbb{C} \tag{4.4b}$$

The solution of (4.3) and (4.4) can be obtained with the help of the Green function as shown in Section 3.4.

$$N_i(z,t) = \iint_{\mathbb{D}} G_i(z,t:\xi,\tau) F_i(\xi,\tau)\, d\xi\, d\tau \tag{4.5}$$

$G_i(z,t;\xi,\tau)$ should satisfy

$$\eta_i \frac{\partial^2 G_i}{\partial z^2} - v_i \frac{\partial G_i}{\partial z} - \frac{\partial G_i}{\partial t} - \lambda_i G_i = -\delta(z-\xi)\delta(t-\tau) \tag{4.6a}$$

$$G_i(z,t:\xi,\tau) = 0 \quad , \quad z, t \in \mathbb{C} \tag{4.6b}$$

The solution for $G_i(z,t;\xi,\tau)$ can be obtained with the aid of Fourier integral. Delta function is expressed by Fourier integral

$$\delta(z-\xi) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{ik(z-\xi)} dk \, , \, \delta(t-\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega(t-\tau)} d\omega \tag{4.7}$$

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Then, the right hand side of (4,6a) can be written as

$$-\delta(z-\xi)\delta(t-\tau)=\frac{-1}{4\pi^2}\int_{-\infty}^{\infty}e^{ik(z-\xi)}dk\int_{-\infty}^{\infty}e^{-i\omega(t-\tau)}d\tau \qquad (4.8)$$

We prescribe the solution for G_i as

$$G_i(z,t:\xi,\tau)=\frac{1}{4\pi^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}e^{ik(z-\xi)-i\omega(t-\tau)}S(k,\omega)dk\,d\omega \qquad (4.9)$$

Introducing (4.9) into (4.6a), S(k,w) takes fhe form;

$$S(k,\omega)=-\frac{1}{i\omega-(\eta_i k^2+\lambda_i)-iv_i k} \qquad (4.10)$$

Then,

$$G_i(z,t:\xi,\tau)=-\frac{1}{4\pi^2}\int_{-\infty}^{\infty}e^{ik(z-\xi)}I(k)\,dk \qquad (4.11)$$

where

$$I(k)=\int_{-\infty}^{\infty}\frac{e^{-i\omega(t-\tau)}}{i\omega-(\eta_i k^2+\lambda_i)-iv_i k}d\omega \qquad (4.12)$$

Now, we consider the integral, I(k). The function in the integral is analytical except for

$$\omega=-i(\eta_i k^2+\lambda_i)+v_i k \qquad (4.13)$$

Then, we can obtain I(k) in the range, $t-\tau<0$;

$$I(k) = \lim_{R\to\infty} \int_{-R}^{R} \frac{e^{-i\omega(t-\tau)}}{i\omega-(\eta_i k^2+\lambda_i)-i v_i k} \cdot d\omega = 0 \quad (4.14)$$

because

$\lim_{\rho\to\infty} \mathrm{Re}(e^{-iw(t-\tau)}) \to 0$ when $t-\tau<0$, see Fig. 4.1, the closed curve C'_ρ.

When $t-\tau>0$, we can integrate along the closed curve, C_ρ.

In this case,

$$\lim_{\rho\to\infty} \mathrm{Re}\,(e^{-i\omega(t-\tau)}) \longrightarrow 0$$

Then, the integral is given by

$$\begin{aligned} I(k) &= -2\pi i \left[\mathrm{Res.}\right]_{\omega=-i(\lambda_i+\eta_i k^2)+v_i k} \\ &= -2\pi \exp\left[-(t-\tau)(\lambda_i+\eta_i k^2)-i v_i k(t-\tau)\right] \\ &\quad , \quad (t-\tau>0) \end{aligned} \quad (4.15)$$

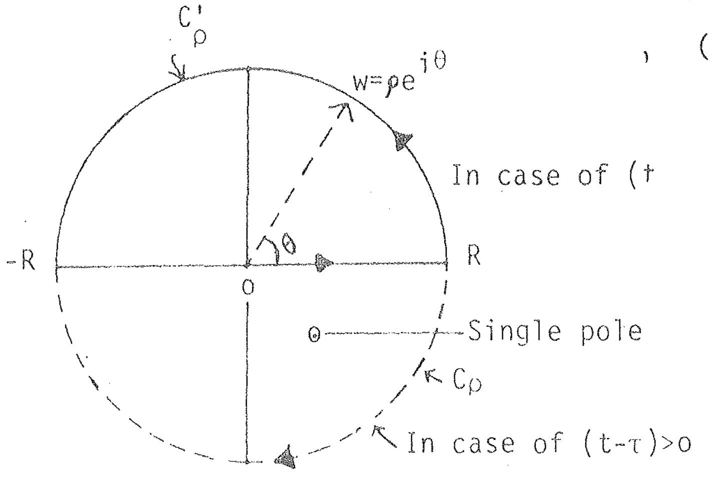

Figure 4.1
Domain of Complex Variable

From (4.14) and (4.15), the integral can be given by

$$I(k) = \begin{cases} 0 & , t-\tau<0 \\ -2\pi e^{-(t-\tau)(\lambda_i+\eta_i k^2) - ik v_i (t-\tau)} & , t-\tau>0 \end{cases} \tag{4.16}$$

Introducing (4.16) into (4.11), the $G_i(z,t;\xi,\tau)$ is expressed as:

$$G_i(z,t:\xi,\tau) = \begin{cases} \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp\left[ik(z-\xi-v_i(t-\tau)) - (\lambda_i+\eta_i k^2)(t-\tau)\right]dk \\ \qquad , \ t-\tau>0 \\ 0 \qquad . \ t-\tau<0 \end{cases} \tag{4.17}$$

For the integration of (4.17) we can use the formula:

$$\int_{-\infty}^{\infty} e^{-a^2k^2+bk} dk = \exp(b^2/4a)\sqrt{\pi}/a \quad , \ a>0 \tag{4.18}$$

Then, the $G_i(z,t;\xi,\tau)$ function is explicitly obtained by

$$G_i(z,t:\xi,\tau) = \begin{cases} \dfrac{e^{-\lambda_i(t-\tau)}}{\sqrt{4\pi\eta_i(t-\tau)}} \exp\left[-\dfrac{\{z-\xi-v_i(t-\tau)\}^2}{4\eta_i(t-\tau)}\right] & , \ t-\tau>0 \\ 0 & . \ t-\tau<0 \end{cases} \tag{4.19}$$

The solution for (4.3) and (4.4) is obtained by introducing (4.19) into (4.5).

$$N_i(z,t)=\int_{-\infty}^{t}\int_{-\infty}^{\infty}\left[\frac{f_i(\xi,\tau)}{K_i}+\lambda_{i-1}\frac{K_{i-1}}{K_i}N_{i-1}(\xi,\tau)\right]\times\frac{e^{-\lambda_i(t-\tau)}}{\sqrt{4\pi\eta_i(t-\tau)}}\exp\left[-\frac{\{z-\xi-v_i(t-\tau)\}^2}{4\eta_i(t-\tau)}\right]d\xi\,d\tau \tag{4.20}$$

This is the general solution for arbitrary release modes.

When $f_i(Z,t)$ is expressed as:

$$f_i(z,t)/K_i=\phi_i(z,t)h(t) \tag{4.21}$$

$N_{i-1}(z,t)$ is zero in the range, $t < 0$.

Then, $N_i(z,t)$ is given as

$$N_i(z,t)=\int_{0}^{t}\int_{-\infty}^{\infty}\left[\frac{f_i(\xi,\tau)}{K_i}+\lambda_{i-1}\frac{K_{i-1}}{K_i}N_{i-1}(\xi,\tau)\right.\times\frac{e^{-\lambda_i(t-\tau)}}{\sqrt{4\pi\eta_i(t-\tau)}}\exp\left[-\frac{\{z-\xi-v_i(t-\tau)\}^2}{4\eta_i(t-\tau)}\right]d\xi\,d\tau \tag{4.22}$$

Furthermore, we postulate that the nuclide is released out at $z = 0$. In this case, $f_i(z,t)$ is expressed by

$$f_i(z,t)/K_i=\Phi_i(t)\delta(z)h(t)\equiv f_i(t)\delta(z) \tag{4.23}$$

Therefore, the nuclide concentration is reduced to a simplified form:

$$N_i(z,t) = \int_0^t f_i(t-\tau)\frac{e^{-\lambda_i\tau}}{\sqrt{4\pi\eta_i\tau}}\exp\left[-\frac{(z-v_i\tau)^2}{4\eta_i\tau}\right]d\tau$$

$$+\frac{\lambda_{i-1}K_{i-1}}{K_i}\int_0^t\int_{-\infty}^{\infty}N_{i-1}(\xi,t-\tau)\frac{e^{-\lambda_i\tau}}{\sqrt{4\pi\eta_i\tau}}\exp\left[-\frac{(z-v_i\tau-\xi)^2}{4\eta_i\tau}\right]d\xi\, d\tau \qquad (4.24)$$

The physical meaning of (4.24) is clear . The first term of the right hand side of (4.24) means the contribution of the ith nuclide which is released out from the repository. The amount of the ith nuclide released from the repository at time t − τ during the time interval, dτ, are given by $f_i(t-\tau)d\tau$. The probability density that the ith nuclide released at time t − τ locates on the position, z, after time τ elapsed, is given by

$$\frac{1}{\sqrt{4\pi\eta_i t}}\exp\left[-\frac{(z-v_i\tau)^2}{4\eta_i\tau}\right] \qquad (4.25)$$

During τ, the amount of the ith nuclide decreases due to decay. The amount of the nuclide which has been released at the time t − τ becomes $f_i(t-\tau)$. $\exp(-\lambda_i\tau)d\tau$ after the time interval, τ. Thus, the contribution of the ith nuclide released from the repository is given by the first term on the right hand side of (4.24).

The second term of the right hand side of (4.24) arises from the i^{th} nuclide generated from the (i-1)-th nuclide, which has been distributed through the whole region of the water path due to the dispersion. The amount of the i^{th} nuclide generated from the (i-1)-th nuclide during the time interval, $d\tau$, at the position, ξ, and at the time $(t-\tau)$, is given by:

$$\frac{\lambda_{i-1} K_{i-1}}{K_i} \cdot N_{i-1}(\xi, t-\tau)\, d\tau \tag{4.26}$$

The probability that the i^{th} nuclide generated in the range of distance, $\xi \sim \xi + d\xi$, locates on the position, z, during the time interval, τ, is given by

$$\frac{1}{\sqrt{4\pi \eta_i t}} \exp\left[-\frac{(z-\xi-v_i\tau)^2}{4\eta_i \tau}\right] d\xi \tag{4.27}$$

Thus the contribution of the i^{th} nuclide generated from the (i-1)-th nuclide for a time interval $d\tau$ and a distance $d\xi$ is given by the product of (4.26) and (4,27).

When the dispersion coefficient becomes zero, the general solution (4.22) yields the solution without dispersion, with the aid of the relation:

$$\lim_{\eta_i \to 0} \frac{1}{\sqrt{4\pi\eta_i\tau}} \exp\left[-\frac{(z-v_i\tau)^2}{4\eta_i\tau}\right] = \delta(z-v_i\tau) \qquad (4.28)$$

The resultant solution is expressed by

$$\begin{aligned} N_i(z,t) = & \int_0^t \frac{f_i(z-v_i\tau,\, t-\tau)}{K_i} e^{-\lambda_i\tau} d\tau \\ & + \int_0^t \frac{\lambda_{i-1}K_{i-1}}{K_i} N_{i-1}(z-v_i\tau,\, t-\tau) e^{-\lambda_i\tau} d\tau \end{aligned} \qquad (4.29)$$

In case of the release given by (4.23), (4.24) is reduced to

$$\begin{aligned} N_i(z,t) = & \frac{1}{v_i} f_i(t-z/v_i)\, e^{-z\lambda_i/v_i} h(t-z/v_i) \\ & + \frac{\lambda_{i-1}K_{i-1}}{K_i} \int_0^{\mathrm{Min}(t,\, z/v_i)} N_{i-1}(z-v_i t,\, t-\tau) e^{-\lambda_i\tau} d\tau \end{aligned} \qquad (4.30)$$

4.2.2 Concentration of the i^{th} nuclide in a decay chain

We can obtain the concentration of the i^{th} nuclide from the recursive formula, (4.26). From (4.3b) the following relations can be derived from setting $D_i = D$

$$\eta_i = D v_i/v \equiv K v_i \quad , \quad K_{i-1}/K_i = v_i/v_{i-1} \qquad (4.31)$$

Introducing (4.31) into (4.24), one can obtain

$$N_i(z,t) = \int_0^t f_i(t-\theta_i)\, e^{-\lambda_i\theta_i} F(v_i\theta_i,\, z-v_i\theta_i)\, d\theta_i + \frac{\lambda_{i-1} v_i}{v_{i-1}} \int_0^t \int_{-\infty}^{\infty} N_{i-1}(\xi,\, t-\theta_i)\, F(v_i\theta_i,\, z-\xi-v_i\theta_i)\, e^{-\lambda_i\theta_i}\, d\xi\, d\theta_i \qquad (4.32)$$

where

$$F(\alpha,\beta) = \frac{1}{\sqrt{4\pi\kappa\alpha}} \exp\left(-\frac{\beta^2}{4\pi\alpha}\right) \qquad (4.33)$$

For the mother nuclide, i = 1, $\lambda_0 = 0$.

$$N_1(z,t) = \int_0^t f_1(t-\theta)\, e^{-\lambda_1\theta} F(v_1\theta,\, z-v_1\theta)\, d\theta \qquad (4.34)$$

With the help of (4.32), $N_2(z,t)$ can be obtained as:

$$N_2(z,t) = \int_0^t f_2(t-\theta_2)\, e^{-\lambda_2\theta_2} F(v_2\theta_2,\, z-v_2\theta_2)\, d\theta_2 + \frac{\lambda_1 v_2}{v_1} \int_0^t d\theta_2 \int_0^{t-\theta_2} d\theta_1\, f_1(t-\theta_1-\theta_2)\, e^{-\lambda_1\theta_1-\lambda_2\theta_2} \times \int_{-\infty}^{\infty} d\xi\, F(v_1\theta_1,\, \xi-v_1\theta_1)\, F(v_2\theta_2,\, z-\xi-v_2\theta_2) \qquad (4.35)$$

The integral term with respect to ξ is reduced to

$$\int_{-\infty}^{\infty} F(a,\, \xi-a)\, F(b,\, z-\xi-b)\, d\xi = F(a+b,\, z-(a+b)) \qquad (4.36)$$

Using the above equation, $N_2(z,t)$ can be rewritten by

$$N_2(z,t) = \int_0^t d\theta_2 f_2(t-\theta_2) e^{-\lambda_2\theta_2} F(v_2\theta_2, z-v_2\theta_2)$$
$$+ \frac{\lambda_1 v_2}{v_1} \int_0^t d\theta_2 \int_0^{t-\theta_2} d\theta_1 f_1(t-\theta_1-\theta_2) e^{-\lambda_1\theta_1-\lambda_2\theta_2} \qquad (4.37)$$
$$\times F(v_1\theta_1+v_2\theta_2, z-(v_1\theta_1+v_2\theta_2))$$

Generally, the $N_i(z,t)$ is calculated by the same procedure.

$$N_i(z,t) = \int_0^t f_i(t-\theta_i) e^{-\lambda_i\theta_i} F(v_i\theta_i, z-v_i\theta_i)\, d\theta_i$$
$$+ \sum_{j=1}^{i-1} \left(\frac{v_i}{v_j} \prod_{l=j}^{i-1} \lambda_l \right) \int_0^t d\theta_i \int_0^{t-\theta_i} d\theta_{i-1} \cdots\cdots \int_0^{t-\sum_{k=j+1}^{i}\theta_k} d\theta_j \qquad (4.38)$$
$$\times f_j(t-\sum_{h=j}^{i}\theta_k)\, e^{-\sum_{k=j}^{i}\lambda_k\theta_k} F(\sum_{k=j}^{i} v_k\theta_k, z-\sum_{k=j}^{i} v_k\theta_k)$$

4.3 Recursive Solution of the Transport Equation with a Concentration Boundary Condition

Consider the canonical system

$$K_1 \frac{\partial N_1}{\partial t} + v \frac{\partial N_1}{\partial z} - D \frac{\partial^2 N_1}{\partial z^2} + \lambda_1 K_1 N_1 = 0$$
$$K_2 \frac{\partial N_2}{\partial t} + v \frac{\partial N_2}{\partial z} - D \frac{\partial^2 N_2}{\partial z^2} + \lambda_2 K_2 N_2 = \lambda_1 K_1 N_1 \qquad (4.39)$$
$$- - - - - -$$
$$K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} - D \frac{\partial^2 N_i}{\partial z^2} + \lambda_i K_i N_i = \lambda_{i-1} K_{i-1} N_{i-1}$$

which is to be solved for $N_i = N_i(z,t)$ in the domain $o<z<\infty$, $o<t<\infty$ subject to the

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Initial conditions: $N_i(z,o) = 0,\ Z > 0$ (4.40)

Boundary conditions [a1]:

$$-D^* \frac{\partial N_i}{\partial z} + v N_i = v N_i^0 \phi_i(t) \quad \text{for } z = 0,\ t > 0 \tag{4.41}$$

$N_i(z,t)$ and its derivatives must tend to zero with a suitable exponential order as $z \to \infty$.

The boundary condition includes a contribution due to diffusion effects and if this term is not desired one can formally set D* equal to zero in the final solution with certain additional changes which we indicate later. The boundary data $\phi_i(t)$ are arbitrary prescribed integrable functions.

It is convenient to introduce the following parameters:

$$\alpha_i = \lambda_i K_i,\ \beta_i = -\lambda_i - \gamma^2 \eta_i,\ \gamma = \frac{v}{2D},\ \eta_i = \frac{D}{K_i} \tag{4.42}$$

With the first of these one rewrites the general equation (4.39) as

$$K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} - D \frac{\partial^2 N_i}{\partial z^2} + \alpha_i N_i = \alpha_{i-1} N_{i-1},\ \alpha_0 = 0 \tag{4.43}$$

We assume that $N_{i-1}(z,t)$ is known and show how to compute $N_i(z.t)$. First one simplifies (4.43) by removing the

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[a1] See section 3.3

convection and radio-active decay term on the left hand side. For this let:

$$N_i(z,t) = e^{\gamma z + \beta_i t} U_i(z,t) \tag{4.44}$$

where $U_i(z,t)$ is as yet an unknown function. Substitution of this into (4.43) results in the following partial differential equation for U_i.

$$\frac{\partial U_i}{\partial t} = \eta_i \frac{\partial^2 U_i}{\partial z^2} + S_{i-1}(z,t) \quad , \quad z>0\,,\ t>0 \tag{4.45a}$$

where

$$S_{i-1}(z,t) = \frac{\alpha_{i-1}}{K_i} e^{-(\beta_i - \beta_{i-1})t} U_{i-1}(z,t) \quad , \quad \alpha_0 = 0 \tag{4.45b}$$

The initial and boundary conditions, (4.40) and (4.41), transform into

$$U_i(z,0) = 0 \quad , \quad z>0 \tag{4.46}$$

and

$$\frac{\partial U_i}{\partial z} - \gamma U_i = -2 N_i^0 \gamma g_i(t) \quad , \quad \text{at } z=0\,,\ t>0 \tag{4.47a}$$

where

$$g_i(t) \equiv e^{-\beta_i t} \phi_i(t) \tag{4.47b}$$

We exclude in the following the cases when $\gamma = 0$ and $\eta_i = 0$. In view of (4.44), U_i and its derivatives must tend to zero faster than $e^{-\gamma z}$ as $z \to \infty$. The governing equations of the problem have thus been reduced to (4.45) ∿ (4.47). We solve this problem in two different forms.

A-Integral Transform Method

One defines a modified Fourier transform and its inverse with help of

$$u_i(p,t) = \int_0^\infty T(p,z)\, U_i(z,t)\, dz \tag{4.48}$$

and

$$U_i(z,t) = \int_0^\infty T(p,z)\, u_i(p,t)\, dp \tag{4.49}$$

This transform kernel has the form $^{(C1)}$

$$T(p,z) = \sqrt{\frac{2}{\pi}}\, \frac{p\cos(pz) + \gamma \sin(pz)}{\sqrt{p^2+\gamma^2}} \tag{4.50}$$

and satisfies the Sturm-Liouville problem

$$\frac{d^2T}{dz^2} + p^2 T = 0 \quad , \qquad 0 < z < \infty \tag{4.51a}$$

$$\frac{dT}{dz} - \gamma T = 0 \quad , \qquad \text{at} \quad z = 0 \tag{4.51b}$$

If one applies the integral transform (4.48) to equation (4.45) there results

$$\frac{d}{dt} u_i(p,t) = \eta_i \int_0^\infty \frac{\partial^2 U_i(z,t)}{\partial z^2} \cdot T(p,z)\,dz + S_{i-1}(p,t) \qquad (4.52a)$$

where

$$S_{i-1}(p,t) = \int_0^\infty S_{i-1}(z,t)\, T(p,z)\, dz \qquad (4.52b)$$

By integration by parts one obtains with help of equations (4.48) and (4.51a)

$$\int_0^\infty \frac{\partial^2 U_i}{\partial z^2} \cdot T(p,z)\, dz = \left[T \frac{\partial U_i}{\partial z} - U_i \frac{\partial T}{\partial z} \right]_0^\infty - p^2 u_i(p,t) \qquad (4.53)$$

The bracketed term vanishes at the upper limit in view of the fact that U_i and its derivatives must vanish faster than $e^{-\gamma z}$ as $z \to \infty$. At the lower limit $z = 0$, there results with help of (4.47) and (4.51),

$$\begin{aligned} -\left[T \frac{\partial U_i}{\partial z} - U_i \frac{\partial T}{\partial z} \right]_{z=0} &= T(p,0)\gamma \left[2N_i^0 g_i(t) - U_i(0,t) \right] + U_i(0,t)\gamma T(p,0) \\ &= 2T(p,0)\gamma N_i^0 g_i(t) \end{aligned} \qquad (4.54)$$

If one combines (4.53) and (4.54) and substitutes the result into (4.52) one obtains the inhomogenous Bateman equations

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$$\frac{du_i(p,t)}{dt} + \eta_i p^2 u_i(p,t) = r_{i-1,i}(p,t) \tag{4.55a}$$

where

$$r_{i-1,i}(p,t) = \left\{ S_{i-1}(p,t) + T(p,0)\, 2\gamma\eta_i N_i^0\, g_i(t) \right\}, \text{ with } S_0(p,t)=0 \tag{4.55b}$$

The source term $R_{i-1,i}(z,t)$ consists of the transform of the precursor solution and the boundary condition of the i^{th} nuclide. Equation (4.55) is solved subject to the transformed initial conditions (4.46) i.e. $u_i(p,0) = 0$,

$$u_i(p,t) = \int_0^t e^{-\eta_i p^2 (t-\tau)}\, r_{i-1,i}(p,\tau)\, d\tau \tag{4.56}$$

This can finally be inverted with help of (4.49) and yields the formal solution for $U_i(z,t)$, or in terms of (4.44), $N_i(z,t)$.

$$N_i(z,t) = e^{\gamma z + \beta_i t} \int_0^\infty e^{-\eta_i p^2 t}\, T(p,z) \left\{ \int_0^t e^{\eta_i p^2 \tau}\, r_{i-1,i}(p,\tau)\, d\tau \right\} dp \tag{4.57}$$

The source term $r_{i-1,i}$ has the form

$$r_{i-1,i}(p,\tau) = \int_0^\infty S_{i-1}(z,\tau)\, T(p,z)\, dz + 2\gamma\, T(p,0)\, \eta_i N_i^0\, g_i(\tau) \tag{4.58}$$

with

$$S_0(z,t) \equiv 0$$

One can separate the solution into the two basic contributions which stem from the boundary condition of the ith nuclide and its radioactive decay of the precursors

$$N_i(z,t) = e^{\gamma z + \beta_i t} \left[2\gamma\eta_i N_i^0 \int_0^\infty e^{-\eta_i p^2 t} T(p,0)\, T(p,z) \left\{ \int_0^t e^{\eta_i p^2 \tau} g_i(\tau)\, d\tau \right\} dp \right.$$

$$\left. + \int_0^\infty e^{-\eta_i p^2 t} T(p,z) \left\{ \int_0^t e^{\eta_i p^2 \tau} \left[\int_0^\infty S_{i-1}(z,\tau)\, T(p,z)\, dz \right] d\tau \right\} dp \right] , \quad S_0(z,t) \equiv 0$$

(4.59)

Consider now the special case that the diffusive component at the boundary vanishes (i.e., D* = 0) in (4.41). Then one can use a standard Fourier sine transform and the kernel in equation (4.50) is replaced by

$$T(p,z;\ D^* = 0) = \sqrt{\frac{2}{\pi}}\ \mathrm{Sin}(pz) \qquad (4.60)$$

With some additional minor changes the final solution reads

$$N_i(z,t) = e^{\gamma z + \beta_i t} \left[\eta_i N_i^0 \int_0^\infty e^{-\eta_i p^2 t} \cdot T(p,z;D^*{=}0)\ \frac{\partial T(p,0;D^*{=}0)}{\partial z} \right.$$

$$\left\{ \int_0^t e^{\eta_i p^2 \tau} g_i(\tau)\, d\tau \right\} dp + \int_0^\infty e^{-\eta_i p^2 t} T(p,z;D^*{=}0) \left\{ \int_0^t e^{\eta_i p^2 \tau} \cdot \right.$$

$$\left. \left. \left(\int_0^\infty S_{i-1}(z,\tau)\, T(p,z;D^*{=}0)\, dz \right) d\tau \right\} dp \right] \qquad (4.61)$$

B- The Greens Function Method

Let us replace in the solution $U_i(z,t)$ of the equation (4.45) i.e.

$$\frac{\partial U_i}{\partial t} = \eta_i \frac{\partial^2 U_i}{\partial z^2} + S_{i-1}(z,t) \quad , \quad z>0,\ t>0 \tag{4.62}$$

the variables z and t by ξ and τ respectively. Let $G(z,\xi,\tau)$ be the Green's function of the homogeneous equation (4.59) which for boundary conditions of type III, i.e., (4.47) has the form (C1)

$$G(z,\xi,\tau) = \frac{1}{\sqrt{4\pi\eta_i\tau}}\left\{\left[e^{-\frac{(z-\xi)^2}{4\eta_i\tau}} + e^{-\frac{(z+\xi)^2}{4\eta_i\tau}}\right] - 2\gamma\int_0^\infty e^{-\frac{(z+\xi+z')^2}{4\eta_i\tau}-\gamma z'}dz'\right\}$$

$$0<z,\ \xi<\infty,\ z\neq\xi,\ \tau>0 \tag{4.63}$$

Now replace in G the argument τ by $t-\tau$. Then G satisfies the equation

$$-\frac{\partial G}{\partial \tau} = \eta_i \frac{\partial^2 G}{\partial \xi^2} \quad , \quad \xi>0,\ \tau>0 \tag{4.64}$$

and the homogeneous boundary condition

$$\frac{\partial G}{\partial \xi} - \gamma G = 0 \quad , \quad \xi=0 \tag{4.65}$$

Consider now

$$\frac{\partial(GU_i)}{\partial\tau} = G\frac{\partial U_i}{\partial\tau} + U_i\frac{\partial G}{\partial\tau} = \eta_i\left[G\frac{\partial^2 U_i}{\partial\xi^2} - U_i\frac{\partial^2 G}{\partial\xi^2}\right] + GS_{i-1} \qquad (4.66)$$

using (4.62) and (4.64). If one integrates this with respect to ξ from 0 to ∞ and then with respect to τ from 0 to $t-\theta$ where $0<\theta<t$ one obtains

$$\int_0^\infty (GU_i)_{\tau=t-\theta}\,d\xi - \int_0^\infty (GU_i)_{\tau=0}\,d\xi = \eta_i\int_0^{t-\theta} d\tau\int_0^\infty\left[G\frac{\partial^2 U_i}{\partial\xi^2} - U_i\frac{\partial^2 G}{\partial\xi^2}\right]d\xi + \int_0^{t-\theta} d\tau\int_0^\infty GS_{i-1}\,d\xi \qquad (4.67)$$

If the first integral on the right hand side is integrated by parts this yields

$$\int_0^\infty (GU_i)_{\tau=t-\theta}\,d\xi = \int_0^\infty (GU_i)_{\tau=0}\,d\xi + \eta_i\int_0^{t-\theta}\left\{\left(G\frac{\partial U_i}{\partial\xi}\right)\Big|_{\xi=0}^{\xi=\infty} - \left(U_i\frac{\partial G}{\partial\xi}\right)\Big|_{\xi=0}^{\xi=\infty} - \int_0^\infty\left[\frac{\partial G}{\partial\xi}\frac{\partial U_i}{\partial\xi} - \frac{\partial U_i}{\partial\xi}\frac{\partial G}{\partial\xi}\right]d\xi\right\}d\tau + \int_0^{t-\theta} d\tau\int_0^\infty GS_{i-1}\,d\xi \qquad (4.68)$$

On the right hand side the first term vanishes on account of the initial condition, (4.46) and so does the third integral. The second integral vanishes at the upper limit $\xi = \infty$ for the reasons started after (4.47) for the behavior of U_i. At the lower limit

$$\left(G\frac{\partial U_i}{\partial \xi} - U_i\frac{\partial G}{\partial \xi}\right)\Big|_{\xi=0} = G\left[\gamma U_i - 2N_i^0\gamma g_i(t)\right]_{\xi=0} - (U_i\gamma G)_{\xi=0}$$

$$= -G(z,0,t-\tau)\,2\gamma N_i^0 g_i(t) \tag{4.69}$$

on account of (4.47) and (4.65). Hence

$$\int_0^\infty G(z,\xi,\theta)U_i(\xi,t-\theta)\,d\xi = 2\gamma\eta_i N_i^0\int_0^{t-\theta} G(z,0,t-\tau)\,g_i(\tau)\,d\tau$$

$$+\int_0^t d\tau\int_0^\infty G(z,\xi,t-\tau)\,S_{i-1}(\xi,\tau)\,d\xi \tag{4.70}$$

We now pass to the limit $\Theta\to 0$ and recall the delta function character of the Green's function $G\ (z,\xi,\Theta)$ when $\Theta\to 0$. The integral on the left hand side sifts out the value of U_i at x,t so that the solution to our problem is given by

$$U_i(z,t) = 2\gamma\eta_i N_i^0\int_0^t G(z,0,t-\tau)\,g_i(\tau)\,d\tau$$

$$+\int_0^t d\tau\int_0^\infty G(z,\xi,t-\tau)\,S_{i-1}(\xi,\tau)\,d\xi \tag{4.71}$$

where G is defined by (4.63).

In the case that the diffusion term in the boundary condition (4.41) or its equivalent (4.47) is absent and G must then satisfy a boundary condition of type I i.e. in place of equation (4.65) we have

$$G = 0, \quad \xi = 0. \tag{4.72}$$

The proper form of G is then

$$G(z,\xi,t) = \frac{1}{\sqrt{4\pi\eta_i\tau}}\left\{ e^{-\frac{(z-\xi)^2}{4\eta_i\tau}} - e^{-\frac{(z+\xi)^2}{4\eta_i\tau}} \right\}, \tag{4.73}$$

$$0 < z < \infty, \quad 0 < \xi < \infty, \quad z \neq \xi, \quad \tau > 0$$

and (4.71) takes on the form

$$\begin{aligned} U_i(z,t) = {} & \eta_i N_i^\circ \int_0^t \frac{\partial G(z,0,t-\tau)}{\partial \xi} g_i(\tau)\, d\tau \\ & + \int_0^t d\tau \int_0^\infty G(z,\xi,t-\tau)\, S_{i-1}(\xi,\tau)\, d\tau \end{aligned} \tag{4.74}$$

4.4 Recursive and General Solutions of Transport Equation without Dispersion

4.4.1 Recursive solution of transport equation without dispersion and some properties of the solution.

Consider the canonical system

$$\begin{aligned} K_1\frac{\partial N_1}{\partial t} + v\frac{\partial N_1}{\partial z} + \lambda_1 K_1 N_1 &= 0 \\ K_2\frac{\partial N_2}{\partial t} + v\frac{\partial N_2}{\partial z} + \lambda_2 K_2 N_2 &= \lambda_1 K_1 N_1 \\ \vdots \qquad\quad \vdots \qquad\quad \vdots \quad &\quad \vdots \\ K_i\frac{\partial N_i}{\partial t} + v\frac{\partial N_i}{\partial z} + \lambda_i K_i N_i &= \lambda_{i-1} K_{i-1} N_{i-1} \end{aligned} \tag{4.75}$$

which is to be solved for $N_i = N_i(z,t)$ in the domain $0 < Z < \infty$, $0 < t < \infty$ subject to the side conditions that

$$N_i(0,t) = N_i^0 \phi_i(t), \; t > 0, \quad \text{with } \phi_i(t) \equiv 0, \; t < 0 \tag{4.76a}$$

$$N_i(z,0) = 0 \quad , \quad z > 0 \tag{4.76b}$$

The boundary data $\phi_i(t)$ are arbitrary integrable functions which describe the release of the members of the chain from the repository.

The general form of the recursive equation system (4.75) is

$$\frac{\partial N_i}{\partial t} + v_i\frac{\partial N_i}{\partial z} + \lambda_i N_i = \frac{K_{i-1}}{K_i}\lambda_{i-1} N_{i-1} \quad \text{with } \lambda_0 = 0 \tag{4.77}$$

where $v_i = v/K_i$

We assume that $N_{i-1}(z,t)$ is known and show how to compute $N_i(z,t)$. We construct the solution in two different forms, each of which exhibits certain advantages in demonstrating certain physical properties of the solution.

A. Integral Transform Method

Consider the Laplace transform of $N_i(z,t)$ with respect to z and define

$$n_i(p,t) = \int_0^\infty e^{-pz} N_i(z,t)\,dz$$

$$n_{i-1}(p,t) = \int_0^\infty e^{-pz} N_{i-1}(z,t)\,dz \qquad (4.78)$$

The application of the transform operator to the equation (4.77) and use of equation (4.76a) yields

$$\frac{dn_i(p,t)}{dt} + (pV_i + \lambda_i)n_i(p,t) = \frac{K_{i-1}}{K_i}\lambda_{i-1}n_{i-1}(p,t) + v_i N_i^0 \phi_i(t) \qquad (4.79)$$

This ordinary differential equation is solved subject to the initial condition $n_i(p,o)$ which results from the application of equation (4.76b). The solution is

$$n_i(p,t) = \int_0^t \left\{ \frac{K_{i-1}}{K_i}\lambda_{i-1}n_{i-1}(p,t-\tau) + v_i N_i^0 \phi_i(t-\tau) \right\} e^{-(pV_i+\lambda_i)\tau}\,d\tau \qquad (4.80)$$

To invert $n_i(p,t)$ we make use of the "shift" rule and apply it to the first term on the right hand side.

$$\mathcal{L}^{-1}\left\{ \int_0^t \frac{K_{i-1}}{K_i} \lambda_{i-1} n_{i-1}(p, t-\tau) e^{-pv_i\tau} e^{-\lambda_i\tau} d\tau \right\}$$

$$= \frac{K_{i-1}}{K_i} \lambda_{i-1} \int_0^t e^{-\lambda_i\tau} N_{i-1}(z - v_i\tau, t-\tau) d\tau$$

Provided $v_i > 0$

The second term in (4.80) is seen to be a Laplace transform if as stated above $\phi_i(t) \equiv 0$ for $t < 0$. Then:

$$v_i N_i^0 \int_0^t \phi_i(t-\tau) e^{-pv_i\tau} e^{-\lambda_i\tau} d\tau = v_i N_i^0 \int_0^\infty e^{-pv_i\tau} e^{-\lambda_i\tau} \phi_i(t-\tau) d\tau$$

$$= N_i^0 \int_0^\infty e^{-pz} e^{-\frac{\lambda_i}{v_i} z} \phi_i\left(t - \frac{z}{v_i}\right) dz$$

This shows that the second contribution in (4.80) has the inverse

$$\mathcal{L}^{-1}\left\{ v_i N_i^0 \int_0^t \phi_i(t-\tau) e^{-(pv_i + \lambda_i)\tau} d\tau = N_i^0 e^{-\frac{\lambda_i}{v_i} z} \phi_i\left(t - \frac{z}{v_i}\right)\right.$$

Thus the complete inverse of (4.80) is

$$N_i(z,t) = N_i^0 e^{-\frac{\lambda_i}{v_i}z}\phi_i\left(t-\frac{z}{v_i}\right) + \frac{K_{i-1}}{K_i}\lambda_{i-1}\int_0^t e^{-\lambda_i\tau} N_{i-1}(z-v_i\tau, t-\tau)\, d\tau \tag{4.81}$$

Now the precursor concentration $N_{i-1}(z,t) \equiv 0$ for $t < 0$, $z < o$. Hence, the upper limit in the integral can be changed to $\mathrm{Min}(t,z/v_i)$ so that the final result reads

$$N_i(z,t) = N_i^0 e^{-\frac{\lambda_i}{v_i}z}\phi_i\left(t-\frac{z}{v_i}\right) + \frac{K_{i-1}}{K_i}\lambda_{i-1}\int_0^t e^{-\lambda_i\tau} N_{i-1}(z-v_i\tau, t-\tau)\, d\tau \tag{4.82}$$

The equation yields the concentration of the i^{th} member of the chain in terms of the $(i-1)^{th}$ member and can hence be used in a recursive manner. Starting with the mother concentration $N_1(z,t)$ solved from the first of the equations (4.75) one computes from (4.82), $N_2(z,t)$ etc.

The physical interpretation of the solution is simple. The first term on the right hand side represents those i^{th} nuclides that have been convected from the boundary ($z = 0$) and have escaped decay (the exponential attenuation term) while propagating along the characteristic $t-Z/v_i$ = constant.

The second term represents those i^{th} nuclides which are contributed from the $(i-1)^{th}$ nuclides lying along the characteristic $z - v_i\tau$ = constant with their exponential attenuation.

B. Classical Method

We observe from (4.77) that the equation $N_1(z,t)$ has the simple solution

$$N_1(z,t) = N_1^0 e^{-\frac{\lambda_i}{v_1}} \phi_1(t-\frac{z}{v_1}) \tag{4.83}$$

This can be verified by direct substitution into (4.76) and (4.77). This in turn suggests that we try for the i^{th} nuclide concentration the generalization

$$N_i(z,t) = \sum_{r=1}^{i} e^{-\frac{\lambda_r}{v_r}z} N_{i,r}(t-\frac{z}{v_r}) \tag{4.84}$$

The functions $N_{i,r}\,(t-z/v_r)$ are at this point unknown and are to be found. For $i = 1$, (4.84) reduces to

$$N_1(z,t) = e^{-\frac{\lambda_1}{v_1}z} N_{1,1}(t-\frac{z}{v_1}) \tag{4.85}$$

which identifies

$$N_{1,1}(t-\frac{z}{v_1}) = N_1^0 \phi_1(t-\frac{z}{v_1}) \tag{4.86}$$

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To find the functions $N_{i,r}(t-z/v_r)$ lets substitute the proposed solution form (4.84) into (4.74) with $N_{i,r}' \equiv \frac{dN_{i,r}}{d(t-z/v_r)}$.

$$\sum_{r=1}^{i} e^{-\frac{\lambda_r}{v_r}z}\left[1-\frac{v_i}{v_r}\right]N_{i,r}'\left(t-\frac{z}{v_r}\right)+\sum_{r=1}^{i} e^{-\frac{\lambda_r}{v_r}z}\left[\lambda_i-\frac{v_i}{v_r}\lambda_r\right]N_{i,r}\left(t-\frac{z}{v_r}\right)$$

$$=\frac{K_{i-1}}{K_i}\lambda_{i-1}\sum_{r=1}^{i-1} e^{-\frac{\lambda_r}{v_r}z}N_{i-1,r}\left(t-\frac{z}{v_r}\right)$$

(4.87)

For r = i, the terms on the left hand side of (4.87) vanish so with $t-z/v_r = T$ there results on equating the coefficients of the exponential terms,

$$\left[\frac{1}{v_i}-\frac{1}{v_r}\right]N_{i,r}'(\tau)+\left[\frac{\lambda_i}{v_i}-\frac{\lambda_r}{v_r}\right]N_{i,r}(\tau)=\frac{K_{i-1}\lambda_{i-1}}{v}N_{i-1,r}(\tau)$$

for $r = 1, 2, \ldots, (i-1)$ (4.88)

The initial condition (4.76b) is satisfied in view of (4.84) if one chooses

$$N_{ir}(\tau) = 0 \quad ; \quad \tau \leq 0 \tag{4.89}$$

The boundary condition (4.76a) is satisfied if

$$\sum_{r=1}^{i} N_{i,r}(t) = N_i^0\,\phi_i(t) \tag{4.90}$$

when

$$t > 0 \quad \text{and} \quad z = 0$$

We now solve the equation system (4.88) - (4.90) recursively. Let

$$\frac{\frac{\lambda_i}{v_i} - \frac{\lambda_r}{v_r}}{\frac{1}{v_i} - \frac{1}{v_r}} = \Delta_{ir} \tag{4.91}$$

then (4.88) can be written as

$$\left(\frac{1}{v_i} - \frac{1}{v_r}\right)\frac{d}{d\tau}\left[e^{\Delta_{ir}\tau} N_{i,r}(\tau)\right] = \lambda_{i-1}\frac{K_{i-1}}{v}\left[e^{\Delta_{ir}\tau} N_{i-1,r}(\tau)\right] \tag{4.92}$$

which has the solution subject to the initial condition (4.89).

$$N_{i,r}(\tau) = \frac{\lambda_{i-1}K_{i-1}}{K_i - K_r} e^{-\Delta_{ir}\tau} \int_0^\tau N_{i-1,r}(\tau') e^{\Delta_{ir}\tau'} d\tau' \,, \quad r = 1, 2, \cdots, (i-1) \tag{4.93}$$

The remaining member $N_{i,i}(\tau)$ is then calculated from (4.84) and (4.90) as

$$N_{i,i}(t) = N_i^0 \phi_i(t) - \sum_{r=1}^{i-1} N_{i,r}(t) \tag{4.94}$$

(4.93) and (4.94) determine the $N_{i,r}$ functions recursively from those of $N_{i-1,r}$ and with this the solution for $N_i(z,t)$ is completed. The variables τ and t in these equations are dummy variables. In order to keep the conditions in equation (4.89) clearly in mind, one must multiply the $N_{i,r}$ functions by unit step functions. The finial solution then takes on the form

$$N_i(z,t) = \sum_{r=1}^{i-1} \left\{ e^{-\left(\frac{\lambda_r}{v_r}\right)z} N_{i,r}\left(t-\frac{z}{v_r}\right) h\left(t-\frac{z}{v_r}\right) - e^{-\left(\frac{\lambda_i}{v_i}\right)z} N_{i,r}\left(t-\frac{z}{v_i}\right) h\left(t-\frac{z}{v_i}\right) \right\}$$

$$+ N_i^0 \, e^{-\left(\frac{\lambda_i}{v_i}\right)z} \phi_i\left(t-\frac{z}{v_i}\right) \qquad (4.95)$$

The physical interpretation of the solution is identical to that discussed in Section A. The first term (sum) represents those i^{th} nuclides which are contributed from all nuclides r = 1,2,..., (i-1) with their exponential attenuation while the last term represents those i^{th} nuclides that have been convected from the boundary and have escaped decay.

An important additional feature of the solution (4.95) is that it allows one to find the range in the t and z variables in which the solution is non-trivial. For this consider the first term on the right hand side of (4.95). The term in the bracket vanishes when both step functions vanish. This will occur for a time value smaller than the smallest of the values, (Z/v_λ), k = 1, 2, .., f, ...i). Suppose this smallest value occurs for the migration velocity v_f, then Z/v_f will represent the first arrival time of the nuclides at position z. If furthermore, $v_f > v_i$, the second term on the right hand side of (4.95) is also zero. Consequently, we have

$$N_i(z,t) = 0 \quad \text{for} \quad t \leq \frac{z}{v_f} \tag{4.96}$$

This can be represented graphically at a fixed z position as shown by Figure 4.2.

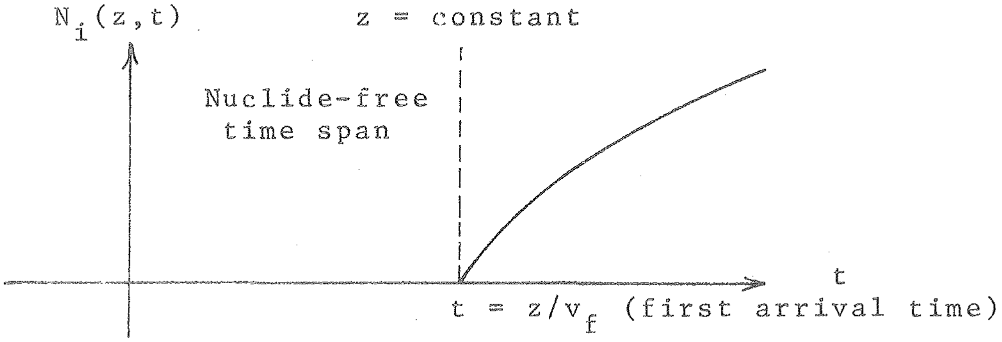

Figure 4.2 A scheme of the concentration against time at fixed position

On the other hand, at a fixed time the above result can be written as

$$N_i(z,t) = 0 \quad , \qquad z \geq v_f t \tag{4.97}$$

Physically $z < v_f t$ defines the region that has been contaminated at time t and $z = v_f t$ represents the leading edge of the contaminant wave. This is illustrated by Fig. 4.3.

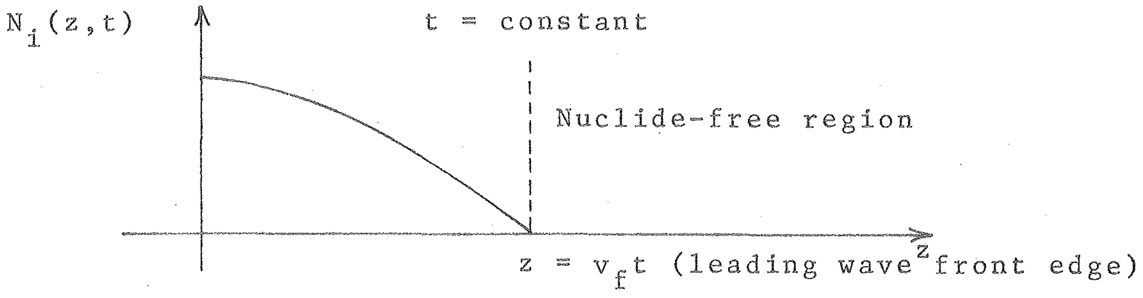

Figure 4.3 A scheme of the concentration profile against distance at fixed time

Furthermore, the rate at which $N_i(z,t)$ rises (from zero) at the first arrival time at z = constant or at the leading wave front edge at t = constant is controlled by the term

$$N_{i,f}\left(t-\frac{z}{v_f}\right) e^{-\left(\frac{\lambda_f}{v_f}\right)z} h\left(t-\frac{z}{v_f}\right)$$

The numerical calculation for three member chain shown in the later chapter verify all of the above conclusion.

In the case of an impulse release at z = 0 one can, in addition, also determine the cut-off in the arrival time (at fixed z) and the trailing wave front edge (at constant t) of the contaminant wave quite easily. The solution for the mother concentration is in view of (4.85) and (4.86).

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$$N_1(z,t) = N_1^0 e^{-\left(\frac{\lambda_1}{v_1}\right)z} \delta\left(t - \frac{z}{v_1}\right) \tag{4.98}$$

With $N_{1,1}$ given by (4.86), (4.91) yields

$$N_{2,1}(\tau) = N_1^0 \frac{\lambda_1 K_1}{K_2 - K_1} e^{-\Delta_{12}\tau} \tag{4.99}$$

so that

$$N_2(z,t) = N_1^0 \frac{\lambda_1 K_1}{K_2 - K_1} e^{-\left(\frac{\lambda_1 - \lambda_2}{v_1 - v_2}\right)z - \Delta_{12} t} \times \left\{ h\left(t - \frac{z}{v_1}\right) - h\left(t - \frac{z}{v_2}\right) \right\} + N_2^0 e^{-\left(\frac{\lambda_2}{v_2}\right)z} \delta\left(t - \frac{z}{v_2}\right) \tag{4.100}$$

In a comparable way, one can write down at once the expression for $N_3(z,t)$. Since the bracketed term divided by $(K_2 - K_1)$ is always positive, the contribution to N_2 from the mother will be non-trivial for

$$v_1 t < z < v_2 t \quad \text{or} \quad v_2 t < z < v_1 t \tag{4.101}$$

Outside this range $N_2(z,t) \equiv 0$. The edges of the wave fronts are immediately determined from $v_1 \lessgtr v_2$.

Finally we note that the wave front slopes are also completely determined in terms of the exponential attenuation factors.

4.4.2 The general solution of the transport equations without dispersion

Consider the canonical system

$$K_1 \frac{\partial N_1}{\partial t} + v \frac{\partial N_1}{\partial z} + \lambda_1 K_1 N_1 = 0$$

$$K_2 \frac{\partial N_2}{\partial t} + v \frac{\partial N_2}{\partial z} + \lambda_2 K_2 N_2 = \lambda_1 K_1 N_1$$

$$\cdots$$

$$K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} + \lambda_i K_i N_i = \lambda_{i-1} K_{i-1} N_{i-1}$$

$$\cdots$$

$$(4.102)$$

which is to be solved for $N_i = N_i(z,t)$ in the domain $0<z<\infty$, $o<t<\infty$ subject to the side conditions that

$$N_i(o,t) = N_i^0 \phi_i(t),\ t>o,\ \text{with } \phi_i(t) \equiv 0,\ t<0 \qquad (4.103a)$$

$$N_i(z,0) = 0,\quad z>0 \qquad (4.103b)$$

The boundary data $\phi_i(t)$ are arbitrary integrable functions which describe the release of the members of the chain from the repository.

The general form of the equation system (4.102) is

$$\frac{\partial N_i}{\partial t} + v_i \frac{\partial N_i}{\partial z} + \lambda_i N_i = \frac{K_{i-1}}{K_i} \lambda_{i-1} N_{i-1} \text{, with } \lambda_0 = 0 \tag{4.104}$$

where

$$v_i = \frac{v}{K_i}$$

The aim is to obtain the general (i.e, non-recursive) solution for $N_i(z,t)$. For this purpose, consider the following i-sets of boundary conditions.

Nuclide chain member / Boundary Condition	$\ell = 1$	$\ell = 2$ ⋯	$\ell = \ell$ ⋯	$\ell = i$
$j=1$	$N_1^{(1)}(0,t)=N_1^0\phi_1(t)$	$N_2^{(1)}(0,t)=0$ ⋯	⋯ $N_\ell^{(1)}(0,t)=0$ ⋯	⋯ $N_i^{(1)}(0,t)=0$
$j=2$	$N_1^{(2)}(0,t)=0$	$N_2^{(2)}(0,t)=N_2^0\phi_2(t)$ ⋯	⋯ $N_\ell^{(2)}(0,t)=0$ ⋯	⋯ $N_i^{(2)}(0,t)=0$
⋮	⋮	⋮	⋮	⋮
$j=\ell$	$N_1^{(\ell)}(0,t)=0$	$N_2^{(\ell)}(0,t)=0$ ⋯	⋯ $N_\ell^{(\ell)}(0,t)=N_\ell^0\phi_\ell(t)$ ⋯	⋯ $N_i^{(\ell)}(0,t)=0$
⋮	⋮	⋮	⋮	⋮
$j=j$	$N_1^{(i)}(0,t)=0$	$N_2^{(i)}(0,t)=0$ ⋯	⋯ $N_\ell^{(i)}(0,t)=0$ ⋯	⋯ $N_i^{(i)}(0,t)=N_i^0\phi_i(t)$

(4.105)

The solutions to (4.103b) and (4.104) subject to the conditions on a line j of the above set of boundary conditions i denoted by $N_\ell^{(j)}(z,t)$, $\ell = 1,2,\ldots,i$.

On account of the linearity of (4.104) the sum of the solutions for all these individual boundary value problems for each value of (j) will be the solution to the complete problem described by (4.103a,b) and (4.104), i.e.,

$$N_\ell(z,t) = N_\ell^{(\ell)}(z,t) + \sum_{j=1}^{\ell-1} N_\ell^{(j)}(z,t) \quad , \; \ell = 1,2,\cdots,i \qquad (4.106)$$

The reader should note that some of the $N_\ell^{(j)}$ functions vanish. For instance, if say $N_1^{(2)}(o,t) = 0$ but not $N_2^{(2)}(o,t)$, which is the case for the second set of boundary data (j = 2) in (4.105) , then $N_1^{(2)}(z,t) \equiv 0$ for all $z,t>0$. Analogously if $N_1^{(3)}(o,t)$ and $N_2^{(3)}(o,t)$ are both zero then $N_1^{(3)}(z,t) \equiv N_2^{(3)}(z,t) \equiv 0$ for $z,t>0$. In general there will be no precursor for the functions $N_\ell^{(j)}(z,t)$ thus $N_{\ell-1}^{(j)}(z,t) = 0$ for $\ell \leq j$. For each set $j = 1,2,\ldots,i$ of (4.105) the equation to be solved for each nuclide $N_\ell^{(j)}$ is

$$\frac{\partial N_\ell^{(j)}}{\partial t} + v_\ell \frac{\partial N_\ell^{(j)}}{\partial z} + \lambda_\ell N_\ell^{(j)} = \frac{K_{\ell-1}}{K_\ell} \lambda_{\ell-1} N_{\ell-1}^{(j)} \qquad (4.107a)$$

where $\ell = 1,2,\ldots,i$ and $N_{\ell-1}^{(j)} = 0$ when $\ell \leq j$. (4.107b)

The boundary conditions (4.105) can be expressed in a more compact form as

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$$N_\ell^{(j)}(0,t) = \begin{cases} 0 \quad , & \ell \neq j \\ N_j^0 \phi_j(t), & \ell = j \,,\ t > 0, \ (\phi_j(t) \equiv 0, t < 0) \end{cases} \tag{4.107c}$$

while the initial condition is

$$N_\ell^{(j)}(z,0) = 0 \quad , \quad z > 0 \tag{4.107d}$$

The functions $N_\ell^{(j)}(z,t)$ are obtained by a repeated Laplace transform with respect to t and z of (4.107 a,b).

Let

$$n_\ell^{(j)}(z,s) = \int_0^\infty e^{-st} N_\ell^{(j)}(z,t)\,dt, \quad \phi_j(s) = \int_0^\infty e^{-st} \phi_j(t)\,dt \tag{4.108}$$

$$n_\ell^{(j)}(p,s) = \int_0^\infty e^{-pz} n_\ell^{(j)}(z,s)\,dz \tag{4.109}$$

There results from (4.107 a,b,c,d)

$$p\,n_\ell^{(j)}(p,s) - N_\ell^0 \phi_\ell(s) + \mu_\ell n_\ell^{(j)}(p,s) = \nu_{\ell-1} n_{\ell-1}^{(j)}(p,s), \qquad j, \ell = 1, 2, \cdots, i \tag{4.110a}$$

$$n_{\ell-1}^{(j)}(p,s) \equiv 0 \quad , \quad \ell \leqslant j \tag{4.110b}$$

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where

$$\mu_\ell = \frac{s+\lambda_\ell}{v_\ell} \quad , \nu_{\ell-1} = \frac{K_{\ell-1}}{K_\ell}\frac{\lambda_{\ell-1}}{v_\ell} \quad , \lambda_0 = 0 \tag{4.110c}$$

Equation (4.107c) implies that

$$N_\ell^0 = \begin{cases} 0, & \ell \neq j \\ N_j^0, & \ell = j \end{cases} \tag{4.110d}$$

We solve (4.110a) in a recursive manner from $\ell = 1$ to $\ell = i$ for each value of j; j runs from j = 1 to j = i since we are interested in the i^{th} nuclide of a given chain.

Starting with j = 1 and letting ℓ run through the values 1,2,...,i one takes from (4.110b) $n_o^{(1)}$ (p,s) = 0 from (4.110d) $N_2^o = N_3^o = ------ = N_i^o = 0$. Therefore, (4.110a) yields in succession

$$n_1^{(1)}(p,s) = \frac{N_1^0 \phi_1(s)}{p+\mu_1}$$

$$n_2^{(1)}(p,s) = \frac{\nu_1 n_1^{(1)}(p,s)}{p+\mu_2} = \frac{\nu_1 N_1^0 \phi_1(s)}{(p+\mu_1)(p+\mu_2)}$$

$$\vdots$$

$$n_i^{(1)}(p,s) = \frac{\nu_1\nu_2\cdots\nu_{i-1} \quad N_1^0\phi_1(s)}{(p+\mu_1)(p+\mu_2)\cdots(p+\mu_{i-1})} \tag{4.111}$$

Next one takes $j = 2$ and again lets ℓ run through the values 1,2, ---, i. From (4.110) and (4.110d) one has $n_o^{(2)}(p,s) = n_1^{(2)}(p,s) = 0$ and $N_3{}^o = N_4{}^o = \text{---} = N_i{}^o = 0$ respectively. Hence (4.110a) yields

$$
\begin{aligned}
n_2^{(2)}(p,s) &= \frac{N_2^0 \phi_2(s)}{p + \mu_2} \\
n_3^{(2)}(p,s) &= \frac{\nu_2 n_2^{(2)}(p,s)}{p + \mu_3} = \frac{\nu_2 N_2^0 \phi_2(s)}{(p+\mu_2)(p+\mu_3)} \\
&\vdots \\
n_i^{(2)}(p,s) &= \frac{\nu_2 \nu_3 \cdots \nu_{i-1} \quad N_2^0 \phi_2(s)}{(p+\mu_2)(p+\mu_3)\cdots(p+\mu_i)}
\end{aligned}
\tag{4.112}
$$

Continuing in this manner one shows that in general

$$
n_i^{(j)}(p,s) = \frac{A_i^{(j)} N_j^0 \phi_j(s)}{\prod_{m=j}^{i} (p + \mu_m)} \quad , \quad j \neq i \tag{4.113}
$$

where $A_i^{(j)} = \prod_{r=j}^{i-1} \nu_r$, while for $j = i$

$$
n_i^{(i)}(p,s) = \frac{N_i^0 \phi_i(s)}{p + \mu_i} \tag{4.114}
$$

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By partial fraction expansion one can rewrite (4.113) as

$$n_i^{(j)}(p,s) = A_i^{(j)} N_j^0 \phi_j(s) \sum_{m=j}^{i} \frac{C_m^{(j)}(s)}{p+\mu_m} \qquad (4.115)$$

provided that the μ_m are all unequal. Here

$$C_m^{(j)}(s) = \left[\prod_{\substack{r=j \\ r\neq m}}^{i} (\mu_r - \mu_m) \right]^{-1} \qquad (4.116)$$

On inversion of (4.114) and (4.115) with respect to z one obtains

$$n_i^{(i)}(z,s) = N_i^0 e^{-\mu_i z} \phi_i(s) \qquad (4.117)$$

$$n_i^{(j)}(z,s) = A_i^{(j)} N_j^0 \phi_j(s) \sum_{m=j}^{i} C_m^{(j)}(s) e^{-\mu_m z} \qquad (4.118)$$

Since

$$\mu_r - \mu_m = \Gamma_{rm}(s + \Delta_{rm}) \qquad (4.119)$$

where

$$\Gamma_{rm} = \frac{v_m - v_r}{v_m v_r}, \quad \Delta_{rm} = \frac{\lambda_r v_m - \lambda_m v_r}{v_m - v_r}$$

(4.118) becomes with help of (4.112), (4.116) and (4.119)

$$n_i^{(j)}(z,s) = A_i^{(j)} N_i^0 \sum_{m=j}^{i} \frac{\phi_j(s)\, e^{-\left(\frac{s+\lambda_m}{v_m}\right)z}}{\prod\limits_{\substack{r=j\\ r\neq m}}^{i} \Gamma_{rm} \prod\limits_{\substack{r=j\\ r\neq m}}^{i} (s+\Delta_{rm})} \tag{4.120}$$

We transform the right hand side prior to inversion with respect to t. Let

$$\prod_{\substack{r=j\\ r\neq m}}^{i} \Gamma_{rm} = B_m^{(j)} \quad , \quad \frac{1}{\prod\limits_{\substack{r=j\\ r\neq m}}^{i} (s+\Delta_{rm})} = \sum_{\substack{r=j\\ r\neq m}}^{i} \frac{D_{rm}^{(j)}}{s+\Delta_{rm}} \tag{4.121}$$

where by partial fraction expansion

$$D_{rm}^{(j)} = \left[\prod_{\substack{q=j\\ q\neq m\neq r}}^{i} \left(\Delta_{qm} - \Delta_{rm} \right) \right]^{-1} \tag{4.122}$$

where $D_{r,m}^{(i-1)} = 1$.

With these (4.120) reads:

$$n_i^{(j)}(z,s) = A_i^{(j)} N_j^0 \sum_{m=j}^{i} \frac{e^{-(\lambda_m/v_m)z}}{B_m^{(j)}} \sum_{\substack{r=j\\ r\neq m}}^{i} \frac{D_{rm}^{(j)} \phi_i(s)\, e^{-(z/v_m)s}}{s+\Delta_{rm}} \qquad (4.123)$$

By the shift rule

$$\mathcal{L}^{-1}\left\{ \frac{e^{-(z/v_m)s}}{s+\Delta_{rm}} \right\} = g_{rm}(t) \equiv \begin{cases} 0 & , \; t < z/v_m \\ e^{-\Delta_{rm}(t - z/v_m)} & , \; t > \frac{z}{v_m} \end{cases}$$

(4.124)

so that by the convolution rule

$$N_i^{(j)}(z,t) = A_i^{(j)} N_j^0 \sum_{m=j}^{i} \frac{e^{-(\lambda_m/v_m)z}}{B_m^{(j)}} \sum_{\substack{r=j\\ r\neq m}}^{i} D_{rm}^{(j)} \left(g_{rm}(t) \otimes \phi_j(t)\right)$$

(4.125)

The inverse of (4.117) is

$$N_i^{(i)}(z,t) = N_i^0 \, e^{-(\lambda_i/v_i)z} \, \phi_i(t - z/v_i) \qquad (4.126)$$

In view of the restriction placed on $\phi_i(t)$ in (4.103). The substitution of (4.125) and (4.126) into (4.106) yields the general (non-recursive) solution

$$N_i(z,t) = N_i^0 e^{-(\lambda_i/v_i)z} \phi_i(t - z/v_i)$$
$$+ \sum_{j=1}^{i-1} A_i^{(j)} N_j^0 \sum_{m=j}^{i} \frac{e^{-(\lambda_m/v_m)z}}{B_m^{(j)}} \sum_{\substack{r=j \\ r \neq m}}^{i} D_{rm}^{(j)} \left(g_{rm}(t) \circledast \phi_j(t) \right) \tag{4.127}$$

Again the first term represents those ith nuclides that have been convected from the boundary (z = o) and have escaped decay. The triple sum represents those ith nuclides contributed from all precursors.

4.5 Local Secular Equilibrium

There are some decay chains in which very rapidly decaying nuclides are involved. For example, one of the most important decay chains is

$$^{241}\mathrm{Pu}_{13y} \rightarrow {}^{241}\mathrm{Am}_{458y} \rightarrow {}^{237}\mathrm{Np}_{2.14 \times 10^6 y} \rightarrow {}^{233}\mathrm{Pa}_{27d} \rightarrow {}^{233}\mathrm{U}_{1.62 \times 10^5 y} \rightarrow {}^{229}\mathrm{Th}_{7340y} \rightarrow {}^{225}\mathrm{Ra}_{15d} \rightarrow \tag{4.128}$$

For long term prediction of nuclide migration, almost all ^{241}Pu and ^{241}Am are reduced to ^{237}Np. Thus in this decay chain nuclide ^{237}Pa and ^{225}Ra are much smaller than those of the others. For this chain, we know that the secular equilibrium state in the decay process can be attained without migration. In this section, we will verify that the secular state is also attained in the migration process under some conditions.

The solution of migration equation in recursive form is given by (4.32).

$$N_i(z,t) = \int_0^t f_i(t-\theta_i)\, e^{-\lambda_i\theta_i} F(v_i\theta_i,\, z-v_i\theta_i)\, d\theta_i$$

$$+ \frac{v_i\lambda_{i-1}}{v_{i-1}} \int_0^t \int_{-\infty}^{\infty} N_{i-1}(\xi, t-\theta_i)\, e^{-\lambda_i\theta_i} F(v_i\theta_i,\, z-\xi-v_i\theta_i)\, d\xi\, d\theta_i \qquad (4.32)$$

where

$$F(v_i\theta_i, z-\xi-v_i\theta_i) = \frac{1}{4\pi\kappa v_i\theta_i} \exp\left[-\frac{(z-\xi-v_i\theta_i)^2}{4\kappa v_i\theta_i}\right]$$

Let us consider the case that the decay constant for the i^{th} nuclide is much larger than that for the (i-1)-th nuclide:

$$\lambda_i \gg \lambda_{i-1} \qquad (4.129)$$

The time interval, t, of our interest is of the order $1/\lambda_{i-1}$. Then, the following equation is satisfied.

$$1/\lambda_i \ll t\ ,\quad 1/\lambda_{i-1} \qquad (4.130)$$

The $\exp(-\lambda_i\Theta_i)$ value in (4.32) is nearly zero in the range except for the shorter time $0<\Theta_i<O(1/\lambda_i)$, because of the high value of λ_i. This is illustrated in Figure 4.4. The function, $F(V_i\Theta_i, Z-\xi-V_i\Theta_i)$, in (4.32) is approximated by a delta function, $\delta(z-\xi)$. This approximation is valid under the conditions, $\overline{AB} >> \overline{BC}$ and $4\kappa v_i\overline{OA} << Z^2$, where point A is a location having an order $(1/\lambda_i)$ at $\xi = 0$ and the point B is a point, $(z, \overline{OA})$. C is a point on the straight line, $z - v_i\Theta_i = \xi$ at $\Theta_i = \overline{OA}$. The effect of the above conditions means that $F(v_i\Theta_i, z-\xi-v_i\Theta_i)$ can be approximated by a delta function at the point C. Since $\overline{OA}$ is of the order $1/\lambda_i$, $\overline{AB} = z$ and $\overline{BC} = v_i\overline{OA}$, the above two conditions are rewritten as

$$z \gg v_i/\lambda_i \;, \quad 4\kappa v_i/\lambda_i \ll z^2 \qquad (4.131)$$

Under the conditions, (4.131), the second term in (4.32) is given by

$$\frac{v_i\lambda_{i-1}}{v_{i-1}}\int_0^t\int_{-\infty}^{\infty} N_{i-1}(\xi, t-\theta_i)e^{-\lambda_i\theta_i}F(v_i\theta_i, z-\xi-v_i\theta_i)\,d\xi\,d\theta_i$$

$$\simeq \frac{v_i\lambda_{i-1}}{v_{i-1}}\int_0^t N_{i-1}(z, t-\theta_i)\,e^{-\lambda_i\theta_i}\,d\theta_i$$

$$\simeq \frac{v_i\lambda_{i-1}}{v_{i-1}}\; N_{i-1}(z,t)\int_0^{\infty} e^{-\lambda_i\theta_i}d\theta_i = \frac{v_i\lambda_{i-1}}{v_{i-1}\lambda_i}N_{i-1}(z,t) \qquad (4.132)$$

In the above derivation, we used the relation that $N_{i-1}(z,t)$ is almost constant in the range of the order, $1/\lambda_i$. The first term in the right hand side of (4.32) can be reduced to

$$\int_0^t f_i(t-\theta_i)\, e^{-\lambda_i\theta_i} F(v_i\theta_i, Z-v_i\theta_i)\, d\theta_i = \delta(Z)\int_0^t e^{-\lambda_i\theta_i} f_i(t-\theta_i)\, d\theta_i \qquad (4.133)$$

because $F(v_i\Theta_i, Z-v_i\Theta_i)$ is approximated as $\delta(z)$. Then the first term can be neglected in the range, $z>0$.

From (4.132) and (4.133), we can obtain

$$\frac{\lambda_i N_i(z,t)}{v_i} = \frac{\lambda_{i-1} N_{i-1}(z,t)}{v_{i-1}} \qquad (4.134)$$

or

$$\lambda_i K_i N_i(z,t) = \lambda_{i-1} K_{i-1} N_{i-1}(z,t)$$

(4.134) shows that the local secular equilibrium is attained between the i^{th} and the (i-1)-th nuclides. The necessary conditions are summarized as:

$$\lambda_{i-1} \ll \lambda_i, \quad z \gg v_i/\lambda_i, \quad \lambda_i z^2/v_i \gg 4\kappa, \quad t\lambda_i \gg 1 \qquad (4.135)$$

$N_{i+1}(z,t)$ is obtained from (4.32):

$$N_{i+1}(z,t) = \int_0^t f_{i+1}(t-\theta)\, e^{-\lambda_{i+1}\theta} F(v_{i+1}\theta, z-v_{i+1}\theta)\, d\theta$$

$$+ \frac{\lambda_i v_{i+1}}{v_i}\int_0^t\int_{-\infty}^{\infty} N_i(\xi, t-\theta)\, e^{-\lambda_{i+1}\theta} F(v_{i+1}\theta, z-\xi-v_{i+1}\theta)\, d\xi\, d\theta \qquad (4.136)$$

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Introduction of (4.134) into (4.136) yields

$$N_{i+1}(z,t) = \int_0^t f_{i+1}(t-\theta)\, e^{-\lambda_i \theta} F(v_{i+1}\theta,\, z - v_{i+1}\theta)\, d\theta$$

$$+ \frac{v_{i+1}\lambda_{i-1}}{v_{i-1}} \int_0^t d\theta \int_{-\infty}^{\infty} d\xi\, N_{i-1}(\xi, t-\theta)\, e^{-\lambda_{i+1}\theta} F(v_{i+1}\theta,\, z-\xi-v_{i+1}\theta) \qquad (4.137)$$

(4.137) shows that the n member decay chain (...,i-1, i, i+1, ..., n) is reduced to the (n-1) member decay chain (..., i-1, i+1, ... n).

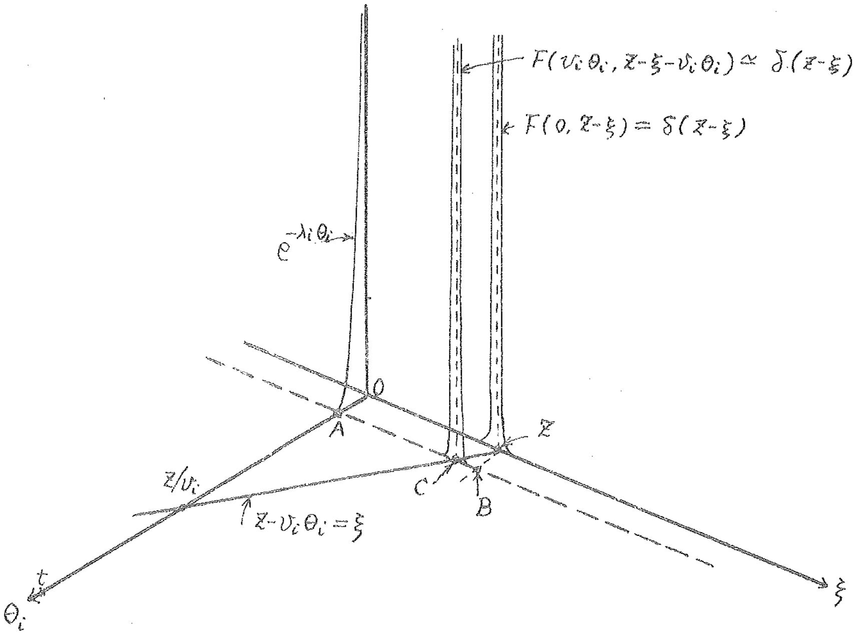

Figure 4.4 Illustrative functional behavior for local secular equilibrium.

5. APPLICATION OF THE GENERAL SOLUTION

5.1 Introduction

In the preceding chapters, we have discussed the different release modes and the relationship among them and also have obtained the general solutions of the transport equation for arbitrary release modes.

In this chapter, the solutions for various release modes will be shown with the help of the general solution. Section 5.2 is concerned with the dispersion free case. In this section, the solutions governing the migration of an i - member decay chain will be shown for several release modes. In sections 5.3 and 5.4, the solutions for a three member decay chain with dispersion are shown for the step and the band release modes. Sections 5.5 and 5.6 will discuss the numerical results based on these solutions. The migration features of some important decay chains will be discussed in detail.

5.2 Application of the General Solution of the Transport Equation Without Dispersion -- Solutions for Several Release Modes --

In the preceding chapter, the general solution of the transport equation without dispersion was shown in an explicit form:

$$N_i(z,t) = e^{-\lambda_i z/v_i} N_i\left(t-\frac{z}{v_i}\right) + \sum_{l=1}^{i-1}\left(\prod_{r=l}^{i-1}\frac{\lambda_r}{v_r}\right)\sum_{m=l}^{i}\frac{e^{-\lambda_m z/v_m}}{\prod_{\substack{r=l\\r\neq m}}^{i}\Gamma_{rm}}\sum_{\substack{r=l\\r\neq m}}^{i} D_{rm}^{il}\, g_{rm}(t)\otimes N_l(t) \tag{5.1}$$

where

$$g_{rm}(t) = \begin{cases} 0 & , \quad t < z/v_m \\ e^{-\Delta_{rm}(t-z/v_m)} & , \quad t > z/v_m \end{cases}$$

In this section, we propose the solutions for several release modes with the help of (5.1) and the relationship among these modes which has been discussed in section 3.2.

5.2.1 Solution for preferential release mode

The preferential release mode has been defined in section 3.2 and is given by:

$$N_i^p(0,t) = N_i(t) = \sum_{j=1}^{i} B_{ij}' \, e^{-\Lambda_j t}\, h(t) \tag{3.22}$$

$$B_{ij}' = \frac{\sum_{m=1}^{j}\left(\frac{k_i}{k_m}N_m^0\right)\frac{1}{\lambda_i}\prod_{r=m}^{i}\lambda_r}{\sum_{\substack{l=m\\l\neq j}}^{i}(\Lambda_l-\Lambda_j)} \tag{3.23}$$

The concentration of the i-th member can be obtained by introducing (3.22) into (5.1)

$$N_i^p(z,t) = e^{-\lambda_i z/v_i} h(t - z/v_i) \sum_{j=1}^{i} B'_{ij} e^{-\Lambda_j (t - z/v_i)}$$
$$+ \sum_{l=1}^{i-1} \left(\prod_{r=l}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=l}^{i} \frac{e^{-\frac{\lambda_m}{v_m} z} h(t - z/v_m)}{\prod_{\substack{r=l \\ r \neq m}}^{i} \Gamma_{rm}}$$
$$\times \sum_{\substack{r=l \\ r \neq m}}^{i} \sum_{j=1}^{l} \frac{D_{rm}^{il} B'_{lj}}{\Lambda_j - \Delta_{rm}} \left\{ e^{-\Delta_{rm}(t - z/v_m)} - e^{-\Lambda_j (t - z/v_m)} \right\}$$

(5.2)

For three members, the above solution is as follows:

$$N_1^p(z,t) = B'_{11} e^{-\Lambda_1 (t - z/v_1) - \lambda_1 z/v_1} \times h(t - \frac{z}{v_1}) \qquad (5.3)$$

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$$
\begin{aligned}
N_2^p(z,t) = {} & e^{-\lambda_2 \frac{z}{v_2}} h\left(t-\frac{z}{v_2}\right)\Big[B'_{21}\, e^{-\Lambda_1\left(t-\frac{z}{v_2}\right)} + B'_{22}\, e^{-\Lambda_2\left(t-\frac{z}{v_2}\right)}\Big] \\
& + \frac{\lambda_1}{v_1}\Big\{ h\left(t-\frac{z}{v_1}\right) \frac{e^{-\lambda_1\frac{z}{v_1}}}{\Gamma_{21}} \frac{D^{21}_{21} B'_{11}}{\Lambda_1-\Delta_{21}} \Big\{ e^{-\Delta_{21}\left(t-\frac{z}{v_1}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_1}\right)} \Big\} \\
& \quad + h\left(t-\frac{z}{v_2}\right) \frac{e^{-\lambda_2\frac{z}{v_2}}}{\Gamma_{12}} \frac{D^{21}_{12} B'_{11}}{\Lambda_1-\Delta_{12}} \Big\{ e^{-\Delta_{12}\left(t-\frac{z}{v_2}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_2}\right)} \Big\}\Big\}\Big)
\end{aligned}
\tag{5.4}
$$

$$
\begin{aligned}
N_3^p(z,t) = {} & e^{-\lambda_3\frac{z}{v_3}} h\left(t-\frac{z}{v_3}\right)\Big\{ B'_{31}\, e^{-\Lambda_1\left(t-\frac{z}{v_3}\right)} + B'_{32}\, e^{-\Lambda_2\left(t-\frac{z}{v_3}\right)} + B'_{33}\, e^{-\Lambda_3\left(t-\frac{z}{v_3}\right)} \Big] \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2} h\left(t-\frac{z}{v_1}\right) \frac{e^{-\lambda_1\frac{z}{v_1}}}{\Gamma_{21}\Gamma_{31}} \Big(\frac{D^{31}_{21} B'_{11}}{\Lambda_1-\Delta_{21}} \Big\{ e^{-\Delta_{21}\left(t-\frac{z}{v_1}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_1}\right)} \Big\} \\
& \qquad + \frac{D^{31}_{31} B'_{11}}{\Lambda_1-\Delta_{31}} \Big\{ e^{-\Delta_{31}\left(t-\frac{z}{v_1}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_1}\right)} \Big\}\Big) \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2} h\left(t-\frac{z}{v_2}\right) \frac{e^{-\lambda_2\frac{z}{v_2}}}{\Gamma_{12}\Gamma_{32}} \Big(\frac{D^{31}_{12} B'_{11}}{\Lambda_1-\Delta_{12}} \Big\{ e^{-\Delta_{12}\left(t-\frac{z}{v_2}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_2}\right)} \Big\} \\
& \qquad + \frac{D^{31}_{32} B'_{11}}{\Lambda_1-\Delta_{32}} \Big\{ e^{-\Delta_{32}\left(t-\frac{z}{v_2}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_2}\right)} \Big\}\Big) \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2} h\left(t-\frac{z}{v_3}\right) \frac{e^{-\lambda_3\frac{z}{v_3}}}{\Gamma_{13}\Gamma_{23}} \Big(\frac{D^{31}_{13} B'_{11}}{\Lambda_1-\Delta_{13}} \Big\{ e^{-\Delta_{13}\left(t-\frac{z}{v_3}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_3}\right)} \Big\} \\
& \qquad + \frac{D^{31}_{23} B'_{11}}{\Lambda_1-\Delta_{23}} \Big\{ e^{-\Delta_{23}\left(t-\frac{z}{v_3}\right)} - e^{-\Lambda_1\left(t-\frac{z}{v_3}\right)} \Big\}\Big)
\end{aligned}
$$

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$$+ \frac{\lambda_2}{v_2} h\left(t-\frac{z}{v_2}\right) \frac{e^{-\lambda_2 \frac{z}{v_2}}}{\Gamma_{32}} \left(\frac{D^{32}_{32} B'_{21}}{\Lambda_1 - \Delta_{32}} \left\{ e^{-\Delta_{32}(t-\frac{z}{v_2})} - e^{-\Lambda_1(t-\frac{z}{v_2})} \right\} \right.$$

$$\left. + \frac{D^{32}_{32} B'_{22}}{\Lambda_2 - \Delta_{32}} \left\{ e^{-\Delta_{32}(t-\frac{z}{v_2})} - e^{-\Lambda_2(t-\frac{z}{v_2})} \right\} \right)$$

$$+ \frac{\lambda_2}{v_2} h\left(t-\frac{z}{v_3}\right) \frac{e^{-\lambda_3 \frac{z}{v_3}}}{\Gamma_{23}} \left(\frac{D^{31}_{23} B'_{21}}{\Lambda_1 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t-\frac{z}{v_3})} - e^{-\Lambda_1(t-\frac{z}{v_3})} \right\} \right.$$

$$\left. + \frac{D^{32}_{23} B'_{22}}{\Lambda_2 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t-\frac{z}{v_3})} - e^{-\Lambda_2(t-\frac{z}{v_3})} \right\} \right) \tag{5.5}$$

where

$$B'_{11} = N^0_1 \;,\; B'_{21} = \frac{k_2}{k_1} \frac{\lambda_1 N^0_1}{\Lambda_2 - \Lambda_1} \;,\; B'_{22} = N^0_2 - \frac{k_2}{k_1} \frac{\lambda_1 N^0_1}{\Lambda_2 - \Lambda_1} \;,\; B'_{31} = \frac{k_3}{k_1} \frac{\lambda_1 \lambda_2 N^0_1}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1)}$$

$$B'_{32} = \frac{k_3}{k_1} \frac{\lambda_1 \lambda_2 N^0_1}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2)} + \frac{k_3}{k_2} \frac{\lambda_2 N^0_2}{\Lambda_3 - \Lambda_2}$$

$$B'_{33} = \frac{k_3}{k_1} \frac{\lambda_1 \lambda_2 N^0_1}{(\Lambda_1 - \Lambda_3)(\Lambda_2 - \Lambda_3)} + \frac{k_3}{k_2} \frac{\lambda_2 N^0_2}{\Lambda_2 - \Lambda_3} + N^0_3$$

$$\Lambda_i = k_i + \lambda_i \;;\; \Gamma_{rm} = -\Gamma_{mr} = \frac{v_m - v_r}{v_r v_m} \;;\; \Delta_{rm} = \Delta_{mr} = \frac{\lambda_r v_m - \lambda_m v_r}{v_m - v_r}$$

$$D^{31}_{21} = -D^{31}_{31} = \frac{1}{\Delta_{31} - \Delta_{21}} \;;\; D^{31}_{12} = -D^{31}_{32} = \frac{1}{\Delta_{32} - \Delta_{12}} \tag{5.6}$$

$$D^{31}_{13} = -D^{31}_{23} = \frac{1}{\Delta_{23} - \Delta_{13}} \;;\; D^{32}_{32} = D^{32}_{23} = 1 \;,\; D^{21}_{21} = D^{21}_{12} = 1$$

5.2.2 Solution for exponential release mode

The solution for the exponential release mode is expressed by setting $k_i = k$ in (3.22).

$$N_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t - kt} h(t)$$

$$B_{ij} = \sum_{m=1}^{j} N_m^0 \frac{\prod_{r=m}^{i} \lambda_i}{\lambda_i} \Big/ \prod_{\substack{l=m \\ (l \neq j)}}^{i} (\lambda_l - \lambda_j) \tag{5.7}$$

Then, the concentration of the i-th nuclide can be obtained by setting $k_i = k$ in (5.2) and replacing B'_{ij} by B_{ij}:

$$\begin{aligned} N_i^e(z,t) = {} & e^{-\lambda_i z/v_i} h\left(t - \frac{z}{v_i}\right) \sum_{j=1}^{i} B_{ij} e^{-(\lambda_j + k)(t - z/v_i)} \\ & + \sum_{l=1}^{i-1} \left(\prod_{r=l}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=l}^{i} \frac{e^{-\lambda_m z/v_m} h(t - z/v_m)}{\prod_{\substack{r=l \\ r \neq m}}^{i} \Gamma_{rm}} \\ & \times \sum_{\substack{r=l \\ r \neq m}}^{i} \sum_{j=1}^{l} \frac{D_{rm}^{il} B_{lj}}{\lambda_j + k - \Delta_{rm}} \left\{ e^{-\Delta_{rm}(t - z/v_m)} - e^{-(\lambda_j + k)(t - z/v_m)} \right\} \end{aligned} \tag{5.8}$$

For three members, we have

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$$N_1^e(z,t) = B_{11}\, h(t - z/v_1)\, e^{-(\lambda_1+k)t - kz/v_1}$$

(5.9)

$$N_2^e(z,t) = e^{-\lambda_2 z/v_2}\, h(t - z/v_2)\left[B_{21} e^{-(k+\lambda_1)(t - z/v_2)} + B_{22} e^{-(k+\lambda_2)(t - z/v_2)}\right]$$

$$+ \frac{\lambda_1}{v_1} h(t - z/v_1) \frac{e^{-\lambda_1 z/v_1}}{\Gamma_{21}} \frac{D_{21}^{21}\, B_{11}}{k+\lambda_1-\Delta_{21}}$$

$$\times \left[e^{-\Delta_{21}(t - z/v_1)} - e^{-(\lambda_1+k)(t - z/v_1)}\right]$$

$$+ \frac{\lambda_1}{v_1} h(t - z/v_2) \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{12}} \frac{D_{12}^{21}\, B_{11}}{k+\lambda_1-\Delta_{12}}$$

$$\times \left[e^{-\Delta_{12}(t - z/v_2)} - e^{-(\lambda_1+k)(t - z/v_2)}\right]$$

(5.10)

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$$
\begin{aligned}
N_3^e(z,t) = {} & e^{-\lambda_3 z/v_3} h(t - z/v_3)\Big[B_{31} e^{-(\lambda_1+k)(t-z/v_3)} + B_{32} e^{-(\lambda_2+k)(t-z/v_3)} \\
& \qquad + B_{33} e^{-(\lambda_3+k)(t-z/v_3)}\Big] \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2} h(t-z/v_1) \frac{e^{-\lambda_1 z/v_1}}{\Gamma_{21}\Gamma_{31}} \Big[\frac{D_{21}^{31} B_{11}}{\lambda_1+k-\Delta_{21}} \Big\{ e^{-\Delta_{21}(t-z/v_1)} - e^{-(\lambda_1+k)(t-z/v_1)} \Big\} \\
& \qquad + \frac{D_{31}^{31} B_{11}}{\lambda_1+k-\Delta_{31}} \Big\{ e^{-\Delta_{31}(t-z/v_1)} - e^{-(\lambda_1+k)(t-z/v_2)} \Big\} \Big] \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2} h(t-z/v_2) \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{12}\Gamma_{32}} \Big[\frac{D_{12}^{31} B_{11}}{\lambda_1+k-\Delta_{12}} \Big\{ e^{-\Delta_{12}(t-z/v_2)} - e^{-(\lambda_1+k)(t-z/v_2)} \Big\} \\
& \qquad + \frac{D_{32}^{31} B_{11}}{\lambda_1+k-\Delta_{32}} \Big\{ e^{-\Delta_{32}(t-z/v_2)} - e^{-(\lambda_1+k)(t-z/v_2)} \Big\} \Big] \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2} h(t-z/v_3) \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{23}\Gamma_{13}} \Big[\frac{D_{13}^{31} B_{11}}{\lambda_1+k-\Delta_{13}} \Big\{ e^{-\Delta_{13}(t-z/v_3)} - e^{-(\lambda_1+k)(t-z/v_3)} \Big\} \\
& \qquad + \frac{D_{23}^{31} B_{11}}{\lambda_1+k-\Delta_{23}} \Big\{ e^{-\Delta_{23}(t-z/v_3)} - e^{-(\lambda_1+k)(t-z/v_3)} \Big\} \Big] \\
& + \frac{\lambda_2}{v_2} h(t-z/v_1) \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{32}} \Big[\frac{D_{32}^{32} B_{21}}{\lambda_1+k-\Delta_{32}} \Big\{ e^{-\Delta_{32}(t-z/v_2)} - e^{-(\lambda_1+k)(t-z/v_2)} \Big\} \\
& \qquad + \frac{D_{32}^{32} B_{22}}{\lambda_2+k-\Delta_{32}} \Big\{ e^{-\Delta_{32}(t-z/v_2)} - e^{-(\lambda_2+k)(t-z/v_2)} \Big\} \Big] \\
& + \frac{\lambda_2}{v_2} h(t-z/v_3) \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{32}} \Big[\frac{D_{23}^{31} B_{21}}{\lambda_1+k-\Delta_{23}} \Big\{ e^{-\Delta_{23}(t-z/v_3)} - e^{-(\lambda_1+k)(t-z/v_3)} \Big\} \\
& \qquad + \frac{D_{23}^{32} B_{22}}{\lambda_1+k-\Delta_{23}} \Big\{ e^{-\Delta_{23}(t-z/v_3)} - e^{-(\lambda_2+k)(t-z/v_3)} \Big\} \Big]
\end{aligned}
\tag{5.11}
$$

Where $B_{11} = N_1^0$, $B_{21} = \dfrac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1}$, $B_{22} = N_2^0 - \dfrac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1}$

$$B_{31} = \frac{\lambda_1 \lambda_2 N_1^0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} , \quad B_{32} = \frac{\lambda_1 \lambda_2 N_1^0}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{\lambda_2 N_2^0}{\lambda_3 - \lambda_2}$$

$$B_{33} = \frac{\lambda_1 \lambda_2 N_1^0}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} + \frac{\lambda_2 N_2^0}{\lambda_2 - \lambda_3} + N_3^0$$

5.2.3 Solution for step release mode

The solution for step release mode is easily obtained from the solution of exponential release mode by setting k = 0.

$$N_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} h(t) \tag{5.12}$$

From (5.8) the concentration for the i-th nuclide is given by:

$$\begin{aligned} N_i^s(z,t) = {} & e^{-\lambda_i z/v_i} h(t - z/v_i) \sum_{j=1}^{i} B_{ij} e^{-\lambda_j (t - z/v_i)} \\ & + \sum_{l=1}^{i-1} \left(\prod_{r=l}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=l}^{i} \frac{e^{-\lambda_m z/v_m}}{\prod\limits_{\substack{r=l \\ r \neq m}}^{i} \Gamma_{rm}} h(t - z/v_m) \\ & \times \sum_{\substack{r=l \\ r \neq m}}^{i} \sum_{j=1}^{l} \frac{D_{rm}^{il} B_{lj}}{\lambda_j - \Delta_{rm}} \left\{ e^{-\Delta_{rm}(t - z/v_m)} - e^{-\lambda_j (t - z/v_m)} \right\} \end{aligned} \tag{5.13}$$

For three members, we have:

$$N_1^s(z,t) = N_1^0 e^{-\lambda_1 t} h(t - z/v_1) \tag{5.14}$$

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$$
\begin{aligned}
N_2^s(z,t) = {} & e^{-\lambda_2 z/v_2} h(t - z/v_2) \left[B_{21} e^{-\lambda_1 (t - z/v_2)} + B_{22} e^{-\lambda_2 (t - z/v_2)} \right] \\
& + \frac{\lambda_1}{v_1} h(t - z/v_1) \frac{e^{-\lambda_1 z/v_1}}{\Gamma_{21}} \frac{D^{21}_{21} B_{11}}{\lambda_1 - \Delta_{21}} \left[e^{-\Delta_{21}(t - z/v_1)} - e^{-\lambda_1 (t - z/v_1)} \right] \\
& + \frac{\lambda_1}{v_1} h(t - z/v_2) \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{12}} \frac{D^{21}_{12} B_{11}}{\lambda_1 - \Delta_{12}} \left[e^{-\Delta_{21}(t - z/v_2)} - e^{-\lambda_1 (t - z/v_2)} \right]
\end{aligned}
\tag{5.15}
$$

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$$
\begin{aligned}
N_3^s(z,t) = {} & e^{-\lambda_3 z/v_3} h(t - z/v_3) \left[B_{31} e^{-\lambda_1 (t - z/v_3)} + B_{32} e^{-\lambda_2 (t - z/v_3)} + B_{33} e^{-\lambda_3 (t - z/v_3)} \right] \\
& + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t - z/v_1) \frac{e^{-\lambda_1 z/v_1}}{\Gamma_{21} \Gamma_{31}} \left[\frac{D_{21}^{31} B_{11}}{\lambda_1 - \Delta_{21}} \left\{ e^{-\Delta_{21}(t - z/v_1)} - e^{-\lambda_1 (t - z/v_1)} \right\} \right. \\
& \qquad \left. + \frac{D_{31}^{31} B_{11}}{\lambda_1 - \Delta_{31}} \left\{ e^{-\Delta_{31}(t - z/v_1)} - e^{-\lambda_1 (t - z/v_1)} \right\} \right] \\
& + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t - z/v_2) \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{12} \Gamma_{32}} \left[\frac{D_{12}^{31} B_{11}}{\lambda_1 - \Delta_{12}} \left\{ e^{-\Delta_{12}(t - z/v_2)} - e^{-\lambda_1 (t - z/v_2)} \right\} \right. \\
& \qquad \left. + \frac{D_{32}^{31} B_{11}}{\lambda_1 - \Delta_{32}} \left\{ e^{-\Delta_{32}(t - z/v_2)} - e^{-\lambda_1 (t - z/v_2)} \right\} \right] \\
& + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t - z/v_3) \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{13} \Gamma_{23}} \left[\frac{D_{13}^{31} B_{11}}{\lambda_1 - \Delta_{13}} \left\{ e^{-\Delta_{13}(t - z/v_3)} - e^{-\lambda_1 (t - z/v_3)} \right\} \right. \\
& \qquad \left. + \frac{D_{23}^{31} B_{11}}{\lambda_1 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t - z/v_1)} - e^{-\lambda_1 (t - z/v_3)} \right\} \right] \\
& + \frac{\lambda_2}{v_2} h(t - z/v_2) \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{32}} \left[\frac{D_{32}^{32} B_{21}}{\lambda_1 - \Delta_{32}} \left\{ e^{-\Delta_{32}(t - z/v_2)} - e^{-\lambda_1 (t - z/v_2)} \right\} \right. \\
& \qquad \left. + \frac{D_{32}^{32} B_{22}}{\lambda_2 - \Delta_{32}} \left\{ e^{-\Delta_{32}(t - z/v_2)} - e^{-\lambda_2 (t - z/v_2)} \right\} \right] \\
& + \frac{\lambda_2}{v_2} h(t - z/v_3) \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{23}} \left[\frac{D_{23}^{32} B_{21}}{\lambda_1 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t - z/v_3)} - e^{-\lambda_1 (t - z/v_3)} \right\} \right. \\
& \qquad \left. + \frac{D_{23}^{32} B_{22}}{\lambda_2 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t - z/v_3)} - e^{-\lambda_2 (t - z/v_3)} \right\} \right]
\end{aligned}
$$

(5.16)

5.2.4 Solution for band release mode

The band release mode is defined by

$$N_i(t) = B_i(t)\left[h(t) - h(t-T)\right] \tag{5.17}$$

The solution for this release mode can be obtained from the solution of step release mode by using the superposition relation given by (3.60).

$$N_i^b(t) = N_i^s(z,t;B_{ij}) - N_i^s(z,t-T;B_{ij}e^{-\lambda_j T}) \tag{3.60}$$

The first term in the right hand side is given by (5.13). The second term can be obtained by replacing B_{ij} in (5.13) by $B_{ij} \times \exp(-\lambda_j T)$ and also replacing t by (t - T). The resultant solution is expressed as:

$$\begin{aligned}
N_i^b(z,t) = {} & e^{-\lambda_i z/v_i} \sum_{j=1}^{i} B_{ij} e^{-\lambda_j (t - z/v_i)} \left\{ h(t - z/v_i) - h(t - T - z/v_i) \right\} \\
& + \sum_{l=1}^{i-1} \left(\prod_{r=l}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=l}^{i} \frac{e^{-\lambda_m z/v_m}}{\prod\limits_{\substack{r=l \\ r \neq m}}^{i} \Gamma_{rm}} \sum_{\substack{r=l \\ r \neq m}}^{i} \sum_{j=1}^{l} \frac{D_{rm}^{il} B_{lj}}{\lambda_j - \Delta_{rm}} \\
& \times \Big[e^{-\Delta_{rm}(t - z/v_m)} \left\{ h(t - z/v_m) - e^{-(\lambda_j - \Delta_{rm})T} h(t - T - z/v_m) \right\} \\
& - e^{-\lambda_j (t - z/v_m)} \left\{ h(t - z/v_m) - h(t - T - z/v_m) \right\} \Big\}
\end{aligned} \tag{5.18}$$

For three members, the solutions can be obtained with the help of (5.14) - (5.16).

5.2.5 Solution for impulse release mode

The impulse release mode is a limiting case which can be obtained by applying the condition, $\lim_{T\to 0}\frac{1}{T}\left[h(t-\frac{z}{v_i})-h(t-T-\frac{z}{v_i})\right]=\delta(t-\frac{z}{v_i})$ on the solution of band release mode. Then, the concentration of the i-th nuclide is expressed as:

$$
\begin{aligned}
N_i^I(z,t) &= (N_i^0 T)\, e^{-\lambda_i z/v_i}\, \delta(t-\frac{z}{v_i}) \\
&\quad + \sum_{\ell=1}^{i-1}\left(\prod_{r=\ell}^{i-1}\frac{\lambda_r}{v_r}\right)\sum_{m=\ell}^{i}\frac{N_\ell^0 T\, e^{-\lambda_m z/v_m}}{\prod\limits_{\substack{r=\ell\\ r\neq m}}^{i}\Gamma_{rm}}\, h(t-\frac{z}{v_m}) \\
&\quad \times \sum_{\substack{r=\ell\\ r\neq m}}^{i} D_{rm}^{i\ell}\, e^{-\Delta_{rm}(t-z/v_m)}
\end{aligned}
$$

(5.19)

For three members, we have:

$$N_1^I(z,t)=(N_1^0 T)\,e^{-\lambda_1 z/v_1}\,\delta(t-z/v_1) \qquad (5.20)$$

$$
\begin{aligned}
N_2^I(z,t) &= (N_2^0 T)\,e^{-\lambda_2 z/v_2}\,\delta(t-z/v_2) \\
&\quad + \frac{\lambda_1}{v_1}\frac{N_1^0 T}{\Gamma_{21}}\,e^{-\lambda_1 z/v_1}\,h(t-z/v_1)\,D_{21}^{21}\,e^{-\Delta_{21}(t-z/v_1)} \\
&\quad + \frac{\lambda_1}{v_1}\frac{N_1^0 T}{\Gamma_{12}}\,e^{-\lambda_2 z/v_2}\,h(t-z/v_2)\,D_{12}^{21}\,e^{-\Delta_{12}(t-z/v_2)}
\end{aligned}
\qquad (5.21)
$$

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$$
\begin{aligned}
N_3^I(z,t) = {} & (N_3^0 T)\, e^{-\lambda_3 z/v_3}\, \delta(t - z/v_3) \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2}\, \frac{N_1^0 T e^{-\lambda_1 z/v_1}}{\Gamma_{21}\,\Gamma_{31}}\, h\left(t-\frac{z}{v_1}\right)\left\{ D_{21}^{31} e^{-\Delta_{21}\left(t-\frac{z}{v_1}\right)} + D_{31}^{31} e^{-\Delta_{31}\left(t-\frac{z}{v_1}\right)} \right\} \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2}\, \frac{N_1^0 T e^{-\lambda_2 z/v_2}}{\Gamma_{12}\,\Gamma_{32}}\, h\left(t-\frac{z}{v_2}\right)\left\{ D_{12}^{31} e^{-\Delta_{12}\left(t-\frac{z}{v_2}\right)} + D_{32}^{31} e^{-\Delta_{32}\left(t-\frac{z}{v_2}\right)} \right\} \\
& + \frac{\lambda_1\lambda_2}{v_1 v_2}\, \frac{N_1^0 T e^{-\lambda_3 z/v_3}}{\Gamma_{13}\,\Gamma_{23}}\, h\left(t-\frac{z}{v_3}\right)\left\{ D_{13}^{31} e^{-\Delta_{13}\left(t-\frac{z}{v_3}\right)} + D_{23}^{31} e^{-\Delta_{23}\left(t-\frac{z}{v_3}\right)} \right\} \\
& + \frac{\lambda_2}{v_2}\, \frac{N_2^0 T e^{-\lambda_2 z/v_2}}{\Gamma_{32}}\, h\left(t-\frac{z}{v_2}\right)\left\{ D_{12}^{32} e^{-\Delta_{12}\left(t-\frac{z}{v_2}\right)} + D_{32}^{32} e^{-\Delta_{32}\left(t-\frac{z}{v_2}\right)} \right\} \\
& + \frac{\lambda_2}{v_2}\, \frac{N_2^0 T}{\Gamma_{23}}\, e^{-\lambda_3 z/v_3}\, h\left(t-\frac{z}{v_3}\right)\left\{ D_{13}^{32} e^{-\Delta_{13}\left(t-\frac{z}{v_2}\right)} + D_{23}^{32} e^{-\Delta_{23}\left(t-\frac{z}{v_3}\right)} \right\}
\end{aligned}
\tag{5.22}
$$

5.3 Application of General Solution -- Solutions of the Transport Equation for Three Member Decay Chain with Plane Source and Dispersion -- (H1)

The general solution for the transport equation with plane source and dispersion is given by (4.38).

$$
\begin{aligned}
N_i(z,t) = {} & \int_0^t f_i(t-\theta_i)\, e^{-\lambda_i\theta_i}\, F(v_i\theta_i,\, z - v_i\theta_i)\, d\theta_i \\
& + \sum_{j=1}^{i-1}\left(\frac{v_i}{v_j}\prod_{l=j}^{i-1}\lambda_l\right)\int_0^t d\theta_i \int_0^{t-\theta_i} d\theta_{i-1} \cdots \int_0^{t-\sum_{k=j+1}^{i}\theta_k} d\theta_j\, f_j\left(t-\sum_{k=j}^{i}\theta_k\right) \\
& \times e^{-\sum_{k=j}^{i}\lambda_k\theta_k}\, F\left(\sum_{k=j}^{i} v_k\theta_k,\; z - \sum_{k=j}^{i} v_k\theta_k\right)
\end{aligned}
\tag{4.38}
$$

Applying this equation to three members, the concentrations are given by:

$$N_1(z,t) = \int_0^t f_1(t-\theta_1) e^{-\lambda_1\theta_1} F(v_1\theta_1, z-v_1\theta_1)\, d\theta_1 \tag{5.23}$$

$$\begin{aligned} N_2(z,t) = & \int_0^t f_2(t-\theta_2) e^{-\lambda_2\theta_2} F(v_2\theta_2, z-v_2\theta_2)\, d\theta_2 \\ & + \frac{\lambda_1 v_2}{v_1} \int_0^t d\theta_2 \int_0^{t-\theta_2} d\theta_1\, f_1(t-\theta_1-\theta_2) e^{-\lambda_1\theta_1-\lambda_2\theta_2} \\ & \times F(v_1\theta_1+v_2\theta_2, z-v_1\theta_1-v_2\theta_2) \end{aligned} \tag{5.24}$$

$$\begin{aligned} N_3(z,t) = & \int_0^t f_3(t-\theta_3) e^{-\lambda_3\theta_3} F(v_3\theta_3, z-v_3\theta_3)\, d\theta_3 \\ & + \frac{\lambda_2 v_3}{v_2} \int_0^t d\theta_3 \int_0^{t-\theta_3} d\theta_2\, f_2(t-\theta_2-\theta_3) e^{-\lambda_3\theta_3-\lambda_2\theta_2} F(v_2\theta_2+v_3\theta_3, z-v_2\theta_2-v_3\theta_3) \\ & + \frac{\lambda_1\lambda_2 v_3}{v_1} \int_0^t d\theta_3 \int_0^{t-\theta_3} d\theta_2 \int_0^{t-\theta_3-\theta_2} d\theta_1 f_1(t-\theta_1-\theta_2-\theta_3) e^{-\lambda_1\theta_1-\lambda_2\theta_2-\lambda_3\theta_3} \\ & \times F(v_1\theta_1+v_2\theta_2+v_3\theta_3, z-v_1\theta_1-v_2\theta_2-v_3\theta_3) \end{aligned} \tag{5.25}$$

where $f_i(t)$ is the source term at the repository and can be related with the concentration at the repository with the use of (3.33) and (4.23).

$$f_i(t) = N_i(t) v_i \tag{5.26}$$

In this section we apply (5.23) - (5.25) to the three member chain and propose explicit solutions for step and band releases modes.

The decaying step release mode asserts that $N_i(t)$ is expressed by the Bateman equation. In this case, $f_i(t)$ for three members can be obtained as:

$$f_1(t) = v_1 N_1(t) = v_1 N_1^0 e^{-\lambda_1 t} \equiv v_1 B_{11} e^{-\lambda_1 t}$$

$$\begin{aligned} f_2(t) &= v_2 N_2(t) = v_2\left[\left(N_2^0 + N_1^0 \frac{\lambda_1}{\lambda_1-\lambda_2}\right) e^{-\lambda_2 t} + N_1^0 \frac{\lambda_1}{\lambda_2-\lambda_1} e^{-\lambda_1 t}\right] \\ &\equiv v_2 \left[B_{22} e^{-\lambda_2 t} + B_{21} e^{-\lambda_1 t}\right] \end{aligned}$$

$$\begin{aligned} f_3(t) = v_3 N_3(t) &= v_3 \left[\left(N_3^0 + \frac{N_2^0 \lambda_2}{\lambda_2-\lambda_3} + \frac{N_1^0 \lambda_1 \lambda_2}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)} \right) e^{-\lambda_3 t} \right. \\ &\quad + \left(N_2^0 \frac{\lambda_2}{\lambda_3-\lambda_2} + \frac{N_1^0 \lambda_1 \lambda_2}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} \right) e^{-\lambda_2 t} \\ &\quad \left. + \frac{N_1^0 \lambda_1 \lambda_2}{(\lambda_1-\lambda_3)(\lambda_1-\lambda_2)} e^{-\lambda_1 t} \right] \\ &\equiv v_3 \left[B_{33} e^{-\lambda_3 t} + B_{32} e^{-\lambda_2 t} + B_{31} e^{-\lambda_1 t} \right] \end{aligned}$$

(5.27)

The concentrations for the three members can be obtained by introducing (5.27) into (5.23) - (5.25) and then by using integration by parts and the following relation:

$$\frac{2}{\sqrt{\pi}}\int_{a/b}^{\infty} e^{-(\lambda^2+\frac{a^2}{\lambda^2})}\,d\lambda = \frac{1}{2}\left[e^{-2a}\,\mathrm{erfc}\left(b-\frac{a}{b}\right) - e^{2a}\,\mathrm{erf}\left(b+\frac{a}{b}\right)\right] \tag{5.28}$$

The resultant expressions for the concentrations are:

$$N_1^S(z,t) = B_{11}E(1,1;1) \tag{5.29}$$

$$\begin{aligned} N_2^S(z,t) = {} & B_{22}E(2,2;2) + B_{21}E(1,1;2) \\ & + \frac{B_{11}\lambda_1}{\nu_1(\Lambda_{12}-\lambda_1\Gamma_{12})}\Big[E(1,1;2) - E(1,1;1) \\ & \qquad + E(1,2;1) - E(1,2;2)\Big] \end{aligned} \tag{5.30}$$

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\begin{aligned}
N_3^{S}(z,t) = {} & B_{33}E(3,3;3) + B_{32}E(2,2;3) + B_{31}E(1,1;3) \\
& + \frac{B_{22}\lambda_2}{v_2(\Lambda_{23}-\lambda_2\Gamma_{23})}\left[E(2,2;3)-E(2,3;3)-E(2,2;2)+E(2,3;2)\right] \\
& + \frac{B_{21}\lambda_2}{v_2(\Lambda_{23}-\lambda_1\Gamma_{23})}\left[E(1,1;3)-E(2,3;3)-E(1,1;2)+E(2,3;2)\right] \\
& + \frac{B_{11}\lambda_1\lambda_2}{v_1v_2(\Lambda_{13}-\lambda_1\Gamma_{13})(\Lambda_{23}-\lambda_1\Gamma_{23})}E(1,1;3) + \frac{B_{11}\lambda_1\lambda_2\Gamma_{13}E(1,3;3)}{v_1v_2(\Lambda_{13}-\lambda_1\Gamma_{13})(\Gamma_{23}\Lambda_{13}-\Gamma_{13}\Lambda_{23})} \\
& + \frac{B_{11}\lambda_1\lambda_2\Gamma_{23}E(2,3;3)}{v_1v_2(\Lambda_{23}-\lambda_1\Gamma_{23})(\Gamma_{13}\Lambda_{23}-\Gamma_{23}\Lambda_{13})} \\
& + \frac{B_{11}\lambda_1\lambda_2\;E(1,1;2)}{v_1v_2(\Lambda_{32}-\lambda_1\Gamma_{32})(\Lambda_{12}-\lambda_1\Gamma_{12})} + \frac{B_{11}\lambda_1\lambda_2E(2,3;2)\;\Gamma_{32}}{v_1v_2(\Lambda_{32}-\lambda_1\Gamma_{32})(\Gamma_{12}\Lambda_{32}-\Gamma_{32}\Lambda_{12})} \\
& + \frac{B_{11}\lambda_1\lambda_2\;\Gamma_{12}E(1,2;2)}{v_1v_2(\Lambda_{12}-\lambda_1\Gamma_{12})(\Gamma_{32}\Lambda_{12}-\Gamma_{12}\Lambda_{32})} \\
& + \frac{B_{11}\lambda_1\lambda_2\;E(1,1;1)}{v_1v_2(\Lambda_{31}-\lambda_1\Gamma_{31})(\Lambda_{21}-\lambda_1\Gamma_{21})} + \frac{B_{11}\lambda_1\lambda_2\Gamma_{31}E(1,3;1)}{v_1v_2(\Lambda_{31}-\lambda_1\Gamma_{31})(\Lambda_{31}\Gamma_{21}-\Lambda_{21}\Gamma_{31})} \\
& + \frac{B_{11}\lambda_1\lambda_2\Gamma_{21}E(1,2;1)}{v_1v_2(\Lambda_{21}-\lambda_1\Gamma_{21})(\Lambda_{21}\Gamma_{31}-\Lambda_{31}\Gamma_{21})}
\end{aligned}
\qquad (5.31)
$$

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where

$$\Lambda_{kj} = \frac{\lambda_k}{v_k} - \frac{\lambda_j}{v_j}, \quad \Gamma_{kj} = \frac{1}{v_k} - \frac{1}{v_j}$$

$$\beta_{ij} = \begin{cases} \lambda_i, & i=j \\ \Lambda_{ij}/\Gamma_{ij}, & i \neq j \end{cases}, \quad \gamma_{ijk} = 1 + 4\kappa(\lambda_k - \beta_{ij})/v_k \tag{5.32}$$

$$E(i,j;k) = e^{\frac{z}{2\kappa} - \beta_{ij} t} \int_0^{\sqrt{v_k t/4\kappa}} \frac{2}{\sqrt{\pi}} \exp\left[-\left(\gamma_{ijk} y^2 + \frac{z^2}{16\kappa^2 y^2}\right)\right] dy$$

$$= \frac{e^{-\beta_{ij} t + \frac{z}{2\kappa}}}{2\sqrt{\gamma_{ijk}}} \left[e^{-\frac{|z|}{2\kappa}\sqrt{\gamma_{ijk}}} \operatorname{erfc}\left(\frac{|z| - v_k t\sqrt{\gamma_{ijk}}}{\sqrt{4\kappa v_k t}}\right) - e^{\frac{|z|}{2\kappa}\sqrt{\gamma_{ijk}}} \operatorname{erfc}\left(\frac{|z| + v_k t\sqrt{\gamma_{ijk}}}{\sqrt{4\kappa v_k t}}\right)\right], \quad (\gamma_{ijk} > 0)$$

(5.33)

In compact form, the above equations can be described as:

$$N_1^S(z,t) = B_{11} E(1,1;1)$$

$$N_2^S(z,t) = \sum_{j=1}^{2} B_{2j} E(j,j:2) + \frac{B_{11}\lambda_1}{v_1(\Lambda_{12} - \lambda_1\Gamma_{12})}\left[E(1,1;2) - E(1,1;1) + E(1,2:1) - E(1,2:2)\right]$$

$$N_3^S(z,t) = \sum_{j=1}^{3} B_{3j} E(j,j:3) + \frac{\lambda_2}{v_2}\sum_{j=1}^{2} \frac{B_{2j}}{\Lambda_{23} - \lambda_j\Gamma_{23}}\left[E(j,j:3) - E(j,j:2) + E(2,3;2) - E(2,3;3)\right]$$

$$+ \frac{\lambda_1\lambda_2 B_{11}}{v_1 v_2}\sum_{j=1}^{3}\left[\frac{E(1,1;j)}{(\Lambda_{kj} - \lambda_1\Gamma_{kj})(\Lambda_{\ell j} - \lambda_1\Gamma_{\ell j})} + \frac{\Gamma_{kj} E(k,j;j)}{(\lambda_{kj} - \lambda_1\Gamma_{kj})(\Gamma_{\ell j}\Lambda_{kj} - \Gamma'_{kj}\Lambda_{\ell j})} + \frac{\Gamma_{\ell j} E(\ell,j:j)}{(\Lambda_{\ell j} - \lambda_1\Gamma_{\ell j})(\Gamma'_{kj}\Lambda_{\ell j} - \Gamma'_{\ell j}\Lambda_{kj})}\right] \tag{5.34}$$

$$(k < \ell, \quad k \neq j, \ell \neq j, \ell, k = 1, 2, 3)$$

Let us consider the solutions for band release mode. The superposition relation is described by (3.60) as

$$N_i^b(z,t) = N_i^s(z,t;B_{ij}) - N_i^s(z,t-T;B_{ij}\bar{e}^{\lambda_j T}) \qquad (5.35)$$

The first term in the right hand side is given by (5.34). The second term is obtained by replacing B_{ij} in (5.34) by B_{ij} exp $(-\lambda_j T)$ and also replacing t by (t-T). Then, the concentrations for the band release can be obtained from (5.34) and (5.35).

When $\kappa \to 0$, (5.34) agrees with the solutions without dispersion. Then, it can be said that (5.34) includes the case of no dispersion.

5.4 Application of General Solution -- Solution of Transport Equation (F2) for 3 member chain with dispersion and the concentration boundary condition --

The governing equation of migration which we will treat in this section is

$$\eta_i \frac{\partial^2 N_i}{\partial z^2} - v_i \frac{\partial N_i}{\partial z} - \frac{\partial N_i}{\partial t} - \lambda_i N_i = -\lambda_{i-1} K_{i-1} N_{i-1} / K_i$$

$$\lambda_0 = 0, \quad i = 1, 2, \cdots \qquad (4.2)$$

The initial and the boundary conditions are

$$N_i(z,0)=0\ ,\ N_i(\infty,t)=\text{Order of } e^{-kz},\ N_i(0,t)=N_i^0\phi_i(t) \tag{5.36}$$

The recursive solution of (4.2) and (5.36) is given in the preceding chapter by (4.61), which can be rewritten as

$$N_i(z,t)=\exp\left[\frac{z}{2\kappa}-\left(\lambda_i+\frac{v_i}{4\kappa}\right)t\right]$$
$$\times\int_0^\infty e^{-\kappa v_i p^2 t}\sqrt{\frac{2}{\pi}}\sin(pz)\int_0^t e^{\kappa v_i p^2\theta}\,R_{i-1}(p,\theta)\,d\theta\,dp \tag{5.37}$$

where

$$R_{i-1}(p,\theta)=\kappa v_i S_{i-1}(p,t)+v_i\kappa p\cos(pz)\exp\left\{\left(\lambda_i+\frac{v_i}{4\kappa}\right)\theta\right\}$$
$$\times N_i^0\phi_i(\theta)$$

$$S_{i-1}(p,t)=\int_0^\infty\sqrt{\frac{2}{\pi}}\,\sin(pz)\,e^{-\beta_i t-\frac{z}{2\kappa}}\,N_{i-1}(z,t)\,\frac{\lambda_{i-1}}{v_{i-1}\kappa}\,dz$$

$$\beta_i=-\lambda_i-\frac{v_i}{4\kappa}$$

(5.38)

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This section is concerned with the concentrations of three members for the step and the band release modes.

For step release, the boundary condition at the repository is expressed as

$$N_i(0,t) = N_i^0 \phi_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} h(t) \tag{5.39}$$

Then, the concentrations of the three members can be calculated from (5.37) - (5.39) by the iterative procedure with the help of Fourier Integral Table. The resultant concentrations are reduced to the analytical solution (5.34) for the step release in the preceding section, but with the function E(i,j:k) defined as

$$\begin{aligned} E(i,j:k) &= e^{\frac{z}{2\kappa} - \beta_{ij} t} \int_{\frac{z}{\sqrt{4\kappa v_k t}}}^{\infty} \frac{2}{\sqrt{\pi}} \exp\left[-\left(y^2 + \frac{z^2 \gamma_{ijk}}{16\kappa^2 y^2}\right)\right] dy \\ &= \frac{1}{2} e^{\frac{z}{2\kappa} - \beta_{ij} t} \left[e^{z\sqrt{\gamma_{ijk}}/2\kappa} \operatorname{erfc}\left\{ \frac{z + v_k t\sqrt{\gamma_{ijk}}}{\sqrt{4\kappa v_k t}} \right\} \right. \\ &\qquad \left. + e^{-z\sqrt{\gamma_{ijk}}/2\kappa} \operatorname{erfc}\left\{ \frac{z - v_k t\sqrt{\gamma_{ijk}}}{\sqrt{4\kappa v_k t}} \right\} \right], \quad \gamma_{ijk} > 0 \end{aligned} \tag{5.40}$$

The band release solution can be obtained by the same procedure as in the preceding section.

When the diffusion or dispersion are neglected in the water path, the functions E(k,j;k) i.e., (5.40) and (5.33) agree with each other. Thus, the solutions for concentration boundary condition agree with the ones for the transport equation with source term.

5.5 $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ Decay Chain Migration Behavior (H4)

One of the important nuclide chains to be considered in the predictive modeling of hydrogeological transport of radionuclides in high-level wastes is:

$$^{242m}Am \rightarrow {}^{242}Cm \rightarrow {}^{238}Pu \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$$

The most important species is usually ^{226}Ra, because of its relatively high biological hazard and its relatively high mobility in geologic media. The important time scales for the appearance of ^{226}Ra are of the order of tens of thousands of years, as controlled by the long life of its precursor ^{230}Th. Therefore, because of the relatively short lives of the first three members of the above chain, the analysis of ^{226}Ra transport can be reduced to the analysis of the last three members of the above chain, with the assumption that the first three members have already decayed to form the long-lived ^{234}U.

In this section, the migration feature for this three member decay chain will be elucidated with the help of the solutions of transport equations (5.29) through (5.31) which correspond to the step and band release modes. The computer code is shown in the Appendix.

5.5.1 Input data and parameters

5.5.1.1 Release Mode

In Chapter 3, several release modes have been discussed. In this calculation, we select the band release mode as a typical release mode. The band release mode requires the unknown initial concentrations for three members, N_i^o (i=1,2,3) (atoms/cm^3 of H$_2$O). The amount of the i-th nuclide $M_i(t')$ (atoms) in the repository changes with time according to the Bateman's equation.

$$M_i(t') = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t'}, \quad B_{ij} = \sum_{m=1}^{j} M_m^0 \frac{1}{\lambda_i} \prod_{\ell=m}^{i} \lambda_\ell \Big/ \prod_{\substack{\ell=1 \\ \ell \neq j}}^{i} (\lambda_\ell - \lambda_j) \quad (5.41)$$

$$i = 1, 2, 3$$

Where t' is the time measured from the moment of initial emplacement of the nucludes in the repository, and we assumed that there is no daughter in the repository at t'=0. The change of the amount of each nuclide is calculated as shown in Figure 5.1. The amounts of ^{230}Th and ^{226}Ra increase with time first, and pass a maximum at $t' = 2\times10^5$ year. Finally, the ratios of the amounts of the second and the third members relative to that of the first member approach to a steady-state (transient equilibrium):

$$\lim_{t\to\infty} \frac{M_2(t)}{M_1(t)} = \frac{\lambda_1}{\lambda_2 - \lambda_1}$$

$$\lim_{t\to\infty} \frac{M_3(t)}{M_1(t)} = \frac{\lambda_1\lambda_2}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} \tag{5.42}$$

In order to simplify the calculation, we can assume two kinds of initial conditions, i.e., 1) The release of nuclides from the repository to the groundwater phase occurs at an early stage, 2) The release occurs after the nuclides have attained the state of transient equilibrium. In the first case pure ^{234}U is the only nuclide present in the repository, and no daughters are present at the beginning of the release, i.e.,

$$N_2^o = N_3^o = 0 \tag{5.43a}$$

In the second case the initial concentrations, N_2^o and N_3^o are assumed from (5.42) as

$$N_2^0 = N_1^0\lambda_1/(\lambda_2-\lambda_1)\ ,\quad N_3^0 = N_1^0\lambda_1\lambda_2/(\lambda_2-\lambda_1)(\lambda_3-\lambda_1) \tag{5.43b}$$

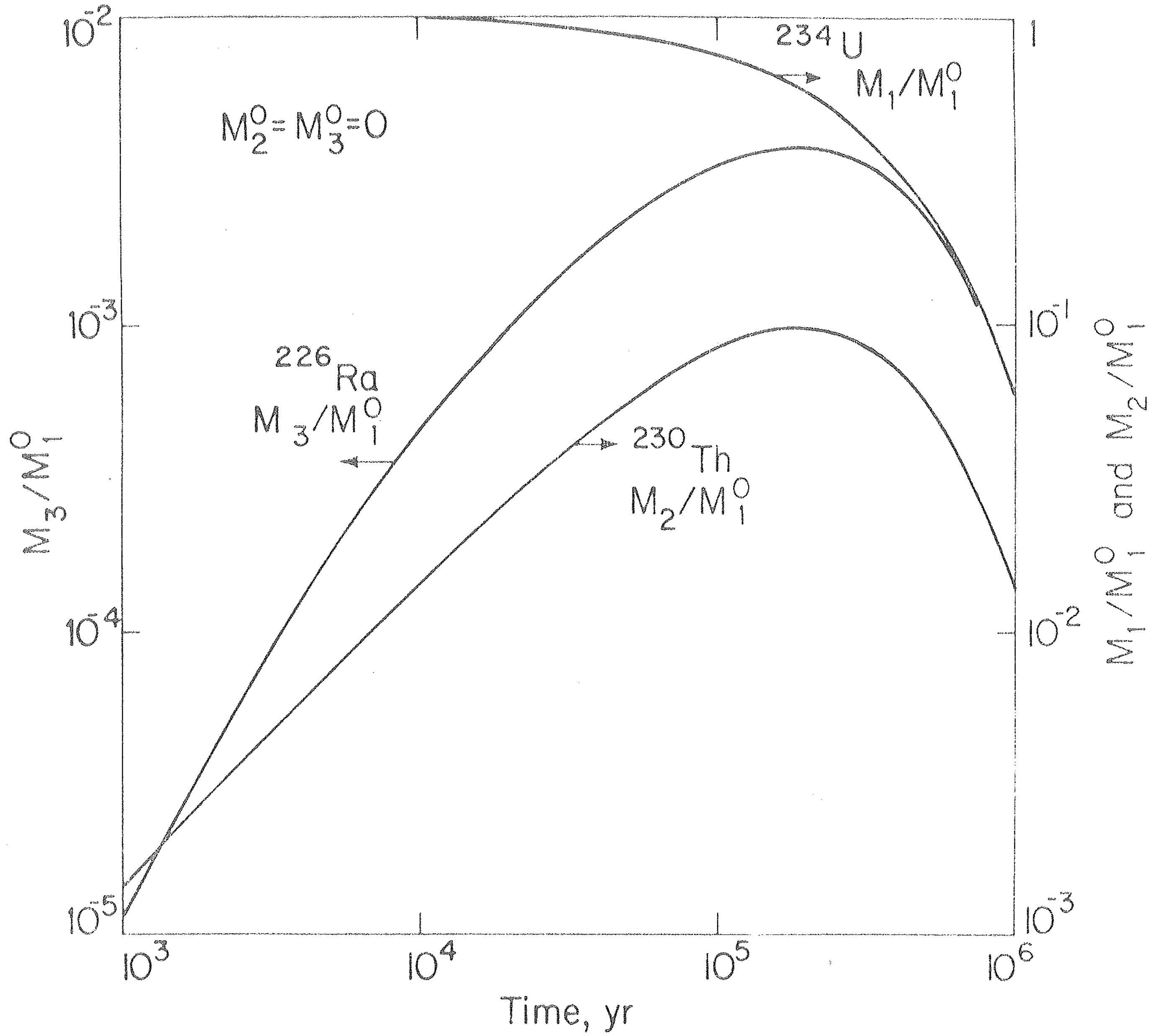

Figure 5.1 - Nuclide concentrations against time in the repository (z=0) for the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain. (Pure ^{234}U source at t=0. M_i =atoms of i^{th} nuclide. M_1^0 = atoms at ^{234}U at z=0, t=0.)

5.5.1.2 Data of Parameters

Nuclear data and sorption equilibrium constants for the nuclides are shown in Table 5.1.

Table 5.1

	^{234}U	^{230}Th	^{226}Ra
Half-life (yr)	2.44×10^{5}	7.7×10^{4}	1.6×10^{3}
Decay Constant λ_i (1/yr)	2.84×10^{-6}	9.00×10^{-6}	4.33×10^{-4}
Sorption Equilibrium Constant, K_i (-)	1×10^{4}	5×10^{4}	5×10^{2}

The values of the parameters, K_i, v and D have essentially defined the nature of the sorption medium through which the nuclides migrate. The K_i values used in this calculation are taken from the most comprehensive set of data available, which are obtained from the values evaluated by Burkholder (B3). The velocity of groundwater is assumed to be 100 m/yr. This is of the same order as the measured value for cracked rock and deep aquifers(H5). With this assumption for the velocity of the groundwater, the migration speed of each nuclide ($v_i = v/K_i$) is:

$$U; \quad v_1 = 1.0 \times 10^{-2} \text{ m/yr.}$$
$$Th; \quad v_2 = 2.0 \times 10^{-3} \text{ m/yr.}$$
$$Ra; \quad v_3 = 2.0 \times 10^{-1} \text{ m/yr.}$$

In this decay chain, the migration speed of ^{234}U is faster than that of ^{230}Th, and that of ^{226}Ra is much faster than its precursors. Half-lives decrease in the order of ^{234}U, ^{230}Th, and ^{226}Ra.

In this section, the successive developments of the nuclide profiles are graphically shown for the case of

$$\text{leach time (T)} = 3 \times 10^4 \text{ yr}$$

This leach time is much longer than the half-life of ^{226}Ra (1600 yr) but much shorter than the time for the occurrence of maximum M_3/M_1^o in Figure 5.1.

During the period of leach time, each nuclide migrates by the distance of

$$^{234}U; \quad v_1T = 300 \text{ m}$$
$$^{230}Th; \quad v_2T = 60 \text{ m}$$
$$^{226}Ra; \quad v_3T = 6000 \text{ m}$$

Each nuclide migrates in its half-life the distance shown in Table 5.2.

Table 5.2

Migration length in each half-life
(half-life) x (migration velocity)

^{234}U	2,440 m
^{230}Th	154 m
^{226}Ra	320 m

In spite of the fast migration speed of ^{226}Ra, the survival distance is not too long because of its shorter half-life. On the other hand, ^{234}U can migrate along a relatively long distance without significant decay. From the above Table 5.2, we may expect that large radioactive effect of this decay chain will extend over a few kilometers (when V = 100 m/yr) because of the long survival distance of the first (parent) nuclide, ^{234}U.

The value of the dispersion coefficient is depending greatly on the water velocity, porosity of the soil column and the pore radius. The characteristic dispersive length, D/V, may range from several to several tenth order (H5). In this calculation, an average value of 10^3 m^2/yr is selected as the dispersion coefficient. The value corresponding to the usual molecular diffusion coefficient, 1×10^{-1} m^2/yr, is also used for comparison.

It should be noted that the relative situation of the development of distributions changes in a variety of ways depending on leach time. In the series of graphs presented in this section for $T = 3 \times 10^4$ yr, however, most of the important features are expected to be seen.

5.5.2 Concentration Profiles of $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$

5.5.2.1 Comparison of the solution of transport equation for the case of plane source and dispersion and the case of concentration boundary condition and dispersion.

Figure 5.2 shows the plotting of the concentration profile of each nuclide at $t=5x10^4$ yr in the case of pure ^{234}U source in the repository at t = 0 for two different solutions of the transport equations, i.e.,(1) the solution of transport equation with plane source and dispersion (plane source solution) (5.33) and (5.34); (2) the solution of transport equation with concentration boundary condition and dispersion (concentration boundary solution) (5.34) and (5.40). In this figure, the broken lines show the solution for the plane source with $D = 1x10^3$ m^2/yr, the dash-dotted lines show the solution for the concentration boundary also with $D = 1x10^3$ m^2/yr, and the solid lines show the solution both for the plane source with $D = 1x10^{-1}$ m^2/yr and for the concentration boundary $D = 1x10^{-1}$ m^2/yr.

In the case of small dispersion coefficient, $D = 1x10^{-1}$, both solutions for the plane source and the concentration boundary agree with each other. Furthermore, it was found by the preliminary calculation (H2,H3) that the calculated concentration profiles almost agreed with those of the dispersion free case. However, in the case that the dispersion coefficient is larger, $D = 1x10^3$, the concentration profiles near the repository are much more affected by the difference in boundary conditions of the transport equation. As shown by the curves in broken lines and dash-dotted lines, the concentrations of each nuclide for the concentration boundary are much lower than those for the plane source. The solution for the plane source takes into account the effect of dispersion in the negative z

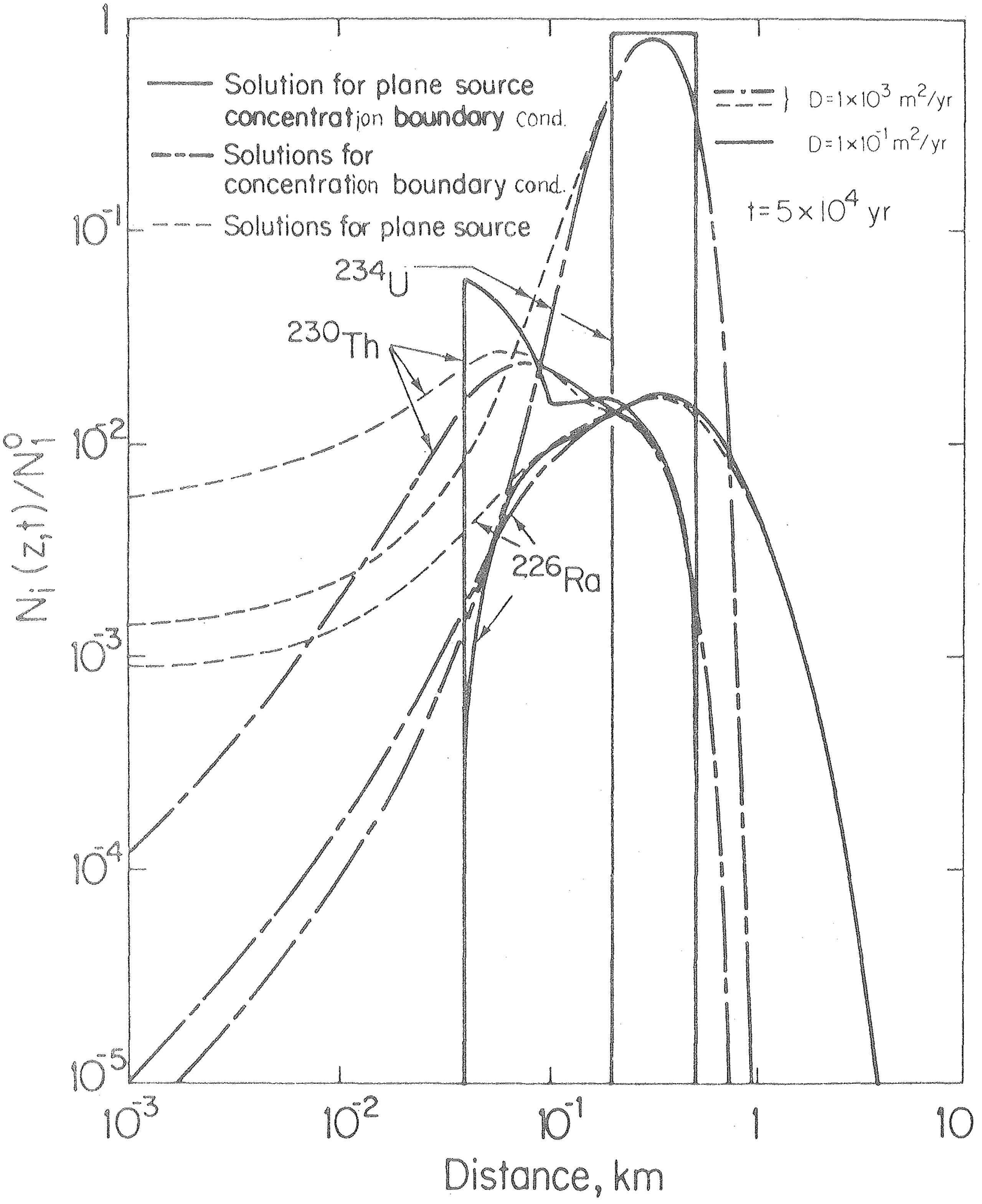

Figure 5.2 Comparison of concentration profiles for different source boundary conditions for the $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at $t=5x10^4$ yr (Pure ^{234}U source at t=0. $N_i(z,t)$=atom/cm^3. N_i^o=atom/cm^3 of U at z=0, t=0. v=100m/yr, $T=3x10^4$ yr., $K_U=1x10^4$, $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

domain, but the solution for the concentration boundary condition does not take this effect into account. Also, the solution for the concentration boundary condition at the repository is governed by the Bateman's equation for $t<T$. So, for a time $t = 5x10^4$ yr $> T$, the concentration of each nuclide has to satisfy the boundary condition so that the curve of its concentration profile has to start from zero at the repository. Thus, the evaluation of the distribution of each nuclide near the repository using the solution for the concentration boundary is less conservative due to this artificial concentration boundary condition.

On the other hand, the solutions for the two different boundary conditions at locations far from the repository almost agree with each other.

Thus, it should be noticed that the solution for the transport equation with the plane source represents a more realistic model of the transport phenomenon than the solution with the concentration boundary condition which has been usually used by many other researchers.

5.5.2.2 Concentration profiles of nuclides in the case of pure ^{234}U source in repository at t=0.

Figures 5.3 through 5.6 show the successive development of concentration profiles of each nuclide for the case that the leaching begins at t = 0 without any daughter nuclides in the repository. $N_i(z,t)$, $i=1,2,3$ is the concentration of the i^{th} nuclide in water phase at a location z and a time t.

N_1^0 is the concentration of ^{234}U at $z = 0$ just after beginning of leaching ($N_2^0 = N_3^0 = 0$). N_1^0 should be inversely proportional to the leach time, T. The concentrations of N_i are all relative to N_1^0, shown by the ratio N_i/N_1^0. In these figures, the solid lines show the concentration profiles of nuclides for the dispersion coefficient $D = 1\times10^{-1}$ m^2/yr, and the broken lines are for $D = 1\times10^3$ m^2/yr.

In Figure 5.3, t is less than the leach time, $T = 3\times10^4$ yr. ^{230}Th which is first leached out from the repository is at the location of v_2t. Let us first consider the case that the dispersion coefficient is small. All ^{230}Th in the region $v_2t < z < v_1t$ has been produced by the decay of ^{230}U outside the repository except a small amount from the contribution of small dispersion. In the region $v_2t < z < v_1t$, the concentration of ^{230}Th decreases with distance, and ends at the leading edge of ^{234}U band, i.e., $Z = v_1t$.

In the region of $z < v_2t$, there are two contributions for the concentration of ^{230}Th. One of them is the decay of ^{234}U that has been leached out from the repository. The other one is ^{230}Th which is from the decay of ^{234}U at the repository and leaches out. As seen in Figure 5.1, $N_2(o,t)$, the concentration of ^{230}Th at $z = o$, increases with time.

^{226}Ra has been migrating from the repository just after the beginning of leaching and arrives at the location around $z = v_3t$.

As shown in broken lines, due to the effect of the larger dispersion coefficient ($D = 1x10^3 \ m^2/yr$), the concentration profiles of ^{234}U and ^{230}Th are smoothed out, and the dent of ^{230}Th at $Z = V_2t$ disappears. But the dispersion hardly affects the leading edge of the ^{226}Ra profile, because ^{226}Ra has a much larger velocity along the migration path and λ_3 is large.

Figures 5.4 and 5.5 show the concentration profiles of the nuclides after the end of leaching. Before the time of $t=2x10^5$ yr, the total amount of ^{226}Ra in the migration column of soil continues to increase (see Figure 5.1). Since the width of the curve of ^{226}Ra profile becomes broader and broader with time, and the shape of the profile also changes with time, the highest peak does not appear at $t=2x10^5$ yr.

Figure 5.4 shows the concentration profiles after the time when the trailing edge of ^{234}U-band catches up with the first ^{230}Th from the repository. ^{230}Th behind the trailing edge of ^{234}U has to migrate without any contribution from the decaying ^{234}U-band, and decreases with time by its own decay. The distribution of ^{226}Ra is almost completely controlled by ^{226}Ra which has been produced by the decay of ^{230}Th outside the repository. In Figure 5.5 at $t=1x10^5$, another peak of ^{230}Th is growing around the trailing edge of ^{234}U. The peak of ^{226}Ra at about 800 m is still increasing compared to that of Figure 5.4. Figure 5.6 shows the concentration profiles of the nuclides at the time when the overall total amount of ^{226}Ra is a maximum. However, the height of its peak is lower than that in Figure 5.2.

The two peaks of ^{230}Th have almost same height. The time of $2x10^5$ yr is around the half-life of ^{233}U, at which the concentration of ^{234}U is about half of its initial value.

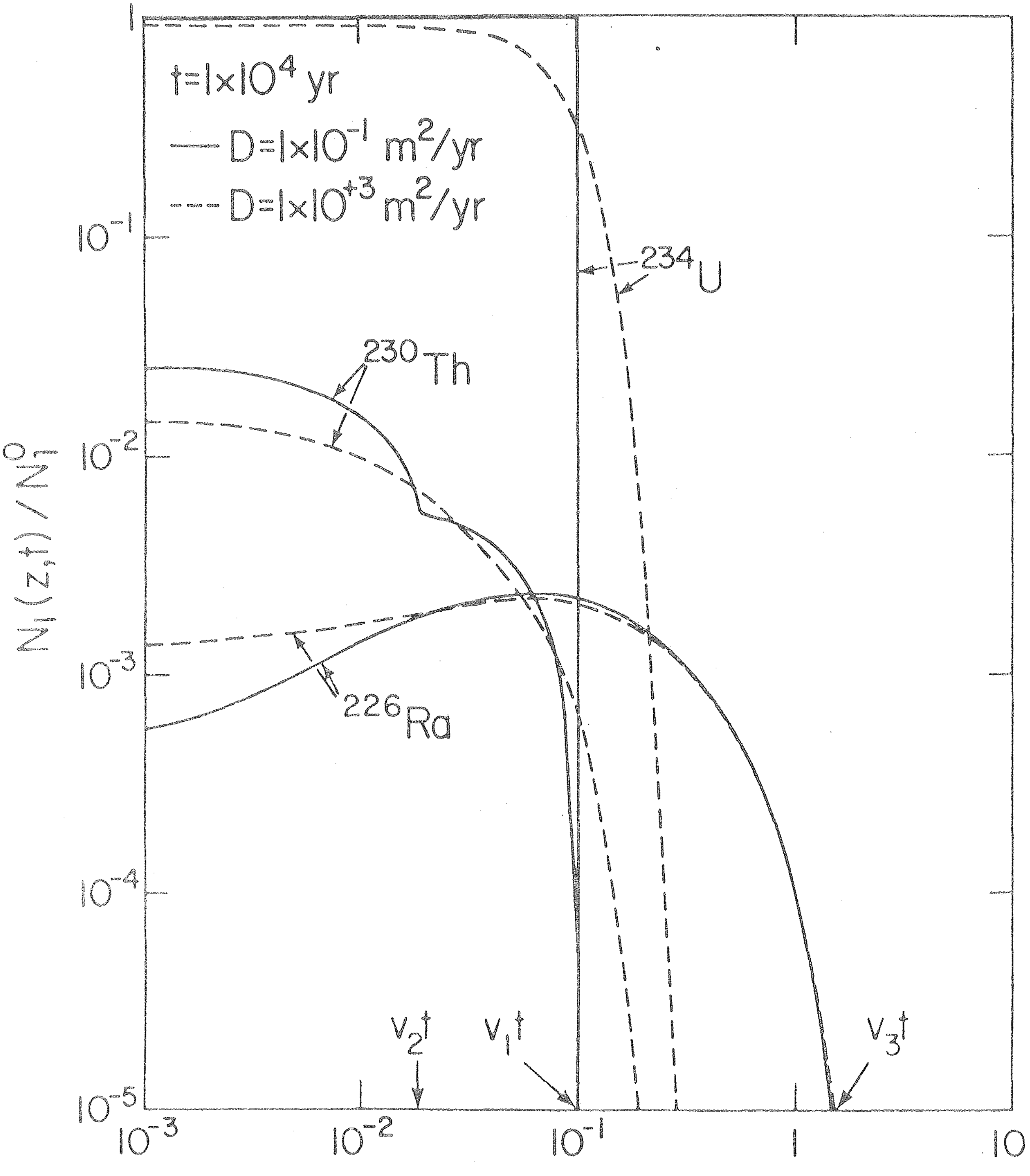

Figure 5.3 - Concentration profiles of $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at t = 1x10^4 yr. (Pure ^{234}U source at t=0. $N_i(z,t)$ = atoms/cm^3. N_1^0 = atoms/cm^3 of ^{234}U at z=0, t=0. T = 3x10^{4}yr. K_U = 1x10^4. K_{Th} = 5x10^4. K_{Ra} = 5x10^2).

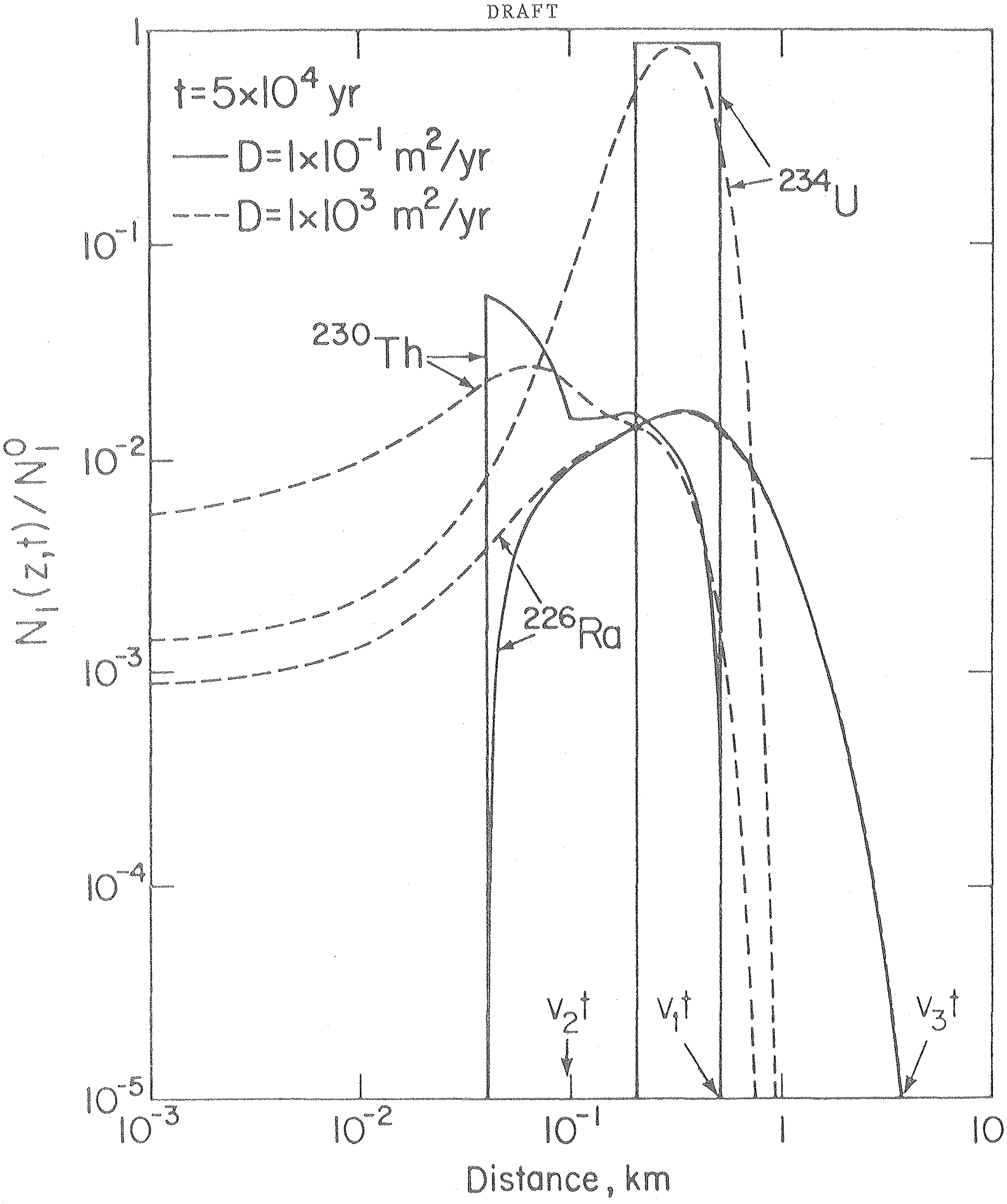

Figure 5.4 - Concentration profiles of $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain at t = 5×10^4 yr. (Pure ^{234}U source at t=0. $N_i(z,t)$ = atoms/cm^3. $N_1{}^o$ = atoms/cm^3 of ^{234}U at z=0, t=0. V=100m/yr. T=3×10^4 yr. K_U = 1×10^4. K_{Th}=5×10^4, K_{Ra} = 5×10^2).

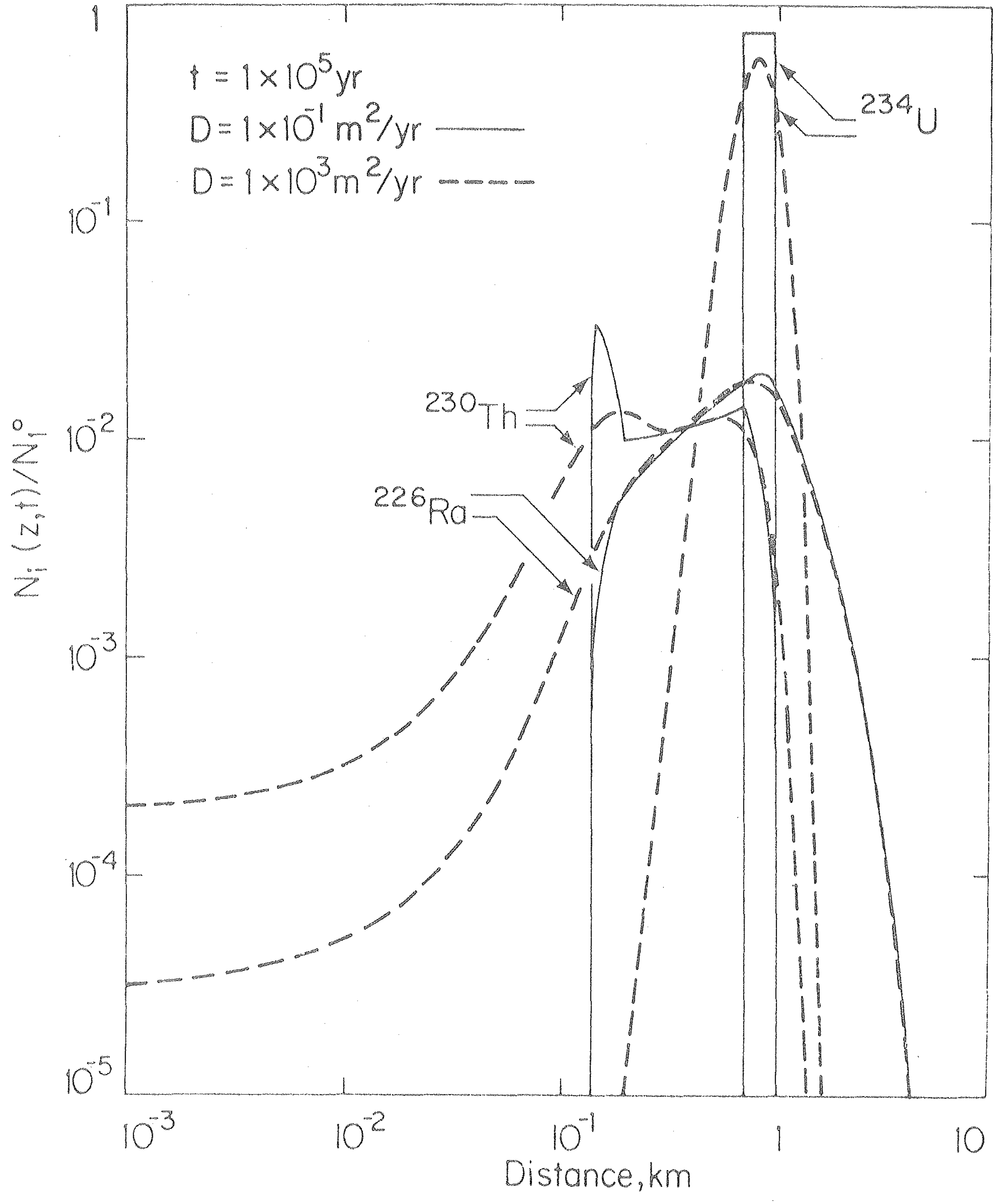

Figure 5.5 - Concentration profiles of $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at $t=1x10^5$ yr. (Pure ^{234}U source at t=0. $N_i(z,t)$= atoms/cm^3, N_o^1 = atoms/cm^3 of ^{234}U at z=0, t=0. V=100/m yr. $T=3x10^4$ yr. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

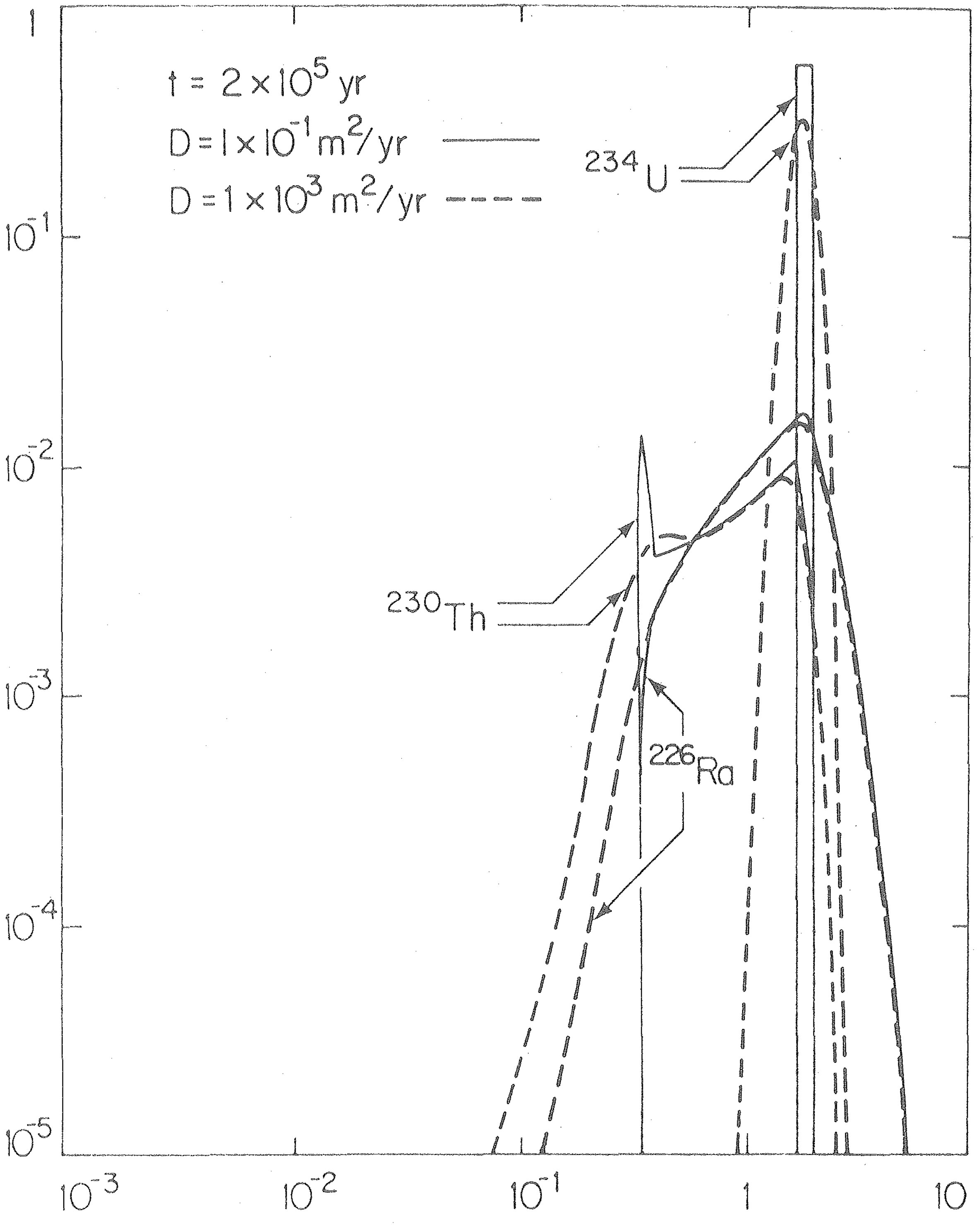

Figure 5.6 - Concentration profiles of $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at $t=2x10^5$ yr. (Pure ^{234}U source at t=0, $N_i(z,t)$ = atoms/cm^3. $N_1{}^o$=atoms/cm^3 of ^{234}U at z=0, t=0. V = 100m/yr. $T=3x10^4$ yr. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra} = 5x10^2$).

5.5.2.3 Concentration profiles of nuclides in the case of transient equilibrium sources in the repository at t=0.

In the case of transient equilibrium, the relatively large concentration of daughters (^{230}Th and ^{226}Ra) in the repository will have considerably large effect on the profile curves until around the time of the half-life of ^{234}U. (Half-life of ^{234}U is about three times longer than that of ^{230}Th).

In the transient equilibrium case, the total amount of nuclides involved in the repository decreases monotonously with time. Then, the tendency may be expected that the maximum concentration of ^{226}Ra along the migration path will occur at an earlier time in comparison with the case of initially pure ^{234}U source in the repository.

Figure 5.7 shows the concentration profiles of the nuclides at the time before the end of leaching. In the present case of $T = 3\text{x}10^4$ yr, the total amount of ^{226}Ra outside the repository increases with time until the end of leaching. As seen in the figure, for the case of small dispersion coefficient, the highest concentration of ^{226}Ra is realized at $z = v_2t$, at which the first ^{230}Th nuclide from the repository arrives. Because of the large concentration of ^{230}Th between the repository and $z = v_2 t$, and the condition of $v_2 < v_3$, a hump in the profile of ^{226}Ra occurs. The contribution of ^{230}Th to the concentration profile of ^{226}Ra between the leading edge of ^{234}U-band and $z = v_2 t$ is relatively not so large. The general effect of the large dispersion on migration pattern is almost the same as the case of pure ^{234}U source as shown in Figure 5.3 through 5.6. Figures 5.8 through 5.10 show the concentration profiles for times after the end of leaching. The highest peak of ^{226}Ra occurs for the case of small D at the time when the last ^{226}Ra from the repository catches up the first ^{230}Th nuclide from the repository at $z = v_2 t$. After that, the height of the peak decreases with time.

As seen in Figure 5.10, a new peak of ^{226}Ra is developed at the trailing edge of ^{234}U-band. The height of the peak at ^{234}U-band decreases more slowly with time than the peak at $z = v_2 t$ does, because the half-life of ^{234}U is longer than that of ^{230}Th.

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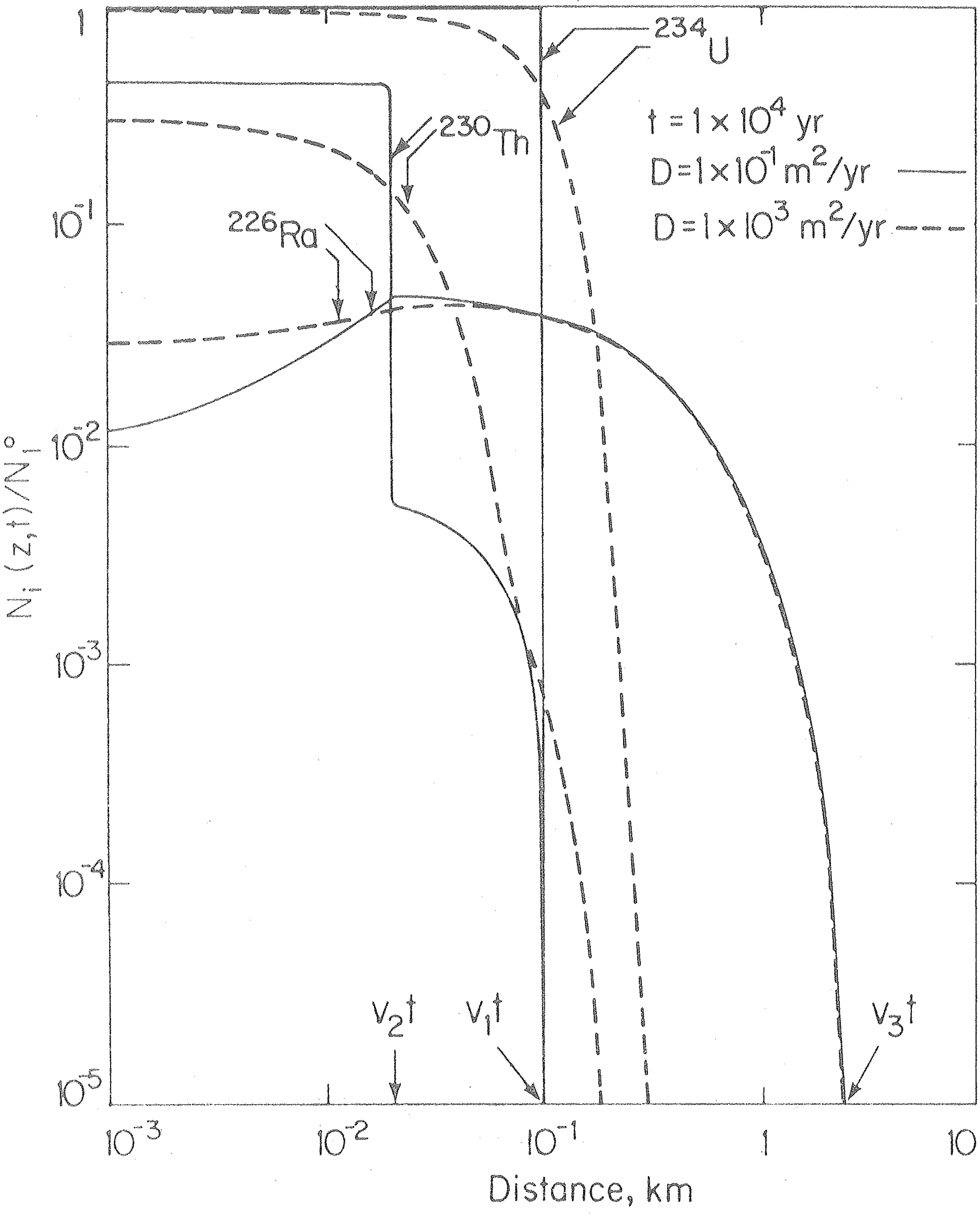

Figure 5.7 - Concentration profiles of $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain at $t=1x10^4$yr. (Transient equilibrium sources at t=0. $N_i(z,t)$ = atoms/cm^3, $N_1{}^o$ = atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. $T=3x10^4$yr. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

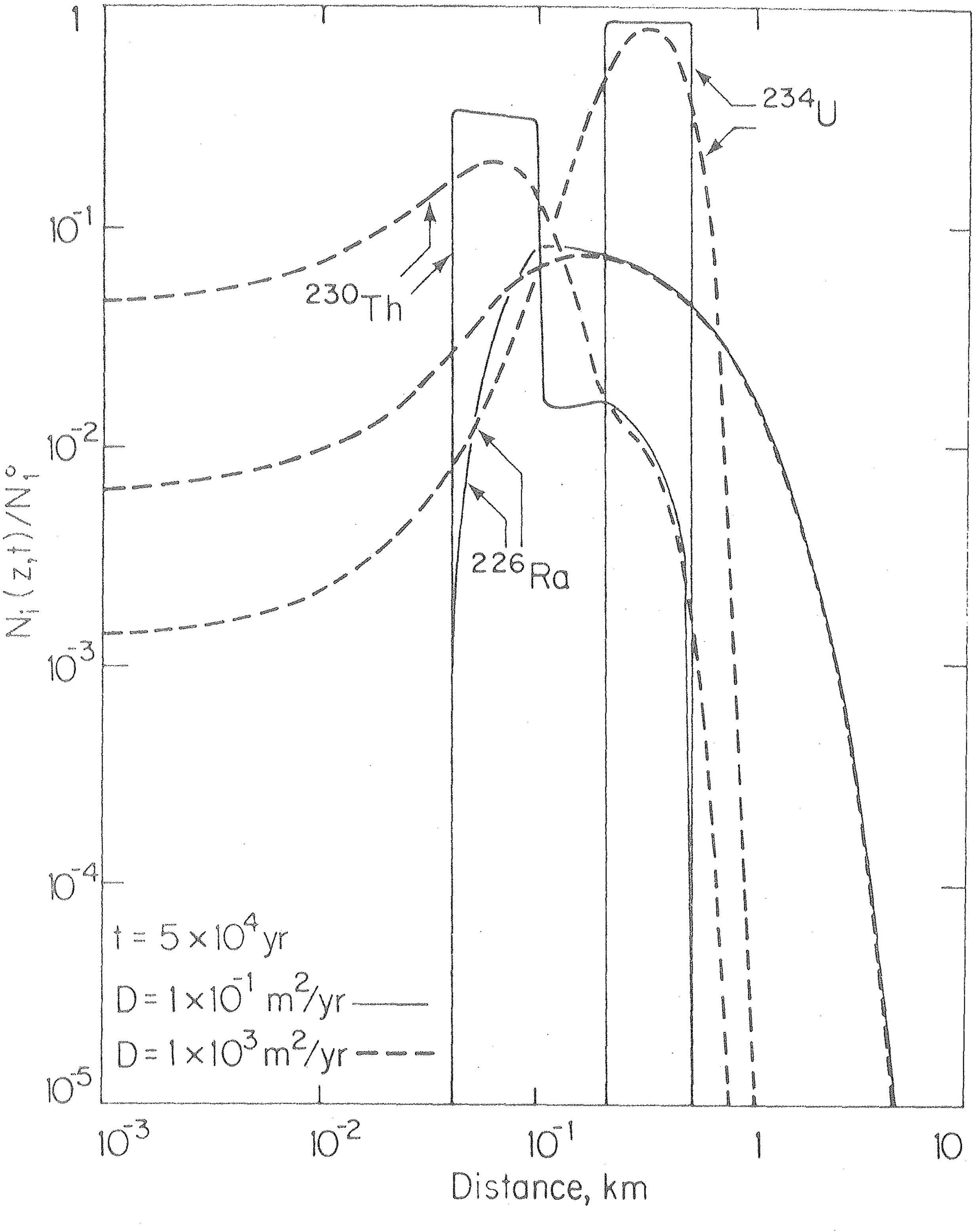

Figure 5.8 - Concentration profiles of $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at $t = 5\times10^4$ yr. (Transient equilibrium sources at t=0. $N_i(z,t)$ = atoms/cm^3. $N_1{}^o$=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. T=3×10^4yr. $K_U=1\times10^4$, $K_{Th} = 5\times10^4$, $K_{Ra}=5\times10^2$).

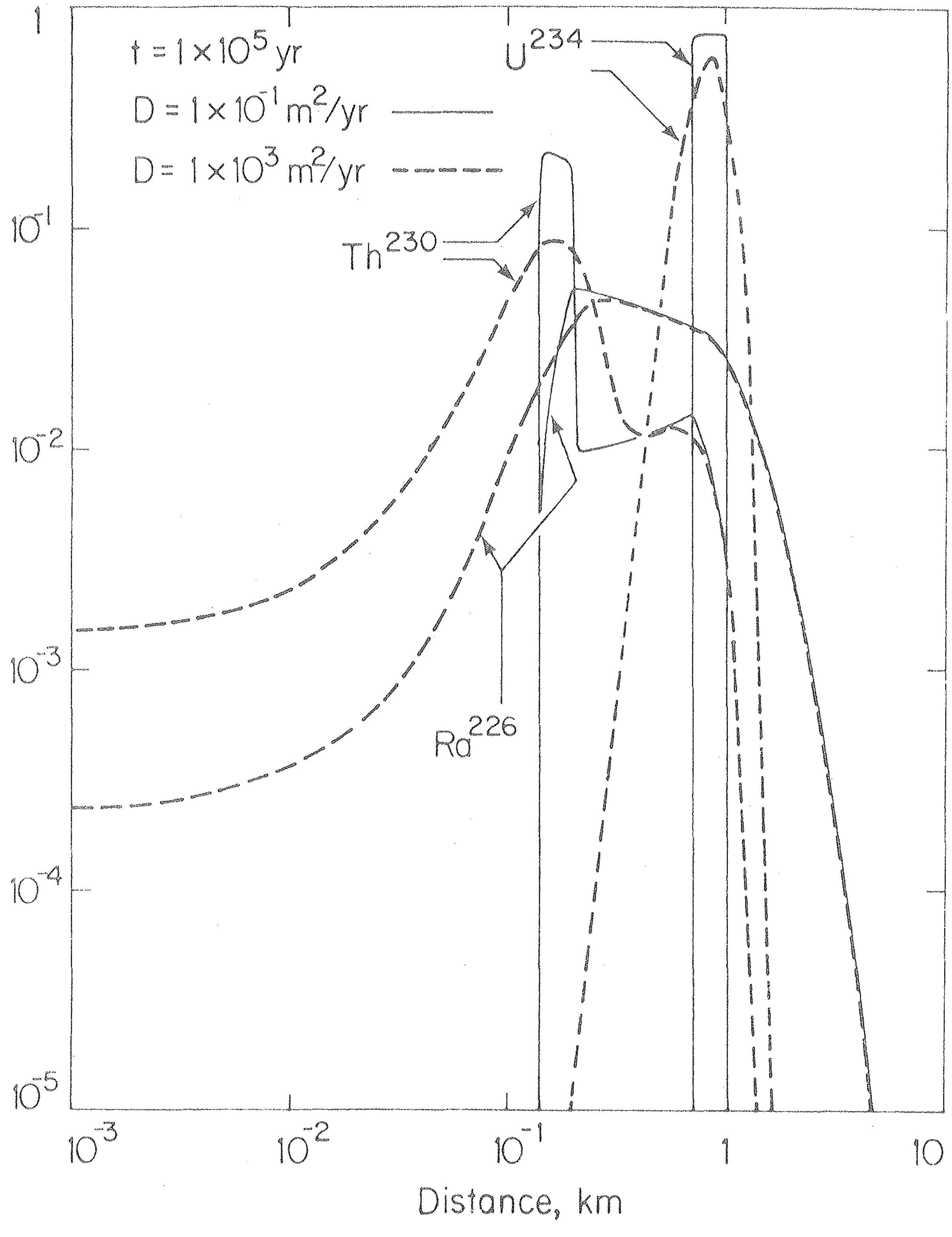

Figure 5.9 - Concentration profiles of $^{234}U \xrightarrow{} {}^{230}Th \xrightarrow{} {}^{226}Ra$ decay chain at t=1x10^{5}yr. (Transient equilibrium sources at t=0. $N_i(z,t)$ = atoms/cm^3. $N_1{}^o$=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. T=3x10^{4}yr., K_U=1x10^4. K_{Ra}=5x10^2).

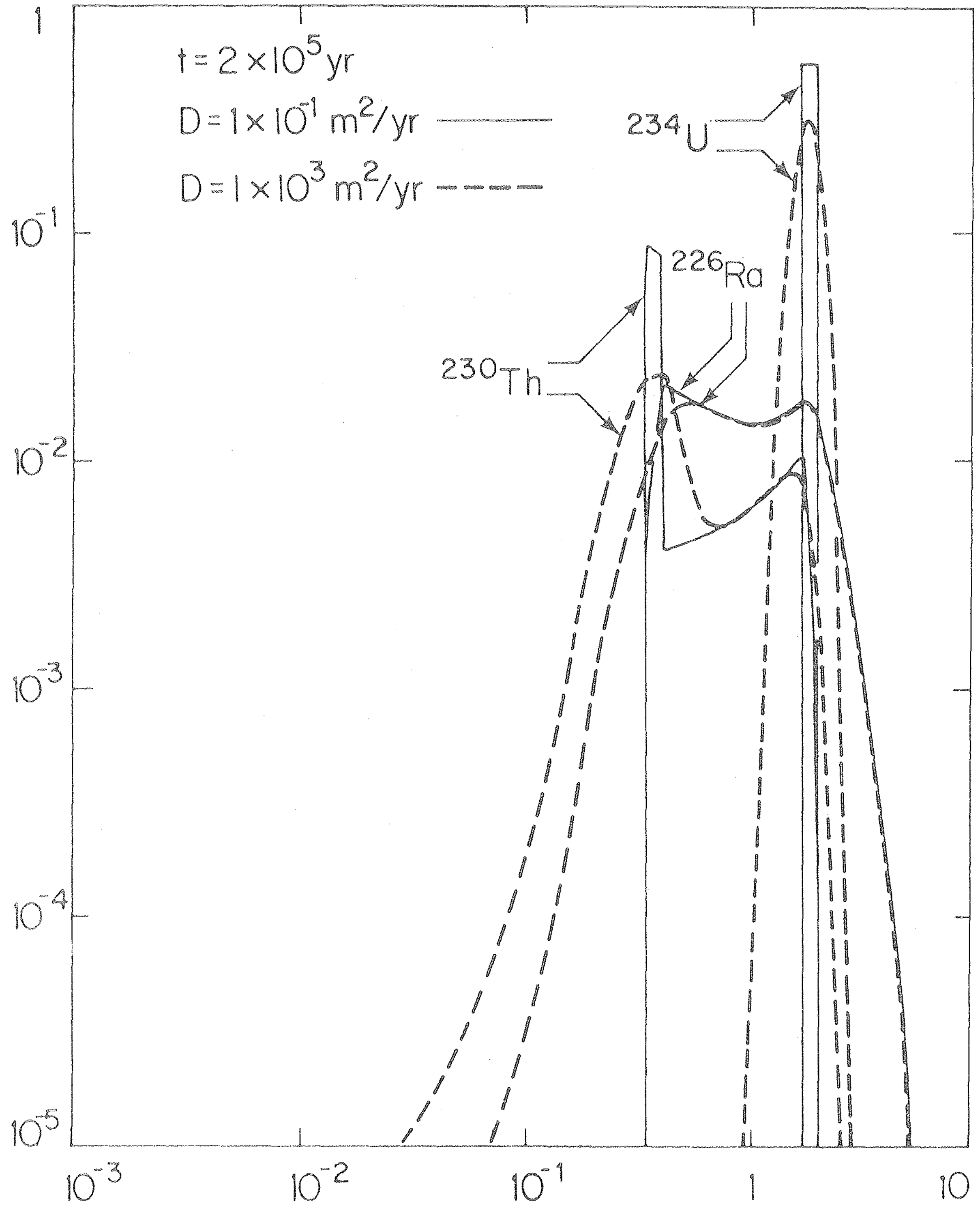

Figure 5.10 - Concentration profiles of $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at $t=2x10^5$yr. (Transient equilibrium sources at t=0. $N_i(z,t)$=atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. $T=3x10^4$yr. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

5.5.3 Maximum Concentration and Isopleth of ^{226}Ra

Figures 5.11 and 5.12 are the plots of the maximum ^{226}Ra concentration occurring at various locations along the migration path for the cases of initial pure ^{234}U source in the repository and initial transient equilibrium sources. The two solid curves represent cases of different dispersion coefficients. Each point on the curve means the maximum ^{226}Ra concentration an observer will eventually experience by sitting at a location long enough. Thus, the time when the maximum concentration occurs at each location is different. Each point is obtained by choosing a fixed location Z and compute the ^{226}Ra curve numerically for various times until a maximum concentration is obtained by plotting the concentration versus time for that fixed location. All these curves have a hump, that means starting from the repository at Z = 0, one sees an increasing maximum ^{226}Ra concentration as one goes farther away from the site, until one passes a certain location where the highest concentration occurs, the nuclide concentration then decreases. This happens both for the case of initial pure ^{234}U source and the case of initial transient equilibrium sources in the repository so that the decaying of the parents does not completely explain this increasing maximum ^{226}Ra concentration phenomenon. In particular for the case of initial transient

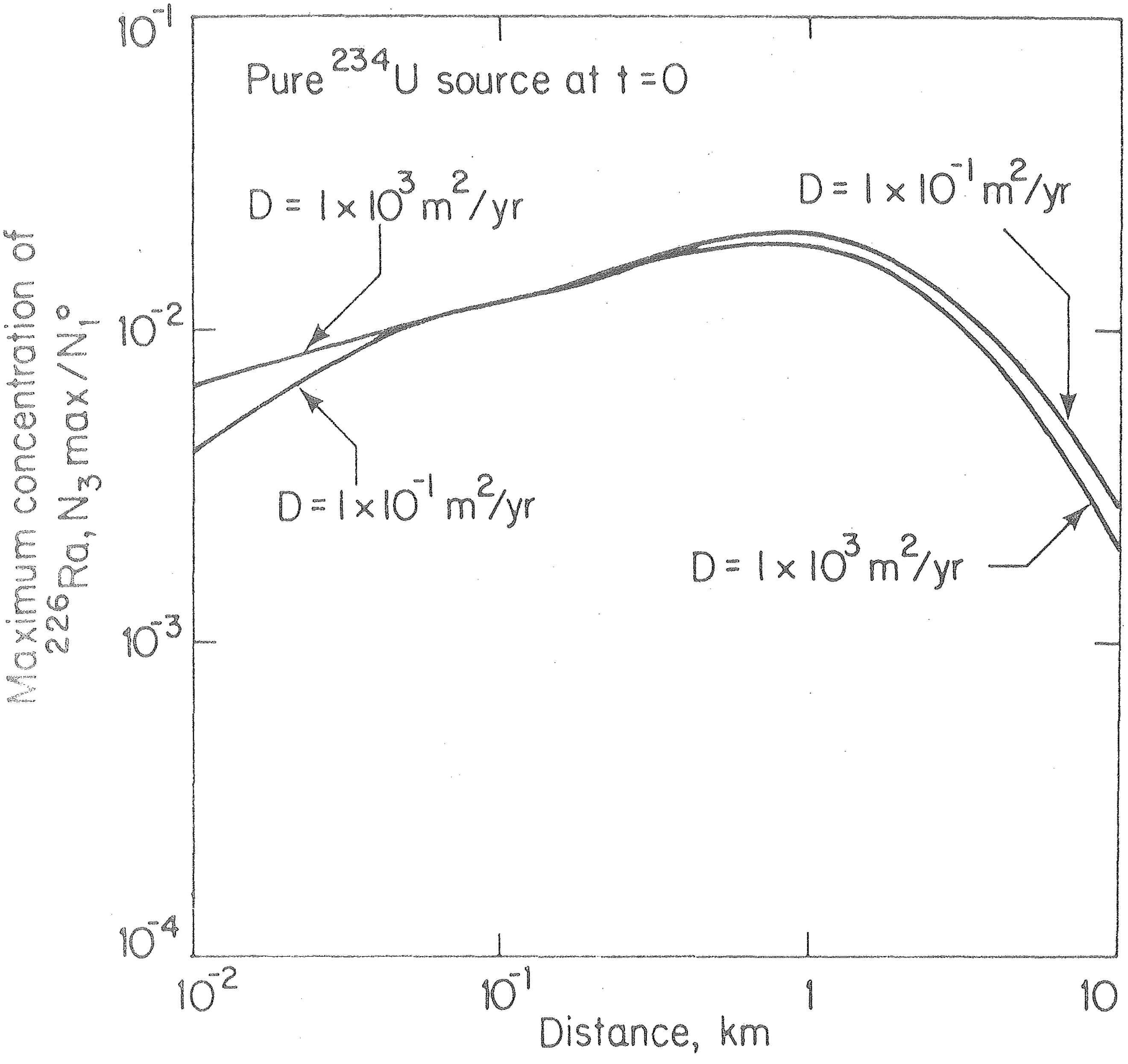

Figure 5.11 - Effect of dispersion coefficient on the maximum concentration of ^{226}Ra for the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain. (Pure ^{234}U source at t=0. $N_i(z,t)$ = atoms/cm^3. N_1^{o}=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. T=3x10^4 yr. K_U=1x10^4. K_{Th}=5x10^4. $K_R a$ = 5x10^2).

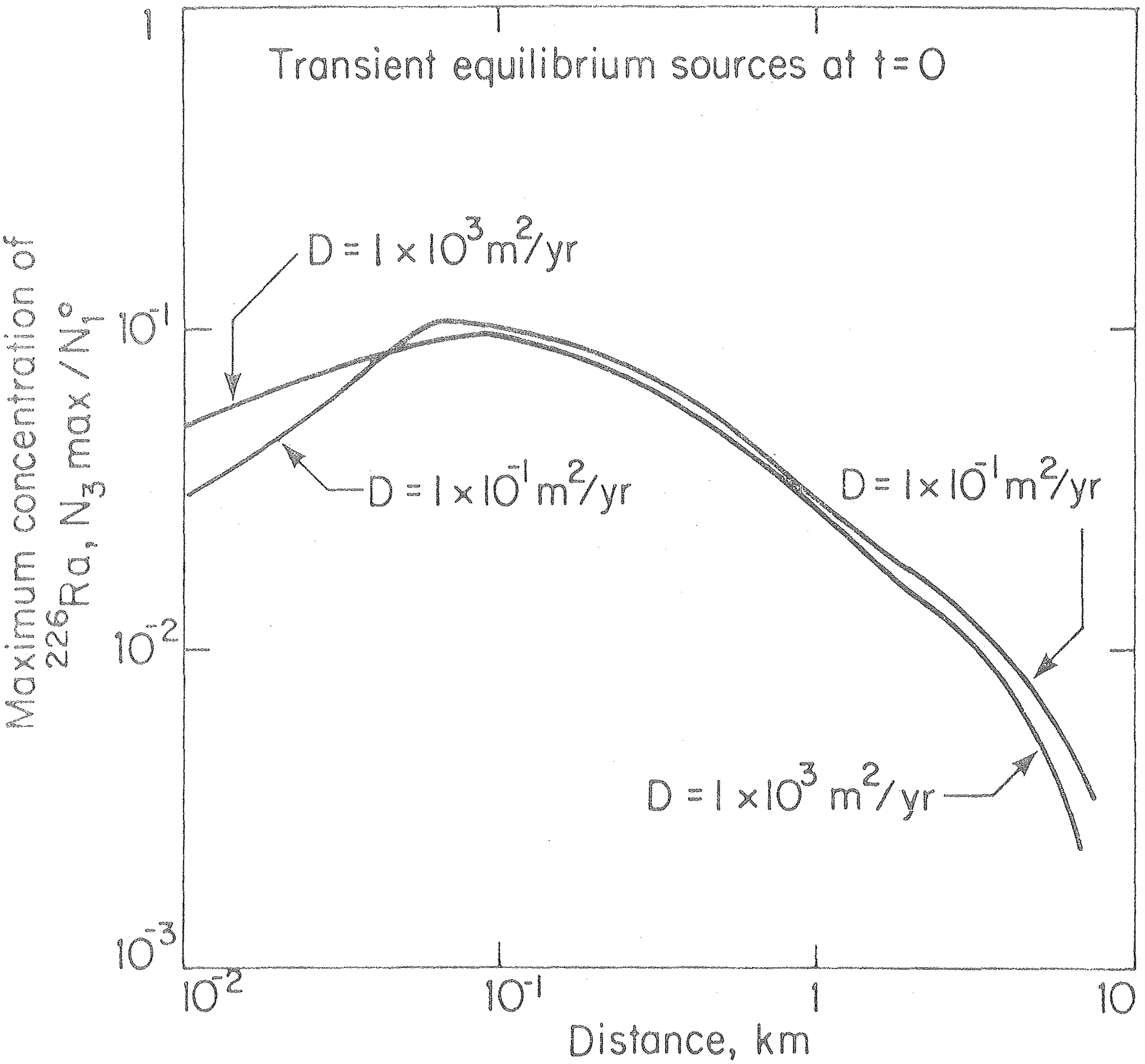

Figure 5.12- Effect of dispersion coefficient on the maximum concentration of ^{226}Ra for the $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain. (Transient equilibrium sources at t=0. $N_I(z,t)$ = atoms/cm^3. $N_1{}^o$=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. T=3x10^{4}yr. K_U = 1x10^4. K_{Th}=5x10^4. K_{Ra} = 5x10^2).

equilibrium sources, this phenomenon is called the "Reconcentration" phenomenon of ^{226}Ra(B4). This reconcentration phenomenon of ^{226}Ra occurs also for the two member decay chain ^{230}Th→^{226}Ra; since for a three member chain the uranium is providing an additional source by decaying to ^{230}Th, the approach to the explanation should be similar for both cases of two and three member chains.

Consider a simple model that only the decay of ^{230}Th is significant in contributing to the source of ^{226}Ra, and an observer is sitting near the repository. When the band of ^{230}Th reaches him, he will detect ^{226}Ra from the immediate decay of ^{230}Th. At the same time, ^{226}Ra which is from the decay of ^{230}Th at an earlier time also catches up to reach him because ^{226}Ra is two orders of magnitude faster than ^{226}Th. As the observer is near the site and the nuclides are always migrating in the forward direction, it is the band of ^{230}Th between the site and the observer that contributes to the concentration of ^{226}Ra at the observer's position, so that when he is farther away, a longer band of ^{230}Th is contributing to a larger concentration of ^{226}Ra, therefore it explains the increasing slope of the curves in the above figures. Because ^{230}Th undergoes decay also, when the distance of the observer is increased, ^{226}Ra has to take a longer time to reach him and so more ^{226}Ra may have to decay away before reaching him. On the other hand the number of ^{230}Th decaying at an earlier instant is greater because less of the original ^{230}Th has decayed at that earlier instant. Therefore the contribution to ^{226}Ra are not linear with increasing distance from the repository.

Eventually when the observer is at an even farther distance from the repository, the band of ^{230}Th will have a lower concentration due to decay even though the band width of ^{230}Th may increase and so from there on the ^{226}Ra concentration decreases with distance as shown by the negative gradient of the curves in the figures. Thus it is seen that one of the main reasons for this reconcentration phenomenon to occur is due to the fact that the daughter nuclides are migrating forward faster than the parent nuclides.

It is observed that in both Figures the dotted curves for the larger dispersion coefficients have their highest concentrations always under those of the solid ones of lower dispersion coefficients. Since for a certain instant, after certain time of leaching, there is a certain amount of nuclides along the path, if the nuclides are more dispersive, they will spread out more along the path, thus sacrificing their highest magnitude of concentration along the path.

In both Figures 5.11 and 5.12 the locations where the curves reach their highest point are 840 m (for $D=1x10^{-1}m^2/yr$) and 800 m (for $D=1x10^3m^2/yr$) for initial no daughter case and are 70 m (for $D=1x10^{-1}m^2/yr$ and 86 m(for $D=1x10^3m^2/yr$) for initial transient equilibrium case. The locations of highest point of the former case are much farther from the repository than that of the latter case. As for the magnitude of the highest point, the latter case is about five times larger than that of the former, because of the initial inventories of ^{230}Th and ^{226}Ra at $t = 0$ in the repository. The effect of the large dispersion on the distribution appears significant near the repository, in making the curve flatter and slightly lower in magnitude. One can see the effect is more clearly shown from the plots of isopleths of ^{226}Ra concentration given in Figure 5.13 and 5.14 for initial pure ^{234}U case and in Figure 5.15 and 5.16 for initial transient equilibrium case. The abscissa shows distance (km) and the ordinate shows time (yr). The isopleths are written for the value of $N_3(Z,t)/N_1^0$ from $5x10^{-3}$ to $2x10^{-2}$ with an interval of $5x10^{-3}$ for each isopleth for the former case, and from $2x10^{-2}$ to $8x10^{-2}$ with an interval of $2x10^{-2}$ for the latter case. They indicate that the time-dependent maximum ^{226}Ra concentration at any given location increases with distance until the highest concentration is reached, after which it decreases monotonically with distance. This is mainly due to: a) the reconcentration phenomenon due to the distribution of sorbed ^{230}Th through the media, and b) gradual growth of ^{226}Ra in the waste material in the case of initial pure ^{234} source. Let us consider Figure 5.13 and Figure 5.14 which are for $D=1x10^{-1}$ and $D=1x10^{3}m^{2}/yr$. As shown in these figures the effect of dispersion on the slope of the isopleth is less important except near the repository. The broken lines in these figures indicate the time when the concentration of ^{226}Ra at any given location reaches its maximum value. One can see that the broken lines agree with the ridge line of the isopleths. Each ridge line lies on the line of $T = 3x10^{4}$ (leach time) near the repository, until a distance around 0.1 km, and finally at large distance it lies asymptotically along the line $t=Z/V_1$ which is the locus of the leading edge of ^{234}U band. The behavior of the ridge line is not so much affected by the change of the dispersion coefficient for the case of pure ^{234}U source. Contrarily, the effect of the dispersion is much more important in the case of initial transient equilibrium as shown in Figure 5.15 and 5.16. First, comparing the plots in Figure 5.15 and Figure 5.16, the slopes of each isopleth on the side of earlier time are almost the same, but the slope of the isopleth on the side at a later time in Figure

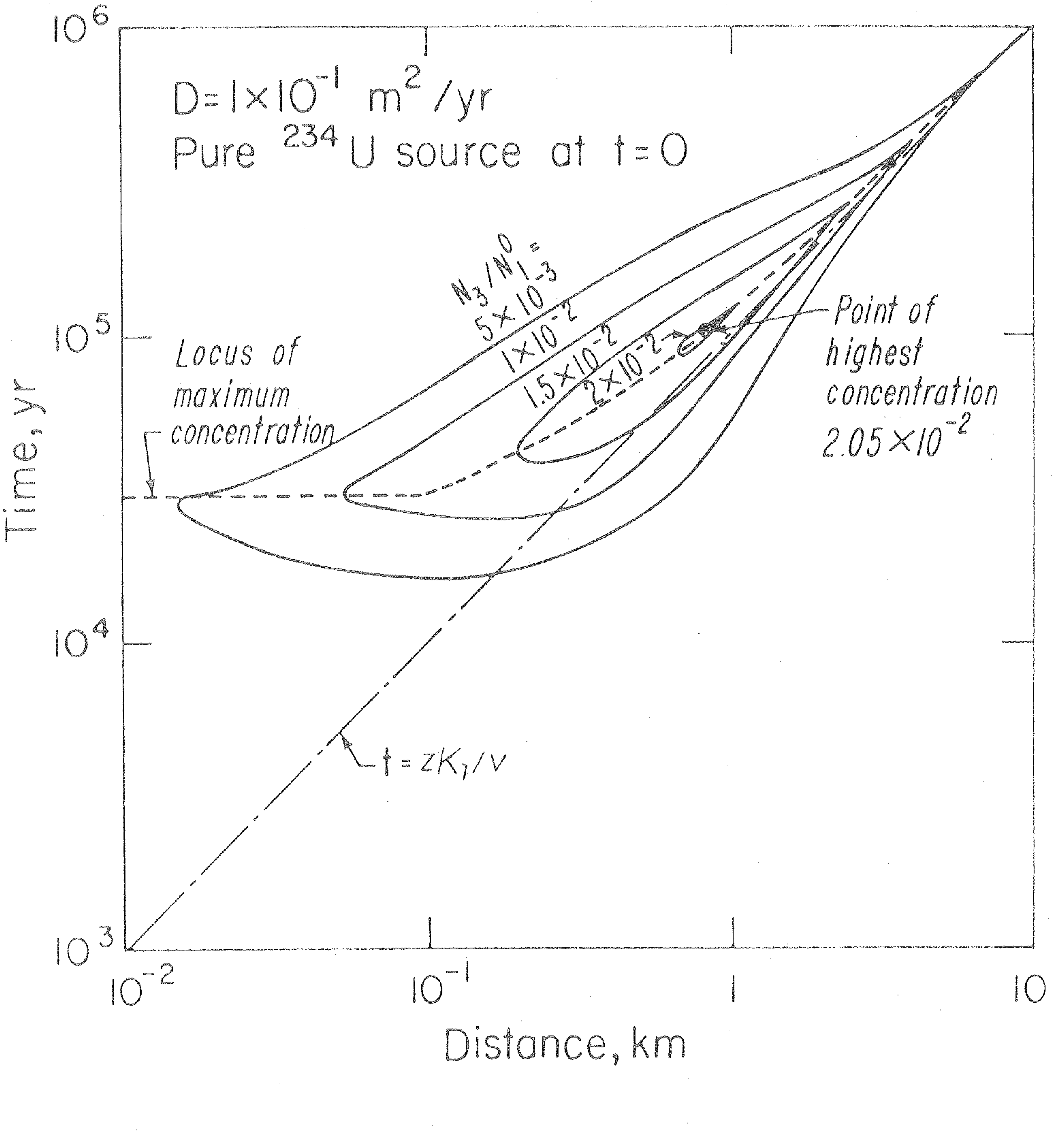

Figure 5.13 - Isopleths of ^{226}Ra for the $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain. (Pure ^{234}U source at t=0. $N_i(z,t)$ = atoms/cm^3 N_i^o=atoms/cm^3 of ^{234}U at z=0, t=0. V=100 m/yr. D=1×10^{-1}m^2/yr. T=3×10^4 yr. $K_U=1\times10^4$. $K_{Th}=5\times10^4$. $K_{Ra} = 5\times10^2$.

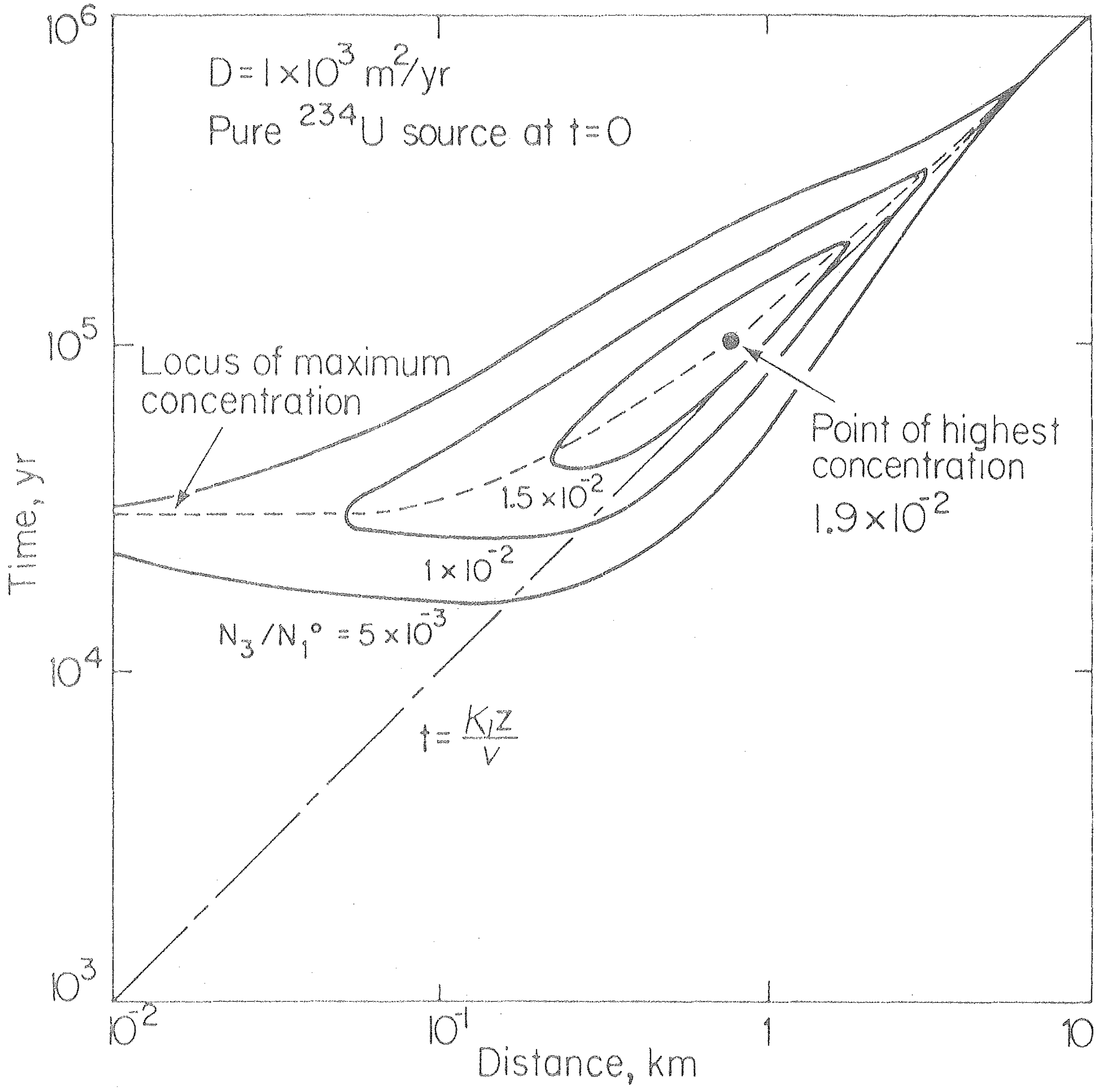

Figure 5.14 - Isopleths of ^{226}Ra for the $^{234}U\rightarrow^{230}Th\rightarrow^{226}Ra$ decay chain. (Pure ^{234}U source at t=0, $N_i(z,t)$ = atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. D=1x10^{3}m^2/yr. T=3x10^{4}yr. K_U=1x10^4. K_{Th}=5x10^4. K_{Ra}=5x10^2).

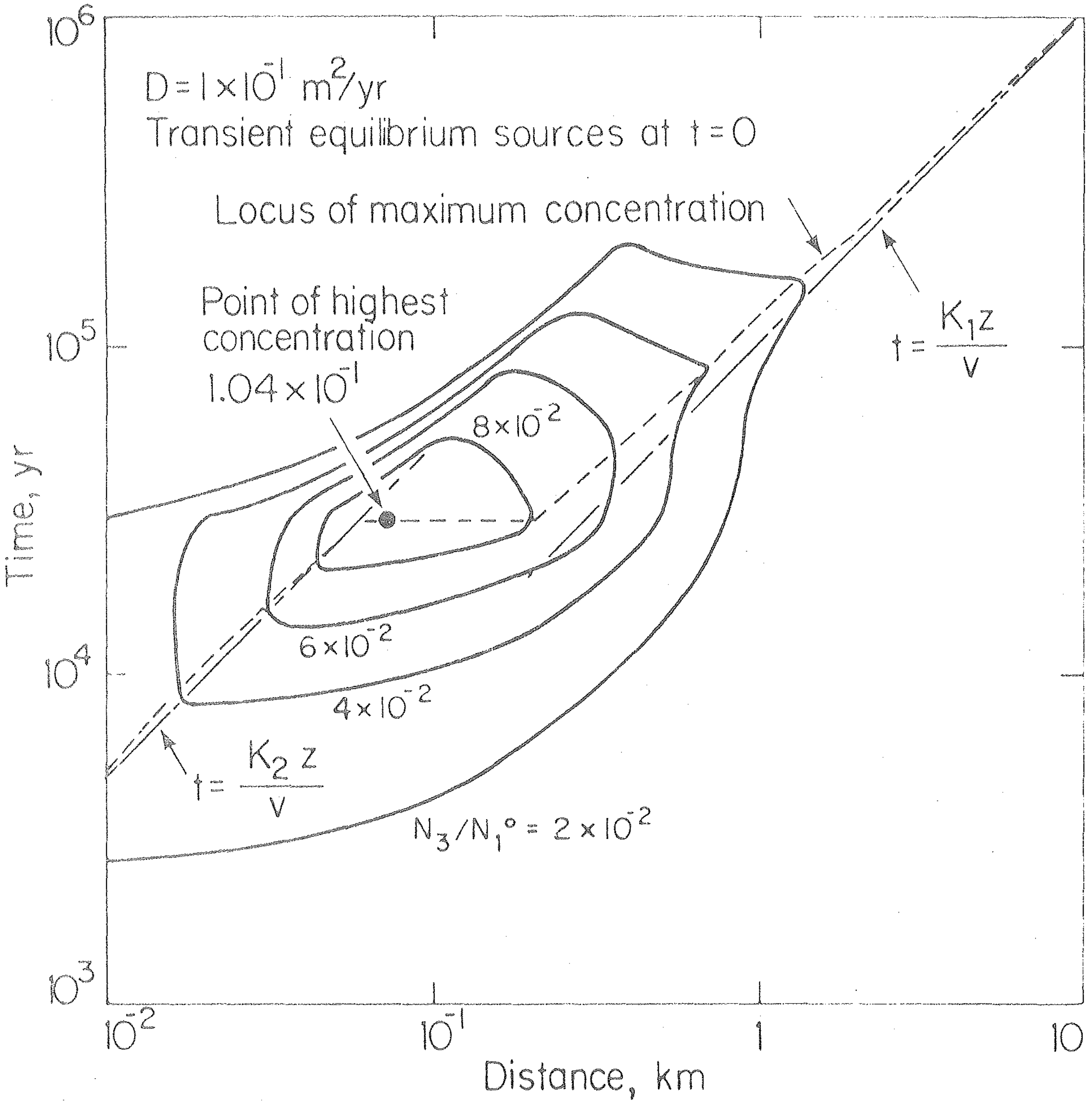

Figure 5.15 - Isoplethds of ^{226}Ra for the $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain. (Transient equilibrium sources at t=0. $N_i(z,t)$= atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. $D=1x10^{-1} m^2/yr$. $T=3x10^4$ yr., $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

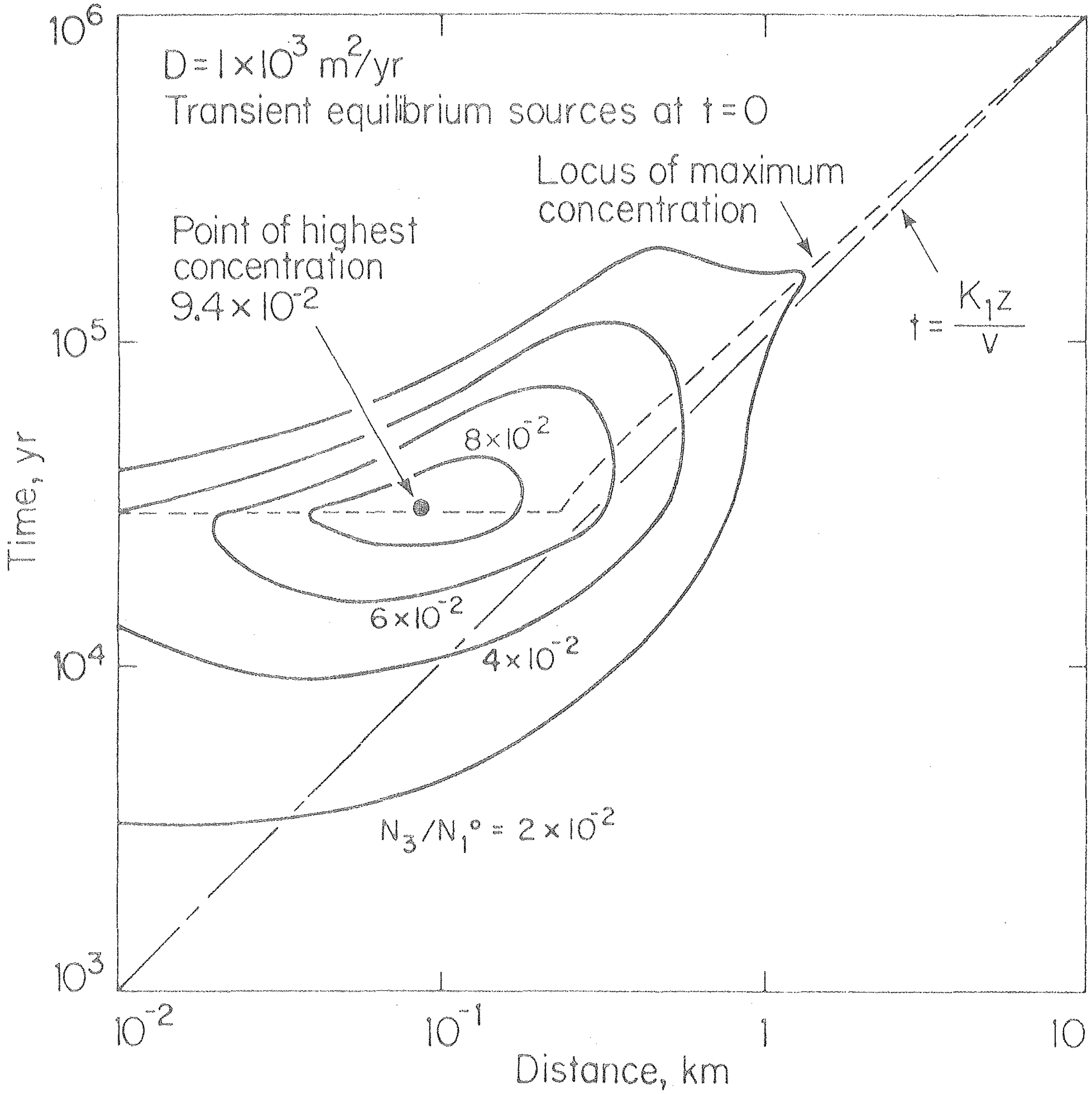

Figure 5.16 - Isopleths of ^{226}Ra for the $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain. (Transient equilibrium sources at t=0. $N_i(z,t)$-atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. D=1×10^3m^2/yr. T=3×10^4yr. $K_U = 1 \times 10^4$. $K_{Th} = 5 \times 10^4$. $K_{Ra} = 5 \times 10^2$).

5.16 is more gentle than that of Figure 5.15, especially around the region 10 ∿ 200 m from the repository. This means that the effect of ^{226}Ra lasts longer in the case of larger dispersion. For example, the time interval for the concentration of ^{226}Ra to reach the value of 2×10^{-2} is about 62,000 yr for the small D case, and is about 73,000 yr for the large D case. It is because ^{234}U and ^{230}Th, parent nuclides of ^{226}Ra, have long tails towards the direction of the repository due to the large D after the end of leaching $(t > T)$. Second, more important difference appears in the behavior of the line of maximum concentration of ^{226}Ra. For the small D case, the line almost agrees with the line $t=Z/v_2$ which is the locus of the leading edge of ^{230}Th, from the repository site up to 60 m from the repository. Then it lies on the line $t = 3\text{x}10^4$ yr (leach time) up to Z=200 m, and finally it comes asymptotically to the line $t=Z/v_1$. For the large D case, however, the locus of the highest concentration does not lie on the line $t = Z/V_2$ but instead it lies on the line $t = 3\text{x}10^4$ yr from the repository site up to a distance of Z = 240 m. This behavior may come from the shape of the $t=Z/v_2$ edge of the ^{230}Th band. For the small D case, it is so called box shape, and the peak of the highest ^{226}Ra concentration is seen at $Z = V_2t$ in Figure 5.15. But for the large D case, this peak is shifted to the right because the $t = Z/v_2$ edge of ^{230}Th band is being smoothly spread out. Thus, one can find that the effect of dispersion to the isopleth profile of ^{226}Ra concentration is more important in the case of initial transient equilibrium sources in the repository. Furthermore, as seen in the isopleths, the slope of the isopleths around the summit (highest point) is not steep, so for evaluating the hazardous effect of ^{226}Ra, it is insufficient just to find the maximum concentration of ^{226}Ra at each location, but one also has to examine the distribution of ^{226}Ra concentration around each location using the isopleth plotting.

5.5.4 Parametric Study

5.5.4.1 The effect of sorption equilibrium constant on the concentration profiles of ^{226}Ra.

Figure 5.17 shows the concentration profiles of ^{226}Ra at various locations for different sorption equilibrium constants of ^{226}Ra ($K_3/10$, K_3, $10K_3$) in the case of initially pure ^{234}U source, at $t=5x10^4$ yr and with $D=1x10^{-1} m^2/yr$. In the figure, the abscissa is a linear scale in units of Z/v_3yr and the ordinate is the concentration of ^{226}Ra in logarithmic scale. The K_3 and $K_3/10$ curves decrease with ZK_3/V linearly,

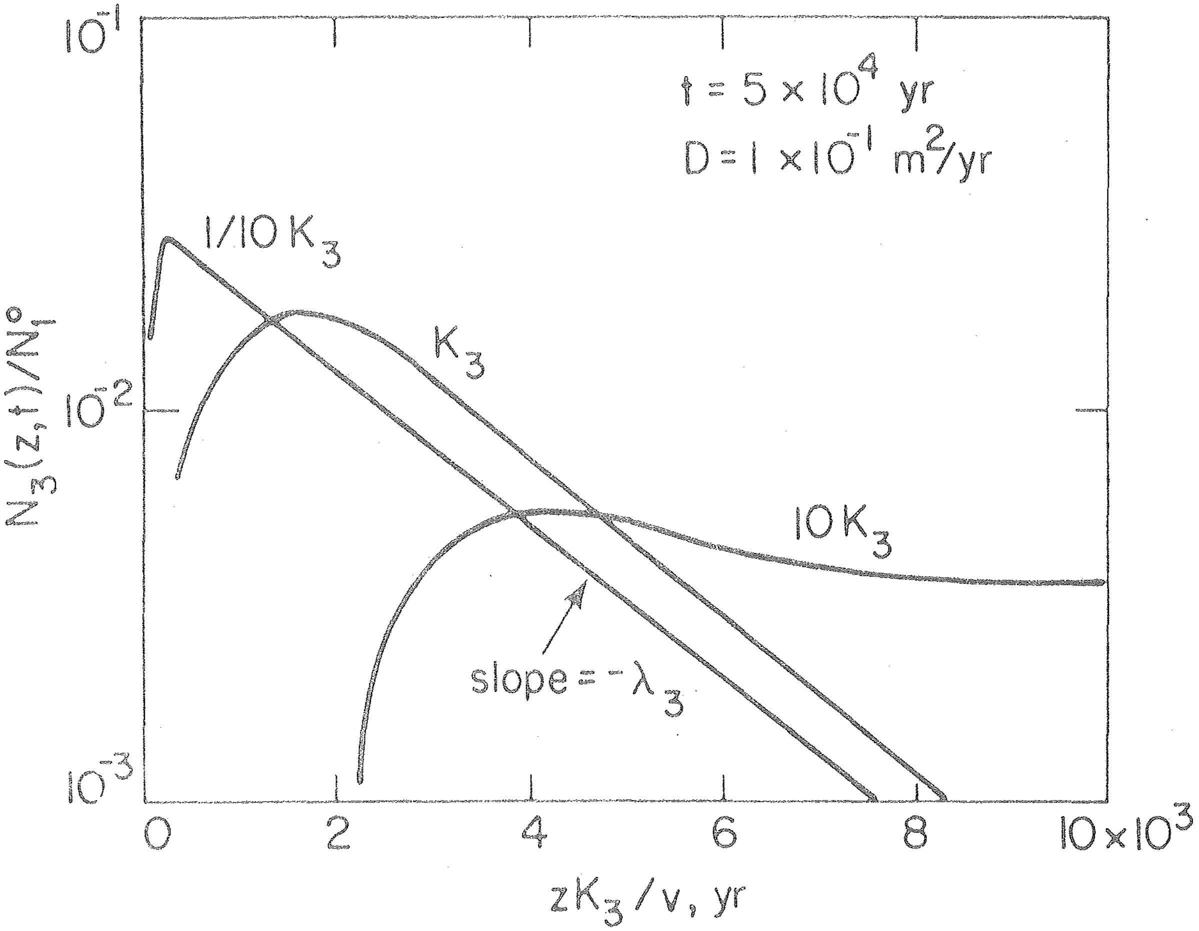

Figure 5.17 - Effect of sorption equilibrium constant on concentration profiles of ^{226}Ra at t = $5\text{x}10^4$/yr for the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain. (Pure ^{234}U source at t=0. $N_i(z,t)$ = atoms/cm^3. $N_1{}^o$=atoms/cm^3 of ^{234}U at z=0, t=0. v=100m/yr. D=$1\text{x}10^{-1}$m^2/yr. T=$3\text{x}10^4$/yr. K_U=$1\text{x}10^4$. K_{Th}=$5\text{x}10^4$. K_{Ra} =$4\text{x}10^2$)

and their slopes are -4.69×10^{-4} yr^{-1} for the K_3 curve, and -4.33×10^{-4}yr^{-}1 for the $K_3/10$ curve. Since the decay constant of ^{226}Ra(λ_3) has the value of 4.332×10^{-4} yr^{-1}, same as the value of the slope for the $K_3/10$ curve, so one can find that at distance far from the repository the concentration of ^{226}Ra decreases exponentially according to $e^{-\lambda_3(Z/v_3)}$ for the case that the velocity of ^{226}Ra ($v_3 = v/K_3$) is much larger than that of its parent nuclides. This characteristic can be explained by the explicit solution for the dispersion free case and step release mode (5.16). For $v_1 t < Z < v_3 t$, the solution is as follows:

$$\begin{aligned}
N_3^s(z,t) = {} & e^{-\lambda_3 z/v_3}\left[B_{31} e^{-\lambda_1(t-z/v_3)} + B_{32} e^{-\lambda_2(t-z/v_3)} + B_{33} e^{-\lambda_3(t-z/v_3)}\right] \\
& + \frac{\lambda_1 \lambda_2}{v_1 v_2} \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{13}\Gamma_{23}} \left[\frac{D_{13}^{31} B_{11}}{\lambda_1 - \Delta_{13}} \left\{ e^{-\Delta_{13}(t-z/v_3)} - e^{-\lambda_1(t-z/v_3)} \right\} \right. \\
& \qquad \left. + \frac{D_{23}^{31} B_{11}}{\lambda_1 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t-z/v_3)} - e^{-\lambda_1(t-z/v_3)} \right\} \right] \\
& + \frac{\lambda_2}{v_2} \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{23}} \left[\frac{D_{23}^{32} B_{21}}{\lambda_1 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t-z/v_3)} - e^{-\lambda_1(t-z/v_3)} \right\} \right. \\
& \qquad \left. + \frac{D_{23}^{32} B_{22}}{\lambda_2 - \Delta_{23}} \left\{ e^{-\Delta_{23}(t-z/v_3)} - e^{-\lambda_2(t-z/v_3)} \right\} \right]
\end{aligned} \tag{5.44}$$

Let's consider the case that $\lambda_3 >> \lambda_1, \lambda_2, v_3 >> v_1, v_2$ and $t >> 1/\lambda_3$. These conditions yield that $\lambda_3 - \lambda_i \cong \lambda_3, v_3 - v_1 \cong v_3$ and $e^{-\lambda_3 t} \cong 0$ where i = 1 or 2. With this approximation, one can rewrite (5.44) as

$$N_3^s(z,t) \cong G(t) \cdot e^{-\lambda_3 z/v_3} \tag{5.45}$$

where

$$\begin{aligned} G(t) = {} & B_{31} e^{-\lambda_1 t} + B_{32} e^{-\lambda_2 t} \\ & + \frac{\lambda_1 \lambda_2}{v_1 v_2} \frac{1}{\Gamma_{13} \Gamma_{23}} \left[\frac{D_{13}^{31} B_{11}}{\lambda_1 - \Delta_{13}} \left(e^{-\Delta_{13} t} - e^{-\lambda_1 t} \right) \right. \\ & \qquad \left. + \frac{D_{23}^{31} B_{11}}{\lambda_1 - \Delta_{23}} \left(e^{-\Delta_{23} t} - e^{-\lambda_1 t} \right) \right] \\ & + \frac{\lambda_2}{v_2} \frac{1}{\Gamma_{23}} \left[\frac{D_{23}^{32} B_{21}}{\lambda_1 - \Delta_{23}} \left(e^{-\Delta_{23} t} - e^{-\lambda_1 t} \right) \right. \\ & \qquad \left. + \frac{D_{23}^{32} B_{22}}{\lambda_2 - \Delta_{23}} \left(e^{-\Delta_{23} t} - e^{-\lambda_2 t} \right) \right] \end{aligned} \tag{5.46}$$

Using the superposition theorem, one can obtain the solution of ^{226}Ra, $N_3^b(Z,t)$, for the band release.

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$$N_3^b(z,t) = N_3^s(z,t\,;B_{ij}) - N_3^s(z,t-T\,;B_{ij}e^{-\lambda_j T})$$

$$= \left\{ G(t;B_{ij}) - G(t-T\,;B_{ij}e^{-\lambda_j T}) \right\} e^{-\lambda_3 z/v_3}$$

(5.47)

Thus, the solution for the band release shows that the concentration of ^{226}Ra decreases with distance exponentially according to $e^{-\lambda_3 Z/V_3}$.

5.5.4.2 The effect of sorption equilibrium constant on the maximum discharge rate of ^{226}Ra.

Figure 5.18 and 5.19 show the effect of varying the sorption equilibrium constants of ^{226}Ra on the maximum discharge rate of ^{226}Ra for $D=1\times10^{-1}\,m^2/yr$ and $D=1\times10^{3}\,m^2/yr$ under the condition of initial pure ^{234}U source and $T = 3\times10^4$. An estimation of initial activity for ^{234}U produced in a nuclear reactor after being cooled for ten years is taken to be 7.67 (Ci/GWe-yr)(B2). As smaller values of K_3 (5, 50, 500, 5000) than that of K_2 (50,000) are taken in all curves, the curves have their highest point at locations away from the repository. The values of the highest point and the extent of the curves

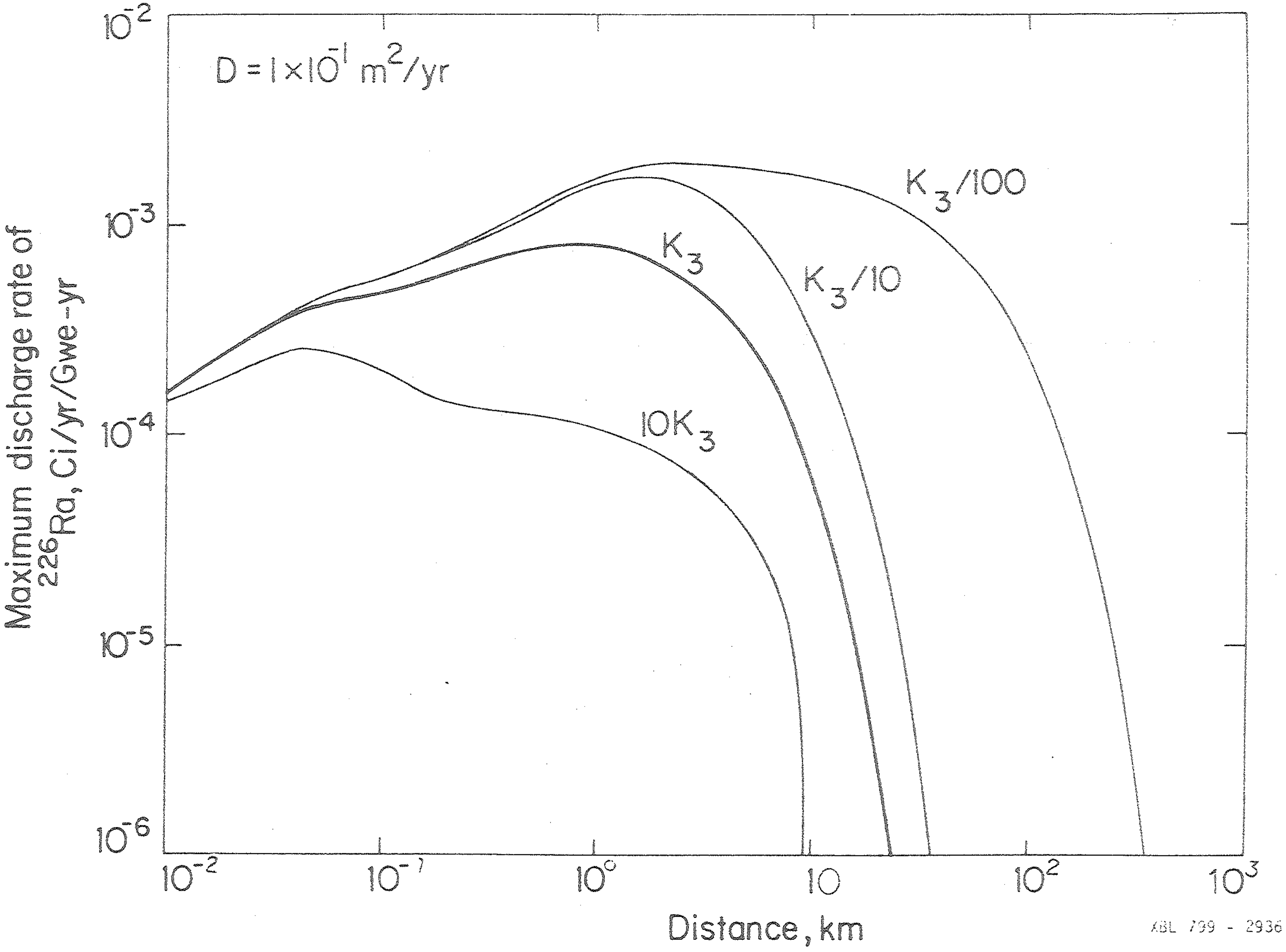

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Figure 5.18 - Effect of sorption equilibrium constant on maximum discharge rate of ^{226}Ra for the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain, with $D=1\times10^{-1} m^2/yr$. (Pure ^{234}U source at t=0. $N_i(z,t)$ = atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. V=100m/yr. T=3×10^4 yr. $K_U=1\times10^4$. $K_{Th} = 5\times10^4$. $K_{Ra}=5\times10^2$)

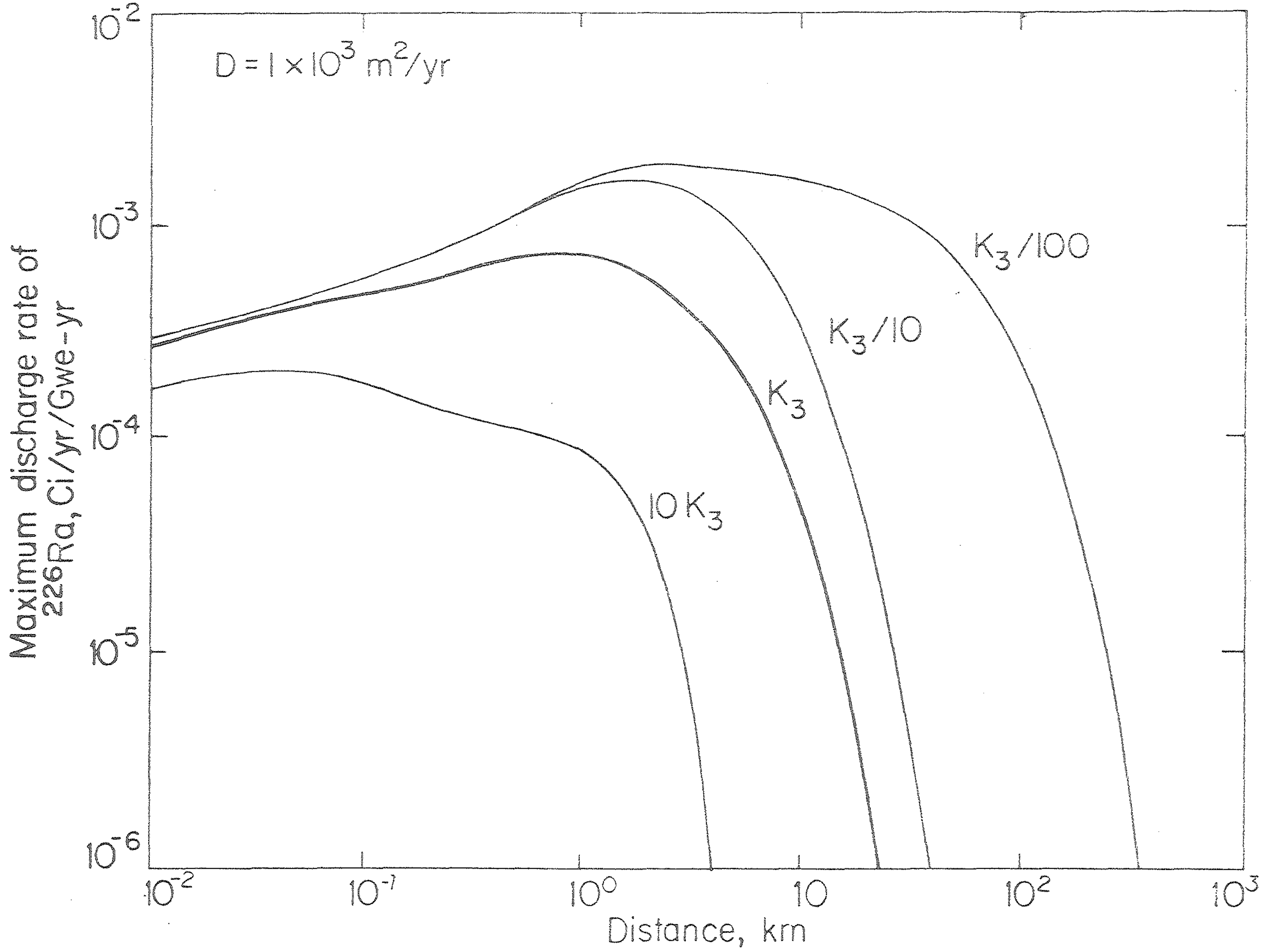

Figure 5.19 - The effect of sorption equilibrium constant on maximum discharge rate of ^{226}Ra for the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain with $D=1x10^3 m^2/yr$. (Pure ^{234}U source at t=0. $N_i(z,t)$=atoms/cm^3. $N_1{}^o$=atoms/cm^3 of ^{234}U at z=0, t=0. V=100m/yr. T=3x10^4/yr. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

increase as K_3 decreases. However, in spite that the concentration of ^{226}Ra in the water phase increases proportionally as K_3 decreases, the highest magnitude and its location are not shifted proportionally. Comparison of Figures 5.18 and 5.19 in regard to different dispersion coefficients show that the $10K_3$ curve in Figure 5.19 is affected by large D in its magnitude and range. But the effect of the dispersion on the curves for K_3, $K_3/10$ and $K_3/100$ is slight except near the repository. This can be explained by the fact that for small K_3, that is large velocity V_3, the behavior of ^{226}Ra is mainly controlled by the convection term of the transport equations (4.2).

5.5.4.3 Effect of leach time on the maximum discharge rate of ^{226}Ra.

Figure 5.20 and 5.21 show the effect of varying leach time on the maximum discharge rate of ^{226}Ra for $D = 1x10^{-1}$ and 1×10^3 m^2/yr at various locations for the condition of initially pure ^{234}U source. Comparison of the $T=3x10^3$ curve and the $T = 3x10^4$ curve in the figures shows that both the value of the highest discharge rate and the discharge rate after the highest point of the $T = 3x10^3$ curve is slightly larger than those of the $T = 3x10^4$ curve. However, the discharge rate of the $T = 3x10^3$ curve near the repository is much less than that

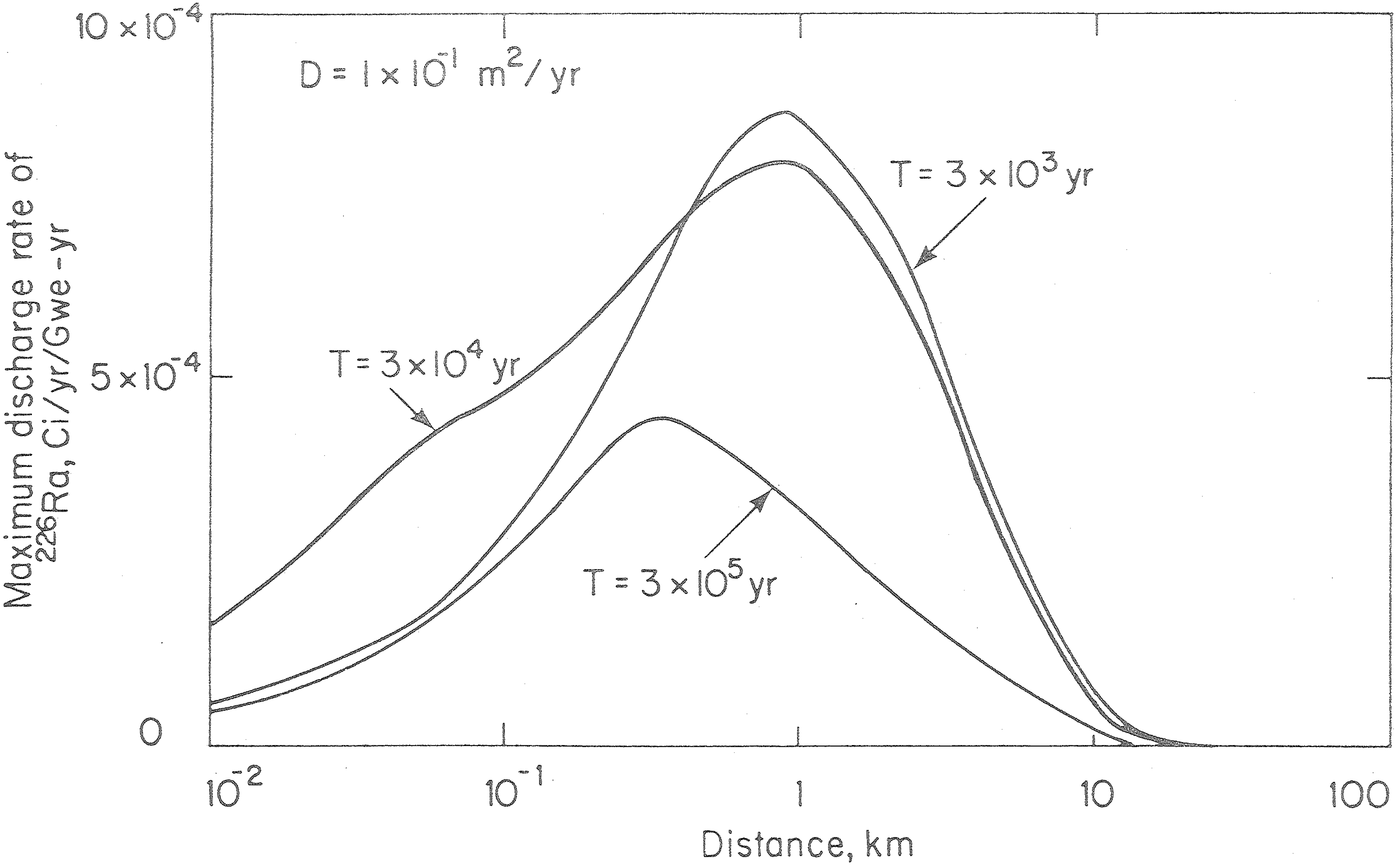

Figure 5.20 - Effect of leach time on the maximum discharge rate of ^{226}Ra for the ^{234}U→^{230}Th→^{226}Ra decay chain, with D=1x10^{-1}m^2/yr. (Pure ^{234}U source at t=0. N_i(z,t)=atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. V=100m/yr. T=3x10^4 yr. K_U = 1x10^4. K_{Th}=5x10^4. K_{Ra}=5x10^2.

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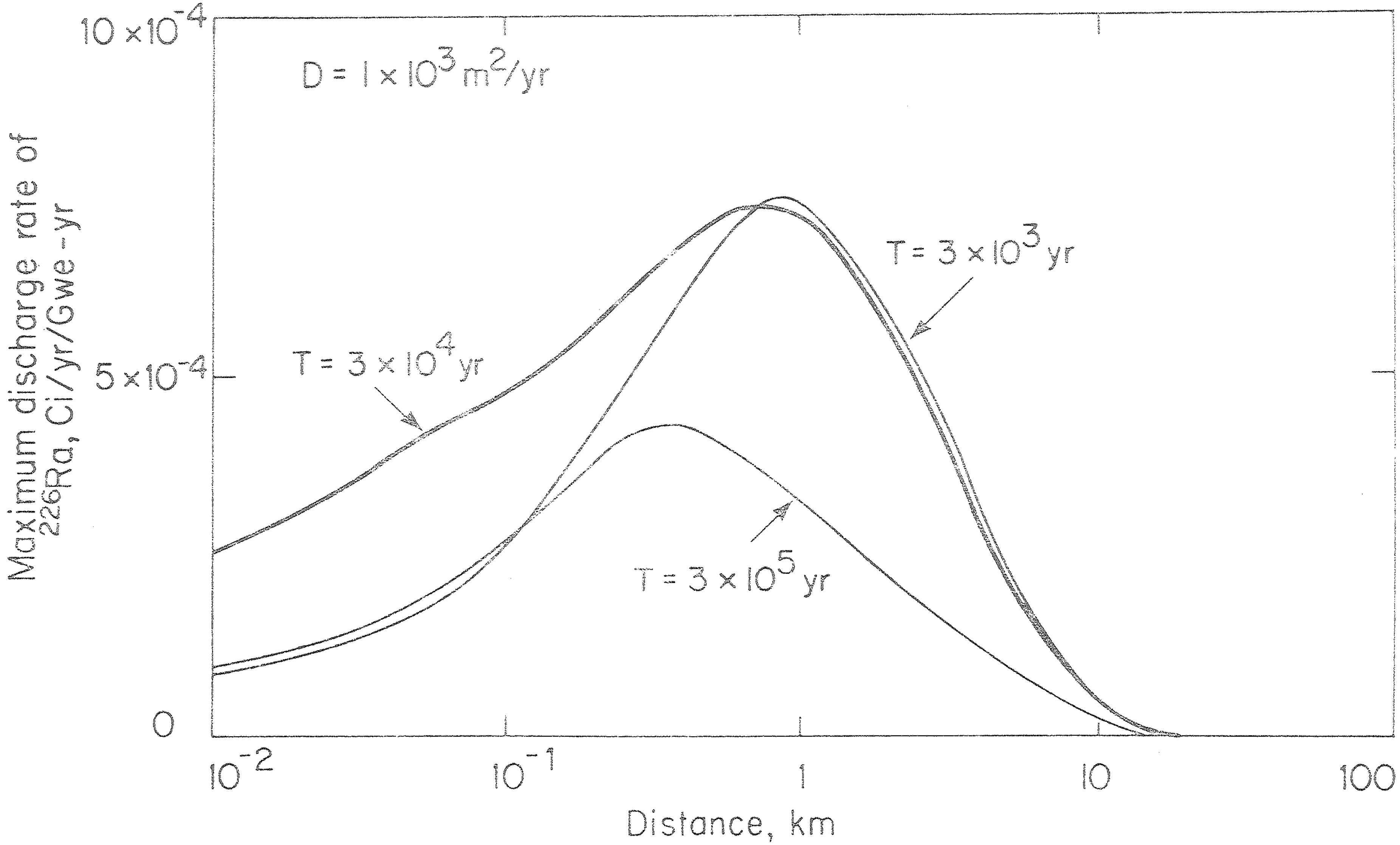

Figure 5.21 - Effect of leach time on the maximum discharge rate of ^{226}Ra for the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain, with $D=1\times10^3 m^2/yr$. (Pure ^{234}U source at t=0. $N_i(z,t)$=atoms/cm^3. N_1^o=atoms/cm^3 of ^{234}U at z=0, t=0. V=100m/yr. $T=3\times10^4$ yr. $K_U=1\times10^4$. $K_{Th}=5\times10^4$. $K_{Ra}=5\times10^2$).

of the T=3x10^4 curve. Due to the small leach time of 3x10^3 and the initial condition in the repository at t = 0, ^{230}Th produced from the decaying ^{234}U near the repository is of lesser amount, so that at a location near the repository one sees a smaller maximum discharge rate of ^{226}Ra. In the case of T = 3x10^5 yr, the concentration of each nuclide which leaches from the repository is rather low, because it is inversely proportional to the leach time, and some of ^{226}Ra produced in the repository has decayed away without coming out of the repository. Hence, the magnitude and range of the influence of ^{226}Ra for T = 3x10^5 yr are less than those for T = 3x10^3 and 3x10^4. It should be noted that the location of the highest point does not shift much in spite of the large variation in the leach time. The effect of dispersion is significant only for the curve of T = 3x10^3 yr, because it has a sharper shape in its distribution. In the band release mode, the leach time may have a close relation with the tightness of the radioactive waste container in the repository. The highest discharge rate of ^{226}Ra depends on the leach time.

Figure 5.22 shows the dependence of the highest discharge rate of ^{226}Ra on leach time. As seen in the figure, the dependence is quite different between the two cases of pure ^{234}U source at t = 0 and initial transient equilibrium sources in the repository. For the former case, very broad

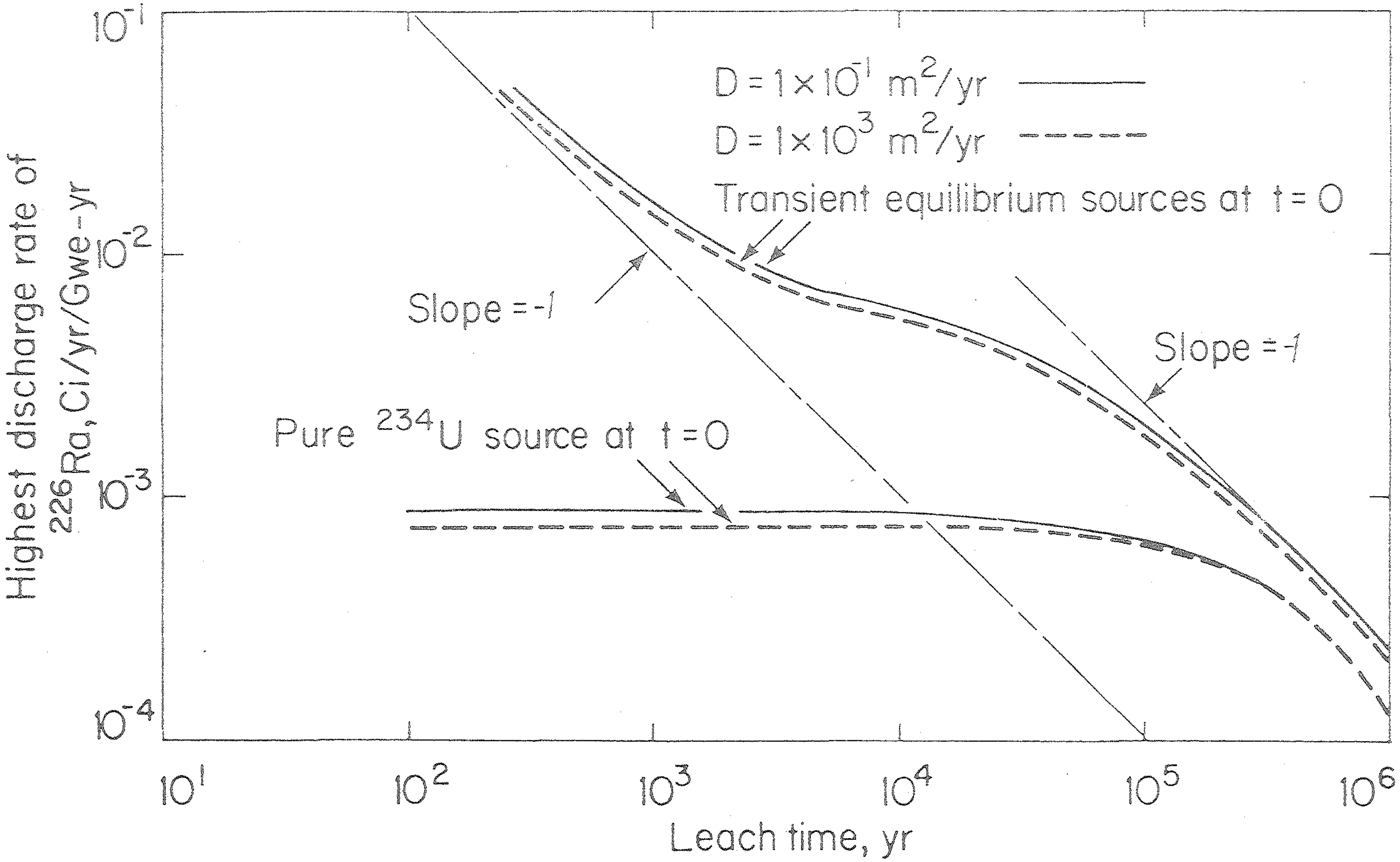

Figure 5.22 - Effect of leach time on the highest concentration of ^{226}Ra for the $^{234}U\rightarrow{}^{230}Th\rightarrow{}^{226}Ra$ decay chain. $N_i(z,t)$=atoms/cm^3. N_1^o atoms/cm^3 of ^{234}U at z=0, t=0. V=100m/yr. T=3x 10^4yr. K_U=1x10^4. K_{Th}=5x10^4. K_{R_a} =5x10^4).

plateau is seen in Figure 5.22. It means that the highest discharge rate of ^{226}Ra has little dependence on the container tightness when only pure ^{234}U source is present at t = 0. Contrarily for the latter case of initial transient equilibrium sources, the highest discharge rate depends strongly on the leach time except around the region of $T = 10^4$ yr. Both of the right hand side and left hand side of the curve in the Figure for the case of initial transient equilibrium source has a slope close to -1. It means that the tightness of the container for radioactive waste is very important for the highest discharge rate of ^{226}Ra in the case of initial transient equilibrium sources in the repository.

5.6 $^{237}Np \rightarrow {}^{233}Pa \rightarrow {}^{233}U \rightarrow {}^{229}Th \rightarrow {}^{225}Ra$ Decay Chain Migration Behavior

5.6.1 Reduction to a three member decay chain.

Half-life, sorption equilibrium constant, migration speed, and migration distance in a half-life time are shown in Table 5.2.

Table 5.2

	^{237}Np	^{233}Pa	^{233}U	^{229}Th	^{225}Ra
Half-life, $T_{1/2}$ (yr)	2.14×10^{6}	7.4×10^{-2}	1.62×10^{5}	7.34×10^{3}	4.1×10^{-2}
K_i (-)	1×10^{2}	1.7×10^{4}	1×10^{4}	5×10^{4}	5×10^{2}
v_i * (m/yr)	1×10^{0}	6×10^{-3}	1×10^{-2}	2×10^{-3}	2×10^{-1}
$v_i \times T_{1/2}$ (m)	2.14×10^{6}	4.44×10^{-4}	1.62×10^{3}	1.47×10^{1}	8.2×10^{-3}

*Assuming V = 100 m/yr

As seen in the above Table, ^{233}Pa and ^{225}Ra have very short half-lives, and the distance of migration in their half-lives, v_ix (half-life), are very short in comparison with those for other three nuclides. So, we may consider as an approximation that ^{233}Pa and ^{225}Ra are in secular equilibrium with ^{237}Np and ^{229}Th, at every location and at any time.

Then, we can apply the solution for three-member decay chain for this five-member decay chain, as if it were the decay chain of $^{237}\text{Np}\rightarrow{}^{233}\text{U}\rightarrow{}^{229}\text{Th}$. As this approximation is already verified in Section 4.5, we may expect that the results will be close enough to that obtained by the exact solution for five-member decay chain.

This decay chain is also very important in the discussion of waste management because it leads to the predicting of ^{225}Ra. As seen in Table 5.2, ^{237}Np migrates a very long distance in its half-life. It means the possibility of a wide hazardous spread due to Ra-225.

5.6.2 Discharge rate profiles of $^{237}NP \rightarrow {}^{233}U \rightarrow {}^{229}Th$ (^{225}Ra) decay chain

Figure 5.23 shows the space-dependence of discharge rate of each nuclide at $t = 5x10^4$ yr under the conditions of the band release of $T = 3x10^4$ yr and initial pure ^{237}Np source. An estimation of the initial activity of ^{237}Np produced in a nuclear power reactor having cooled for 10 years, is taken to be 14.4 Ci/GWe yr.(B2). ^{233}Pa and ^{225}Ra are in secular equilibrium state with ^{237} Np and ^{229}Th, respectively. The figure shows also the effect of varying the dispersion coefficient on the discharge rate profile of each nuclide. ^{237}Np migrates very fast, in this hypothetical three member decay chain, the migration speeds are in the increasing order of v_1, v_2 and v_3, respectively. Thus, the first two nuclides have the tendency to leave their daughters behind them. The dip of ^{225}Ra between $Z=v_2(t-T)$ in the $D=1x10^{-1}$ curve, is due to the competition between the increase in concentration of ^{225}Ra in the repository with time and the decrease in concentration of ^{225}Ra behind its parent nuclide ^{233}U because of its decay. The effect of the large dispersion is to smooth and spread out the profiles of each nuclide. Figure 5.24 is the plots of the maximum discharge rate of ^{225}Ra versus location under the same conditions as those of Figure 5.23. The maximum discharge rate decreases monotonously with distance from the repository. The small D curve might be discussed in three separate regions along the migration path. In the first region, $Z < \sim 0.2$ km, the maximum discharge rate at any given location happens to be at the time when the last ^{229}Th produced inside the repository passes the location. In the second region, $0.2 \text{ km} < Z < 9 \text{ km}$, the maximum discharge rate of ^{225}Ra occurs around the same time at which the peak of ^{233}U, the parent of ^{225}Ra (^{229}Th), occurs. In the last region, the flat part, $9 \text{ km} < Z$, the time when the maximum discharge rate appears is $Z/V_1 + 1.7 \times 10^5$ yr. It is seen that this flat discharge rate extends very far from the repository because the first nuclide, ^{237}Np, has a large migration velocity and a long half life. In this case, the reconcentration phenomenon of ^{225}Ra does not occur at any location because of the sorption equilibrium constant of ^{229}Th is assumed larger than that of parent nuclides so that the migration speed of ^{229}Th is smaller than that of ^{233}U. One should notice the differences for the occurrence of reconcentration between $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ ($v_{Ra} > v_{U} > v_{Th}$) decay chain and $^{237}Np \rightarrow {}^{233}U \rightarrow {}^{229}Th$ ^{225}Ra ($v_{Np} > v_{U} > v_{Th}$) decay chain.

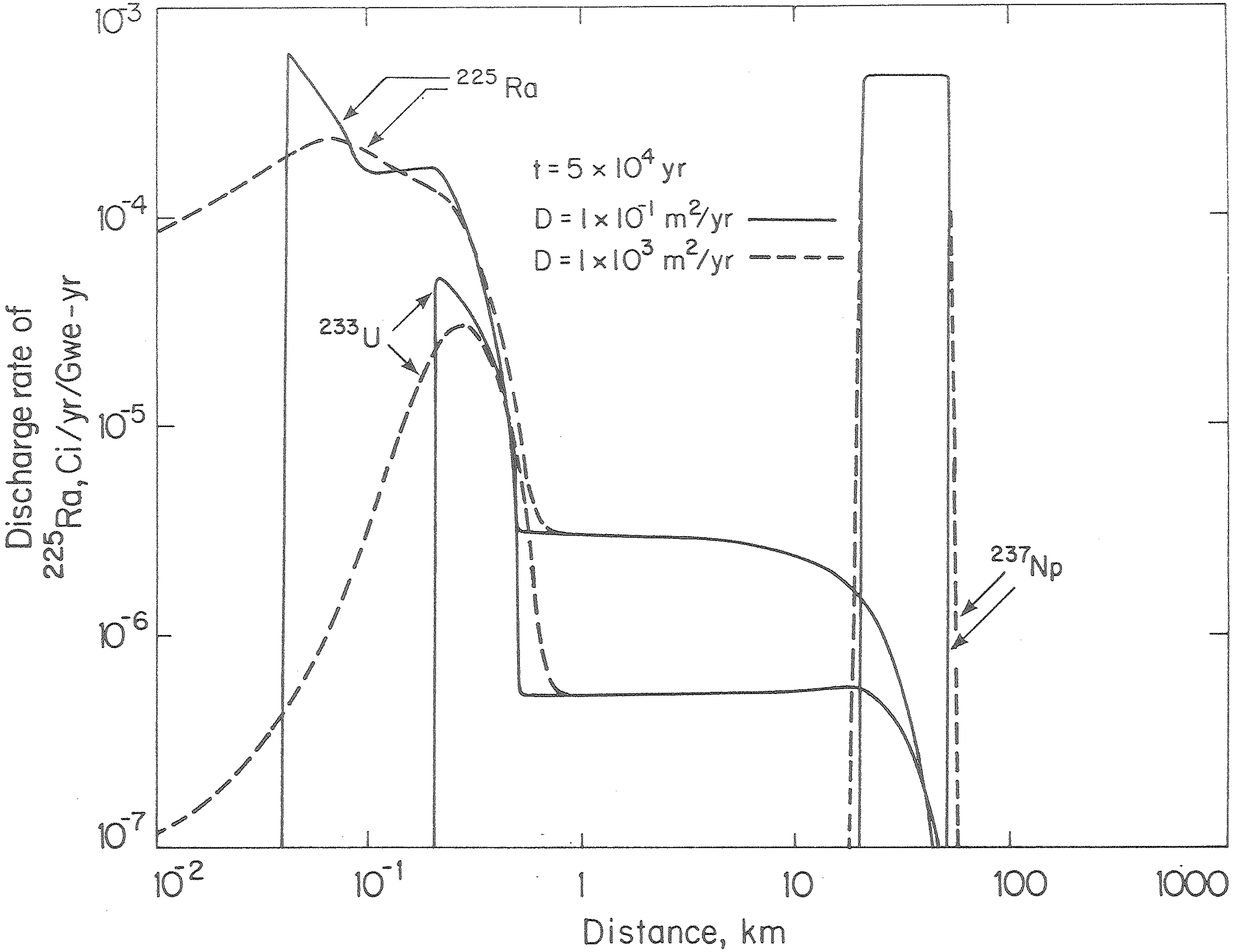

Figure 5.23 - Concentration profiles of ^{225}Ra of ^{237}Np→^{233}Pu→^{233}U→^{229}Th→ ^{225}Ra decay chain at t = 5x10^{4}yr. (Pure ^{237}Np source at t=0. V=100m/yr. T=3x10^4, $K_{Np}=10^2$. $K_{Pu}=1x10^4$. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

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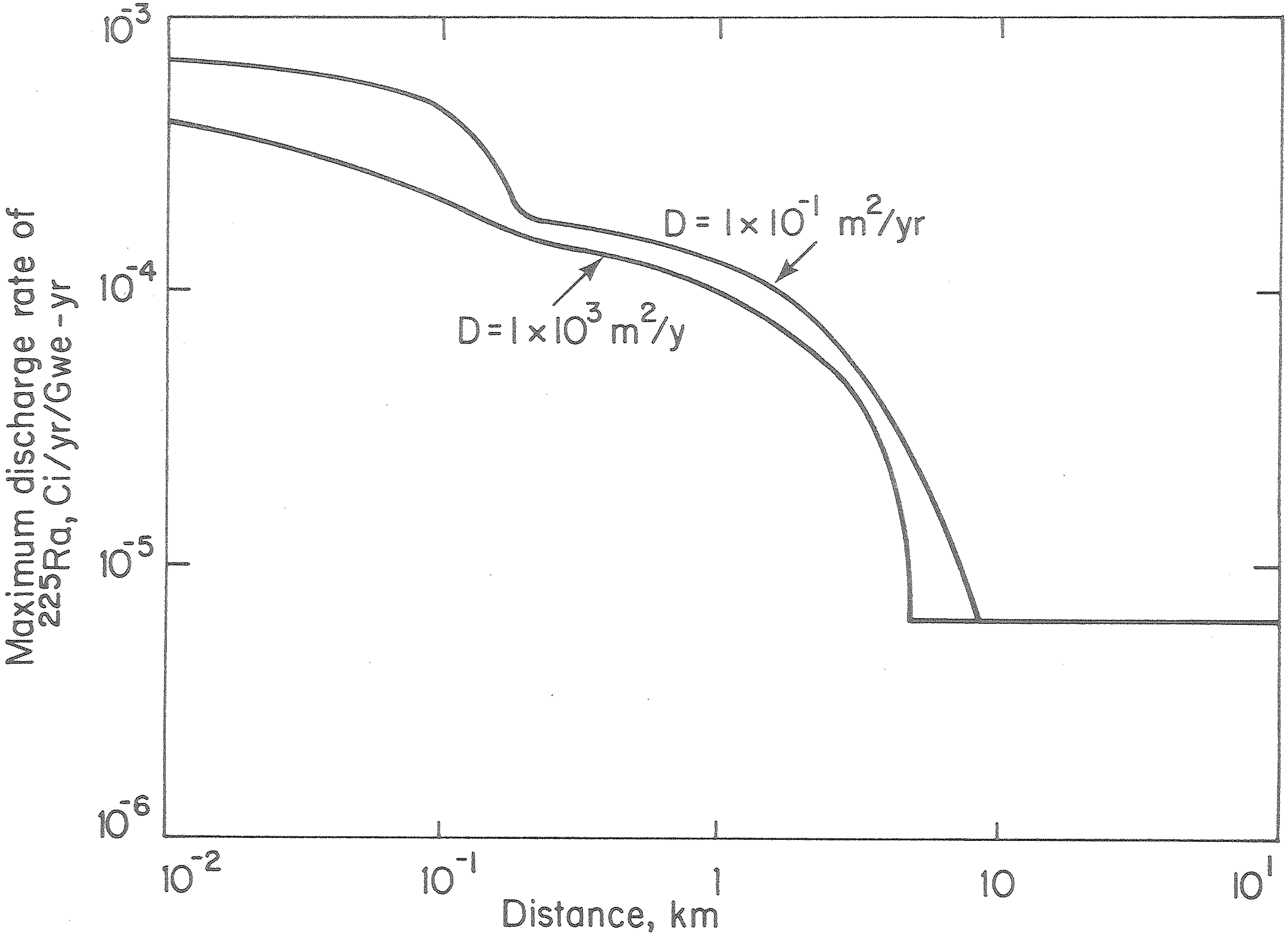

Figure 5.24 - Effect of dispersion coefficient on maximum discharge rate of ^{225}Ra for the ^{237}Np→^{233}Pu→^{233}U→^{229}Th→^{225}Ra decay chain. (Pure ^{237}Np source at t=0. V=100m/yr. T=3x10^4yr. $K_{Np}=10^2$. $K_{Pu}=1x10^4$. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

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6. NUCLIDE MIGRATION THROUGH A GEOLOGICAL MEDIUM OF MULTI-LAYERS

6.1 Introduction

This chapter deals with the problem posed by a multi-layer system. It seems indeed very likely that the nuclides will encounter different media, while being transported in the water. These media may be characterized by different values of:

v = water velocity

K_i= the sorption constant, this reflects the different behavior of the nuclides when adsorbed on chemically different media,

S_f= the cross sectional area of the water phase,

ε = the porosity

D_i= the dispersion coefficient, etc.

Each layer is supposed to have constant properties of v, k_i, ε, etc. The transport equation discussed in the preceding chapters is valid in each layer. The Boundary Condition at the boundary of two layers is given by the continuity of nuclide flux and the nuclide concentration. For the case of no axial dispersion, a recursive solution can be easily found by the use of the general solution for a one medium system (see Chapter 4). Some applications will be given for a two media system:

a) A general formula, for a general release mode at the respitory, and an i members chain,

b) A general formula for a step release and an i members chain,

c) A step release formula for the first, second and third members of a radioactive chain.

The solution for the band release can be found by the superposition principle, which has been derived in Chapter 3.

For the case with axial dispersion, a recursive formula is given for a two media system and a general release mode at the repository.

6.2 Transport Equation and Conditions at the Layer Boundary

In this chapter we consider the case of a multilayer system. Lets characterize each layer by the superscript, ℓ, at the left, e.g.,

${}^{\ell}N_i(z,t)$ = concentration of nuclide i, at z and t in the ℓ^{th} layer,

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${}^{\ell}D_i$ = axial dispersion coefficient of nuclide i, in the ℓ^{th} layer,

${}^{\ell}v$ = water velocity in the ℓ^{th} layer, and so on.

Lets furthermore characterize by ${}^{\ell}Z$ the coordinate between the ℓ^{th} and the $(\ell + 1)^{th}$ layers.

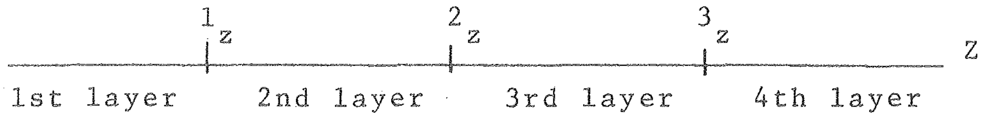

Figure 6.1 Multi-layer Pathway

We assume the following parameters to be constant in each layer:

$${}^{\ell}D_i,\ {}^{\ell}K_i,\ {}^{\ell}v_i,\ {}^{\ell}V,\ {}^{\ell}\varepsilon,\ {}^{\ell}S_f,\ {}^{\ell}S_s$$

Under these assumptions the transport equation, as discussed in Chapter 2 is still valid in each layer. Transport equation in the ℓ^{th} layer

$${}^{\ell}\hat{L}_i\,{}^{\ell}N_i(z,t) = {}^{\ell}K_{i-1}\lambda_{i-1}\,{}^{\ell}N_{i-1}\ ; \quad {}^{\ell-1}z \le z \le {}^{\ell}z \qquad (6.1)$$

where the operator ${}^{\ell}\hat{L}_i$ is defined by

$${}^{\ell}\hat{L}_i \equiv -{}^{\ell}D_i\frac{\partial^2}{\partial z^2} + {}^{\ell}v\frac{\partial}{\partial z} + {}^{\ell}K_i\frac{\partial}{\partial t} + {}^{\ell}K_i\lambda_i \qquad (6.2)$$

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At the interface ${}^{\ell}z$ we must have the continuity of the nuclide flux, that the number of atoms of nuclide i leaving the layer ℓ per second is equal to the number of atoms of isotope i entering the layer $(\ell + 1)$, per second:

$$ {}^{\ell}S_f\left[-{}^{\ell}D_i\frac{\partial\,{}^{\ell}N_i}{\partial z}+{}^{\ell}v\,{}^{\ell}N_i\right]_{z={}^{\ell}z} = {}^{\ell+1}S_f\left[-{}^{\ell+1}D\frac{\partial\,{}^{\ell+1}N_i}{\partial z}+{}^{\ell+1}v\,{}^{\ell+1}N_i\right]_{z={}^{\ell+1}z} \tag{6.3} $$

The term ${}^{\ell}D_i\ \partial\,{}^{\ell}N_i/\partial z$ represents the transport by diffusion and the term ${}^{\ell}V\,{}^{\ell}N_i$ by convection. By introducing

$$ {}^{\ell}d_i \equiv {}^{\ell}D_i/{}^{\ell}v \tag{6.4} $$

and by the use of the conservation of mass of water,

$$ {}^{\ell}S_f\,{}^{\ell}v\,{}^{\ell}\rho = {}^{\ell+1}S_f\,{}^{\ell+1}v\,{}^{\ell+1}\rho \tag{6.5} $$

we can rewrite (6.3) as

$$ {}^{\ell}I\left[-{}^{\ell}d_i\frac{\partial\,{}^{\ell}N_i}{\partial z}+{}^{\ell}N_i\right] = -{}^{\ell+1}d_i\frac{\partial\,{}^{\ell+1}N_i}{\partial z}+{}^{\ell+1}N_i\ ;\ \text{at}\ z={}^{\ell}z \tag{6.6} $$

where

$$ {}^{\ell}I \equiv {}^{\ell+1}\rho/{}^{\ell}\rho \tag{6.7} $$

In most cases, the water density can be considered a constant, therefore the term ${}^{\ell}I$ can be set equal to unity. Since the transport by diffusion is independent of the transport by convection, (6.6) can be split into two equations, and we get (${}^{\ell}I$ is now considered to be equal to unity):

$$ {}^{\ell}N_i = {}^{\ell+1}N_i \quad ; \quad \text{at} \quad z = {}^{\ell}z \tag{6.8}$$

and

$$ {}^{\ell}d_i \frac{\partial\, {}^{\ell}N_i}{\partial z} = {}^{\ell+1}d_i \frac{\partial\, {}^{\ell+1}N_i}{\partial z} \quad ; \quad \text{at} \quad z = {}^{\ell}z \tag{6.9}$$

6.3 Analytical Solutions for Non-Dispersion Case

We have the following problem

Solve

$$ {}^{\ell}v \frac{\partial\, {}^{\ell}N_i}{\partial z} + {}^{\ell}K_i \frac{\partial\, {}^{\ell}N_i}{\partial t} + {}^{\ell}K_i \lambda_i {}^{\ell}N_i = {}^{\ell}K_{i-1} \lambda_{i-1} {}^{\ell}N_{i-1} \; ; \; {}^{\ell-1}z \leq z \leq {}^{\ell}z \tag{6.10}$$

with

I.C. $${}^{\ell}N_i(z,0) = 0 \quad ; \quad \text{all } \ell\text{'s} \tag{6.11}$$

B.C. $${}^{1}N_i(0,t) = {}^{1}\phi_i(t) \; ; \; \text{at the repository} \tag{6.12}$$

$$ {}^{\ell-1}I \; {}^{\ell-1}N_i({}^{\ell-1}z, t) = {}^{\ell}N_i({}^{\ell-1}z, t) \tag{6.13}$$

Let's define for each interval ℓ the new variable:

$$ {}^{\ell}\tilde{z} \equiv z - {}^{\ell-1}z \tag{6.14}$$

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In terms of this variable, we have now the problem:

Solve

$$ {}^{\ell}v \frac{\partial\, {}^{\ell}N_i(\tilde{z},t)}{\partial\, {}^{\ell}\tilde{z}} + {}^{\ell}K_i \frac{\partial\, {}^{\ell}N_i}{\partial t} + {}^{\ell}K_i \lambda_i\, {}^{\ell}N_i = {}^{\ell}K_{i-1}\lambda_{i-1}\, {}^{\ell}N_{i-1} \quad ; \quad 0 \leq \tilde{z} \leq {}^{\ell}z - {}^{\ell-1}z \tag{6.15} $$

with

I.C. $${}^{\ell}N_i({}^{\ell}\tilde{z},0) = 0 \quad ; \quad \text{all } \ell\text{'s} \tag{6.16}$$

B.C. $${}^{\ell}N_i(0,t) = {}^{\ell}\phi_i(t) \quad ; \quad \text{all } \ell\text{'s} \tag{6.17}$$ [a1]

where ${}^{1}\phi_i(t)$ is given by the repository release (6.12)

and

$$ {}^{\ell}\phi_i(t) \equiv {}^{\ell-1}I\, {}^{\ell-1}N_i({}^{\ell-1}z,t) \quad ; \quad \ell \geq 2 \tag{6.18} $$

and

$$ {}^{\ell}\phi_i(t) = 0 \quad ; \quad t < 0, \ \text{all } \ell\text{'s} $$

If (6.15), (6.16) and (6.17) are compared with the problem posed in Chapter 4.4 we see that we have the same problem. We can therefore use the general solution (4.117) shown in 4.4 for each interval. For clarity's sake, lets note that solution by ${}^{\ell}N_i(z,t\,;{}^{\ell}\Phi_i(t))$.

$$ {}^{\ell}N_i(z,t\,;{}^{\ell}\Phi_i(t)) \equiv e^{-(\lambda_i/{}^{\ell}v_i)z}\, {}^{\ell}\phi_i(t - z/{}^{\ell}v_i) $$

$$ + \sum_{j=1}^{i-1} {}^{\ell}A_i^{(j)} \sum_{m=j}^{i} \frac{1}{{}^{\ell}B_m^{ij}}\, e^{-(\lambda_m/{}^{\ell}v_m)z} \sum_{\substack{r=j \\ r\neq m}}^{i} {}^{\ell}D_{rm}^{ij}\, {}^{\ell}g_{rm}(t) \circledast {}^{\ell}\phi_j(t) \tag{6.19} $$

[a1] The parameter N_i^o, which appeared explicitly in the previous chapters has now been assimilated with the symbol ${}^{1}\phi(t)$.

where $g \otimes \phi$ represents the convolution product and ${}^{\ell}\Phi_i(t)$ represents the dependence on ${}^{\ell}\phi_i(t)$, ${}^{\ell}\phi_2(t)$, ... ${}^{\ell}\phi_i(t)$.

The other symbols are the ones used in Chapter 4. The superscript, ℓ, means that we must take ${}^{\ell}v_i = \frac{{}^{\ell}V}{{}^{\ell}K_i}$ whenever the value v_i appears. The solution for each interval is then:

$$ {}^{\ell}N_i({}^{\ell}\tilde{z},t) = {}^{\ell}\mathcal{N}_i({}^{\ell}\tilde{z},t;{}^{\ell}\Phi_i(t)) \ ; \quad 0 \le {}^{\ell}\tilde{z} \le {}^{\ell}z - {}^{\ell-1}z \qquad (6.20) $$

the ${}^{\ell}\phi_i$'s being given by (6.18).

Finally, in terms of z we have

$$ {}^{\ell}N_i(z,t) = {}^{\ell}\mathcal{N}_i(z - {}^{\ell-1}z, t; {}^{\ell}\Phi_i(t)) \ ; \quad {}^{\ell-1}z \le z \le {}^{\ell}z \qquad (6.21) $$

where ${}^{1}\phi_i(t)$ is given by the nuclide release mode at the repository and

$$ {}^{\ell}\phi_i(t) \equiv \frac{{}^{\ell}\rho}{{}^{\ell-1}\rho}\,{}^{\ell-1}N_i({}^{\ell-1}z,t) \qquad \text{for} \quad \ell \ge 2 $$

Equation (6.21) is a "doubly recursive" solution, i.e. it is recursive both in space and in the nuclide. One must calculate first ${}^{1}N_j({}^{1}z,t)$, for j = 1,2,3,..i, using equation (6.21), then calculate ${}^{2}N_j({}^{2}z,t)$ for j = 1,2,...i, using the previous results and (6.21), and so on up to ${}^{\ell}N_i(z,t)$.

6.4 Application of Recursive Solution to a Two Media System

6.4.1 General release mode at the repository: ${}^{1}\phi_i(t)$

We have:

$$ {}^{1}N_i(z,t) = {}^{1}\mathcal{N}_i(z,t\,;\,{}^{1}\Phi_i(t)) \tag{6.22} $$

and (6.21) yields:

$$ \begin{aligned} {}^{2}N_i(z,t) = {} & e^{-\lambda_i(z-z_1)/{}^{2}v_i} \cdot \frac{{}^{2}\rho}{{}^{1}\rho} \Bigg\{ e^{-\lambda_i z_1/{}^{1}v_i}\, \phi_i\left(t - \frac{z-z_1}{2v_i} - \frac{z_1}{{}^{1}v_i}\right) + \\ & + \sum_{j=1}^{i-1} {}^{1}A_i^{(j)} \sum_{m=j}^{i} \frac{1}{{}^{1}B_m^{ij}}\, e^{-\lambda_m z_1/{}^{1}v_m} \cdot \sum_{\substack{r=j \\ (r\neq m)}}^{i} {}^{1}D_{r,m}^{i,j} \cdot \\ & \cdot \left[{}^{1}g_{r,m}(t') \otimes {}^{1}\phi_j(t') \right]_{t'=t-(z-z_1)/{}^{2}v_i} \Bigg\} + \\ & + \sum_{j=1}^{i-1} {}^{2}A_i^{(j)} \sum_{m=j}^{i} \frac{1}{{}^{2}B_m^{ij}}\, e^{-\lambda_m (z-z_1)/{}^{2}v_m} \cdot \sum_{\substack{r=j \\ r\neq m}}^{i} {}^{2}D_{r,m}^{ij}\, {}^{2}g_{r,m}(t) \otimes \\ & \otimes \Bigg\{ \frac{{}^{2}\rho}{{}^{1}\rho}\, e^{-\lambda_j z_1/{}^{1}v_j}\, {}^{1}\phi_j(t - z_1/{}^{1}v_j) + \frac{{}^{2}\rho}{{}^{1}\rho} \sum_{\alpha=1}^{j-1} {}^{1}A_j^{(\alpha)} \sum_{\beta=\alpha}^{j} \frac{1}{{}^{1}B_\beta^{j,\alpha}} \cdot \\ & \cdot e^{-\lambda_\beta z_1/{}^{1}v_\beta} \cdot \sum_{\substack{\omega=\alpha \\ \omega\neq\beta}}^{j} {}^{1}D_{\omega,\beta}^{j,\alpha}\, {}^{1}g_{\omega,\beta}(t) \otimes {}^{1}\phi_\alpha(t) \Bigg\} \end{aligned} \tag{6.23} $$

$\{ {}^{1}g_{r,m}(t') \otimes {}^{i}\phi_{j}(t') \,]_{t'=t-(z-z_1)/2v_i}$ means that one must first perform the convolution product using t', then set $t'=t-(z-z_1)/{}^{2}v_i$.

6.4.2 Step release

${}^{1}N_i^{S}(z,t)$ is given by (5.13).

Using ${}^{1}\phi_i(t) = B_i(t)$ in eq. (6.23) we get

$$
\begin{aligned}
{}^{2}N_i^{s}(z,t) = {} & \sum_{q=1}^{i} B_{iq} \, {}^{2}G_i^{i}(z,t) \, {}^{2}F_i^{i}(z,t,\lambda_q) + \\
& + \sum_{j=1}^{i-1} \sum_{m=j}^{i} \sum_{\substack{r=j \\ r \neq m}}^{i} \sum_{q=1}^{j} B_{jq} \, {}^{1}H_{r,m}^{i,j,q} \, {}^{2}G_i^{m}(z,t) \cdot \\
& \qquad \cdot \left[{}^{2}F_i^{m}(z,t,\lambda_q) - {}^{2}F_i^{m}(z,t,{}^{1}\Delta_{r,m}) \right] + \\
& + \sum_{j=1}^{i-1} \sum_{m=j}^{i} \sum_{\substack{r=j \\ r \neq m}}^{i} \sum_{q=1}^{j} B_{jq} \, {}^{2}H_{r,m}^{i,j,q} \, {}^{2}G_m^{j}(z,t) \left[{}^{2}F_m^{j}(z,t,\lambda_q) - {}^{2}F_m^{j}(z,t,{}^{2}\Delta_{r,m}) \right] + \\
& + \sum_{j=1}^{i-1} \sum_{m=j}^{i} \sum_{\substack{r=j \\ r \neq m}}^{i} \sum_{\alpha=1}^{j-1} \sum_{\beta=\alpha}^{j} \sum_{\substack{\omega=\alpha \\ \omega \neq \beta}}^{j} \sum_{q=1}^{\alpha} B_{\alpha q} \, {}^{2}H_{r,m}^{i,j,q} \; {}^{1}H_{\omega,\beta}^{j,\alpha,q} \, {}^{2}G_m^{\beta}(z,t) \cdot \\
& \cdot \left\{ \left[{}^{2}F_m^{\beta}(z,t,\lambda_q) - {}^{2}F_m^{\beta}(z,t,{}^{2}\Delta_{r,m}) \right] + \left(\frac{{}^{2}\Delta_{r,m} - \lambda_q}{{}^{2}\Delta_{r,m} - {}^{1}\Delta_{\omega\beta}} \right) \cdot \right. \\
& \quad \left. \cdot \left[{}^{2}F_m^{\beta}(z,t,{}^{2}\Delta_{rm}) - {}^{2}F_m^{\beta}(z,t,{}^{1}\Delta_{\omega\beta}) \right] \right\} \qquad (6.24)
\end{aligned}
$$

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where

$$ {}^2F_p^j(z,t,\lambda_q) \equiv e^{-\lambda_q}\,\theta_{jp}(z,t) \tag{6.25}$$

$$ {}^2F_p^j(z,t,{}^\ell\Delta_{m,r}) = e^{-{}^\ell\Delta_{r,m}}\,\theta_{jp}(z,t) \tag{6.26}$$

$$ {}^2G_p^j(z,t) \equiv \frac{{}^2\rho}{{}^1\rho}\, e^{-\lambda_p(z-z_1)/{}^2v_p} \cdot e^{-\lambda_j z_1/{}^1v_j}\; h(\theta_{jp}(z,t)) \tag{6.27}$$

$$ {}^\ell H_{rm}^{ijq} \equiv {}^\ell A_i^{(j)} \frac{1}{{}^\ell B_m^{ij}}\; {}^\ell D_{r,m}^{ij} \frac{1}{{}^\ell\Delta_{rm} - \lambda_q} \tag{6.28}$$

$$ \theta_{jp}(z,t) \equiv t - \frac{z-z_1}{{}^2v_p} - \frac{z_1}{{}^1v_j} \tag{6.29}$$

B_{iq} are the coefficients in the Bateman equation: See Chapter 3.

6.4.2 Step solution in a two media system for a three members chain, no dispersion

By substituting i = 1,2,3 into (6.13) and (6.24) one gets:

First member:

$$ {}^1J_1^S(z,t) = {}^1G_1^1(z,t)\,{}^1F_1^1(z,t,\lambda_1)\,B_{11}\;;\quad 0 \le z \le z_1 \tag{6.30}$$

$$ {}^{2}N_{1}^{s}(z,t) = {}^{2}G_{1}^{1}(z,t)\,{}^{2}F_{1}^{1}(z,t,\lambda_{1})\,B_{11} \quad ; \quad z \geq z_{1} \qquad (6.31) $$

Second member:

$$ {}^{1}N_{2}^{s}(z,t) = {}^{1}\mathcal{A}_{2}^{s}(z,t) \quad ; \quad 0 \leq z \leq z_{1} \qquad (6.32) $$

$$ {}^{2}N_{2}^{s}(z,t) = {}^{2}\mathcal{A}_{2}^{s}(z,t) + {}^{2}\mathcal{B}_{2}^{s}(z,t) \quad ; \quad z \geq z_{1} \qquad (6.33) $$

Third member:

$$ {}^{1}N_{3}^{s}(z,t) = {}^{1}\mathcal{A}_{3}^{s}(z,t) \quad ; \quad 0 \leq z \leq z_{1} \qquad (6.34) $$

$$ {}^{2}N_{3}^{s}(z,t) = {}^{2}\mathcal{A}_{3}^{s}(z,t) + {}^{2}\mathcal{B}_{3}^{s}(z,t) + {}^{2}\mathcal{C}_{3}^{s}(z,t) ; \quad z \geq z_{1} \qquad (6.35) $$

where:

$$ \begin{aligned} {}^{\ell}\mathcal{A}_{2}^{s}(z,t) \equiv {}& {}^{\ell}G_{2}^{2}(z,t)\left[B_{21}\,{}^{\ell}F_{2}^{2}(z,t,\lambda_{1}) + B_{22}\,{}^{\ell}F_{2}^{2}(z,t,\lambda_{2})\right] + \\ & + {}^{1}H_{7}\,B_{11}\,{}^{\ell}G_{2}^{1}(z,t)\left[{}^{\ell}F_{2}^{1}(z,t,{}^{1}\Delta_{21}) - {}^{\ell}F_{2}^{1}(z,t,\lambda_{1})\right] + \\ & + {}^{1}H_{7}\,B_{11}\,{}^{\ell}G_{2}^{2}(z,t)\left[{}^{\ell}F_{2}^{2}(z,t,\lambda_{1}) - {}^{\ell}F_{2}^{2}(z,t,{}^{1}\Delta_{21})\right] \end{aligned} \qquad (6.36) $$

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$$
\begin{aligned}
{}^{\ell}A_3^s(z,t) = {}^{\ell}G_3^3(z,t)\Big[&B_{31}\,{}^{\ell}F_3^3(z,t,\lambda_1) + B_{32}\,{}^{\ell}F_3^3(z,t,\lambda_2) + B_{33}\,{}^{\ell}F_3^3(z,t,\lambda_3)\Big] \\
&+ {}^{1}H_2 B_{11}\,{}^{\ell}G_3^1(z,t)\Big[{}^{\ell}F_3^1(z,t,{}^{1}\Delta_{21}) - {}^{\ell}F_3^1(z,t,\lambda_1)\Big] + \\
&+ {}^{1}H_3 B_{11}\,{}^{\ell}G_3^1(z,t)\Big[{}^{\ell}F_3^1(z,t,{}^{1}\Delta_{31}) - {}^{\ell}F_3^1(z,t,\lambda_1)\Big] + \\
&+ {}^{1}H_2 B_{11}\,{}^{\ell}G_3^2(z,t)\Big[{}^{\ell}F_3^2(z,t,\lambda_1) - {}^{\ell}F_3^2(z,t,{}^{1}\Delta_{21})\Big] + \\
&+ {}^{1}H_4 B_{11}\,{}^{\ell}G_3^2(z,t)\Big[{}^{\ell}F_3^2(z,t,{}^{1}\Delta_{32}) - {}^{\ell}F_3^2(z,t,\lambda_1)\Big] + \\
&+ {}^{1}H_3 B_{11}\,{}^{\ell}G_3^3(z,t)\Big[{}^{\ell}F_3^3(z,t,\lambda_1) - {}^{\ell}F_3^3(z,t,{}^{1}\Delta_{31})\Big] + \\
&+ {}^{1}H_4 B_{11}\,{}^{\ell}G_3^3(z,t)\Big[{}^{\ell}F_3^3(z,t,\lambda_1) - {}^{\ell}F_3^3(z,t,{}^{1}\Delta_{23})\Big] + \\
&+ {}^{1}H_5 B_{21}\,{}^{\ell}G_3^2(z,t)\Big[{}^{\ell}F_3^2(z,t,{}^{1}\Delta_{32}) - {}^{\ell}F_3^2(z,t,\lambda_1)\Big] + \\
&+ {}^{1}H_6 B_{22}\,{}^{\ell}G_3^2(z,t)\Big[{}^{\ell}F_3^2(z,t,{}^{1}\Delta_{32}) - {}^{\ell}F_3^2(z,t,\lambda_2)\Big] + \\
&+ {}^{1}H_5 B_{21}\,{}^{\ell}G_3^3(z,t)\Big[{}^{\ell}F_3^3(z,t,\lambda_1) - {}^{\ell}F_3^3(z,t,{}^{1}\Delta_{32})\Big] + \\
&+ {}^{1}H_6 B_{22}\,{}^{\ell}G_3^3(z,t)\Big[{}^{\ell}F_3^3(z,t,\lambda_2) - {}^{\ell}F_3^3(z,t,{}^{1}\Delta_{32})\Big]
\end{aligned}
\tag{6.37}
$$

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$$
\begin{aligned}
{}^2\mathcal{B}_2^s(z,t) \equiv {} & {}^2H_7 B_{11}\,{}^2G_1^1(z,t)\left[{}^2F_1^1(z,t,{}^2\Delta_{21}) - {}^2F_1^1(z,t,\lambda_3)\right] + \\
& + {}^2H_7 B_{11}\,{}^2G_2^1(z,t)\left[{}^2F_2^1(z,t,\lambda_1) - {}^2F_2^1(z,t,{}^2\Delta_{21})\right]
\end{aligned}
$$

(6.38)

$$
\begin{aligned}
{}^2\mathcal{B}_3^s(z,t) = {} & {}^2H_2 B_{11}\,{}^2G_1^1(z,t)\left[{}^2F_1^1(z,t,{}^2\Delta_{21}) - {}^2F_1^1(z,t,\lambda_1)\right] + \\
& + {}^2H_3 B_{11}\,{}^2G_1^1(z,t)\left[{}^2F_1^1(z,t,{}^2\Delta_{31}) - {}^2F_1^1(z,t,\lambda_1)\right] + \\
& + {}^2H_2 B_{11}\,{}^2G_2^1(z,t)\left[{}^2F_2^1(z,t,\lambda_1) - {}^2F_2^1(z,t,{}^2\Delta_{21})\right] + \\
& + {}^2H_4 B_{11}\,{}^2G_2^1(z,t)\left[{}^2F_2^1(z,t,{}^2\Delta_{32}) - {}^2F_2^1(z,t,\lambda_1)\right] + \\
& + {}^2H_3 B_{11}\,{}^2G_3^1(z,t)\left[{}^2F_3^1(z,t,\lambda_1) - {}^2F_3^1(z,t,{}^2\Delta_{31})\right] + \\
& + {}^2H_4 B_{11}\,{}^2G_3^1(z,t)\left[{}^2F_3^1(z,t,\lambda_1) - {}^2F_3^1(z,t,{}^2\Delta_{32})\right] + \\
& + {}^2H_5 B_{21}\,{}^2G_2^2(z,t)\left[{}^2F_2^2(z,t,{}^2\Delta_{32}) - {}^2F_2^2(z,t,\lambda_1)\right] + \\
& + {}^2H_6 B_{22}\,{}^2G_2^2(z,t)\left[{}^2F_2^2(z,t,{}^2\Delta_{32}) - {}^2F_2^2(z,t,\lambda_2)\right] +
\end{aligned}
$$

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$$+ {}^2H_5 B_{21} {}^2G_3^2(z,t)\left[{}^2F_3^2(z,t,\lambda_1) - {}^2F_3^2(z,t,{}^2\Delta_{32})\right] +$$

$$+ {}^2H_6 B_{22} {}^2G_3^2(z,t)\left[{}^2F_3^2(z,t,\lambda_2) - {}^2F_3^2(z,t,{}^2\Delta_{32})\right]$$

(6.39)

$${}^2\mathcal{C}_3^S(z,t) = {}^2H_5 {}^1H_7 B_{11} {}^2G_2^1(z,t)\left[M_{32}^1(z,t,\lambda_1) + M_{32}^1(z,t,{}^1\Delta_{21})\right] +$$

$$+ {}^2H_5 {}^1H_7 B_{11} {}^2G_3^2(z,t)\left[M_{23}^2(z,t,\lambda_1) + M_{23}^2(z,t,{}^1\Delta_{21})\right] -$$

$$- {}^2H_5 {}^1H_7 B_{11} {}^2G_2^2(z,t)\left[M_{32}^2(z,t,{}^1\Delta_{21}) + M_{12}^2(z,t,\lambda_3)\right] -$$

$$- {}^2H_5 {}^1H_7 B_{11} {}^2G_3^1(z,t)\left[M_{23}^1(z,t,\lambda_1) + M_{23}^1(z,t,{}^1\Delta_{21})\right]$$

(6.40)

and where:

$$\Delta_{r,m} = \frac{\lambda_m/v_m - \lambda_r/v_r}{1/v_m - 1/v_r} \tag{6.41}$$

$${}^1F_1^j(z,t,\lambda_k) = {}^1F_2^j(z,t,\lambda_k) = {}^1F_3^j(z,t,\lambda_k) = e^{-\lambda_k\left(\frac{z}{{}^1v_j} - t\right)} \tag{6.42}$$

$$ {}^1F_1^j(z,t,{}^1\Delta_{rm}) = {}^1F_2^j(z,t,{}^1\Delta_{rm}) = {}^1F_3^j(z,t,{}^1\Delta_{rm}) = e^{{}^1\Delta_{rm}\left(\frac{z}{{}^1v_j}-t\right)} \qquad (6.43) $$

$$ {}^2F_p^j(z,t,\lambda_K) = e^{-\lambda_K \Theta_{jp}(z,t)} \qquad (6.44) $$

$$ {}^2F_p^j(z,t,{}^l\Delta_{rm}) = e^{-{}^l\Delta_{rm}\,\Theta_{jp}(z,t)} \qquad (6.45) $$

$$ {}^1G_1^j(z,t) = {}^1G_2^j(z,t) = {}^1G_3^j(z,t) = e^{-z\lambda_j/{}^1v_j}\, h\left(t-\frac{z}{{}^1v_j}\right) \qquad (6.46) $$

$$ {}^2G_p^j(z,t) = e^{-\lambda_p(z-z_1)/{}^2v_p} \cdot e^{-\lambda_j z_1/{}^1v_j}\, h\left(\Theta_{jp}(z,t)\right)\frac{{}^2\rho}{{}^1\rho} \qquad (6.47) $$

h (t) = step function

$$ {}^lH_1 = \lambda_1\lambda_2\,{}^lv_1\,{}^lv_3 \Big/ \left[({}^lv_1-{}^lv_2)({}^lv_1-{}^lv_3)({}^l\Delta_{21}-{}^l\Delta_{31})\right] \qquad (6.48) $$

$$ {}^lH_2 = {}^lH_1 / ({}^l\Delta_{21}-\lambda_1) \qquad (6.49) $$

$$ {}^lH_3 = {}^lH_1 / (\lambda_1-{}^l\Delta_{31}) \qquad (6.50) $$

$$ {}^{\ell}H_4 = {}^{\ell}H_1 / ({}^{\ell}\Delta_{32} - \lambda_1) \qquad (6.51) $$

$$ {}^{\ell}H_5 = \lambda_2 \, {}^{\ell}\upsilon_3 \Big/ \Big[({}^{\ell}\upsilon_2 - {}^{\ell}\upsilon_3)(\lambda_1 - {}^{\ell}\Delta_{32}) \Big] \qquad (6.52) $$

$$ {}^{\ell}H_6 = \lambda_2 \, {}^{\ell}\upsilon_3 \Big/ \Big[({}^{\ell}\upsilon_2 - {}^{\ell}\upsilon_3)(\lambda_2 - {}^{\ell}\Delta_{32}) \Big] \qquad (6.53) $$

$$ {}^{\ell}H_7 = \lambda_1 \, {}^{\ell}\upsilon_2 \Big/ \Big[({}^{\ell}\upsilon_1 - {}^{\ell}\upsilon_2)(\lambda_1 - {}^{\ell}\Delta_{21}) \Big] \qquad (6.54) $$

$$ M^{\beta}_{rm}(z, t, \lambda_1) = {}^{2}F^{\beta}_{m}(z, t, \lambda_1) - {}^{2}F^{\beta}_{m}(z, t, {}^{2}\Delta_{rm}) \qquad (6.55) $$

$$ M^{\beta}_{rm}(z, t, {}^{1}\Delta_{21}) = \frac{({}^{2}\Delta_{rm} - \lambda_1)}{({}^{2}\Delta_{rm} - {}^{1}\Delta_{21})} \Big[{}^{2}F^{\beta}_{m}(z, t, {}^{2}\Delta_{rm}) - {}^{2}F^{\beta}_{m}(z, t, {}^{1}\Delta_{21}) \Big] \qquad (6.56) $$

$$ \Theta_{jp}(z, t) = t - \frac{z - z_1}{{}^{2}\upsilon_p} - \frac{z_1}{{}^{1}\upsilon_j} \qquad (6.57) $$

6.5 Recursive Formula for a Two Media System with Dispersion

For this case we use the transport equation with the source term. We have the following situation as shown in Figure 6.2.

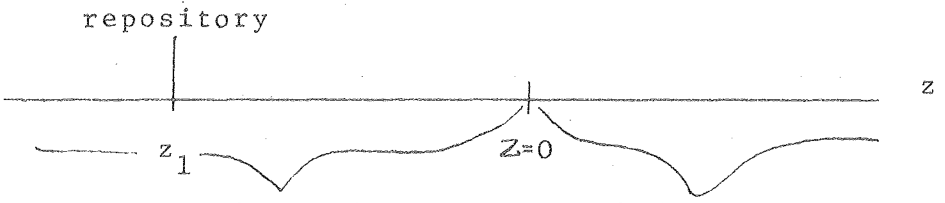

Figure 6.2 Two media system with disperion

The interface between the two media is set at z = 0. The repository is set in the first medium, at z_1.
The boundary condition at the media boundary has been discussed in Chapter 6.2. We have therefore the problem:
Solve

$$ {}^1\hat{L}_i\,{}^1N_i(z,t) = \lambda_{i-1}\,{}^1K_{i-1}\,{}^1N_{i-1} + {}^1\phi(t)\,\delta(z-z_1) \;;\quad z \le 0 \tag{6.58} $$

$$ {}^2\hat{L}_i\,{}^2N_i(z,t) = \lambda_{i-1}\,{}^2K_{i-1}\,{}^2N_{i-1} \;;\quad z \ge 0 \tag{6.59} $$

with

I.C.

$${}^1N_i(z,0) = {}^2N_i(z,0) = 0 \tag{6.60}$$

B.C.

$${}^1N_i(-\infty,t) = \text{bound} \tag{6.61}$$

$${}^2N_i(+\infty,t) = \text{bound} \tag{6.62}$$

$${}^1N_i(0,t) = {}^2N_i(0,t) \tag{6.63}$$

$$\frac{{}^1d_i}{{}^2d_i}\,\frac{\partial\,{}^1N_i}{\partial z} = \frac{\partial\,{}^2N_i}{\partial z} \quad ; \quad \text{at} \quad z = 0 \tag{6.64}$$

This problem is solved by the Green's method. We seek first the solutions for a unit pulse (in time and space) in the first medium, then in the second. We solve

$$\left\{ \begin{array}{ll} {}^1\hat{L}_i\,{}^Ln_i(z,t;\xi,\tau) = \delta(z-\xi)\,\delta(t-\tau) & ;\ \xi<0,\ z\leq 0,\ 0\leq\tau\leq t \\ {}^2\hat{L}_i\,{}^Lm_i(z,t;\xi,\tau) = 0 & ;\ \xi<0,\ z\geq 0,\ 0\leq\tau\leq t \end{array} \right. \tag{6.65}$$

With the initial conditions (6.60) and the boundary conditions (6.61) to (6.64).

Then we solve

$$\begin{cases} {}^1\hat{L}_i\,{}^R n_i(z,t\,;\,\xi,\tau) = 0 & ;\ \xi > 0,\ z \leq 0,\ 0 \leq \tau \leq t \\ {}^2\hat{L}_i\,{}^R m_i(z,t\,;\,\xi,\tau) = \delta(z-\xi)\,\delta(t-\tau) & ;\ \xi > 0,\ z \geq 0,\ 0 \leq \tau \leq t \end{cases} \qquad (6.66)$$

with ditto initial and boundary conditions.

${}^L N_i\ (z,t,\xi,\tau)$ represents the response in the first medium to a pulse happening in the first (Left) medium at $z=\xi$ and $t=\tau$.

${}^L M_i\ (z,t,\xi,\tau)$ represents the response in the second medium to a pulse happening in the first medium at $z = \xi$ and $t = \tau$.

On the same way, ${}^R N_i$ and ${}^R M_i$ represent respectively the responses in the first or second medium to a pulse happening in the second (Right) medium. (6.65) and (6.66) are solved by taking the Laplace transform with respect to t: the equations are now ordinary differential equations of second order inone variable, z. These can be solved, and yield the Laplace transformed solutions ${}^L\tilde{N}_i(z,s)$, ${}^L\tilde{M}_i(z,s)$, ${}^R\tilde{N}_i(z,s)$, ${}^R\tilde{M}_i(z,s)$. By the use of Laplace transform tables, one finds then ${}^Ln_i(z,t)$, ${}^LM_i(z,t)$, etc.

These functions: ${}^LN_i$, ${}^LM_i$, ${}^RN_i$, ${}^RM_i$, play then the role of a Green kernel. The final result is then:

$$ {}^1N_i(z,t) = {}^Rn_i(z,t) + {}^Ln_i(z,t) \quad ; \quad z \leq 0 \tag{6.67} $$

$$ {}^2N_i(z,t) = {}^Rm_i(z,t) + {}^Lm_i(z,t) \quad ; \quad z \geq 0 \tag{6.68} $$

where

$$ {}^Rn_i(z,t) \equiv \int_0^t \left[\int_0^\infty {}^Rn_i(z,t;\xi,\tau)\, {}^2f_i(\xi,\tau)\, d\xi \right] d\tau \quad ; \quad z < 0 \tag{6.69} $$

$$ {}^Ln_i(z,t) \equiv \int_0^t \left[\int_{-\infty}^0 {}^Ln_i(z,t;\xi,\tau)\, {}^1f_i(\xi,\tau)\, d\xi \right] d\tau \quad ; \quad z < 0 \tag{6.70} $$

$$ {}^Rm_i(z,t) \equiv \int_0^t \left[\int_0^\infty {}^Rm_i(z,t;\xi,\tau)\, {}^2f_i(\xi,\tau)\, d\xi \right] d\tau \quad ; \quad z > 0 \tag{6.71} $$

$$ {}^Lm_i(z,t) \equiv \int_0^t \left[\int_{-\infty}^0 {}^Lm_i(z,t;\xi,\tau)\, {}^1f_i(\xi,\tau)\, d\xi \right] d\tau \quad ; \quad z > 0 \tag{6.72} $$

$$
{}^{R}n_i(z,t\,;\xi,\tau) \equiv \frac{1}{G_-} e^{z/2\,{}^{1}d}\, e^{-\xi/2\,{}^{2}d} \cdot \Big\{ {}^{2}\gamma_i \, {}^{2,1}P_5^{i}(\xi,-z,t-\tau) -
$$

$$
- {}^{2,1}P_4^{i}(\xi,-z,t-\tau) \Big\}\, h(t-\tau) \quad ; \quad \xi > 0
$$

(6.73)

$$
{}^{L}n_i(z,t\,;\xi,\tau) \equiv \frac{1}{2\,{}^{1}\gamma_i}\, e^{(z-\xi)/2\,{}^{1}d} \Big\{ {}^{1}P_1^{i}(z-\xi,t-\tau)\, h(\xi-z) +
$$

$$
+ {}^{1}P_1^{i}(z-\xi,t-\tau)\, h(z-\xi) - \frac{G_+}{G_-}\, {}^{1}P_1^{i}(-z-\xi,t-\tau) -
$$

$$
- \frac{1}{2G_-}\, {}^{1}P_2^{i}(-z-\xi,t-\tau) + \frac{2\,{}^{2}\gamma_i\,{}^{1}\gamma_i}{G_-}\, {}^{12}P_3^{i}(-z-\xi,t-\tau) \Big\} \cdot h(t-\tau)
$$

$$
; \quad \xi < 0
$$

(6.74)

$$
{}^{R}m_i(z,t,\xi,\tau) = \frac{1}{2\,{}^{2}\gamma_i}\, e^{(z-\xi)/2\,{}^{2}d}\, h(t-\tau) \Big\{ {}^{2}P_1^{i}(z-\xi,t-\tau)\, h(z-\xi) +
$$

$$
+ {}^{2}P_1^{i}(z-\xi,t-\tau)\, h(\xi-z) + \frac{G_+}{G_-}\, {}^{2}P_1^{i}(z+\xi,t-\tau) +
$$

$$
+ \frac{1}{2G_-}\, {}^{2}P_2^{i}(z+\xi,t-\tau) - \frac{2\,{}^{1}\gamma_i\,{}^{2}\gamma_i}{G_-}\, {}^{2,1}P_3^{i}(z+\xi,t-\tau) \Big\} \quad ; \quad \xi > 0
$$

(6.75)

$$ {}^{L}m_i(z,t,\xi,\tau) = \frac{e^{z/2\,{}^{2}d}}{G_-}\, e^{-\xi/2\,{}^{1}d}\, h(t-\tau)\Big\{ {}^{1,2}P_4^i(-\xi,z,t-\tau) - $$

$$ - {}^{1}\gamma_i\, {}^{1,2}P_5^i(\xi,z,t-\tau)\Big\} \quad ; \quad \xi < 0 \tag{6.76} $$

$$ {}^{1}f_i(\xi,\tau) \equiv \frac{\lambda_{i-1}}{{}^{1}v_{i-1}}\, {}^{1}n_{i-1}(\xi,\tau) + \phi_i(\tau)\,\delta(\xi - z_1) \; ; \; \xi < 0 \tag{6.77} $$

$$ {}^{2}f_i(\xi,\tau) \equiv \frac{\lambda_{i-1}}{{}^{2}v_{i-1}}\, {}^{2}n_{i-1}(\xi,\tau) \quad ; \quad \xi > 0 \tag{6.78} $$

$$ {}^{\ell}P_1^i(z,t) = \frac{1}{\sqrt{\pi t}}\, e^{-\left[{}^{\ell}\omega_i t \,+\, z^2/4t\,{}^{\ell}v_i\,{}^{\ell}d_i\right]} \quad ; \quad t > 0 \tag{6.79} $$

$$ {}^{\ell}P_2^i(z+\xi,t) = \int_0^t e^{-\lambda_i u}\, {}^{\ell}P_1^i(z+\xi,t-u)\,du \quad ; \quad t > 0 \tag{6.80} $$

$$ {}^{\ell k}P_3^i(z,t) \equiv \int_0^t \frac{z}{2(t-u)\sqrt{{}^{\ell}v\,{}^{\ell}d}}\, {}^{\ell}P_1^i(z,t-u)\cdot $$

$$ \cdot\left\{ \frac{e^{-{}^{k}\omega u}}{\sqrt{\pi u}} + \sqrt{\frac{{}^{\ell}v}{{}^{\ell}d}}\,\frac{1}{2}\, e^{-\lambda_i u}\, \mathrm{erf}\left(\frac{1}{2}\sqrt{u\,{}^{k}v/{}^{k}d}\right)\right\} du \quad ; \quad t > 0 \tag{6.81} $$

$$ {}^{\ell k}P_4^i(x,y,t) \equiv \int_0^t \frac{x\,{}^{\ell}P_1^i(x,t-u)}{2\sqrt{{}^{\ell}v\,{}^{\ell}d}\,(t-u)}\left[\frac{1}{2}\,{}^{k}P_7^i(y,u) + {}^{k}\gamma_i\,{}^{k}P_1(y,u)\right]du \qquad ; \quad t>0 \tag{6.82} $$

$$ {}^{\ell k}P_5^i(x,y,t) \equiv \int_0^t \left[\frac{x}{2(t-u)\,{}^{\ell}v\,{}^{\ell}d} - 1\right]\frac{{}^{\ell}P_1^i(x,t-u)}{2(t-u)} \cdot {}^{k}P_6(y,u)\,du \qquad ; \quad t>0 \tag{6.83} $$

$$ {}^{k}P_6^i(z,t) \equiv \frac{e^{-\lambda_i t}}{2}\left\{ e^{-z/2\,{}^{k}d}\,\mathrm{erfc}\left[\frac{z}{2\sqrt{t\,{}^{k}v\,{}^{k}d}} - \frac{1}{2}\sqrt{t\,{}^{k}v/{}^{k}d}\right] + e^{z/2\,{}^{k}d}\,\mathrm{erfc}\left[\frac{z}{2\sqrt{t\,{}^{k}v\,{}^{k}d}} + \frac{1}{2}\sqrt{t\,{}^{k}v/{}^{k}d}\right]\right\} \quad ; \quad t>0 \tag{6.84} $$

$$ {}^{k}P_7^i(z,t) \equiv \frac{e^{-\lambda_i t}}{2}\left\{ e^{-z/2\,{}^{k}d}\,\mathrm{erfc}\left[\frac{z}{2\sqrt{t\,{}^{k}v\,{}^{k}d}} - \frac{1}{2}\sqrt{t\,{}^{k}v/{}^{k}d}\right] - e^{z/2\,{}^{k}d}\,\mathrm{erfc}\left[\frac{z}{2\sqrt{t\,{}^{k}v\,{}^{k}d}} + \frac{1}{2}\sqrt{t\,{}^{k}v/{}^{k}d}\right]\right\} \quad ; \quad t>0 \tag{6.85} $$

Notice: The superscript i in ${}^{\ell}F_1{}^{i}$, ${}^{\ell}F_2{}^{i}$, ..., ${}^{\ell}F_7{}^{i}$ means that one must use ${}^{\ell}v_i$, ${}^{\ell}d_i$, ${}^{\ell}\omega_i$ etc whenever ${}^{\ell}v$, ${}^{\ell}d$, ${}^{\ell}\omega_i$ etc appear in the definition.

$$G_- \equiv \frac{{}^2d\,{}^1v - {}^1d\,{}^2v}{{}^1v - {}^2v} \qquad (6.86)$$

$$G_+ \equiv \frac{{}^2d\,{}^1v + {}^1d\,{}^2v}{{}^1v\,{}^2v} \qquad (6.87)$$

$${}^{\ell}\gamma_i \equiv \sqrt{\frac{{}^{\ell}d_i}{{}^{\ell}v_i}} \qquad (6.88)$$

$${}^{\ell}\omega_i \equiv \lambda_i + \frac{{}^{\ell}v_i}{4\,{}^{\ell}d_i} \qquad (6.89)$$

7. CONCLUSION

The research in FY-1979 is aimed to elucidate the following problems:

1) The fundamental transport equation governing the nuclide migration,
2) The relation among several nuclide release modes from the repository,
3) Recursive and general solutions of an i^{th} member chain migration for one-dimensional water path and arbitrary release modes,
4) Application of the recursive and the general solutions to three-member decay chain and the elucidation of the migration features for some most important decay chains,
5) Nuclide migration through the media of multi-layers.

To summarize, the following results have been obtained:

1. The fundamental transport equations governing the migration of nuclide decay chain were derived based on the phenomenological approach and with help of volume average concept. In case that the sorption and the chemical reaction processes are attained to be equilibrated, the fundamental governing equation is given by equation (1).

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$$K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} - D_i \frac{\partial^2 N_i}{\partial z^2} + \lambda_i K_i N_i$$

$$= \lambda_{i-1} K_{i-1} N_{i-1} + f_i \qquad (1)$$

where

N_i = concentration of i^{th} nuclide in aqueous phase
K_i = overall sorption equilibrium constant for i^{th} nuclide
λ_i = radioactive decay constant
f_i = nuclide source term, z=distance, t=time
D_i = effective dispersion coefficient
v = groundwater velocity

2. The relationship among several release modes has been discussed. The release modes include a) impulse, b) step, c) band, d) linear leach rate, f) fractional leach rate characteristic to individual nuclide. The transformation to yield different solutions from the solution of representative release mode has been discussed. A general superposition theorem has been developed for the propagation of chromatographic bands with radioactive decay. This provides an exact method for constructing the space-time dependent concentration resulting from a source of finite duration (band release) by superposing time-displace solutions for source of infinite duration (step release).

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3. Recursive solutions of the transport equations in one-dimensional pathway with and without dispersion have been developed both for a generalized source boundary condition and for a generalized concentration boundary condition. Solution for any radioactive nuclide in an i member decay chain in one dimension and without dispersion has been evaluated.

4. Explicit solutions for three member chains in one dimensional transport have been developed for a source of constant leach rate, with and without dispersion, using a source boundary condition which specifies a time-dependent concentration of leachant at the source. The solution has been programmed.

Solutions for one-dimensional transport with dispersion have been developed for a time-dependent plane source. The results for a three member decay chain have been programmed. The error due to the usual assumption of a time-dependent concentration in the leachant at the source, which ignored dispersion at the source, have been evaluated.

The explicit solutions for an i member decay chain without dispersion have been developed for the above five release modes.

The migration features for the most important two decay chains, $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ and $^{237}NP \rightarrow {}^{233}Pa \rightarrow {}^{233}U \rightarrow {}^{229}Th \rightarrow {}^{225}Ra$, have been elucidated with the help of the above solutions. The isopleths for ^{226}Ra in the water pathway have been demonstrated, in which the peak concentration of a nuclide in the moving sorption band can increase with distance travelled through the medium. Parametric analysis for the migration features has been carried out for the dispersion coefficient, the leaching time, the sorption equilibrium constant and the initial ratios of nuclide sources at the repository.

5. Analytical expression for the nuclide migration through one-dimensional multi-layered media of different hydrological sorptive properties have been developed. This expression is of recursive type with respect to media and nuclide decay. The analytical explicit solution for two media and three member decay chain has been obtained for dispersion free case. The recursive solution for two media with dispersion has been developed.

The analysis in 2. to 5. has been carried out based in equation (1), which was derived by assuming equilibrium sorption and chemical reaction processes.

8. NOMENCLATURE

A : Interfacial area in the averaging domain

A_i : Coefficient defined by (2,55)

$A_i^{(j)} = \prod_{r=j}^{i-1} (\lambda_r / \upsilon_r)$: Defined by (4.113)

${}^{\ell}\mathcal{A}_i^s$: Function defined by (6.36) and (6.37)

a : Interfacial area per unit volume of geological media

B_i : Coefficient defined by (2.55)

$B_i(t)$: Bateman equation

B_{ij} : Bateman coefficient defined by (3.12)

B'_{ij} : Modified Bateman coefficient defined by (3,23)

$B_m^{(j)} = B_m^{(ij)} = \prod_{\substack{r=j \\ r \neq m}}^{i} \Gamma_{rm}$: Defined by (4.121)

${}^{\ell}\mathcal{B}_i^s$: Function defined by (6.38) and (6.39)

$C_{k\alpha}$: Concentration of k-th chemical species in α phase

$\overset{o}{C}_{k\alpha}$: Fluctuation concentration of k-th chemical species in α-phase

$C_m^{(j)}(s) = \left[\prod_{\substack{r=j \\ r \neq m}}^{i} (\mu_r - \mu_m) \right]^{-1}$: Defined by (4.116)

${}^{2}\mathcal{C}_3^s$: Function defined by (6.40)

D : Axial effective dispersion coefficient (mean value)

D^* : Axial dispersion coefficient defined by (4.41)

D_i : Effective dispersion coefficient for i-th nuclide

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D_f : Dispersion coefficient in fluid phase

$\mathbb{D}_f = \mathbb{D}_{df} + D_{mf}\mathbb{1}$: Diffusion coefficient tensor

D_s : Molecular diffusion coefficient in solid phase

D_{df} : Dispersion coefficient tensor in fluid phase

$D_{k\alpha}$: Molecular diffusion coefficient of k-th species in α-phase

D_{mf} : Molecular diffusion coefficient in fluid phase

$D^{ij}_{rm} = \left[\prod_{\substack{q=j \\ q\neq m,\, q\neq r}}^{i} (\Delta_{qm} - \Delta_{rm})\right]^{-1}$: Defined by (4.122)

${}^{\ell}d_i = {}^{\ell}D_i / {}^{\ell}V$: Defined by (6.4)

$E(i,j,k)$: Function defined by (5.33) or (5.40)

$F(\alpha,\beta)$: Gaussian distribution function defined by (4.33)

$f_{i\alpha}(\chi,t)$: Source term of i-th nuclide in α-phase

$f_i(Z,t)$: Source term of ith nuclide in water phase

[illegible]$_i(t) = \varphi_i(t)\,\delta(z)$: Defined by (4.23)

${}^{\ell}$[illegible]$_i$: Function defined by (6.77) and (6.78)

${}^{\ell}$[illegible]$^{j}_{p}$: Function defined by (6.42) to (6.45) or (6.25) and (6.26)

$G_i(z,t;\xi,\tau), G_i(z;\xi,\tau)$; Green function

G_+, G_- : Function defined by (6.87) and (6.86)

${}^{\ell}G^{j}_{p}$: Function defined by (6.46) and (6.47) or (6.27)

$g_i(t)$: Function defined by (4.47b)

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$g_{rm}(t)$: Function defined by (4.124)

${}^{\ell}H_j$: Function defined by (6.48) to (6.54)

${}^{\ell}H_{rm}^{ij\ell}$: Function defined by (6.28)

$H(x\epsilon x)$: Function defined by (2.5)

$h(t-T)$: Heaviside step function

${}^{\ell}I = {}^{\ell+1}\rho / {}^{\ell}\rho$

$\mathbf{j}_{k\alpha}$: Relative flux of k-th chemical species in α-phase

K_i : Overall sorption equilibrium coefficient defined by (2.53)

$K_{D,i}$: Overall distribution coefficient defined by (2.57)

k : Leaching rate constant

k_i : Leaching rate constant for i-th nuclide species defined by (3.18)

k_{Ak} : Reaction rate constant for k-th chemical species defined by (2.58)

$K_{D,k}$: Distribution coefficient of k-th chemical species between solid and water phase (2.49)

$K_{D,i}$: Distribution coefficient of i-th nuclide between solid and water phases

k_I : Rate coefficient for mineralization reaction

k_{Mk} : Overall mass transfer coefficient for k-th chemical species

$k_{R,k\alpha}^{t}$: Formation constant of t-th chemical species from k-th species in α-phase

L : Length of repository zone

$L_i, \hat{L}_i$: Partial differential operators defined by (3.34) and (6.2)

$\mathbb{I}$: Unit tensor

M_T : Total amount of waste material

$M_i(t)$: Amount of i-th nuclide in the repository at time t

M_i^0 : Initial amount of i-th nuclide at the repository

$M_{rm}^{\hat{p}}$: Function defined by (6.55) and (6.56)

${}^{\alpha}m_i(z,t;\xi,\tau)$: Green function satisfying (6.65) and (6.66) for α = R or L

N_i^0 : Nuclide concentration in water phase, Initial value

$N_i(t)$: Concentration of i-th nuclide in water phase at the exit of the repository

$N_{i\alpha}(Z,t)$: Concentration of i-th nuclide in α - phase

$N_i(z,t)$: Concentration of i-th nuclide in water phase

$N_i^{(j)}(z,t)$: Contribution of j-th nuclide at the repository to concentration of i-th nuclide

$N_{i,r}(t)$: Concentration of i-th nuclide defined by (4,84)

$\mathbf{n}_\alpha$: Unit normal vector pointing out of the interface from α - phase

$n_i(t)$: Amount of nuclide i per unit amount of waste

$n_i(p,t)$:Laplace transform of $N_i(z,t)$ with respect to z

$n_i^{(j)}(p,t)$: Laplace transform of $N_i^{(j)}(z,t)$ with respect to z

${}_{\alpha}n_i(z,t:\xi,\tau)$: Green function satisfying (6.65) and (6.66)

α = R or L

${}_{\ell k}p_j^i$: Function defined by (6.81) and (6.82)

${}_{\ell}p_j^i$:Function defined by (6.79) and (6.80), (6.84) and (6.85)

p : Variable for Laplace transform with respect to z

Q : Volumetric flow rate of water

$R_{k\alpha}$: Birth rate of k-th chemical species in α-phase

$R_{i-1,i}(z,t)$: Inverse transform of $r_{i-1,i}(p,t)$

$r_{i-1,i}(p,t)$: Function defined by (4.55b)

S : Effective cross-sectional area for water phase of the pathway

S_α : Cross-sectional area of α-phase

$S_{i-1}(z,t)$: Function defined by (4.45b)

S_i^k : Atomic number of i-th nuclide in k-th chemical species

s : Variable for Laplace transform with respect to time t

$s_{i-1}(p,t)$: Laplace transform of $S_{i-1}(z,t)$

$s_{i_1}^{i-1}$: Fraction that i_1 chemical form is generated by the decay of (i-1)-th nuclide

t : Time

T ; Leach time or mean residence time of water phase in the repository zone

$T(p,t)$: Transform Kernal defined by (4.50)

$U_i(z,t)$: Function defined by (4.44)

$u_i(p,t)$: Laplace transform of $U_i(z,t)$

V : Volume of averaging domain or water phase volume at repository

v : water phase velocity

$\mathbf{v}_\alpha$: Velocity in α - phase

v_i = v/K_i

v_f : Fluid phase velocity

$\mathbf{w}$: Interface velocity

$\mathbf{X}$: Position vector measured from the outer coordinate

$\mathbf{x}$: Position vector relative to $\mathbf{X}$

$\mathbf{x}_b$: Position vector at interface

z : Distance along water pathway

z_1 : Repository position

${}^{\ell}z$: Length of l-th layer

${}^{\ell}\tilde{z}$: Relative distance from (ℓ-1)-layer boundary

α_i = $\lambda_i k_i$; Defined by (4.42)

β_i = $-\lambda_i - \gamma^2\eta_i$; Defined by (4.42)

β_{ij} : Parameter defined by (5.32)

γ = $v/2D$; Defined by (4.42)

${}^{\ell}\gamma_i$ = $\sqrt{{}^{\ell}d_i/\rho v_i}$: Defined by (6.88)

γ_{ijk} : Parameter defined by (5.32)

Γ_{rm} = $v_r^{-1} - v_m^{-1}$

Δ_{ir} = $(\lambda_i/v_i - \lambda_r/v_r)/(v_i^{-1} - v_r^{-1})$

$\delta(t)$: Dirac delta function

ε : Porosity

ε_α : Volume fraction of α- phase

Θ_{ip} : Function defined by (6.29)

η_i = Di/Ki

κ = D/v

Λ_i = $\lambda_i + k_i$

Λ_{kj} = $\lambda_k/v_k - \lambda_j/v_j$

λ_i : Decay constant of i-th nuclide

μ_ℓ = $(S+\lambda_\ell)/v_\ell$: Defined by (4.110c)

$\nu_{\ell-1}$ = $K_{\ell-1}\lambda_{\ell-1}/K_\ell\lambda_\ell$: Defined by (4.110c)

ξ : Distance

ρ : Density of water phase

ρ_α : Density of α phase

τ : Time

$\phi_i(t)$: Functions defined by (4.21) or (4.41)

ψ : Dynamical property

Ψ_α : Dynamical property in α - phase

Ψ, Ψ_α : Dynamical property defined by (2.6)

${}^{\ell}\omega_i$: Parameter defined by (6.89)

Subscript

i : nuclide i

k : Chemical species k

α : phase

Superscript

b : band release

e : exponential release

I : impulse release

p : preferential release

s : step release

ℓ : ℓ-th layer (superscript at left-hand side)

$\sim$: Volume average defined by (2.8)

$\wedge$: Volume average defined by (2.7)

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APPENDIX - Computer Code

A computer code named MGRAT01 has been developed to calculate the migration of radionuclides with dispersion for both the band and the step release made up to three member chain, in a geologic media using the analytical expressions which are given in Section 5.

The MGRAT01 calcules the distribution either of the relative concentration in a water phase of each nuclide or of the discharge rate, along the time for fixed locations or along the location for fixed times. The code is constructed by a main program MGRAT, and three function programs, ELF, ERFC, ERF as attached lists of programs. Some rational approximation shown below are used to calculate the error function.

$$\mathrm{erf}\ x = 1 - \frac{1}{[1 + a_1 x + a_2 x^2 + \cdots\cdots + a_6 x^6]^{16}} + \varepsilon(x)$$

$$|\varepsilon(x)| \leq 3 \times 10^{-7}$$

$a_1 = .07052\ 30784$ $\quad a_2 = .04228\ 20123$

$a_3 = .00927\ 05272$ $\quad a_4 = .00015\ 20143$

$a_5 = .00027\ 65672$ $\quad a_6 = .00004\ 30638$

and

$$\mathrm{erf}\ x = 1 - (a_1 t + a_2 t^2 + \cdots\cdots + a_5 t^5)\, e^{-x^2} + \epsilon(x)$$

$$t = \frac{1}{1 + px}, \quad |\epsilon(x)| \leq 1.5 \times 10^{-7}$$

$P = .32759\ 11$ $\quad a_1 = .25482\ 9592$

$a_2 = -.28449\ 6736$ $\quad a_3 = 1.42141\ 3741$

$a_4 = -1.45315\ 2027$ $\quad a_5 = 1.06140\ 5429$

Due to the approximation, the absolute error of the code is less equal to $3x10^{-7}$. The development of the code has been made by using the CDC 6600 and 7600 of the Lawrence Berkeley Laboratory Computer Center. However, this code is valid when the value of γ_{ijk} in (5.32) is positive. In the case of $\gamma_{ijk}<0$, we can easily extend the code using a direct numerical integration of the function E(i,j;k) in (5.33). The development of the code is now underway.

```
  **PROGRAM MIGRAT(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

      PROGRAM MIGRAT(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
      DIMENSION ANAME(5),HLIFE(5),CURIN(5),COEFK(5),T(2),Z(2),R(5),AT(5)
     1          ,V(5),C2(5),C3(15),D1(2),D2(5,2),D3(15,2),CN(5),CM(5)
     2          ,RCGW(3)
      READ(5,500) IC,ICAL,IRCG,IDM
      READ(5,501)  (ANAME(I),HLIFE(I),CURIN(I),COEFK(I),RCGW(I),I=1,IC)
      READ(5,502)  DIF,VEL,FLW,TL
      READ(5,503)  IT,T(1),T(2)
      READ(5,503)  IZ,Z(1),Z(2)
      DO 5  I=1,IC
      A=  ALOG(2.)/HLIFE(I)
      R(I)=  A
      AT(I)=  CURIN(I)/(FLW*TL)
      IF(IDM.EQ.1) AT(I)= AT(I)/A
    5 V(I)=  VEL/COEFK(I)
      VMAX=  AMAX1(V(1),V(2),V(3))
      VMIN=  AMIN1(V(1),V(2),V(3))
      CUR=  R(1)*CURIN(1)/1.16683E18
      IF(IDM.EQ.1)  CUR= CURIN(1)
      D=  DIF/VEL
  500 FORMAT(4I1)
  501 FORMAT(A7,4F8.0)
  502 FORMAT(4F8.0)
  503 FORMAT(I2,2F8.0)
      WRITE(6,605) (ANAME(I),HLIFE(I),CURIN(I),COEFK(I),R(I),V(I),AT(I),
     1I=1,IC)
      WRITE(6,606)  DIF,VEL,FLW,TL
  605 FORMAT(10X,A7,1PE15.3,*YR*,5E15.3)
  606 FORMAT(10X,1P4E15.3,1H1)
      R12= R(1)-R(2)
      R23=  R(2)-R(3)
      R31=  R(3)-R(1)
      R21= -R12
      R32= -R23
      R13= -R31
      RR=  R(1)*R(2)*AT(1)
      B11=  AT(1)
      B12= -AT(1)*R(1)/R12
      B22=  AT(2)-B12
      B13= -RR/(R12*R31)
      B23= -RR/(R12*R23)-AT(2)*R(2)/R23
      B33= -RR/(R23*R31)+AT(2)*R(2)/R23+AT(3)
      D4=  4.*D
      S1=  1./SQRT(V(1)*D)
      S2=  1./SQRT(V(2)*D)
      S3=  1./SQRT(V(3)*D)
      F12= (V(1)*R(2)-V(2)*R(1))/(V(1)-V(2))
      F23= (V(2)*R(3)-V(3)*R(2))/(V(2)-V(3))
      F31= (V(3)*R(1)-V(1)*R(3))/(V(3)-V(1))
      V1=  V(1)/D4
      V2=  V(2)/D4
      V3=  V(3)/D4
      IX=  0
      TD=  0.
   10 CONTINUE
      E1=  EXP(-R(1)*TD)
      E2=  EXP(-R(2)*TD)
```

```
      E3=  EXP(-R(3)*TD)
      B11=  B11*E1
      B12=  B12*E1
      B13=  B13*E1
      B22=  B22*E2
      B23=  B23*E2
      B33=  B33*E3
      C1=  B11
      C2(1)=  B12
      C2(2)=  B22
      C2(3)=  B11*V(2)*R(1)/(V(1)*R21)
      C3(1)=  B13
      C3(2)=  B23
      C3(3)=  B33
      C3(4)=  B12*V(3)*R(2)/(V(3)*R21+V(2)*R13)
      C3(5)=  B22*V(3)*R(2)/(V(2)*R32)
      THY=  V(2)*R13+V(3)*R21
      RHO=  THY+V(1)*R32
      CCC=  R(1)*R(2)*V(3)/(V(1)*R21)
      WA=  B11*V(2)*CCC/THY
      WB=  B11*CCC/R31
      WC=  B11*(V(2)-V(1))*CCC/RHO
      WD= -WC
      C3(6)=  WA
      C3(7)= -WA-WB
      C3(8)=  WB
      C3(9)= -WA-WD
      C3(10)=  WD
      C3(11)= -WB-WC
      C3(12)= -WD
      C3(13)=  WB+WC
      C3(14)=  WA+WD
      IF(IX.EQ.1)  GO TO 13
      D1(1)=  C1
      DO 11  I=1,3
   11 D2(I,1)=  C2(I)
      DO 12  I=1,14
   12 D3(I,1)=  C3(I)
      IX=  1
      TD=  TL
      GO TO 10
   13 D1(2)=  C1
      DO 14  I=1,3
   14 D2(I,2)=  C2(I)
      DO 15  I=1,14
   15 D3(I,2)=  C3(I)
      WRITE(6,650) (D1(I),(D2(J,I),J=1,3),I=1,2)
      WRITE(6,650) ((D3(J,I),J=1,14),I=1,2)
  650 FORMAT(10E11.3)
      IF(ICAL.EQ.1)  GO TO 21
      KM=  0
      IZ1=  IZ+1
      IT1=  IT+1
      DO 100  M=1,IT1
      T2=  T(1)*10.**(M-1)
      KM=  1
      GO TO 20
   16 CONTINUE
```

```
      T2=  T(1)*10.**(M-1)*5.
      KM=  0
   20 CONTINUE
      T1=  T2
      WRITE(6,601)  T2,(ANAME(I),I=1,3),(ANAME(I),I=1,3)
  601 FORMAT(//,8X,*T=*,1PE10.3,15X,3(2X,A7,1X),5X,3(2X,A7,1X))
      DO 90. N=1,IZ1
      DO 90 L=1,90
      FL=  L
      Z1=  Z(1)*10.**(N-1)*(1.+.1*(FL-1.))
      Z2=  Z1/D4*2.
      IA=  1
   30 C1=  D1(IA)
      DO 31  I=1,3
   31 C2(I)=  D2(I,IA)
      DO 32  I=1,14
   32 C3(I)=  D3(I,IA)
      IF(IA.EQ.2)  T1= T2-TL
      XA=  ELF(R(1),V1,Z1*S1,Z2,T1)
      XB=  ELF(R(1),R21+V2,Z1*S2,Z2,T1)
      XC=  ELF(R(2),V2,Z1*S2,Z2,T1)
      XD=  ELF(F12,R(1)-F12+V1,Z1*S1,Z2,T1)
      XE=  ELF(F12,R(2)-F12+V2,Z1*S2,Z2,T1)
      XF=  ELF(R(1),R31+V3,Z1*S3,Z2,T1)
      XG=  ELF(R(2),R32+V3,Z1*S3,Z2,T1)
      XH=  ELF(R(3),V3,Z1*S3,Z2,T1)
      XI=  ELF(F23,R(2)-F23+V2,Z1*S2,Z2,T1)
      XJ=  ELF(F23,R(3)-F23+V3,Z1*S3,Z2,T1)
      XK=  ELF(F31,R(1)-F31+V1,Z1*S1,Z2,T1)
      XL=  ELF(F31,R(3)-F31+V3,Z1*S3,Z2,T1)
      CN(1)=  .5*C1*XA
      CN(2)=  .5*(C2(1)*XB+C2(2)*XC+C2(3)*(-XB+XA-XD+XE))
      CN(3)= (C3(1)*XF+C3(2)*XG+C3(3)*XH+C3(4)*(XF- XB+XI- XJ)
     1        +C3(5)*(XC-XG+XJ-XI)+C3(6)*XB+C3(7)*XF+C3(8)*XA
     2        +C3(9)*XI+C3(10)*XE+C3(11)*XK+C3(12)*XD
     3        +C3(13)*XL+C3(14)*XJ)*.5
      IF(T2.LT.TL)  GO TO 36
      IF(IA.EQ.2)  GO TO 34
      DO 33  I=1,3
   33 CM(I)=  CN(I)
      IA=  2
      GO TO 30
   34 DO 35  I=1,3
   35 CN(I)=  CM(I)-CN(I)
   36 CONTINUE
      DO 37  I=1,3
      CM(I)= CN(I)/AT(1)
   37 CN(I)=  CN(I)*R(I)/(AT(1)*R(1))
      CMAX=  AMAX1(CN(1),CN(2),CN(3))
      IF(IRCG.NE.1)  GO TO 75
      DO 38  I=1,3
      CM(I)=  CUR*CN(I)/TL
   38 CN(I)=  CM(I)/RCGW(I)
   75 CONTINUE
      ZMAX=  VMAX*T2
          IF(ZMAX.GT.Z1)  GO TO 80
      IF(CMAX.LT.1.E-100)  GO TO 95
   80 CONTINUE
```

```
      WRITE(6,603) Z1,(CM(I),I=1,3),(CN(I),I=1,3)
  603 FORMAT(20X,1PE10.3,5X,3E10.3,5X,3E10.3)
      T1=  T2
   90 CONTINUE
   95 CONTINUE
      IF(KM.EQ.1)  GO TO 16
  100 CONTINUE
      GO TO 300
   21 KM=  0
      IZ1=  IZ+1
      IT1=  IT+1
      DO 200  M=1,IZ1
      JJ= 1
      DO 250  J=1,4
      JJ= JJ+J-1
      FJ= JJ
      Z1=  Z(1)*10.**(M-1)*FJ
      Z2=  Z1/D4*2.0
      WRITE(6,602)  Z1,(ANAME(I),I=1,3),(ANAME(I),I=1,3)
  602 FORMAT(//,8X,*Z=*,1PE10.3,15X,3(2X,A7,1X),5X,3(2X,A7,1X))
      DO 190  N=1,IT1
      DO 190 L=1,90
      FL=  L
      T2=  T(1)*10.**(N-1)*(1.+.1*(FL-1.))
      DELT=  ABS(T2-TL)
      IF(DELT.LT.10.)  GO TO 190
      T1=  T2
      IA=  1
  301 C1=  D1(IA)
      DO 311 I=1,3
  311 C2(I)=  D2(I,IA)
      DO 321 I=1,14
  321 C3(I)=  D3(I,IA)
      IF(IA.EQ.2)  T1= T2-TL
      XA=  ELF(R(1),V1,Z1*S1,Z2,T1)
      XB=  ELF(R(1),R21+V2,Z1*S2,Z2,T1)
      XC=  ELF(R(2),V2,Z1*S2,Z2,T1)
      XD=  ELF(F12,R(1)-F12+V1,Z1*S1,Z2,T1)
      XE=  ELF(F12,R(2)-F12+V2,Z1*S2,Z2,T1)
      XF=  ELF(R(1),R31+V3,Z1*S3,Z2,T1)
      XG=  ELF(R(2),R32+V3,Z1*S3,Z2,T1)
      XH=  ELF(R(3),V3,Z1*S3,Z2,T1)
      XI=  ELF(F23,R(2)-F23+V2,Z1*S2,Z2,T1)
      XJ=  ELF(F23,R(3)-F23+V3,Z1*S3,Z2,T1)
      XK=  ELF(F31,R(1)-F31+V1,Z1*S1,Z2,T1)
      XL=  ELF(F31,R(3)-F31+V3,Z1*S3,Z2,T1)
      CN(1)=  .5*C1*XA
      CN(2)=  .5*(C2(1)*XB+C2(2)*XC+C2(3)*(-XB+XA-XD+XE))
      CN(3)= (C3(1)*XF+C3(2)*XG+C3(3)*XH+C3(4)*(XF-XB+XI-XJ)
     1       +C3(5)*(XC-XG+XJ-XI)+C3(6)*XB+C3(7)*XF+C3(8)*XA
     2       +C3(9)*XI+C3(10)*XE+C3(11)*XK+C3(12)*XD
     3       +C3(13)*XL+C3(14)*XJ)*.5
      IF(T2.LT.TL)  GO TO 361
      IF(IA.EQ.2)  GO TO 341
      DO 331 I=1,3
  331 CM(I)=  CN(I)
      IA=  2
      GO TO 301
```

```
    **PROGRAM MIGRAT(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

341 DO 351 I=1,3
351 CN(I)=  CM(I)-CN(I)
361 CONTINUE
    DO 371 I=1,3
    CM(I)= CN(I)/AT(1)
371 CN(I)=  CN(I)*R(I)/(AT(1)*R(1))
    CMAX=  AMAX1(CN(1),CN(2),CN(3))
    IF(IRCG.NE.1)  GO TO 175
    DO 381 I=1,3
    CM(I)=  CUR*CN(I)/TL
381 CN(I)=  CM(I)/RCGW(I)
175 CONTINUE
    TMAX=  Z1/VMIN+TL
    IF(TMAX.GT.T2)  GO TO 180
    IF(CMAX.LT.1.E-100)  GO TO 250
180 CONTINUE
    WRITE(6,603) T2,(CM(I),I=1,3),(CN(I),I=1,3)
190 CONTINUE
250 CONTINUE
200 CONTINUE
300 CONTINUE
    STOP
    END
```

```
    **FUNCTION ERF(XX)**

    FUNCTION ERF(XX)

 ERF=2/SQRT(PI)*INTEGRAL OF EXP(-T*T) FROM 0 TO X.
 USING AN APPROXIMATION DUE TO HASTINGS  GOOD TO SEVEN SI
 USING AN APPROXIMATION DUE TO HASTINGS. ABSOLUTE ERROR ABOUT 3E-7

    DIMENSION A(6)
    DATA A/.0000430638,.0002765672,.0001520143,.0092705272,.0422820123
   1,.0705230784 /
    X=ABS(XX)
    T=A(1)*X
    DO 10 I=2,6
    T=(T+A(I))*X
 10 CONTINUE
    T=1./(T+1.)
    ERF=1.-T**16
    IF(XX.LT.0.)ERF=-ERF
    RETURN
    END
```

FUNCTION ERFC(X)

```
      FUNCTION ERFC(X)
      DIMENSION A(5)
      DATA A/.254829592,-.284496736,1.421413741,-1.453152027,1.061405429
     1/,P/.3275911/
      X=  ABS(X)
      T=  1./(1.+P*X)
      ER=  0.0
      DO 10  I=1,5
   10 ER=  ER+A(I)*T**I
      ERFC=  ER
      IF(X.LT.0.)  ERFC= -ER
      RETURN
      END
```

FUNCTION ELF(A,B,C,D,E)

```
      FUNCTION ELF(A,B,C,D,E)
      Y=  D-A*E-C*SQRT(B)
      Y=  EXP(Y)
      W= C/SQRT(4.*E)-SQRT(B*E)
      W=  ERF(W)
      S=  C/SQRT(4.*E)+SQRT(B*E)
      X=  -A*E+D+C*SQRT(B)-S**2
      X=  EXP(X)*ERFC(S)
      ELF=  (Y*(1.-W)-X)*D/(C*SQRT(B))
      RETURN
      END
```

Errata: "Migration of Radionuclides Through Sorbing Media: Analytical Solutions - I", LBL 10500 Draft, by M. Harada, P. L. Chambré, M. Foglia K. Higashi, F. Iwamoto, D. Leung, T. H. Pigford and D. Ting, February 1980.

Errata

	page	line	Eq. or Fig.	erratum	correct
✓	2.22	13	(2.51)	$\tilde{N}_{is} = [\sum_k (k_{D,k} S_i^k / k^t_{R,ks}) / \sum_k S_i^k / k^t_{R,ks}]$ $\cdot \tilde{N}_{if} = \cdots$	$\tilde{N}_{is} = [\sum_k (k_{D,k} S_i^k / k^t_{R,kf}) / \sum_k (S_i^k / k^t_{R,kf})$ $= \cdots$
✓	3.1	20		all of the waste material begins	all of the waste material begin
✓	3.2	8	(3.1)	$\dot{M}_T = M^0 / T$	$\dot{M}_T = M_T^0 / T$
✓	3.3	6	(3.5)	$N_1(t) = N_1^0 e^{-\lambda_1 t}$	$n_1(t) = n_1^0 e^{-\lambda_1 t}$
✓	3.4	7	(3.10)	$B_i(0) = N_i(0)$	$B_i(0) = N_i^0$
✓	3.4	8		$N_i(0)$ is the concentration ...	N_i^0 is the concentration ...
✓	3.4	10		(3.10) is the Bateman	(3.9) is the Bateman ...
✓	3.4	11	(3.11)	$B_i(t) = \sum_{m=1}^{i} \left(\frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \sum \cdots$	$B_i(t) = \sum_{m=1}^{i} \left(\frac{N_i^0}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \sum \cdots$
✓	3.7	16	(3.23)	$B_{ij}' \equiv \sum_{m=1}^{j} (\cdots)$	$B_{ij}' \equiv \sum_{m=1}^{j} (\cdots)$, $N_m^0 = \frac{k_m M_m^0}{Q}$
✓	3.8	14	(3.28)	$N_m^0 = k M_m^0 Q$	$N_m^0 = k M_m^0 / Q$
✓	3.10	1	Fig. 1	$\leftarrow v N_i(0+, t)$	$\rightarrow v N_i(0+, t)$
✓	3.11	4		$M_i^0(0, t)$	$\dot{M}_i(0, t)$
✓	3.11	5	(3.31)	$M_i^0(t) = N_i(t) Q$	$\dot{M}_i(t) = N_i(t) Q$
✓	3.14	6		Where T is ...	where T is ...
✓	3.16	5		(3.44) and (3.43) ...	(3.42) and (3.43) ...
✓	3.19	7		with the help of (3.55) ...	with the help of (3.32) ...
✓	3.21	5		As seen how equation ...	As seen from equation ...
✓	4.4	2	(4.8)	$-\frac{1}{4\pi^2} \int_{-\infty}^{\infty} e^{ik(z-\xi)} dk \int_{-\infty}^{\infty} e^{-i\omega(t-\tau)} d\tau$	$-\frac{1}{4\pi^2} \int_{-\infty}^{\infty} e^{ik(z-\xi)} dk \int_{-\infty}^{\infty} e^{-i\omega(t-\tau)} d\omega$
✓	4.5		Fig. 4.1	In case of $(t$	In case of $(t-\tau) < 0$
✓	4.8	12	(4.25)	$\frac{1}{\sqrt{4\pi \eta_i t}} \exp(\cdots)$	$\frac{1}{\sqrt{4\pi \eta_i \tau}} \exp(\cdots)$
✓	4.9	12	(4.27)	$\frac{1}{\sqrt{4\pi \eta_i t}} \exp(\cdots)$	$\frac{1}{\sqrt{4\pi \eta_i \tau}} \exp(\cdots)$
✓	4.22	10	(4.44)	$U_i(z, t) = \cdots + \int_0^t d\tau \int_0^\infty \cdots d\tau$	$U_i(z, t) = \cdots + \int_0^t d\tau \int_0^\infty \cdots d\xi$
✓	4.26	6	(4.82)	$N_i(z, t) = \cdots + \frac{K_{i-1} \lambda_{i-1}}{K_i} \int_0^t e^{-\lambda_i \tau} \cdots$	$N_i(z, t) = \cdots + \frac{K_{i-1} \lambda_{i-1}}{K_i} \int_0^{\min(t, z/v_i)} e^{-\lambda_i \tau} \cdots$
✓	4.27	8	(4.83)	$N_1(z, t) = N_1^0 e^{-\lambda_1 / v_1} \phi_1 (t - \frac{z}{v_1})$	$N_1(z, t) = N_1^0 e^{-\lambda_1 z / v_1} \phi_1 (t - z/v_1)$

page	line	Eq. or Fig.	erratum	correct
✓ 4.30	20		(z/v_λ), $k=1,2,\cdots$	(z/v_k), $k=1,2,\cdots$
✓ 4.35		(4.105)	$j=\underline{j}$	$j=\underline{i}$
✓ 4.41	3	(4.120)	$n_i^{(j)}(z,s)=A_i^{(j)}N_i^0\sum_{m=j}^{i}\frac{\phi_i(s)e^{-\left(\frac{s+\lambda_m}{\underline{v_m}}\right)z}}{\Pi\cdots}$	$n_i^{(j)}(z,s)=A_i^{(j)}N_i^0\sum_{m=j}^{i}\frac{\phi_j(s)e^{-\frac{s+\lambda_m}{\underline{v_m}}z}}{\Pi\cdots}$
✓ 4.42	9		$\underline{\text{In}}$ view of the restriction …	in view of the restriction …
✓ 4.44	9		, where $F(v_i\theta_i;z-\xi-v_i\theta_i)=\frac{1}{\underline{4\pi k v_i\theta_i}}\exp(\cdots)$	$F(v_i\theta_i,z-\xi-v_i\theta_i)=\frac{1}{\underline{\sqrt{4\pi k v_i\theta_i}}}\exp(\cdots)$
✓ 5.2	14	(3.23)	$B'_{ij}=\frac{\sum_{m=1}^{j}\left(\frac{k_i}{k_m}N_m^0\right)\cdots}{\sum_{\substack{l=m\\l\neq j}}^{i}(\lambda_l-\lambda_j)}$	$B'_{ij}=\underline{\sum_{m=1}^{j}}\frac{\left(\frac{k_i}{k_m}N_m^0\right)\cdots}{\sum_{\substack{l=m\\l\neq j}}^{i}(\lambda_l-\lambda_j)}$
✓ 5.3	1	(5.2)	$N_i^P(z,t)=e^{-\lambda_i z_i/v_i}h\left(t-\frac{z}{\underline{v_1}}\right)\sum_{j=1}^{i}\cdots$ $+\cdots$	$N_i^P(z,t)=e^{-\lambda_i z_i/v_i}h\left(t-\frac{z}{\underline{v_i}}\right)\sum_{j=1}^{i}\cdots$ $+\cdots$
✓ 5.5	3	(5.5)	$+\frac{\lambda_2}{v_2}h\left(t-\frac{z}{v_3}\right)\frac{e^{-\lambda_3 z/\lambda_3}}{\Gamma_{23}}\left[\frac{D_{23}^{31}B'_{21}}{\lambda_1-\Delta_{23}}\{\cdots\}+\cdots\right.$	$+\frac{\lambda_2}{v_2}h\left(t-\frac{z}{v_3}\right)\frac{e^{-\lambda_3 z/v_3}}{\Gamma_{23}}\left[\frac{D_{23}^{32}B'_{21}}{\lambda_1-\Delta_{23}}\{\cdots\}+\cdots\right.$
✓ 5.7	1	(5.9)	$N_1^e(z,t)=B_{11}h(t-z/v_1)$ $\cdot e^{-(\lambda_1+k)t\underline{-}kz/v_1}$	$N_1^e(z,t)=B_{11}h\left(t-\frac{z}{v_1}\right)e^{-(\lambda_1+k)t\underline{+}kz/v_1}$
✓ 5.7	5	(5.9)	$B_{ij}=\sum_{m=1}^{j}N_m^0\frac{\prod_{r=m}^{i}\underline{\lambda_i}}{\lambda_i}\Big/(\cdots)=$	$B_{ij}=\sum_{m=1}^{j}N_m^0\frac{\prod_{r=m}^{i}\underline{\lambda_r}}{\lambda_i}\Big/(\cdots)=$
✓ 5.8	3	(5.11)	$\frac{D_{31}^{31}B_{11}}{\lambda_1+k-\Delta_{31}}\left\{e^{-\Delta_{31}(t-z/v_1)}-e^{-(\lambda_1+k)\left(t-\frac{z}{\underline{v_2}}\right)}\right\}$	$\frac{D_{31}^{32}B_{11}}{\lambda_1+k-\Delta_{31}}\left\{e^{-\Delta_{31}\left(t-\frac{z}{v_1}\right)}-e^{-(\lambda_1+k)\left(t-\frac{z}{\underline{v_1}}\right)}\right\}$
✓ 5.8	10~11	(5.11)	$+\frac{\lambda_2}{v_2}h\left(t-\frac{z}{v_3}\right)\frac{e^{-\lambda_3 z/v_3}}{\Gamma_{\underline{32}}}\left[\frac{D_{23}^{\underline{31}}B_{21}}{\lambda_1+k-\Delta_{23}}\{\cdots\}\right.$ $\left.+\frac{D_{23}^{32}B_{11}}{\underline{\lambda_1}+k-\Delta_{23}}\{\cdots\}\right]$	$+\frac{\lambda_2}{v_2}h\left(t-\frac{z}{v_3}\right)\frac{e^{-\lambda_3 z/v_3}}{\Gamma_{\underline{23}}}\left[\frac{D_{23}^{\underline{32}}B_{21}}{\lambda_1+k-\Delta_{23}}\{\cdots\}\right.$ $\left.+\frac{D_{23}^{32}B_{22}}{\underline{\lambda_2}+k-\Delta_{23}}\{\cdots\}\right]$
✓ 5.17	4	(5.28)	$\frac{2}{\sqrt{\pi}}\int_{a/b}^{\infty}e^{-(\lambda^2+a^2/\lambda^2)}d\lambda=\cdots$	$\frac{2}{\sqrt{\pi}}\int_{0}^{a/b}e^{-(\lambda^2+a^2/\lambda^2)}d\lambda=\cdots$
✓ 5.24	15	(5.41)	$B_{ij}=\sum_{m=1}^{i}N_m^0\frac{1}{\lambda_i}\prod_{l=m}^{i}\lambda_l\Big/\prod_{\substack{\underline{l=1}\\l\neq j}}^{i}(\lambda_l-\lambda_j)$	$B_{ij}=\sum_{m=1}^{j}N_m^0\frac{1}{\lambda_i}\prod_{l=m}^{i}\lambda_l\Big/\prod_{\substack{\underline{l=m}\\l\neq j}}^{i}(\lambda_l-\lambda_j)$
✓ 5.16 ~ 5.17		Fig. 5.23 & Fig. 5.24		To be changed as attached figures.

page	line	Eq.	erratum	correct
✓ 6.3	12		… in each layer. Trans-	… in each layer. The trans-
✓ 6.4	6	(6.3)	${}^{l+1}S_f\left[-{}^{l+1}D\frac{\partial\,{}^{l+1}N_i}{\partial z}+{}^{l+1}v\,{}^{l+1}N_i\right]_{z={}^{l+1}\underline{z}}$	${}^{l+1}S_f\left[-{}^{l+1}D\frac{\partial\,{}^{l+1}N_i}{\partial z}+{}^{l+1}v\,{}^{l+1}N_i\right]_{z={}^{l}\underline{z}}$
✓ 6.6	14		use the general solution (4.$\underline{117}$)…	use the general solution (4.$\underline{127}$)…
✓ 6.6	17	(6.19)	${}^{l}\underline{N_i}(z,t;{}^{l}\Phi_i(t))$ $\equiv\cdots+\cdots\sum_{\substack{r=j\\ r\neq m}}^{i}{}^{l}D^{ij}_{r,m}\;{}^{l}\underline{g_{r,m}(t)}\otimes{}^{l}\phi_j(t)$	${}^{l}\underline{\mathcal{N}_i}(z,t;{}^{l}\Phi_i(t))$ $\equiv\cdots+\cdots\sum_{\substack{r=j\\ r\neq m}}^{i}{}^{l}D^{ij}_{r,m}\;{}^{l}\underline{g_{r,m}(z,t)}\otimes{}^{l}\phi_j(t)$
✓ 6.8		(6.23)		To be changed as the attached equation
✓ 6.9	1		${}^{1}\underline{g_{r,m}(t')}\otimes{}^{1}\phi_j(t')\Big]_{\underline{t'=t-(z-z_1)/{}^{2}v_i}}$	${}^{1}\underline{g_{r,m}({}^{1}z,t')}\otimes{}^{1}\phi_j(t')\Big]_{\underline{t'=t-(z-{}^{1}z)/{}^{2}v_i}}$
✓ 6.9	3		$t'=t-(z-\underline{z_1})/{}^{2}v_i$	$t'=t-(z-{}^{1}\underline{z})/{}^{2}v_i$
✓ 6.10	4	(6.27)	$\underline{z_1}$ in the equation	To be changed from z_1 to ${}^{1}\underline{z}$
✓ 6.10	6	(6.29)	〃	〃
✓ 6.10	11		By substituting $i=1,2,3,$ into ($\underline{6}$.13)	By substituting $i=1,2,3,$ into ($\underline{5}$.13)
✓ 6.10	14	(6.30)	$;\ 0\leq z\leq \underline{z_1}$	$;\ 0\leq z\leq {}^{1}\underline{z}$
✓ 6.11		(6.31), (6.32) (6.33), (6.34) (6.35)	$\underline{z_1}$ in these equations	To be changed from z_1 to ${}^{1}\underline{z}$
✓ 6.13	1	(6.38)	$\mathcal{B}^{S}_{2}(z,t)\equiv\cdots[\cdots-{}^{2}F'_1(z,t,\underline{\lambda_3})]+$ $+\cdots$	$\mathcal{B}^{S}_{2}(z,t)\equiv\cdots+[\cdots-{}^{2}F'_1(z,t,\underline{\lambda_1})]+$ $+\cdots$
✓ 6.14	6	(6.40)	${}^{2}\mathcal{C}^{S}_{j}(z,t)=\cdots+$ $+\cdots-$ $-\cdots[M^{2}_{32}(z,t,{}^{1}\Delta_{31})+M^{2}_{32}(z,t,\underline{\lambda_3})]-$ $-\cdots$	${}^{2}\mathcal{C}^{S}_{3}(z,t)=\cdots+$ $+\cdots-$ $-\cdots[M^{2}_{32}(z,t,{}^{1}\Delta_{31})+M^{2}_{32}(z,t,\underline{\lambda_1})]-$ $-\cdots$
✓ 6.15	5	(6.47)	$\underline{z_1}$ in the equation	To be changed from z_1 to ${}^{1}\underline{z}$.
✓ 6.16	6	(6.56)	$M^{\beta}_{rm}(z,t,{}^{1}\Delta_{21})$ $=\frac{(\cdots)}{(\cdots)}$ $[\cdots]$	$M^{\beta}_{rm}(z,t,{}^{1}\Delta_{21})$ $=\frac{(\cdots)}{(\cdots)}\underline{\times}$ $\underline{\times}\,[\cdots]$

page	line	Eq.	erratum	correct
✓6.19	7,10		${}^LN_j(z,t,\xi,\tau),\ {}^LM_i(z,t,\xi,\tau)$	${}^Ln_i(z,t,\xi,\tau),\ {}^Lm_i(z,t,\xi,\tau)$
✓6.19	12		${}^RN_i$ and ${}^RM_i$	${}^Rn_i$ and ${}^Rm_i$
✓6.19	17		order inone variable, z, ...	order in one variable, z, ...
✓6.20	1,2,3,4		${}^L\tilde{N}_i,\ {}^L\tilde{M}_i,\ {}^R\tilde{N}_i,\ {}^R\tilde{M}_i,\ {}^LN_i,\ {}^LM_i$ ${}^RN_i,\ {}^RM_i$	${}^L\tilde{n}_i,\ {}^L\tilde{m}_i,\ {}^R\tilde{n}_i,\ {}^R\tilde{m}_i,\ {}^Ln_i,\ {}^Lm_i$ ${}^Rn_i,\ {}^Rm_i$
✓6.22	3	(6.77)	${}^1f_i(\xi,\tau)\equiv\cdots+\phi_i(\tau)\delta(\xi-z_1);\ \xi<0$	${}^1f_i(\xi,\tau)\equiv\cdots+\frac{\phi_i(\tau)}{v}\delta(\xi-{}^1z);\ \xi<$
✓6.22	8	(6.81)	${}^{\ell k}P_s^i(z,t)\equiv\int\cdots\cdot$ $\cdot\{\cdots+\sqrt{\frac{{}^\ell v}{{}^\ell d}}\,\frac{1}{2}e^{-\lambda_i v}\operatorname{erf}\left(\frac{1}{2}\sqrt{v\,{}^kv/{}^kd}\right)\}dv$	${}^{\ell k}P_s^i(z,t)\equiv\int\cdots\cdot$ $\cdot\{\cdots+\sqrt{\frac{{}^kv}{{}^kd}}\,\frac{1}{2}e^{-\lambda_i v}\operatorname{erf}\left(\frac{1}{2}\sqrt{\frac{v\,{}^kv}{{}^kd}}\right)\}dv$
✓6.23	3	(6.83)	${}^{\ell k}P_s^i(x,y,t)\equiv\int_0^t\left[\frac{x}{2(t-v)\,{}^\ell v\,{}^\ell d}-1\right]\cdots$	${}^{\ell k}P_s^i(x,y,t)\equiv\int_0^t\left[\frac{x^2}{2(t-v)\,{}^\ell v\,{}^\ell d}-1\right]\cdots$
✓6.24	4	(6.86)	$G_-\equiv\frac{{}^2d\,{}^1v-{}^1d\,{}^2v}{{}^1v-{}^2v}$	$G_-\equiv\frac{{}^2d\,{}^1v-{}^1d\,{}^2v}{{}^1v\cdot{}^2v}$
✓8.2	7		$D_{1m}^{ij}=[\cdots]$	$D_{rm}^{ij}\equiv D_{rm}^{(j)}=[\cdots]$
✓8.3	1		$g_{rm}(t):\cdots$	$g_{rm}(z,t)\equiv g_{rm}(t):\cdots$
✓8.4	19			Insert: ${}^\ell\mathcal{N}_i(z,t;\ {}^\ell\Phi_i(t))$: function defined by (6.19)
✓8.7	7		$\sqrt{{}^\ell d_i/\rho v_i}$	$\sqrt{{}^\ell d_i/{}^\ell v_i}$
✓8.7	20			Insert: ${}^\ell\Phi_i(t)$: represents the functions ${}^\ell\phi_1(t),\ {}^\ell\phi_2(t),\ \ldots,\ {}^\ell\phi_i(t)$ ${}^\ell\phi_i(t)$: functions defined by (6.17) or (6.58)

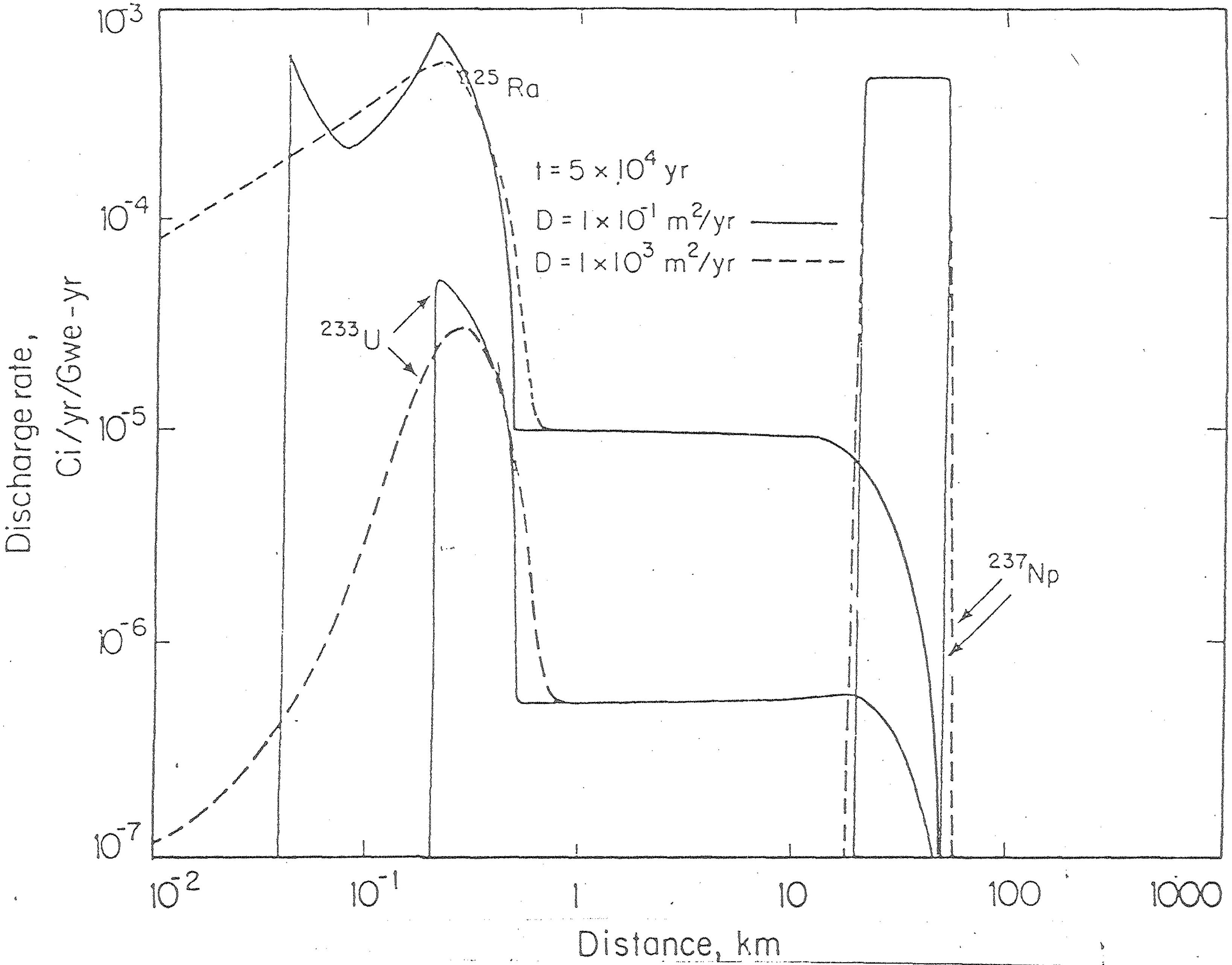

Figure 5.23 – Concentration profiles of ^{225}Ra of $^{237}Np \rightarrow ^{233}P_a \rightarrow ^{233}U \rightarrow ^{229}Th \rightarrow ^{225}Ra$ decay chain at t = 5x10^{4}yr. (Pure ^{237}Np source at t=0. V=100m/yr. T=3x10^4, $K_{Np}=10^2$. $K_{Pa}=1x10^4$. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

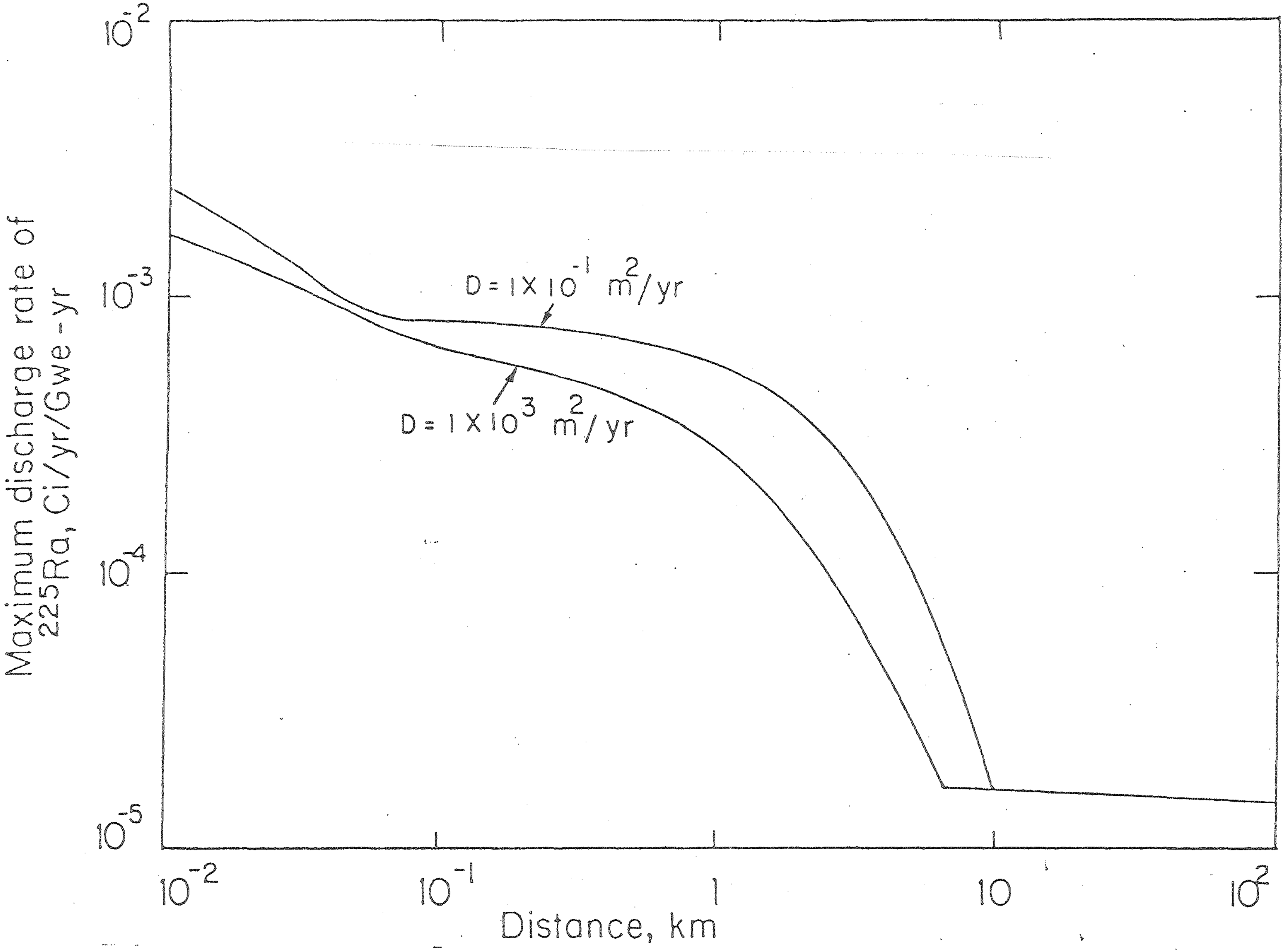

Figure 5.24 - Effect of dispersion coefficient on maximum discharge rate of ^{225}Ra for the $^{237}\text{Np}\rightarrow{}^{233}\text{Pa}\rightarrow{}^{233}\text{U}\rightarrow{}^{229}\text{Th}\rightarrow{}^{225}\text{Ra}$ decay chain. (Pure ^{237}Np source at t=0. V=100m/yr. T=3x10^{4}yr. $K_{Np}=10^2$. $K_{Pa}=1x10^4$. $K_U=1x10^4$. $K_{Th}=5x10^4$. $K_{Ra}=5x10^2$).

previous results and (6.21), and so on up to ${}^{\ell}N_i(z,t)$.

6.4 Application of Recursive Solution to a Two Media System

6.4.1 General release mode at the repository: ${}^{1}\phi_i(t)$

We have:

$$ {}^{1}N_i(z,t) = {}^{1}\mathcal{N}_i(z,t\,;\,{}^{1}\Phi_i(t)) \tag{6.22} $$

and (6.21) yields:

$$
\begin{aligned}
{}^{2}N_i(z,t) = {} & e^{-\lambda_i(z-{}^{1}z)/{}^{2}v_i}\cdot\frac{{}^{2}\rho}{{}^{1}\rho}\Bigg\{ e^{-\lambda_i{}^{1}z/{}^{1}v_i}\,{}^{1}\phi_i\Big(t-\frac{z-{}^{1}z}{{}^{2}v_i}-\frac{{}^{1}z}{{}^{1}v_i}\Big) + \\
& + \sum_{j=1}^{i-1}{}^{1}A_i^{(j)}\sum_{m=j}^{i}\frac{1}{{}^{1}B_m^{ij}}\,e^{-\lambda_m{}^{1}z/{}^{1}v_m}\cdot\sum_{\substack{r=j\\(r\neq m)}}^{i}{}^{1}D_{r,m}^{i,j}\cdot \\
& \cdot\Big[{}^{1}g_{r,m}({}^{1}z,t')\otimes{}^{1}\phi_j(t')\Big]_{t'=t-(z-{}^{1}z)/{}^{2}v_i}\Bigg\} + \\
& + \sum_{j=1}^{i-1}{}^{2}A_i^{(j)}\sum_{m=j}^{i}\frac{1}{{}^{2}B_m^{ij}}\,e^{-\lambda_m(z-{}^{1}z)/{}^{2}v_m}\cdot\sum_{\substack{r=j\\r\neq m}}^{i}{}^{2}D_{r,m}^{ij}\;{}^{2}g_{r,m}(z-{}^{1}z,t)\otimes \\
& \otimes\Bigg\{\frac{{}^{2}\rho}{{}^{1}\rho}\cdot e^{-\lambda_j{}^{1}z/{}^{1}v_j}\,{}^{1}\phi_j(t-{}^{1}z/{}^{1}v_j) + \frac{{}^{2}\rho}{{}^{1}\rho}\sum_{\alpha=1}^{j-1}{}^{1}A_j^{(\alpha)}\cdot\sum_{\beta=\alpha}^{j}\frac{1}{{}^{1}B_\beta^{j,\alpha}}\cdot \\
& \cdot e^{-\lambda_\beta{}^{1}z/{}^{1}v_\beta}\cdot\sum_{\substack{\omega=\alpha\\\omega\neq\beta}}^{j}{}^{1}D_{\omega,\beta}^{j,\alpha}\;{}^{1}g_{\omega,\beta}({}^{1}z,t)\otimes{}^{1}\phi_\alpha(t)\Bigg\}
\end{aligned}
\tag{6.23}
$$

ERRATA