LBL-11616
UC-70
Volume II

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA, BERKELEY

EARTH SCIENCES DIVISION

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Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

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UCB-NE-4003
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Earth Sciences Division, Lawrence Berkeley Laboratory
and
Department of Nuclear Engineering, University of California
Berkeley, California 94720

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*Prepared for the U. S. Department of Energy under contract W-7405-ENG-48 Administered by the Office of Nuclear Waste Isolation, Battelle Memorial Institute under MPO E511-05200.

The authors invite comments and would appreciate being notified of any errors in the report.

T. H. Pigford
Department of Nuclear Engineering
University of California
Berkeley, CA 94720

REPORT OUTLINE

MIGRATION OF RADIONUCLIDES THROUGH SORBING MEDIA, ANALYTICAL SOLUTIONS--II

4. Effect of Transverse Dispersion on Transport in a One-Dimensional Flow Field with Transverse Dispersion

4.1 Transport of Radionuclides Released from a Point Source

Our previous analyses [Sects. 2,3, and (H1)] have been concerned with the migration of radionuclides released from an infinite plane source into a one-dimensional flow field. In this geometry there is no net effect of diffusion and dispersion of radionuclides in directions normal to the direction of water flow, so transverse dispersion was omitted from the differential equations to be solved. However, for a radionuclide source of finite extent the effects of transverse dispersion must be considered.

Here we first consider the time-space-dependent concentration of a radionuclide emitted from a generalized source into a porous medium, with dispersion occurring in any direction. The transport equation is then simplied for the case of local sorption equilibrium and for a point source of dissolving radionuclides. A solution is then developed for the space-time-dependent concentration of the i^{th} member of a radionuclide decay chain in an infinite porous medium in which uniform one-dimensional flow of groundwater occurs.

In Sect. 4.2 the analyses are generalized to apply to any member of a radionuclide decay chain, resulting from dissolution of a plane source of finite extent. Numerical examples are given in Sect. 4.3.

4.1.1 The transport equation

The general equation governing the transport of nuclide $\underline{i}$ through the porous medium is

$$\frac{\partial}{\partial t}[\epsilon N_{i\ell}+(1-\epsilon)N_{is}]=\overset{\Rightarrow}{\nabla}\cdot\overset{\Rightarrow}{D}_{\ell}\cdot\vec{\nabla}(\epsilon N_{i\ell})+\vec{\nabla}\cdot\overset{\Rightarrow}{D}_{s}\cdot\vec{\nabla}[(1-\epsilon)N_{is}]-\vec{\nabla}\cdot(\epsilon\vec{v}N_{i\ell})$$

$$-\lambda_i[\epsilon N_{i\ell}+(1-\epsilon)N_{is}]+\lambda_{i-1}[\epsilon N_{i-1,\ell}+(1-\epsilon)N_{i-1,s}]$$

$$+\epsilon S_{i\ell}+(1-\epsilon)S_{is} \qquad (4.1.1)$$

where N_{il} and N_{is} are the concentrations of the i-th nuclide in liquid and solid phases, respectively. The first two terms on the right-hand side represent dispersion in both phases, with dispersion-coefficient tensors $\overset{\Rightarrow}{D}_1$ and $\overset{\Rightarrow}{D}_s$ in liquid and solid phases. The dispersion coefficient in a given phase is assumed to be the same for all radionuclides.

To simplify, we make three basic assumptions:

(a) There is local chemical equilibrium between radionuclides dissolved in the ground water and radionuclides sorbed on the solid, so that the concentrations in liquid and solid phases are related by a linear equilibrium:

$$N_{is} = K_{Di}\, N_{il} \tag{4.1.2}$$

where K_{Di} is the sorption equilibrium constant for radionuclide i. The retardation coefficient K_i is defined as

$$K_i = 1 + \frac{1-\varepsilon}{\varepsilon} K_{Di} \; . \tag{4.1.3}$$

(b) The values of porosity ε, flow velocity $\vec{v}$, dispersion-coefficient tensors $\vec{\vec{D}}_l$ and $\vec{\vec{D}}_s$, and sorption constants K_{Di} and K_i are constant in space and time.

(c) We can choose a system of coordinates (x,y,z,t) so as to satisfy both of the following conditions:

c.1) The three axes of this system are the principal axes of an overall dispersion tensor, including contribution from both phases, and defined as:

$$\vec{\vec{D}} = \vec{\vec{D}}_l + \frac{1-\varepsilon}{\varepsilon} K_{Di} \vec{\vec{D}}_S \; . \tag{4.1.4}$$

Thus, the tensor $\vec{\vec{D}}$ will include only three dispersion coefficients D_x, D_y, D_z, along the three axes. If diffusion in the solid phase is negligible, $\vec{\vec{D}}$ reduced to $\vec{\vec{D}}_l$.

c.2) The direction of the flow velocity $\vec{v}$, which is constant according to assumption (b), is the direction of one of the axes, e.g., the z axis. Thus

$$\vec{v} = v \cdot \vec{z} \tag{4.1.5}$$

where $\vec{z}$ is the unit vector in the z direction.

Using assumptions (a), (b), and (c), Equation (4.1.1) becomes:

$$\varepsilon K_i \frac{\partial N_{i\ell}}{\partial t} = \varepsilon \left[D_x \frac{\partial^2 N_{i\ell}}{\partial x^2} + D_y \frac{\partial^2 N_{i\ell}}{\partial y^2} + D_z \frac{\partial^2 N_{i\ell}}{\partial z^2} \right] - \varepsilon v \frac{\partial N_{i\ell}}{\partial z}$$

$$- \lambda_i \varepsilon K_i N_{il} + \lambda_{i-1} \varepsilon K_{i-1} N_{i-1,l} + \varepsilon S_i \tag{4.1.6}$$

S_i is a volumetric source term (atoms/m^3 sec) defined as:

$$S_i = S_{il} + \frac{1-\varepsilon}{\varepsilon} S_{is} . \tag{4.1.7}$$

Here again, if the solid-phase source term S_{is} is negligible, S_i reduces to S_{il}.

The change of variables:

$$\tilde{x} = x\sqrt{D_z/D_x} \tag{4.1.8}$$

$$\tilde{y} = y\sqrt{D_z/D_y} \tag{4.1.9}$$

is such that:

$$D_x \frac{\partial^2 N_{il}}{\partial x^2} = D_x \frac{\partial^2 N_{il}}{\partial(\tilde{x}\sqrt{D_x/D_z})^2} = D_z \frac{\partial^2 N_{il}}{\partial \tilde{x}^2} \tag{4.1.10}$$

and

$$D_y \frac{\partial^2 N_{il}}{\partial y^2} = D_z \frac{\partial^2 N_{il}}{\partial \tilde{y}^2} . \tag{4.1.11}$$

The transport equation written in the transposed coordinates $(\tilde{x},\tilde{y},\tilde{z})$, becomes:

$$\varepsilon K_i \frac{\partial N_i}{\partial t} = \varepsilon D_z \left(\frac{\partial^2 N_i}{\partial \tilde{z}^2} + \frac{\partial^2 N_i}{\partial \tilde{y}^2} + \frac{\partial^2 N_i}{\partial \tilde{z}^2}\right) - \varepsilon v \frac{\partial N_i}{} \tag{4.1.12}$$

$$- \lambda_i \varepsilon K_i N_i + \lambda_{i-1} \varepsilon K_{i-1} N_{i-1} + \varepsilon S_i$$

where $\tilde{z} = z$ and $N_i = N_{il}$.

We introduce the following notations:

$$l = \frac{D_z}{v} \tag{4.1.13}$$

where l is a characteristic length of the dispersion process, and represents the ratio of the magnitudes of dispersion and convection, and

$$v_i = \frac{v}{K_i} \tag{4.1.14}$$

where v_i is the convective velocity for a chromatographic band for the ith nuclide, taking sorption into account, as has been previously defined in the one-dimensional models.

Using (4.1.12), (4.1.13), (4.1.14), dividing by K_i, and denoting the liquid phase concentration by N_i for simplicity, Eq. (4.1.6) can be written:

$$\frac{\partial N_i}{\partial t} = lv_i\left(\frac{\partial^2 N_i}{\partial \tilde{x}^2} + \frac{\partial^2 N_i}{\partial \tilde{y}^2} + \frac{\partial^2 N_i}{\partial \tilde{z}^2}\right) - v_i \frac{\partial N_i}{\partial \tilde{z}} -\lambda_i N_i + \lambda_{i-1} \frac{v_i}{v_{i-1}} N_{i-1} + \frac{S_i}{K_i} \quad . \tag{4.1.15}$$

This is the basic transport equation to be used in the present study. Considering that both of the last two terms are sources of the i-th nuclide, Eq. (2.6.15) can alternatively be written as:

$$\frac{\partial N_i}{\partial t} = lv_i\left(\frac{\partial^2 N_i}{\partial \tilde{x}^2} + \frac{\partial^2 N_i}{\partial \tilde{y}^2} + \frac{\partial^2 N_i}{\partial \tilde{z}^2}\right) - v_i \frac{\partial N_i}{\partial \tilde{z}} - \lambda_i N_i + \$_i(\tilde{x},\tilde{y},\tilde{z},t) \tag{4.1.16}$$

where $\$_i(\tilde{x},\tilde{y},\tilde{z},t)$ is a general source term, defined as:

$$\$_i(\tilde{x},\tilde{y},\tilde{z},t) \equiv - \lambda_i \frac{v_i}{v_{i-1}} \cdot N_{i-1}(\tilde{x},\tilde{y},\tilde{z},t) + \frac{S_i(\tilde{x},\tilde{y},\tilde{z},t)}{K_i} \tag{4.1.17}$$

Equation (4.1.15) gives $N_i(\tilde{x},\tilde{y},\tilde{z},t)$ for

$$0 < t < +\infty, \quad -\infty < \tilde{x} < +\infty, \quad -\infty < \tilde{y} < +\infty, \quad -\infty < \tilde{z} < +\infty \; .$$

4.1.2. Initial and boundary conditions

The initial condition is:

$$N_i(\tilde{x},\tilde{y},\tilde{z},0) = 0 \text{ for any } \tilde{x},\tilde{y},\tilde{z} \quad . \tag{4.1.18}$$

The boundary conditions are:

$$N_i(\pm\infty,\tilde{y},\tilde{z},t) = N_i(\tilde{x},\pm\infty,\tilde{z},t) = N_i(\tilde{x},\tilde{y},\pm\infty,t) = 0 \tag{4.1.19}$$

for an infinite medium. When bounded media are considered, appropriate boundary conditions will be specified.

4.1.3 Source terms and release modes

For a discrete point source of radionuclides, releasing ${}^{pt}S_i$ atoms/sec of nuclide i into the ground water at x=y=z=0, the volumetric source term is:

$$S_i = {}^{pt}S_i(t)\cdot\delta(\tilde{x})\cdot\delta(\tilde{y})\cdot\delta(\tilde{z}) \tag{4.1.20}$$

The point-source condition does not limit the applications of the solutions. It is likely that releases from wastes into ground water can be represented by a collection of point sources, each corresponding to the release from a waste canister. Furthermore, even if the sources are assumed to be continuously distributed throughout a repository, the distribution can be treated as a collection of point sources, i.e., the point-source solution can be integrated over the source domain (cf. Appendix D.).

The nature of the function ${}^{pt}S_i(t)$ depends upon the release mode. We consider three of the modes defined in our earlier report (H1):

(a) Impulse release: a quantity Q_i of the i-th nuclide is released instantaneously at time $t = 0$. The source function is then:

$${}^{pt}S_i(t) = Q_i\ \delta(t) \tag{4.1.21}$$

(b) Band release: an initial amount W_0 of total waste material is released at a constant rate W_0/T over a leach time T. The undissolved waste material contains an atom fraction $n_i(t)$ of nuclide i, so that:

$${}^{pt}S_i(t) = \frac{W_o}{T}\,n_i(t)\,[h(t) - h(t-T)] \tag{4.1.22}$$

where h(t) is the Heaviside step function:

$$\begin{aligned} h(t) &= 0, \quad t < 0 \\ h(t) &= 1, \quad t > 0\ . \end{aligned} \tag{4.1.23}$$

If nuclide i is formed by radioactive decay of precursors in the waste, the atom fraction $n_i(t)$ is given by the Bateman equation (B1) [cf. Eqs. (2.1.12), (2.1.13), (2.1.14), and (2.1.15)]. The source term in the liquid then becomes:

$${}^{pt}S_i(t) = \frac{W_o}{T}\ \sum_{j=1}^{i} B_{ij}e^{-\lambda_j t}\ [h(t)-h(t-T)]\ . \tag{4.1.24}$$

(c) Step release: In this case waste material dissolves at a constant total rate W_0/T, as for a band release, but the dissolution continues at this rate without regard to limitation by the finite amount of waste material, so that:

$$^{pt}S_i(t) = \frac{W_0}{T}\left\{\sum_{j=1}^{i} B_{ij}e^{-\lambda_j t}\right\} \quad h(t) \; . \tag{4.1.25}$$

4.1.4. Integrated conservation equation

The total amount $m_i(t)$ of the i-th nuclide present in all the domain β of interest is:

$$m_i(t) = \iiint_\beta N_i(x,y,z,t)dxdydz. \tag{4.1.26}$$

If β is unbounded, or if the flux of radionuclides transported across the boundaries is zero, $m_i(t)$ verifies the following conservation equation:

$$\frac{dm_i}{dt} = - \lambda_i m_i + \left\{\int\int\int_\beta \mathcal{S}_i(\tilde{x},\tilde{y},\tilde{z},t)d\tilde{x}d\tilde{y}d\tilde{z}\right\}. \tag{4.1.27}$$

The solution of this equation is:

$$m_i(t) = \int_0^t e^{-\lambda_i\ (t-\theta)} \left\{\iiint_\beta \mathcal{S}_i(\tilde{x},\tilde{y},\tilde{z},\theta)d\tilde{x}d\tilde{y}d\tilde{z} \quad d\theta\right\}. \tag{4.1.28}$$

Thus, the integrated conservation equation for the i-th nuclide is:

$$\iiint_\beta N_i(\tilde{x},\tilde{y},\tilde{z},t)d\tilde{x}d\tilde{y}d\tilde{z} =$$

$$\int_0^t d\theta \iiint_\beta \quad e^{-\lambda_i(t-\theta)} \quad \mathcal{S}_i(\tilde{x},\tilde{y},\tilde{z},\theta)d\tilde{x}d\tilde{y}d\tilde{z} \; .$$

4.1.5. Concentrations for an impulse point-source release

We now present analytical solutions to the transport equation (4.1.15) corresponding to the different source terms previously discussed. For the impulse point-source release of a single nuclide $^{pt}S_i(t)$ is given by Eq. (4.1.21). We suppose that nuclide i has no precursors. The solution of this classical problem can be derived formally by transform calculus, using Laplace transforms for the time coordinate and exponential Fourier transforms on the space coordinates (C1):

$$N_i(\tilde{x},\tilde{y},\tilde{z},t) = Q_i\ \epsilon^{-\lambda_i t}\ G(\tilde{x}, l v_i t) \cdot G(\tilde{y}, l v_i t) \cdot G(\tilde{z}-v_i t, l v_i t) \tag{4.1.30}$$

where G is the Gaussian function, defined by:

$$G(\alpha,\mu) = \frac{e^{-\frac{\alpha^2}{4\nu}}}{\sqrt{4\pi\ \nu}} \quad (\nu > 0)\ . \tag{4.1.31}$$

If the impulse release occurs at location (u, v, w) and time θ, i.e., if:

$$S_i(\tilde{x},\tilde{y},\tilde{z},t) = Q_i \cdot \delta(\tilde{x}-u)\ \delta(\tilde{y}-v) \cdot \delta(\tilde{z}-w)\ \delta(t-\theta) \tag{4.1.32}$$

then the concentration at $\tilde{x},\tilde{y},\tilde{z}$ due to this impulse is

$$\begin{aligned} N_i(\tilde{x},\tilde{y},\tilde{z},t) = Q_i\ e^{-\lambda_i(t-\theta)}\ & G[\tilde{x}-u, l v_i(t-\theta)] \cdot \\ & G[\tilde{y}-v, l v_i(t-\theta)]\ G[\tilde{z}-w-v_i(t-\theta),\ l v_i(t-\theta)] \end{aligned} \tag{4.1.33}$$

4.1.6. Concentrations for a general point-source release at $\tilde{x} = \tilde{y} = \tilde{z} = 0$

To determine the actual concentration of the i-th member of a chain, we must consider the transport Eq. (4.1.15), with a source term given by Eq. (4.1.20),

$$\begin{aligned} \frac{\partial N_i}{\partial t} = l v_i \left(\frac{\partial^2 N_i}{\partial \tilde{x}^2} + \frac{\partial^2 N_i}{\partial \tilde{y}^2} + \frac{\partial^2 N_i}{\partial \tilde{z}^2}\right) - v_i \frac{\partial\ N_i}{\partial \tilde{z}} - \lambda_i N_i \\ + \lambda_{i-1} \frac{v_i}{v_{i-1}}\ N_{i-1} + \frac{1}{K_i}\ S_i(t)\ \delta(\tilde{x})\delta(\tilde{y})\delta(\tilde{z})\ . \end{aligned} \tag{4.1.34}$$

To find N_i, we can use the previous solution for impulse release, Eq. (4.1.33), as a Green's function. The two source terms in Eq. (4.1.34) are considered as sums of impulse functions, i.e., impulse releases:

$$\begin{aligned} \lambda_{1-1} \frac{v_i}{v_{i-1}} N_{i-1}(\tilde{x},\tilde{y},\tilde{z},t) = \lambda_{i-1} \frac{v_i}{v_{i-1}} \cdot \\ \int_0^t d\theta \int_{-\infty}^{+\infty} du \int_{-\infty}^{+\infty} dv \int_{-\infty}^{+\infty} \Big\{ N_{i-1}(u,v,w,\theta) \cdot \delta(\tilde{x}-u) \cdot \delta(\tilde{y}-v)\ \cdot \\ \delta(\tilde{z}-w) \cdot \delta(t-\theta) \Big\}\ dw \end{aligned} \tag{4.1.35}$$

and

$$\frac{1}{K_i}\ {}^{pt}S_i(t)\ \ \delta(\tilde{x})\ \ \delta(\tilde{y})\ \ \delta(\tilde{z})$$

$$= \frac{1}{K_i}\ \delta(\tilde{x})\ \ \delta(\tilde{y})\ \ \delta(\tilde{z})\int_0^t {}^{pt}S_i(\theta)\ \ \delta(t-\theta)d\theta\ . \qquad (4.1.36)$$

The solution for each impulse release is the Green's function given by Eq. (4.1.33). The complete solution is obtained by superimposing all these solutions for all impulse releases from both source terms. It can be written in forms of two contributions:

$$N_i(\tilde{x},\tilde{y},\tilde{z},t) = N_i^R(\tilde{x},\tilde{y},\tilde{z},t) + N_i^P(\tilde{x},\tilde{y},\tilde{z},t) \qquad (4.1.37)$$

where N_i^R is the concentration of i directly released from the waste:

$$N_i^R\ (\tilde{x},\tilde{y},\tilde{z},t)$$

$$= \int_0^t \frac{{}^{pt}S_i(\theta)}{K_i}\, e^{-\lambda_i(t-\theta)}\ \ G[\tilde{x},lv_i(t-\theta)]\ \ G[\tilde{y},lv_i(t-\theta)]\ \cdot \qquad (4.1.38)$$

$$G[\tilde{z}-v_i(t-\theta),\ lv_i(t-\theta)]d\theta$$

and N_i^P is the concentration of i formed by decay of its precursors previously released from the waste:

$$N_i^P(\tilde{z},\tilde{y},\tilde{z},t) =$$

$$\int_0^t d\theta \int_{-\infty}^{+\infty} du \int_{-\infty}^{+\infty} dv \int_{-\infty}^{+\infty} dw \left\{ \lambda_{i-1}\ \ \frac{v_i}{v_{i-1}}\ N_{i-1}(u,v,w,\theta) \right\} \cdot \qquad (4.1.39)$$

$$e^{-\lambda_i(t-\theta)} \cdot G[\tilde{x}-u,lv_i(t-\theta)]\ \cdot$$

$$G[\tilde{y}-v,lv_i(t-\theta)]\ \cdot G[\tilde{z}-w-v_i(t-\theta),lv_i(t-\theta)]\ .$$

The study of the integrals in N_i^R and N_i^P and the validity of the solution they define are considered in (A1). We simply assume here that they exist for any $t > 0$ and $(\tilde{x},\tilde{y},\tilde{z})$ such that $\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2 \neq 0$, and define a solution to Eq. (4.1.34) with the boundary conditions of Sect. 4.1.2.

We will deal separately with the terms N_i^R and N_i^P in the following section on step release.

4.1.7 Concentrations for a step release from a point source

To evaluate the term N_i^R resulting from a step release, we must replace ${}^{pt}S_i(t)$ in Eq. (4.1.38) by the corresponding expression, given by Eq. (4.1.25). Using the definition of the function G from Eq. (4.1.31), there results:

$$N_i^R = \frac{W_o}{TK_i} \sum_{j=1}^{i} B_{ij}\, e^{\frac{z}{2l} - \lambda_j t} \int_0^t \frac{e^{-\left\{\left[\frac{v_i}{4l} + (\lambda_i - \lambda_j)\right](t-\theta) + \frac{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}{4lv_i(t-\theta)}\right\}}}{[4\pi l v_i(t-\theta)]^{3/2}}\, d\theta \,. \tag{4.1.40}$$

The change of variable: $\tau \equiv lv_i(t-\theta)$ transforms this expression into:

$$N_i^R = \frac{W_o}{TK_i l v_i} \sum_{j=1}^{i} B_{ij}\, e^{-\frac{z}{2l} - \lambda_j t} \int_0^{lv_i t} \frac{e^{-\left\{\frac{\tau}{4l^2}\left[1 + \frac{4l(\lambda_i - \lambda_j)}{v_i}\right] + \frac{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}{4\tau}\right\}}}{(4\pi\tau)^{3/2}}\, d\tau \,. \tag{4.1.41}$$

We will use the following notations (H1):

$$\left.\begin{aligned} \Lambda_{ij} &= \frac{\lambda_i}{v_i} &&, \ j = 0 \\ \Lambda_{ij} &= \frac{\lambda_i}{v_i} - \frac{\lambda_j}{v_j} &&, \ j \neq 0 \end{aligned}\right\} \tag{4.1.42}$$

$$\left.\begin{aligned} \Gamma_{ij} &= \frac{1}{v_i} &&, \ j = 0 \\ \Gamma_{ij} &= \frac{1}{v_i} - \frac{1}{v_j} &&, \ j \neq 0 \end{aligned}\right\} \tag{4.1.43}$$

$$\left.\begin{aligned} \beta_{ij} &= \lambda_i \qquad , \; j = 0 \\ \beta_{ij} &= \frac{\Lambda_{ij}}{\Gamma_{ij}} \qquad , \; j \neq 0 \end{aligned}\right\} \tag{4.1.44}$$

$$\gamma_{ijk} = 1 + 4l\,\frac{\lambda_k - \beta_{ij}}{v_k} \tag{4.1.45}$$

Thus, using $K_i v_i = v$, Eq. (4.1.42) becomes:

$$N_i^R = \frac{W_o}{vTl} \sum_{j=1}^{i} B_{ij}\, e^{-\frac{z}{2l} - \beta_{jo} t} \int_0^{lv_i t} \frac{e^{-\left\{\frac{\gamma_{joi}}{4l^2}\tau + \frac{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}{4\tau}\right\}}}{(4\pi\tau)^{3/2}}\, d\tau \; . \tag{4.1.46}$$

This form suggests the use of two functions F and E defined by:

$$F(U,a,b) = l \int_0^U \frac{e^{-\left\{a^2\tau + \frac{b^2}{\tau}\right\}}}{(4\pi\tau)^{3/2}}\, d\tau \tag{4.1.47}$$

$$E(I,J,K,\tilde{x},\tilde{y},\tilde{z},t) = e^{\frac{z}{2l} - \beta_{IJ} t} \cdot F\left(lv_K t, \frac{\sqrt{\gamma_{IJK}}}{2l} \; . \; \frac{\sqrt{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}}{2}\right) . \tag{4.1.48}$$

When no confusion is possible, we will use the notation E (I, J, K) = E (I, J, K, $\tilde{x}$, $\tilde{y}$, $\tilde{z}$, t). F can be determined by performing the integral of Eq. (4.1.47):

$$F(U,a,b) = \frac{1}{16\pi b} \cdot \left\{ e^{2ab}\, \mathrm{erfc}\left(a\sqrt{U} + \frac{b}{\sqrt{U}}\right) + e^{-2ab}\, \mathrm{erfc}\left(-a\sqrt{U} + \frac{b}{\sqrt{U}}\right) \right\} \tag{4.1.49}$$

where "erfc" represents the complementary error function. Thus, N_i^R can finally be written:

$$N_i^R = \frac{W_o}{vT} \frac{1}{l^2} \sum_{j=1}^{i} B_{ij} e^{\frac{z}{2l} - \beta_{jo} t} \cdot F\left(lv_i t, \frac{\sqrt{\gamma_{joi}}}{2l}, \frac{\sqrt{\tilde{x}^2+\tilde{y}^2+\tilde{z}^2}}{2}\right) \qquad (4.1.50)$$

or

$$N_i^R = \frac{W_o}{vt} \frac{1}{l^2} \sum_{j=1}^{i} B_{ij} \, E\,(j,o,i) \, . \qquad (4.1.51)$$

It can be noted that the functions F and N_i^R are not defined for $\tilde{x} = \tilde{y} = \tilde{z} = 0$, since the form:

$$\int_0^{lv_i t} \frac{e^{-\frac{\gamma}{4l^2}\tau}}{(4\pi\tau)^{3/2}} \, d\tau$$

does not then exist. This results from the fact that the solution for a point-source release is infinite for t = 0, at the source location $\tilde{x} = \tilde{y} = \tilde{z} = 0$.

These same functions E(I,J,K) and F appear in the contribution N_i^P due to precursors previously leached from the waste. As given by Eq. (4.1.39):

$$N_i^P = \int_0^t d\theta \int_{-\infty}^{+\infty} du \int_{-\infty}^{+\infty} dv \int_{-\infty}^{+\infty} dw \left[\lambda_{i-1} \frac{v_i}{v_{i-1}} N_{i-1}(u,v,w,\theta)\right]$$
$$\cdot \; e^{-\lambda_i(t-\theta)} \, G\big(\tilde{x}-u, lv_i(t-\theta)\big) \qquad (4.1.52)$$
$$\cdot \; G\big(\tilde{y}-v, lv_i(t-\theta)\big) \cdot G\big(\tilde{z}-w-v_i(t-\theta), lv_i(t-\theta)\big)$$

Assume that N_{i-i} consists of a sum of terms P_{i-1} of the form:

$$P_{i-1} = \frac{W_o}{vt} \frac{1}{l^2} C \, E(I,J,K,\tilde{x},\tilde{y},\tilde{z},t) \qquad (4.1.53)$$

where C is an arbitrary constant.

Each of these terms will correspond in N_i^P to a term P_i:

$$P_i = \frac{W_0}{vT}\ \frac{C}{l^2}\,\lambda_{i-1}\ \frac{v_i}{v_{i-1}} \int_0^t d\theta \int_{-\infty}^{+\infty} du \int_{-\infty}^{+\infty} dv \int_{-\infty}^{+\infty} dw\ \ E(I,J,K,u,v,w,\theta)$$

$$\cdot\ e^{-\lambda_i(t-\theta)} \cdot G\left(\tilde{x}-u, l v_i(t-\theta)\right) \cdot G\left(\tilde{y}-v, l v_i(t-\theta)\right) \qquad (4.1.54)$$

$$\cdot\ G\left(\tilde{z}-w-v_i(t-\theta), l v_i(t-\theta)\right) .$$

The mathematical treatment of this term given in (A1) shows that P_i can be reduced to:

$$P_i = \frac{W_0}{vT}\ \frac{C}{l^2}\ \lambda_{i-1}\ \frac{v_i}{v_{i-1}}\ \frac{\Gamma_{IJ}}{v_i M_{IJKi}}$$

$$\Big\{ E(I,J,i,\tilde{x},\tilde{y},\tilde{z},t) - E(I,J,K,\tilde{x},\tilde{y},\tilde{z},t) \qquad (4.1.55)$$

$$+ E(K,i,K,\tilde{x},\tilde{y},\tilde{z},t) - E(K,i,i,\tilde{x},\tilde{y},\tilde{z},t) \Big\}$$

where

$$M_{IJKi} = \Gamma_{IJ}\ \Lambda_{Ki} - \Gamma_{Ki}\ \Lambda_{IJ} \qquad (4.1.56)$$

Equations (4.1.53) and (4.1.55) establish a recurrence relation showing that if it is possible to express N_{i-1} with the function E, this is also possible for N_i^P. Equation (4.1.51) shows that N_i^R is a linear function of E for the first nuclide, which has no precursor. However, since $N_1 = N_1^R$, N_1 is also a linear function of E. Therefore, $N_i = N_i(E)$ for any i.

Equations (4.1.53) and (4.1.55) therefore allow the derivation of N_i^P from N_{i-1}.

The recurrence relation defined by Eqs. (4.1.53) and (4.1.55) states that a term E(I,J,K) in the solution for N_{i-1} appears as four different E terms in the solution for N_{i-1}, i.e.,

in N_{i-1}: E (I, J, K) is unchanged,

$$\text{in} \quad N_i: \frac{\lambda_{i-1}}{v_{i-1}}\ \frac{\Gamma_{IJ}}{M_{IJKi}} \cdot \Big\{ E(I,J,i) - E(I,J,K) + E(K,i,K) - E(K,i,i) \Big\} \quad (4.1.57)$$

Using this procedure, the solutions $N_1{}^S$, $N_2{}^S$, $N_3{}^S$ for the step release of a three-member decay chain can be readily determined:

$$N_1^S\ (\tilde{x},\tilde{y},\tilde{z},t) = \frac{W_o}{vTl^2} B_{11}\ E(1,0,1) \tag{4.1.58a}$$

$$\begin{aligned} N_2^S\ (\tilde{x},\tilde{y},\tilde{z},t) = {} & \frac{W_o}{vTl^2} \sum_{j=1}^{2} B_{2j}\ E(j,0,2) \\ & + \frac{W_o}{vTl^2}\ \frac{B_{11}\lambda_1\Gamma_{10}}{v_1\ M_{1012}} \Big\{ E(1,0,2) - E(1,0,1) \\ & \qquad + E(1,2,1) - E(1,2,2) \Big\} \end{aligned} \tag{4.1.58b}$$

$$\begin{aligned} N_3^S\ (\tilde{x},\tilde{y},\tilde{z},t) = {} & \frac{W_o}{vTl^2} \sum_{j=1}^{3} B_{3j}\ E(j,0,3) \\ & + \frac{W_o}{vTl^2} \sum_{j=1}^{2} B_{2j}\ \frac{\lambda_2}{v_2}\ \frac{\Gamma_{j0}}{M_{j023}} \\ & \qquad \cdot \Big\{ E(j,0,3) - E(j,0,2) + E(2,3,2) - E(2,3,3) \Big\} \\ & + \frac{W_o}{vTl^2}\ \frac{B_{11}\ \lambda_1\ \Gamma_{10}}{v_1\ M_{1012}} \\ & \cdot \Big\{ \frac{\lambda_2\ \Gamma_{10}}{v_2\ M_{1023}}\ [E(1,0,3) - E(1,0,2) + E(2,3,2) - E(2,3,3)] \\ & - \frac{\lambda_2\ \Gamma_{10}}{v_2\ M_{1013}}\ [E(1,0,3) - E(1,0,1) + E(1,3,1) - E(1,3,3)] \\ & + \frac{\lambda_2\ \Gamma_{12}}{v_2\ M_{1213}}\ [E(1,2,3) - E(1,2,1) + E(1,3,1) - E(1,3,3)] \\ & - \frac{\lambda_2\ \Gamma_{12}}{v_2\ M_{1223}}\ [E(1,2,3) - (E(1,2,2) + E(2,3,2) - E(2,3,3)] \Big\} \end{aligned} \tag{4.1.58c}$$

Equations (4.1.58a,b,c) and the supplementary Eqs. (4.1.48-49) for E and F are written in terms of the transformed coordinates $(\tilde{x},\tilde{y},\tilde{z},t)$. The corresponding concentrations in real coordinates (x,y,z,t) are written by employing Eqs. (4.18) and (4.19), and noting that $\tilde{z} = z$:

$$N_i(x,y,z,t) = \frac{D_z}{\sqrt{D_x\ D_y}}\quad N_i(\tilde{x},\tilde{y},\tilde{z},t) \tag{4.1.59}$$

The function E in Eqs. (4.1.58a,b,c) for $N_i(\tilde{x},\tilde{y},\tilde{z},t)$ must be evaluated at $\tilde{x},\tilde{y},\tilde{z}$, which are determined by

$$\tilde{x} = x\quad \sqrt{D_z/D_x} \tag{4.1.8}$$

$$\tilde{y} = y\quad \sqrt{D_z/D_y} \tag{4.1.9}$$

$$\tilde{z} = z \tag{4.1.60}$$

4.1.8 Concentrations for a band release from a point-source

Concentration profiles for a band release [Eq. (4.1.22)] are obtained by substituting the step release solutions into the superposition equation (2.1.18).

4.1.9 First Nuclide in Chain, Step Release, No Axial Dispersion

A relatively simple solution for the concentration $N_1(x,y,z,t)$ for a point-source in a one-dimensional flow field, with transverse dispersion ($D_x = D_y \equiv D_T$) and no axial dispersion ($D_z = 0$) can be obtained from Eqs. (4.1.38) and (4.1.59) since $N_1 = N_1^R$.

We assume a step release at x = y = z = t = 0. From Eq. (4.1.25):

$$^{pt}S_1(t) = \frac{W_o n_1^o}{T}\, e^{-\lambda_1 t}\ h(t)\ . \tag{4.1.61}$$

The following terms appear when Eqs. (4.1.8), (4.1.9), and (4.1.59) are substituted into (4.1.38):

$$\sqrt{\frac{D_z}{D_x}}\ G[\tilde{x},1v_1t] = \sqrt{\frac{K_1}{4\pi x D_x}}\ \exp\ -\left(\frac{K_1x^2}{4tD_x}\right) \tag{4.1.62}$$

$$\sqrt{\frac{D_z}{D_y}}\ G[\tilde{y},1v_1t] = \sqrt{\frac{K_1}{4\pi y D_y}}\ \exp\ -\left(\frac{K_1y^2}{4tD_y}\right). \tag{4.1.63}$$

When $D_z \to 0$, a unit impulse originating at $z-v_1t$ appears at z unmodified by axial dispersion, so that the Green's function for transport in the axial direction becomes:

$$\lim_{D_z \to 0} G(\tilde{z}-v,t,|v_1t) = \delta(z-v_1t) . \tag{4.1.64}$$

Substituting Eqs. (4.1.59) and (4.1.60–64) into Eq. (4.1.38) and integrating, we obtain:

$$N_1(x,y,z,t) = \frac{W^0 n_1^0}{4\pi T v}\, e^{-\lambda_1 t}\, \frac{\exp -\left(\dfrac{x^2}{4zD_x/v}\right)}{\sqrt{\dfrac{zD_x}{v}}}\; \frac{\exp\left(-\dfrac{y^2}{4zD_y/v}\right)}{\sqrt{\dfrac{zD_y}{v}}}\, h(v_1t-z) . \tag{4.1.65}$$

Equation (4.1.65) is rewritten with the simplification that $D_x = D_y = D_T$:

$$N_1(x,y,z,t) = \frac{W^0 n_1^0}{4\pi T v\Omega}\, e^{-\lambda_1 t}\, e^{-\left(\dfrac{x^2+y^2}{4\Omega}\right)}\, h(v_1t-z) \tag{4.1.66}$$

where

$$\Omega \equiv \frac{zD_T}{v} . \tag{4.1.67}$$

This solution for the first member, with no axial dispersion, will be used for numerical demonstrations of point-source and plane-source concentration profiles in Sect. 4.3.3.

4.1.10 Application to various far-field boundary conditions

(a) Release In a Semi-Infinite Medium with Zero-Flux Boundary Conditions. We consider a semi-infinite medium bounded by a plane $x = L$ parallel with the flow direction, thus including all points $(\tilde{x}, \tilde{y}, \tilde{z})$ such that:

$$-\infty \le \tilde{x} \le L \qquad (L > 0)$$
$$-\infty \le \tilde{y} \le +\infty$$
$$-\infty \le \tilde{z} \le +\infty .$$

We assume that the flux of any radionuclide i, at any point of the boundary, is zero at any time. This corresponds to a boundary impervious to nuclides, for instance the ground surface. The first of the boundary conditions (4.1.19) is now replaced by:

$$\frac{\partial N_i}{\partial x} (L,\tilde{y},\tilde{z},t) = 0 \ . \tag{4.1.68}$$

Release is assumed to occur at the origin of the axes, but we need not specify its nature. Let $N_i^U (\tilde{x}, \tilde{y}, \tilde{z}, t)$ be the solution in an unbounded medium, for the release we are considering. Then, we can express N_i simply as:

$$N_i(\tilde{x},\tilde{y},\tilde{z},t) = N_i^U(\tilde{x},\tilde{y},\tilde{z},t) + N_i^U(\tilde{x}-2L,\tilde{y},\tilde{z},t) \ . \tag{4.1.69}$$

(b) Release In a Semi-Infinite Medium with Zero-Concentration Boundary Conditions. We now assume that the concentration of nuclide i is zero at any point on the boundary and at any time; i.e., we replace (4.1.68) by:

$$N_i \ (L, \tilde{y}, \tilde{z}, t) = 0 \ . \tag{4.1.70}$$

This implies that there exists a physical mechanism removing all nuclides as soon as they reach the boundary, so as to maintain the concentration equal to zero.

A flow, either of ground water or surface water, can provide such a mechanism if it is fast enough compared to the flow in the initial medium and maintains a concentration much smaller than the concentrations observed there.

With regard to the amount of nuclides escaping through the boundary, this assumption of immediate removal of nuclides is conservative and thus gives maximum values of possible releases out of the repository medium through a lateral boundary.

In this case we can express N_i as:

$$N_i \ (\tilde{x}, \tilde{y}, \tilde{z}, t) = N_i^U \ (\tilde{x}, \tilde{y}, \tilde{z}, t) - N_i^U \ (\tilde{x}-2L,\tilde{y},\tilde{z},t) \tag{4.1.71}$$

(c) Diffusive Current of the i-th Nuclide through a Boundary at x = L. At any point of the boundary, characterized by $\tilde{x} = L$, the waste being at the position $\tilde{x} = \tilde{y} = \tilde{z} = 0$, the dispersion current is equal to:

$$j = - D_z \frac{\partial N_i}{\partial \tilde{x}} (L, \tilde{y}, \tilde{z}, t) . \tag{4.1.72}$$

Equations (4.1.36), (4.1.37) and (4.1.38) show that $\partial N_i^U / \partial x$ is odd, so that, using (4.1.68) we obtain:

$$j = - 2 D_z \frac{\partial N_i^U}{\partial \tilde{x}} (L, \tilde{y}, \tilde{z}, t) . \tag{4.1.73}$$

The total rate J at which atoms diffuse across the boundary at time t is:

$$J = - 2 D_z \int_{-\infty}^{+\infty} dy \int_{-\infty}^{+\infty} d\tilde{z} \quad \frac{\partial N_i^U}{\partial \tilde{x}} (L,\tilde{y},\tilde{z},t) . \tag{4.1.74}$$

One can show mathematically that, if J_{IjK} is defined as:

$$J_{IJK} \equiv - 2 D_z \frac{W_o}{vTl^2} \int_{-\infty}^{\infty} d\tilde{y} \int_{-\infty}^{\infty} d\tilde{z} \quad \frac{\partial}{\partial x} E(I,J,K,L,\tilde{y},\tilde{z},t) \tag{4.1.75}$$

then we can rewrite J_{IJK} as:

$$J_{IJK} = 4\pi \frac{W_o}{T} \frac{L}{T} e^{-\beta_{IJ} t} F\left(lv_K t, \sqrt{\frac{\gamma_{IJK} - 1}{2l}}, \frac{L}{2}\right) \tag{4.1.76}$$

if $\gamma_{IJK} \geq 1$, and:

$$J_{IJK} = 4\pi \frac{W_o}{T} \frac{L}{T} e^{-\beta_{IJ} t} F^*\left(lv_K t, \sqrt{\frac{1 - \gamma_{IJK}}{2l}}, \frac{L}{2}\right) \tag{4.1.77}$$

if $\gamma_{IJK} < 1$, F^* being defined as:

$$F^*(U,a,b) = l \int_0^U \frac{e^{a^2 \tau - \frac{b^2}{\tau}}}{(4\pi\tau)^{3/2}} d\tau . \tag{4.1.78}$$

The preceding formulas are useful for the step-release case.

4.1.11 Comparison with the One-Dimensional Solutions for an Infinite Plane Source

The source of nuclides released into the ground water is a major difference between this model and the one-dimensional models developed earlier in Sect. 2.1 and (H1). In the one-dimensional models for an infinite plane source (H1), the water flow rate per unit amount of waste must be specified for the purpose of determining a concentration. In the three-dimensional model presented here, only the point-source release rate, or the amount released, need be known, together with the velocity of ground water flow.

The concentrations predicted for the point-source with three dimensional dispersion depend upon the values of transverse dispersion coefficients, which now become critical parameters.

In the one-dimensional solution with a boundary condition expressing dissolution from a plane source (H1), there appears an integral F', similar to the F developed here:

$$F'(U,a,b) = \int \frac{e^{-\left\{a^2\tau + \frac{b^2}{\tau}\right\}}}{(4\pi\tau)^{1/2}}\,d\tau \tag{4.1.79}$$

Resulting from the consideration of the z direction only, the exponent in the denominator of F' is $\frac{1}{2}$ instead of $\frac{3}{2}$ in F. Accordingly, the integrated form of F' is different from the integrated form of F in Eq. (4.1.49):

$$F'(U,a,b) = \frac{1}{4a}\left\{-e^{2ab}\,\mathrm{erfc}\left(a\sqrt{U} + \frac{b}{\sqrt{U}}\right) + e^{-2ab}\,\mathrm{erfc}\left(-a\sqrt{U} + \frac{b}{\sqrt{U}}\right)\right\} \tag{4.1.80}$$

and the solutions for source boundary conditions show different functional behaviors in the one-dimensional and three-dimensional cases.

On the contrary, the one-dimensional solution with a concentration boundary condition at the plane source includes the same integral as in the present three-dimensional solution [Eq. (4.1.49)]. As a result, the only differences between both solutions, besides numerical constants, are the following:

a) $|z|$ is replaced by $\sqrt{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}$ everywhere except in the first exponential term, which remains $e^{z/2l}$;

b) the whole expression is divided by $\sqrt{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}$. Therefore, along the $\tilde{z}$ axis, i.e., for $\tilde{x} = \tilde{y} = 0$, the three-dimensional solution differs from the one-dimensional solution with concentration boundary condition only by multiplication by a numerical factor and division by $|\tilde{z}|$. The later modification accounts for the fact that as concentration peaks proceed, lateral diffusion broadens them and so decreases the concentration on the z axis for larger values of $\tilde{z}$.

The maxima of peaks will thus occur for smaller values of $\tilde{z}$, and the peaks will be shifted towards the repository. However, this shift is not an important one because the variation of the term $1/|\tilde{z}|$ (or $1/\sqrt{\tilde{x}^2 + \tilde{y}^2 + \tilde{z}^2}$) is slow compared to variations of other exponential and error function factors.

A much more important difference between the concentrations on the z axis calculated by the two models results from the difference of the numerical coefficients in both cases. The area of the waste is a crucial parameter in the one-dimensional model, and the concentrations become infinite if the area tends towards zero while the dissolution rate remains finite.

On the other hand, the concentrations on the z axis calculated by the three-dimensional model depend largely on the value of the dispersion coefficient, and become infinite if dispersion tends towards zero.

Finally, as shown in Appendix D, the three-dimensional solution can be used to obtain the one-dimensional solution for a plane source. To do so, the plane source is represented by an infinite distribution of point sources, and the three-dimensional solution for a point source is integrated with respect to this distribution.

4.1.8 Application to the transport of $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$

As an illustration, we will now apply the previous results to the migration of the chain:

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

We assume no ^{234}U daughters at the beginning of leaching. The migration characteristics are summarized in Table 4.1.1.

TABLE 4.1.1

Data for the ^{234}U – ^{230}Th – ^{226}Ra chain migration.

Medium parameters:	
Flow velocity	v : 100 m / yr
Diffusion coefficient	D : 10^3 m^2 / yr (unless otherwise specified)
Characteristic diffusion length	l : 10 m (unless otherwise specified)
Leach Time	T : 30,000 yrs

Nuclide characteristics:	^{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, yr^{-1}	2.841×10^{-6}	9.002×10^{-6}	4.332×10^{-4}
Assumed sorption retardation retardation constant, K_i	1×10^4	5×10^4	5×10^2
Migration speed $v_i = \frac{v}{K_i}$, m/yr	1.0×10^{-2}	2.0×10^{-3}	2.0×10^{-1}

4.1.8.A Concentration Profiles in an Unbounded Medium

Concentration profiles of the three nuclides migrating in an unbounded medium are given in Figs. 4.1.1 through 4.1.4, at time t = 50,000 yrs. Figure 4.1.1 shows the profiles of the three nuclides along the flow direction. Relative concentrations, expressed per unit amount of ^{234}U initially present in the waste, are plotted versus the distance from the waste.

These profiles can be compared with those given by our previous report (H1) for similar conditions, but for one-dimensional dispersion. It is not meaningful to compare magnitudes, because the two models depend upon different parameters (see Sect. 4.1.7).

The profiles with three-dimensional dispersion appear to be quite similar in shape to those in (H1). The maxima of the peaks expectedly occur for smaller values of z (cf. Sect. 4.1.7), as shown in Table 4.1.2.

TABLE 4.1.2

Location of concentration maxima for a transport time of 5 x 10^4 yr, as computed from one- and three-dimensional dispersion.

	Locations of the maximum global concentration, m		
	^{234}U	^{230}Th	^{226}Ra
One-dimensional dispersion	320	63	360
Three-dimensional dispersion	270	56	110

This last effect is specially important for ^{226}Ra, because the broad peak of this radionuclide actually results from the mingling of two ^{226}Ra peaks, located at the ^{230}Th and ^{234}U peaks. Due to transverse diffusion, the farthest peak is broadened more and thus becomes smaller than the nearest peak, resulting in the important shift of the concentration maximum.

Figure 4.1.2 gives the transverse concentration profiles along a direction perpendicular to the water flow. They correspond to a time t = 50,000 yrs and a distance z = 100 m from the repository.

It can be seen that ^{234}U, more mobile than ^{230}Th, presents a broader peak. However, though ^{226}Ra is more mobile than its precursor ^{230}Th, its peak is not broader because the shorter half-life of ^{226}Ra prevents its build-up in regions containing relatively little ^{230}Th.

Figure 4.1.3 gives the transverse profiles of ^{230}Th at a time of 5 x 10^4 yr, for three different distances from the repository:

$$z = 50, 100, 300 \text{ m.}$$

The decrease in the maximum concentration of ^{230}Th with distance z from the repository is not due to dispersion, but to the initially zero amount of ^{230}Th in the waste at the beginning of dissolution (cf. Figure 4.1.1).

To illustrate the importance of the dispersion coefficient, Figure 4.1.4 shows ^{234}U concentration profiles along the flow direction, for dispersion coefficients 0.5 x 10^{+3}, 1 x 10^{+3}, 2 x 10^{+3} m^2/yr, corresponding to characteristic dispersion lengths 5, 10, 20 m.

Expectedly, increased dispersion broadens peaks and considerably reduces maximum concentrations.

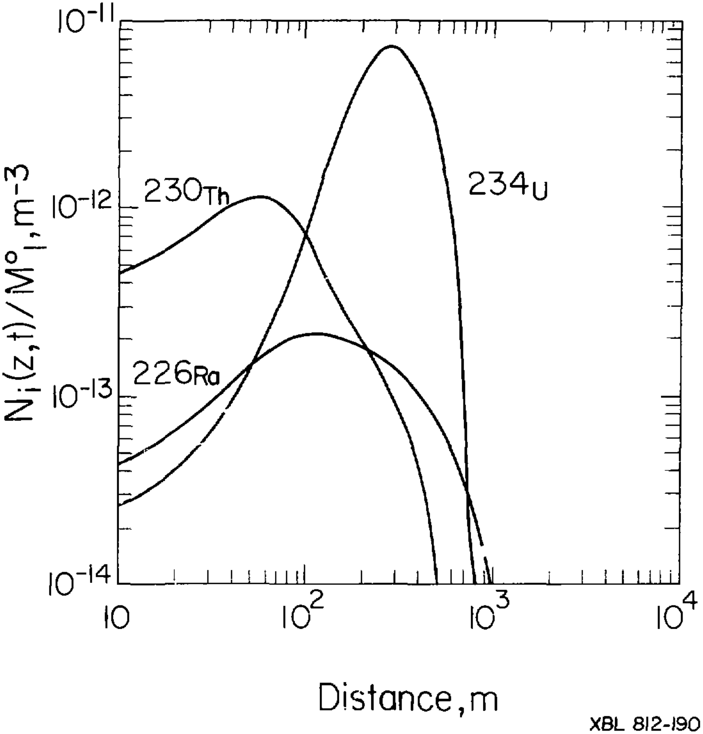

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Figure 4.1.1. Concentration profiles of ^{234}U ^{230}Th ^{226}Ra decay chain along the z axis, at $t = 5\times10^4$ yr.
No daughters at t = 0. $N_i(z,t)$ = atoms/m^3 in the waste at t = 0
M_1^0 = atoms of ^{234}U
$v = 100$ m/yr. $T = 3\times10^4$ yr. $K_U = 1\times10^4$, $K_{Th} = 5\times10^4$
$K_{Ra} = 5\times10^2$. $D = 10^3 m^2/yr$. Point source. One-dimensional flow. Three dimensional dispersion.

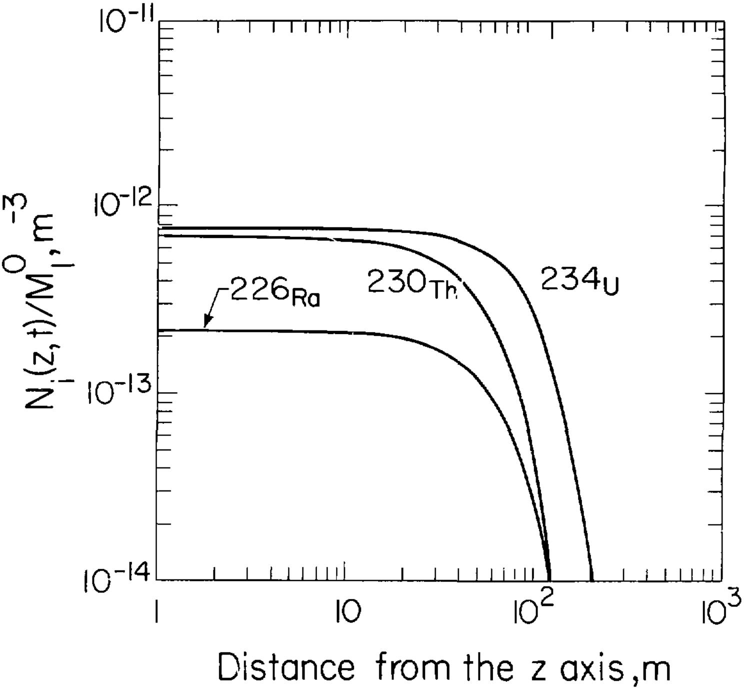

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Figure 4.1.2. Concentration profiles of ^{234}U ^{230}Th ^{226}Ra decay chain along a direction perpendicular to the water flow, at $t = 5x10^4$ yr.

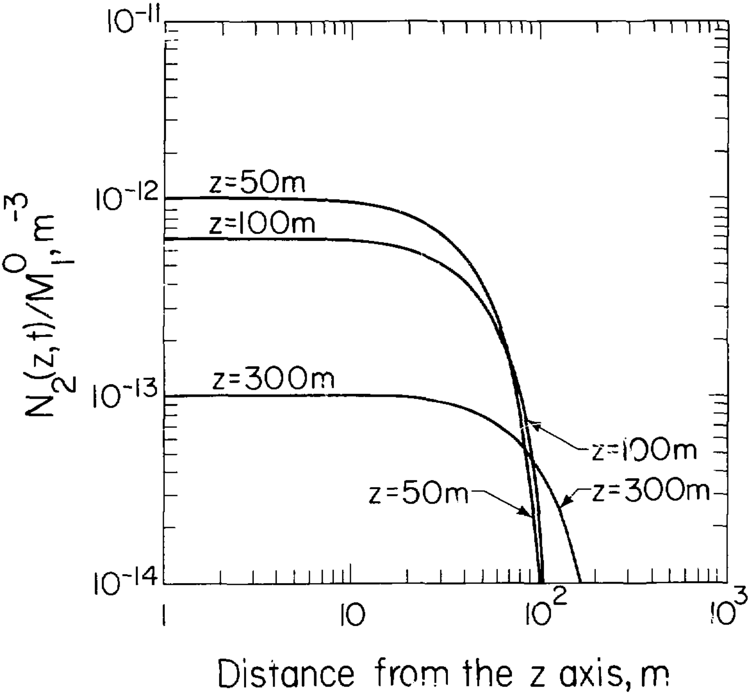

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Figure 4.1.3. ^{230}Th concentration profiles along a direction perpendicular to the water flow, at t = 5×10^4 yr.

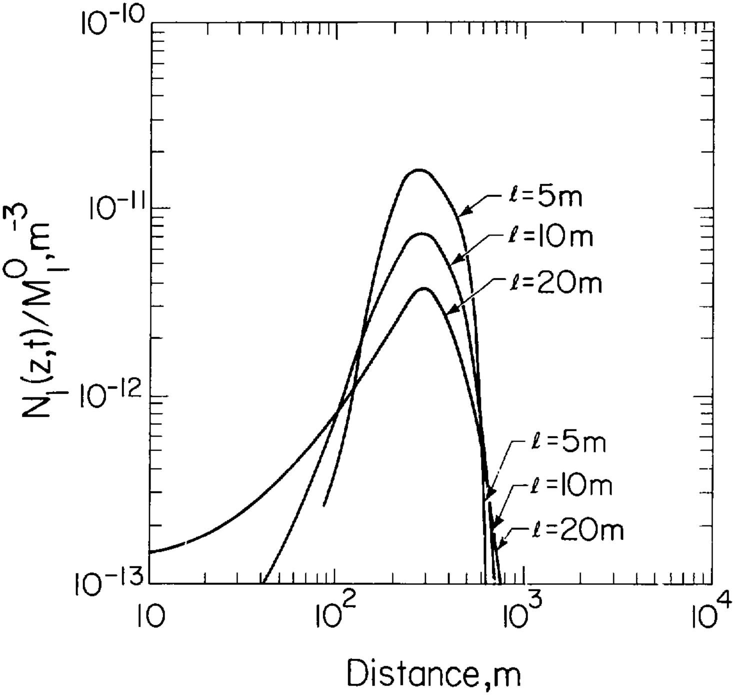

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Figure 4.1.4. ^{234}U concentration profiles along the z axis, at t = 5×10^4 yr. D/v = characteristic length of diffusion $N_1(z,t)$ = atoms of $^{234}U/m^3$. M_1^0 = atoms of ^{234}U in the waste at t = 0. v = 100 m/yr. T = 3×10^4 yr. $K_U = 1 \times 10^4$. Point source. One-dimensional flow. Three-dimensional dispersion.

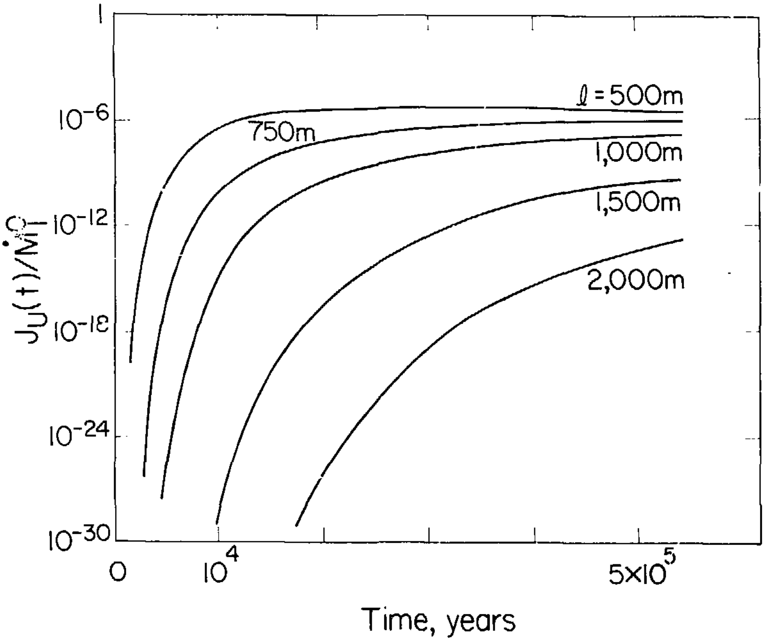

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Figure 4.1.5. Release rate J_U of ^{234}U through boundaries at $x = \pm L$.
M_i^0 = release rate of ^{234}U at $z = 0$ and $t = 0$
$v = 100$ m/yr. $T = 3\times10^4$ yr. $K_U = 1\times10^4$. $D = 10^3 m^2/hr$.
Point source. One-dimensional flow. Three-dimensional dispersion.

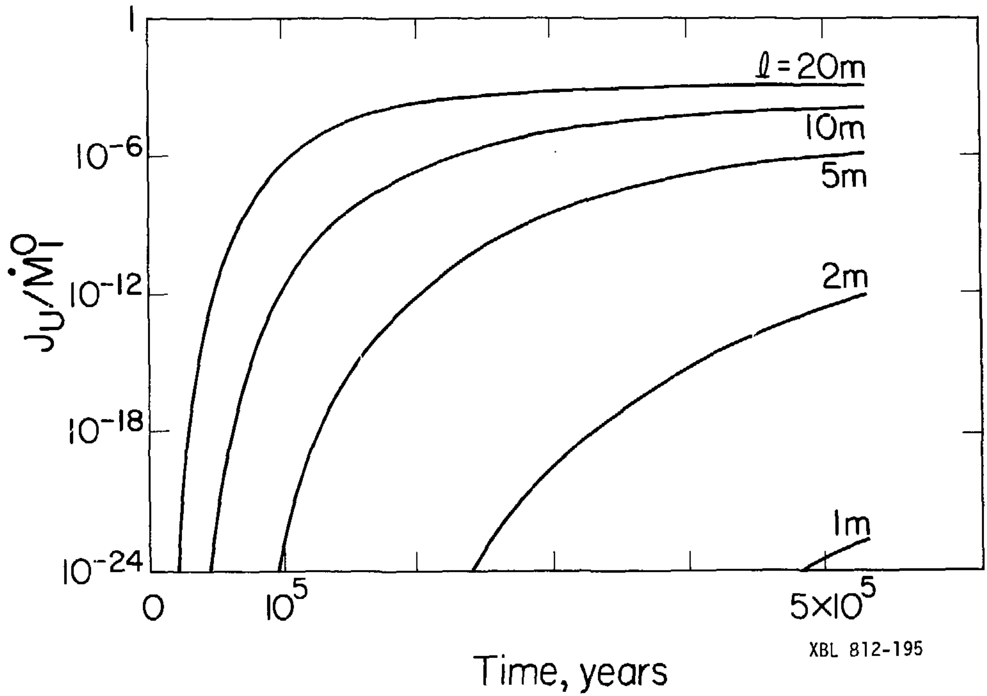

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Figure 4.1.6 Release rate of J_U of ^{234}U at x = 1000 m. D/v = characteristic diffusion length M_i^0 = release rate of ^{234}U at z = 0 and t = 0. V = 100 m/yr, T = 3 x 10^4 yr, K_U = 10^4. Point source. One-dimensional flow. Three-dimensional dispersion.

4.1.8.B Release of Nuclides from a Bounded Medium

Applying the results in Sec. 4.1.6, we now consider the maximum rate of dispersive release out of a medium bounded by planes parallel with the flow direction. Using Eq. 4.1.60, we compute the instantaneous discharge rate across boundaries at $x = \pm L$, with $x = 0$ at the point source of dissolving waste. Aquifers along the boundaries are assumed to give a zero-concentration boundary condition.

The results are presented for ^{234}U, giving a relative release rate defined as:

relative release rate =

$$\frac{\text{total discharge rate of } ^{234}\text{U through the boundary at time } t}{\text{initial } ^{234}\text{U release rate, at time 0, from the waste point source}}$$

Figure 4.1.5 shows a plot of this relative rate versus time, for a characteristic dispersion length $l = 10$ m ($D = 10^3$ m^2/yr), and for various values of the distance L from the repository to the boundary. It can be seen that, as transverse dispersion proceeds, the release rate first increases dramatically with time. Then, as radioactive decay reduces the available amounts of ^{234}U, the release rate increases no more, and would eventually decrease for larger times. It also appears that moderate variations in the distance between the waste and the boundary can change the rate by several orders of magnitude. Figure 4.1.6 shows an equivalent plot for a distance L = 1000 m between the waste and the boundary, and for various values of the characteristic diffusion length. The same variation in time than previously is observed. Expectedly, the influence of the dispersion coefficient is considerable.

4.2. Transport of Radionuclides Released from a Finite Plane Source

4.2.1 Introduction

The analytical studies in Reference (H1) have dealt primarily with the transport of nuclides in a single space dimension in the presence of one-dimensional groundwater flow. The release of nuclides was modelled either by a source term or a concentration boundary condition applied on the plane $z = 0$. In the present research we extend these results to an emitting repository surface of finite dimensions located in a uniform flow field. The primary purpose is to judge the effects of transverse dispersion on the transport of the nuclides which was not considered in the previous work.

The following assumptions are made:

1) The repository surface S is located in a uniform, one-dimensional ground water flow field. This means that S is penetrated with the same groundwater velocity as prevails outside of the repository in the plane of S, see Figure 4.2.1.

2) Dispersion effects in the flow direction are neglected in the first part of the analysis.

A detailed study of the hydrodynamic effects on radionuclide transport is given by the writer in Sect. 5. Work is in progress to deal with a more realistic velocity field in which the (possibly) different permeabilities of soil and repository are taken into account. It is hoped that this will eliminate the first assumption.

The neglect of dispersion effects in the flow direction is one of mathematical convenience only. By the method of solution to be given one can obtain a recursive solution for individual members of a nuclide chain in the presence of longitudinal dispersion. These solutions are of a rather complex character and are costly to evaluate by machine computations.

4.2.2 Without Longitudinal Dispersion

If the longitudinal dispersion is neglected one can obtain the general (non-recursive) solution for a nuclide chain of arbitrary length. This solution is of a relatively simple form and gives physical insight into the effects of diffusion in one-dimensional flow fields.

The starting point is the equation system, $i = 1,2,---$

$$\frac{\partial N_i}{\partial t} + v_i \frac{\partial N_i}{\partial z} + \lambda_i N_i = D_{xi} \frac{\partial^2 N_i}{\partial x^2} + D_{yi} \frac{\partial^2 N_i}{\partial y^2} + \frac{K_{i-1}}{K_i} \lambda_{i-1} N_{i-1}, \lambda_0 = 0 \quad (4.2.1)$$

where

$$v_i = \frac{v}{K_i}, \quad D_{xi} = \frac{D_x}{K_i}, \quad D_{yi} = \frac{D_y}{K_i} \quad (4.2.2)$$

for the determination of the functions $N_i(z,t;x,y)$ in an (x,y,z) cartesian coordinate system, see Figure 4.2.1.

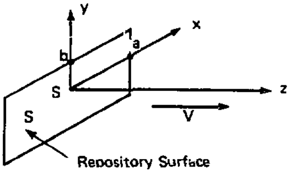

Figure 4.2.1 Repository Surface

The solution is sought for $-\infty < (x,y) < \infty$, $0 < z < \infty$, $t > 0$. The concentration boundary conditions are

$$N_i(0,t;x,y) = N_i^0 S_i(x,y)\phi_i''(t), |x| < a, \ |y| < b, \ t > 0$$
$$= 0, \ |x| > a, \ |y| > b \tag{4.2.3}$$

witn

$$\phi_i''(t) \equiv 0, \ t < 0 \tag{4.2.4}$$

and the initial conditions,

$$N_i(z,0;x,y) = 0, \ -\infty < (x,y) < \infty \ , \ 0 < z < \infty \ . \tag{4.2.5}$$

Equation (4.2.1) allows for the possibility of dissimilar dispersion effect in the x and y directions. Equation (4.2.3) permits the nuclide concentration emitted at the repository surface S to be an arbitrary function of position $S_i(x,y)$ on S. The release rate function $\phi_i''(t)$ is also arbitrary.

In order to obtain the general solution to the equation system, consider the following sets of boundary conditions, where $P \equiv (x,y)$ and $B_i(P,t) \equiv S_i(x,y)\phi_i''(t)$.

(4.2.6)

Boundary Condition	Nuclide Chain Member l=1	l=2	-------	l=l	------	l=i
j=1	$N_1^{(1)}(o,t;P)=N_1^0B_1$	$N_2^{(1)}(o,t;P)=0$		$N_l^{(1)}(o,t;P)=0$		$N_i^{(1)}(o,t;P)=0$
j=2	$N_1^{(2)}(o,t;P)=0$	$N_2^{(2)}(o,t;P)=N_2^0B_2$		$N_l^{(2)}(o,t;P)=o$		$N_i^{(2)}(o,t;P)=0$
j=l	$N_1^{(l)}(o,t;P)=0$	$N_2^{(l)}(o,t;P)=0$		$N_l^{(l)}(o,t;P)=N_l^{(l)}B_l$		$N_i^{(i)}(o,t;P)=0$
j=i	$N_1^{(i)}(o,t;P)=0$	$N_2^{(i)}(o,t;P)=0$		$N_l^{(i)}(o,t;P)=0$		$N_i^{(i)}(o,t;P=N_i^0B_i$

The solutions to equations (4.2.1) and (4.2.5) subject to the conditions on line j of the above set of boundary conditions i is denoted by $N_l^{(j)}(z,t;x,y), l = 1,2, \ldots , i$.

On account of the linearity of the governing equations 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 Eqs. (4.2.1-5) i.e.,

$$N_i(z,t;x,y) = N_i^{(i)}(z,t;x,y) + \sum_{j=1}^{i-1} N_i^{(j)}(z,t;x,y), i = 1,2, \ldots \qquad (4.2.7)$$

It should be noted that some of the $N_l^{(j)}$ functions vanish. For instance, if say $N_1^{(2)}(0,t;x,y) = 0$ but not $N_2^{(2)}(0,t;x,y)$, which is the case for the second set of boundary data (j = 2) in Eq. (4.2.6), then $N_1^{(2)}(z,t;x,y) = 0$ for $Z > 0$ and $t > 0$. In general there will be no precursors for the functions $N_l^{(j)}(z,t;x,y)$ for $l \leq j$ and so $N_{l-1}^{(j)}(z,t;x,y) = 0$ in the space $z > 0$ for $t > 0$ provided $l \leq j$. For each set $j = 1,2, \ldots ,i$ of Eq. (4.2.6) the governing equation to be solved for each nuclide $N_l^{(j)}$ is:

$$\frac{\partial N_l^{(j)}}{\partial t} + v_l \frac{\partial N_l^{(j)}}{\partial z} + \lambda_l N_l^{(j)} = D_{xl} \frac{\partial^2 N_l^{(j)}}{\partial x^2} + D_{yl} \frac{\partial^2 N_l^{(j)}}{\partial y^2} + \frac{K_{l-1}}{K_l} \lambda_{l-1} N_{l-1}^{(j)} \qquad (4.2.8)$$

where $l = 1,2,\ldots i$ and

$$N_{l-1}^{(i)} = 0 \text{ when } l \leq j. \qquad (4.2.9)$$

The boundary conditions are obtained from Eqs. (4.2.3) and (4.2.6)

$$N_l^{(j)}(0,t;x,y) = \begin{cases} 0, & l \neq j \\ N_j^0 S_j(x,y)\phi_j''(t), & |x| < a, |y| < b, l = j \\ 0, & |x| > a, |y| > b, l = j \end{cases} \qquad (4.2.10)$$

where

$$\phi_i''(t) \equiv 0 \quad , \quad t < 0.$$

The initial conditions is

$$N_l^{(j)}(z,0;x,y) = 0. \quad , \qquad -\infty < (x,y) < \infty, \quad 0 < z < \infty. \tag{4.2.11}$$

On applying a double Fourier transformation, defined by

$$\bar{N}(z,t;\omega_1,\omega_2) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} N(z,t;x,y)e^{-\sqrt{-1}\,(\omega_1 x + \omega_2 y)}\,dxdy, \quad (\sqrt{-1} = i) \tag{4.2.12}$$

to Eqs. (4.2.8) (4.2.10) and (4.2.11) one obtains

$$\frac{\partial \bar{N}_l^{(j)}}{\partial t} + v_l \frac{\partial \bar{N}_l^{(j)}}{\partial z} + \lambda_l'' \bar{N}_l^{(j)} = \frac{K_{l-1}}{K_l} \lambda_{l-1} \bar{N}_{l-1}^{(j)} \tag{4.2.13}$$

where

$$\lambda_l'' = \lambda_l + (D_{xl}\omega_1{}^2 + D_{yl}\,\omega_2{}^2)\ . \tag{4.2.14}$$

$$\bar{N}_l(0,t;\omega_1,\omega_2) = \begin{cases} 0, & l \neq j \\ N_j{}^0\,\bar{S}_j(\omega_1,\omega_2)\,\phi_j(t)\ , & l = j \end{cases} \tag{4.2.14a}$$

where

$$\bar{S}_j(\omega_1,\omega_2) = \int_{-a}^{+a}\int_{-b}^{b} S_j(x,y)\,e^{-\sqrt{-1}\,(\omega_1 x + \omega_2 y)}\,dxdy, \quad (\sqrt{-1} = i) \tag{4.2.15}$$

where

$$\bar{N}_l(z,0;\omega_1,\omega_2) = 0, \quad 0 < z < \infty. \tag{4.2.16}$$

The problem has thus been reduced to a one-dimensional transport problem which was studied in detail in Ref. (H1). There the author presented in Sect. 4.4.2 the general solution to the equations (4.2.13-16). There appears to be no need to reproduce the steps in that analysis and an outline of the method might suffice.

The above equation system is subjected to a repeated Laplace transform with respect to t and z. There results an algebraic system of difference equations which can be solved in closed form, Ref. (H1), Eqs. (4.114, 4.115). *These expressions are inverted to recover the z and t dependence.* This yields, see eqs. (4.125a, 4.126)

$$\bar{N}_i^{(j)}(z,t;\,\omega_1,\,\omega_2) = A_i^{(j)}\, N_j^0\, \bar{S}_j(\omega_1,\,\omega_2) \sum_{m=j}^{i} \frac{e^{-\left(\frac{\lambda_m z}{V_m}\right)}}{B_m^{(j)}}$$

$$\times \sum_{\substack{r=j\\ r\neq m}}^{i} D_{rm}^{(j)} \left[g_{rm}(t,\omega) * \phi_j''(t)\right] \quad j\neq i \qquad (4.2.17)$$

and

$$\bar{N}_i^{(i)}(z,t;\,\omega_1,\,\omega_2) = N_i^0\, \bar{S}_i(\omega_1,\,\omega_2)\, e^{-\frac{z}{v_i}\left[\lambda_i + D_{xi}\,\omega_1^2 + D_{yi}\,\omega_2^2\right]}\, \phi_i''\left(t - \frac{z}{v_i}\right) \ . \qquad (4.2.18)$$

The constants have the following meaning

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

$$B_m^{(j)} = \prod_{\substack{r=j\\ r\neq m}}^{i} \Gamma_{rm}$$

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

$$D_{r,m}^{(i-1)} = 1 \qquad (4.2.19)$$

$$\nu_r = \frac{K_r}{K_{r+1}}\,\frac{\lambda_r}{\lambda_{r+1}}\ , \ (\nu_0 = 0)$$

$$\Gamma_{rm} = \frac{V_m - V_r}{V_m\, V_r}\ , \quad \Delta_{rm} = \frac{\lambda_r\, V_m - \lambda_m V_r}{V_m - V_r}\ .$$

The functions $g_{rm}(t,\omega)$ which appear in the convolution integral are

$$g_{rm}(t,\omega) = \begin{cases} 0 \quad , \quad t < z/V_m \\ e^{-(D_{xm}\omega_1^2 + D_{ym}\omega_2^2 + \Delta_{rm})(t - z/V_m)}, \quad t > z/V_m \end{cases} \qquad (4.2.20)$$

There remains the inversion with respect to the transverse coordinate variable (x,y), which is carried out with help of the Fourier inversion formula

$$N(z,t;x,y) = \frac{1}{4\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\overline{N}(z,t;\omega_1,\omega_2)e^{\sqrt{-1}(\omega_1 x+\omega_2 y)}\, d\omega_1 d\omega_2 \ . \qquad (4.2.21)$$

The formal interchange of this double integral in Eq. (4.2.17) with convolution integral yields the function

$$G_{r,m,j}(z,t;x,y)$$

$$= \frac{1}{4\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\overline{S}_j(\omega_1,\omega_2)\exp\Big[-(D_{xm}\omega_1^2 + D_{ym}\omega_2^2 + \Delta_{rm})(t - z/V_m)$$

$$+\sqrt{-1}(\omega_1 x+\omega_2 y)\Big]\, d\omega_1 d\omega_2 \ . \qquad (4.2.22)$$

For the inversion of Eq. (4.2.18) we require

$$H_1(z;x,y) = \frac{1}{4\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\overline{S}(\omega_1,\omega_2)\, e^{-\frac{z}{V_1}\left[D_{x1}\omega_1^2 + D_{y1}\omega_2^2\right]}$$

$$+\sqrt{-1}[\omega_1 x+\omega_2 u]\, d\omega_1 d\omega_2 \qquad (4.2.23)$$

With these functions we obtain the general (non-recursive) solution from Eq. (4.2.7) in the following form:

$$N_l(z,t;x,y) = N_l^0 e^{-\frac{\lambda_l}{v_l}} H_l(z;x,y)\phi_l''(t-\frac{z}{v_l}) \tag{4.2.24}$$

$$+ \sum_{j=1}^{l-1} A_l^{(j)} N_j^0 \sum_{m=j}^{l} \frac{e^{-(\lambda_m/v_m)z}}{B_m^{(j)}} \sum_{\substack{r=j \\ r\neq m}}^{l} D_{rm}^{(j)}[G_{r,m,j}(z,t;x,y) * \phi_j''(t)]$$

$$l = 1,2,\ldots$$

The first term represents those l^{th} nuclides that have been convected from the boundary (z=0) and have escaped decay. The triple sum represents l^{th} nuclides contributed from all precursors.

In order to illustrate the solution and obtain physical insight into its character let us consider the radionuclides to be emitted in a uniform manner from the repository surface. In this case:

$$S_j(x,y) = \begin{cases} 1, |x|<a, |y|<b \\ 0, |x|>a, |y|>b \end{cases} \qquad j = 1,2\ldots l \tag{4.2.25}$$

in Eq. (4.2.3). Furthermore, consider the dispersion identical in the x and y directions, i.e., $D_{xm}=D_{ym}=D_m$. The principal task is the evaluation of the functions G_{rmj} and H_l. The double integral (4.2.22) can be expressed as an iterated integral because $s_j(\omega_1,\omega_2)$ is separable in its variables. To evaluate it one can use the convolution theorem for Fourier transformation. This requires the Fourier inverse transforms:

$$F^{-1}\left\{\overline{S_j}(\omega_1,\omega_2)\right\} = \left[h(x+a)-h(x-a)\right]\left[h(y+b)-h(y-b)\right] \tag{4.2.25a}$$

$$F^{-1}\left\{e^{-D_m(\omega_1^2+\omega_2^2)(t-z/V_m)}\right\} = \frac{1}{4\pi D_m(t-\frac{z}{v_m})} e^{-\frac{(x^2+y^2)}{4D_m(t-z/V_m)}} \tag{4.2.26}$$

where h(x) is the unit step function.

On setting

$$\theta = D_m(t-z/V_m) \tag{4.2.27}$$

$$G_{r,m,j}(z,t;x,y) = \frac{e}{\pi}^{-\Delta_{rm}(t-z/V_m)}\left[\int_{-\infty}^{\infty}\frac{e^{-\eta^2/4\theta}}{2\sqrt{\theta}}[h(x-\eta+a)-h(x-\eta-a)]d\eta.\right]$$
$$\left[\int_{-\infty}^{\infty}\frac{e^{-\eta^2/4\theta}}{2\sqrt{\theta}}[h(y-\eta+b)-h(y-\eta-b)]dn\right]$$

$$= \frac{e}{\pi}^{-\Delta_{rm}(t-z/V_m)}\left\{\int_{-\infty}^{x+a}\frac{e^{-\eta^2/4\theta}}{2\sqrt{\theta}}d\eta-\int_{-\infty}^{x-a}\frac{e^{-\frac{\eta 2}{4\theta}}}{2\sqrt{\theta}}d\eta\right\}\left\{\int_{-\infty}^{y+b}\frac{e^{-\frac{\eta 2}{4\theta}}}{2\sqrt{\theta}}dn\right.$$
$$\left.-\int_{-\infty}^{y-b}\frac{e^{\frac{-\eta^z}{4\theta}}}{2\sqrt{\theta}}d\eta\right\} \tag{4.2.28}$$

To simplify set $\frac{\eta}{2\sqrt{\theta}} = \psi$ then

$$G_{r,m,j}(z,t;x,y) = \frac{e}{\pi}^{-\Delta_{rm}(t-z/V_m)}\left\{\int_0^{\frac{x+a}{2\sqrt{\theta}}}e^{-\psi^2}d\psi+\int_0^{\frac{a-x}{2\sqrt{\theta}}}e^{-\psi^2}d\psi\right\} \tag{4.2.29}$$
$$\left\{\int_0^{\frac{y+b}{2\sqrt{\theta}}}e^{-\psi^2}d\psi+\int_0^{\frac{b-y}{2\sqrt{\theta}}}e^{-\psi^2}d\psi\right\}$$

Each integral can be expressed as the (tabulated) error function. If we define

$$E_m(c\pm u,\theta) = \text{erf}\left(\frac{c+u}{2\sqrt{D_m\theta}}\right)+\text{erf}\left(\frac{c-u}{2\sqrt{D_m\theta}}\right) \tag{4.2.30}$$

Then

$$G_{r,m,j}(z,t;x,y) = \frac{e^{-\Delta_{rm}(t-z/V_m)}}{4}E_m(a\pm x,t-\frac{z}{V_m})\cdot E_m(b\pm y,t-\frac{z}{V_m}),m\neq 1 \tag{4.2.31}$$

Equation (4.2.23) differ from Eq. (4.2.22) in that m=1, Δ_{rm}=o and that $t-z/V_m$ is replaced by Z/V_1. If these changes are made in the last equation and the expressions for $G_{r,m,j}$; and H_1 are substituted into the general solution Eq. (4.2.24) there results finally:

$$N_l(z,t;x,y) = \frac{1}{4} N_l^0 e^{-(\frac{\lambda_l}{V_l})z} \phi_l''(t - \frac{z}{V_l}) \quad E_l(a\pm X, \frac{z}{V_l}) E_l(b\pm y, \frac{z}{V_l}) +$$

$$\frac{1}{4} \sum_{j=1}^{l-1} A_l^{(j)} N_j^0 \sum_{m=j}^{l} \frac{e^{-(\frac{\lambda_m}{V_m})z}}{B_m^{(j)}} \sum_{\substack{r=j \\ r\neq m}}^{l} D_{rm}^{(j)} \int_{z/V_m}^{t} E_r(a\pm x, \tau - \frac{z}{V_r}) \cdot$$

$$E_r(b\pm y, \tau - \frac{z}{V_r}) \, e^{-\Delta_{rm}(\tau - \frac{z}{V_m})} \phi_j''(t-\tau) d\tau \qquad (4.2.32)$$

To obtain some insight into the physical character of this solution let us consider the mother nuclide concentration field which is given by the first term for l=1. Dropping the subscript,

$$N(z,t;x,y) = \frac{N^0}{4} e^{-(\frac{\lambda}{V})z} \phi''(t - \frac{z}{V}) \quad E(a\pm x, \frac{z}{V}) E(b\pm y, \frac{z}{V}) \qquad (4.2.33)$$

The effects of lateral diffusion are contained in the two bracketed terms as can be seen from Eq. (4.2.30). As D→0, the error function tends to unity and the solution (4.2.33) tends to:

$$N(z,t;x,y) = N^0 e^{-(\frac{\lambda}{V})z} \phi''(t - \frac{z}{V}) \qquad (4.2.34)$$

which is the well known solution for the dispersion free case. In this case the repository surface emits the mother nuclide in form of a beam of cross sectional area (2ax2b). This beam retains this cross section with increasing distance z from repository, with the nuclide concentration decreasing exponentially. The effects of lateral diffusion can be judged in terms of a boundary layer which builds around this beam. To see this, consider the concentration profile in the y direction at a fixed x position. It is convenient to rewrite E(b±y,z/V) as follows:

$$E(b \pm y, z/V) = \operatorname{erf}\left(\frac{1-y/b}{2\sqrt{\frac{Dz}{Vb^2}}}\right) + \operatorname{erf}\left(\frac{1+y/b}{2\sqrt{\frac{Dz}{Vb^2}}}\right) \tag{4.2.35}$$

This function is shown in Figure 4.2.2 for a number of parameter values (Dz/Vb^2).

In view of this the concentration profile obtained from Eq. (4.2.33) at the repository surface (z=o) is a sharply defined beam. As z increases, lateral diffusion in the y direction takes place. The point where N vanishes can be taken as the edge of a boundary layer. One can estimate this position as follows:

Since $(y/b) > 1$ and $\operatorname{erf}(-\eta) = \operatorname{erf}(\eta)$

$$E\left(y/b, \frac{Dz}{b^2 v}\right) = \operatorname{erf}\left(\frac{\frac{y}{b} + 1}{2\sqrt{\frac{Dz}{vb^2}}}\right) - \operatorname{erf}\left(\frac{(y/b) - 1}{2\sqrt{\frac{Dz}{vb^2}}}\right) \tag{4.2.36}$$

Now $E \approx 0$ when $\dfrac{(y/b) \pm 1}{2\sqrt{\frac{Dz}{Vb^2}}} \approx 1.8$. In case of the negative sign

$$y/b \approx 1 + 3.6\sqrt{\frac{Dz}{Vb^2}} \tag{4.2.37}$$

which shows the growth of the boundary layer with the distance z and its dependence on the dispersion coefficient and the ground water velocity. The equation can be rewritten in dimensionless variable form

$$y^* \approx 1 + 3.6\sqrt{\frac{z^*}{Pe}} \quad , \quad y^* = \frac{y}{b} \, , \; z^* = \frac{z}{b} \quad , \quad Pe = \frac{Vb}{D} \tag{4.2.38}$$

where Pe is the Peclet number. In a comparable way the growth of the boundary layer in the x direction is given by

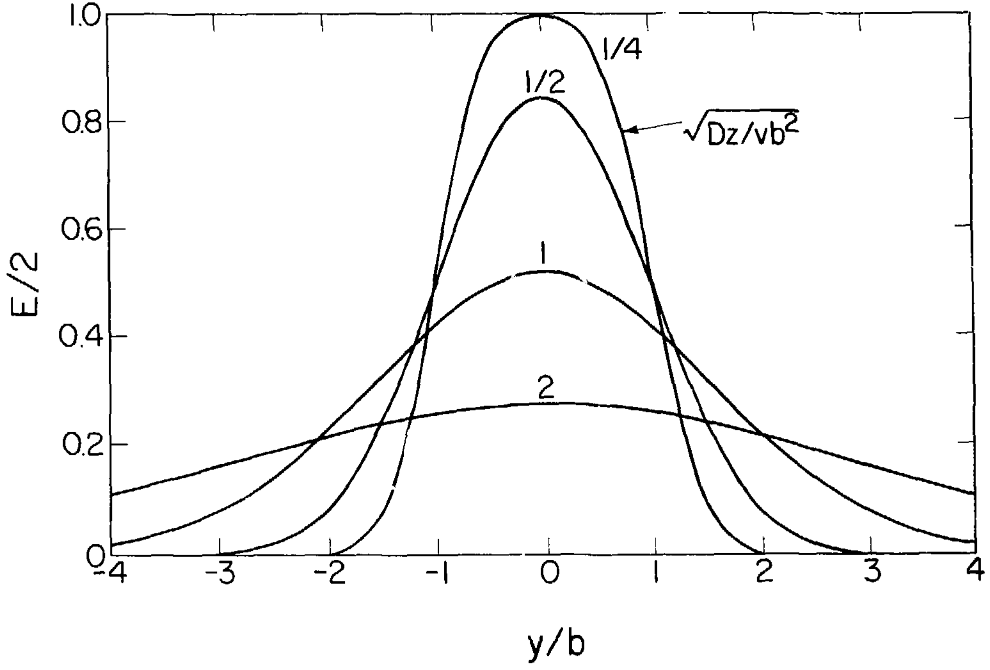

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Figure 4.2.2 E(b±y,z/V) from Equation (4.2.35) as a function of y/b and z/V.

$$x^* \simeq r^* + 3.6\sqrt{\frac{z^*}{Pe}} \quad , \quad r^* = \frac{a}{b} \quad ; \quad x^* = \frac{x}{b} \tag{4.2.39}$$

This enlargement of the effective cross-section due to dispersion causes a decrease in the nuclide concentration over the dispersion free case which can be computed without difficulty from the general solution Eq. (4.2.24).

As discussed above, nuclide transport takes place across the boundary surface of the beam emitted from the repository surface. Suppose the repository wall dimensions are such that b<<a. To estimate when longitudinal dispersion effects become negligible compared to transverse dispersion consider the repository dimensions such that b<<a. In this case, losses from the beam occur primarily in the y direction and those in the z direction can be ignored.

The principal terms in Eq. (4.2.1) which account for dispersion in the y and x direction are of the form

$$D_y \frac{\partial^2 N}{\partial y^2} \quad , \quad D_x \frac{\partial^2 N}{\partial x^2}$$

If dispersion in the transverse direction is characterized by a length scale of order b, dispersion effects in the transverse direction are important compared to longitudinal dispersion if

$$\frac{D_y \frac{\partial^2 N}{\partial y^2}}{D_x \frac{\partial^2 N}{\partial x^2}} > 1 \tag{4.2.40}$$

or for

$$\frac{b^2}{D_y} < \frac{X^2}{D_x} \tag{4.2.41}$$

Treating also longitudinal dispersion effects interior to longitudinal convection yields $\frac{D_x}{v} < x$. Combining this with Eq. (4.2.41) leads to the criterion:

$$\frac{b^2}{D_y} < \frac{D_x}{v^2} \tag{4.2.42}$$

When the inequalities (4.2.41-42) are fulfilled, longitudinal dispersion can be neglected in favor of transverse dispersion. However for small distances from the repository the solution to the complete equation may be needed.

A numerical example of the application of Eq. (4.2.33) is given in Sec. 4.3.2.

4.2.3 Longitudinal and Transverse Dispersion

We now extend the above analysis to include the effect of longitudinal dispersion. In this case the term $D_{zi}\frac{\partial^2 N_i}{\partial z^2}$ is included in Eq. (4.2.1). On setting

$$N_i(z,t;\ x,y) = \exp\left\{\left(\frac{V_i}{2D_{zi}}\right) z - \left(\lambda_i + \frac{V_i^2}{4D_{zi}}\right) t\right\}\ U_i(z,t;x,y) \tag{4.2.43}$$

Eq. (4.2.1) reduces to the simpler form

$$\frac{\partial U_i}{\partial t} = D_{xi}\frac{\partial^2 U_i}{\partial x^2} + D_{yi}\frac{\partial^2 U_i}{\partial y^2} + D_{zi}\frac{\partial^2 U_i}{\partial z^2} +$$

$$+ \frac{K_{i-1}\lambda_{i-1}}{K_i}\ e^{-(\beta_i - \beta_{i-1})t}\ U_{i-1},\ \lambda_o = o,\ i=1,2\ldots \tag{4.2.44}$$

where $\beta_i = -\lambda_i - \frac{V_i^2}{4D_{zi}}$

The boundary conditions are, from Eqs. (4.2.3-4)

$$U_i(o,t;x,y) = N_i^o\ \ S_i(x,y)\ e^{-\beta_i t}\ \phi_i''(t),\ |x|<a, |y|<b, t>o$$

$$= 0,\ |x|>a,\ |y|>b \tag{4.2.45}$$

and the initial conditions

$$U_i(z,o;x,y) = 0 \quad -\infty < (x,y) < \infty, \quad 0 < z < \infty \tag{4.2.46}$$

The solution to this system is obtained with help of the Green's function method discussed by the writer in Section 4.3 of Ref. (H1). In place of Eq. (4.73) the Green's function for this three-dimensional problem is

$$G(z,z',\tau;x-x',y-y') = \frac{1}{8[\pi\tau]^{3/2}[D_{xi}D_{yi}D_{zi}]^{1/2}}\left\{\exp\left(-\frac{1}{4\tau}\left[\frac{(x-x')^2}{D_{xi}}+\frac{(y-y')^2}{D_{yi}}+\frac{(z-z')^2}{D_{zi}}\right]\right)\right.$$
$$\left.-\exp\left(-\frac{1}{4\tau}\left[\frac{(x-x')^2}{D_{xi}}+\frac{(y-y')^2}{D_{yi}}+\frac{(z+z')^2}{D_{zi}}\right]\right)\right\} \tag{4.2.47}$$

Analogous to Eq. (4.74), Ref. (H1) the recursive solution is then given by

$$U_i(z,t;x,y) = [D_{xi}D_{yi}D_{zi}]^{1/3}\int_0^t d\tau\int_{-a}^{a}\int_{-b}^{b} U_i(o,\tau;x',y')\,\frac{\partial G}{\partial z}(o,z',t-\tau,x-x',y-y')dx'\,dy'$$
$$+\frac{K_{i-1}\lambda_{i-1}}{K_i}\int_0^t d\tau\int_0^\infty dz'\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} e^{-(\beta_i-\beta_{i-1})\tau}\,U_{i-1}(z,z',\tau,x',y')G(z,z',t-\tau;x-x',y-y')dx'dy' \,. \tag{4.2.48}$$

The integrations are tedious to carry out and will not be reproduced here. However, the mother concentration is readily evaluated. We consider the case where $D_{x1}=D_{y1}=D_{z1}=D$ and $S_1(x,y)$ is given by Eq. (4.2.25). Only the first term is present in eq. (4.2.48) so,

$$U_1(z,t;x,y) = \frac{zN_1^o}{8(\pi D)^{3/2}}\int_0^t d\tau\int_{-a}^{a}dx'\int_{-b}^{b}dy'\,\frac{e^{-\beta_1\tau}\phi_1''(\tau)}{\tau^{5/2}}\exp\left\{-\frac{(x-x')^2+(y-y')^2+z^2}{4D(t-\tau)}\right\} \tag{4.2.49}$$

In terms of the E function defined in Eq. (4.2.30) this takes on the form

$$U_1(z,t;x,y) = \frac{zN_1^o}{8(\pi D)^{1/2}}\int_0^t e^{-\beta_1(t-\tau)-\frac{z^2}{4D\tau}}\,E(a\pm x,\tau)E(b\pm y,\tau)\phi_1''(t-\tau)\frac{d\tau}{\tau^{3/2}} \tag{4.2.50}$$

If this is combined with Eq. (4.2.43) one obtains, by grouping the exponents as follows

$$-\frac{v}{2D}z + \frac{v^2}{4D}\tau + \frac{z^2}{4D\tau} + \lambda_1\tau = \lambda_1\tau + \left(\frac{z-v\tau}{4D\tau}\right)^2 , \tag{4.2.51}$$

the result

$$N_1(z,t;x,y) = \frac{N_1^0 z}{8(\pi D)^{1/2}}\int_0^t e^{-\frac{(z-v\tau)^2}{4D\tau}-\lambda_1\tau}\,\frac{\phi_1''(t-\tau)}{\tau^{3/2}}E(a\pm x,\tau)E(b\pm y,\tau)d\tau \tag{4.2.52}$$

This answer is in form of a (thermal) dipole which is due to the structure of the Green's function given in Eq. (4.2.47) because G vanishes on the boundary z=o. From a numerical point it is more convenient to represent this result in an alternate form. Let

$$\frac{z}{\sqrt{4D\tau}} = \mu \ , \quad \tau = \frac{z^2}{4D\mu^2} \quad , \quad z\,\frac{d\tau}{\tau^{3/2}} = -4\sqrt{D}\,d\mu \tag{4.2.53}$$

then:

$$N_1(z,t;x,y) = \frac{N_1^0}{2\sqrt{\pi}}\int_{z/2\sqrt{Dt}}^{\infty} e^{-\left(\mu-\frac{vz}{4D\mu}\right)^2-\left(\frac{\lambda_1 z^2}{4D}\right)\frac{1}{\mu^2}}\,\phi_1''\left(t-\frac{z^2}{4D\mu^2}\right)\cdot$$

$$E(a\pm x, \frac{z^2}{4D\mu^2})\,E\left(b\pm y, \frac{z^2}{4D\mu^2}\right)\,d\mu \tag{4.2.54}$$

A comparison with Eq. (4.2.33) shows that now a quadrature is required to find the mother concentration when longitudinal dispersion is present.

4.3 Comparison of Concentrations from an Array of Discrete Sources with Concentrations from an Infinite Plane Source

The purpose of this section is to clarify the effect of transverse dispersion on the migration behavior in a one-dimensional flow field. Parameter studies are made by using the solution in Section 4.1 and 4.2, assuming a radionuclide with no precursor. We compare the concentration fields for a finite plane source, for an array of point sources, and for the infinite plane source.

4.3.1 Parameters Used

The longitudinal and transverse dispersion coefficients D_L and D_T are related to the groundwater velocity v and the molecular-diffusion coefficient Dm by:

$$D_L = \alpha_L v + D_m \tag{4.3.1}$$

$$D_T = \alpha_T v + D_m \tag{4.3.2}$$

Where α_L and α_T are the longitudinal and transverse dispersivities. Usually D_m is of the order of $10^{-2} m^2/yr$, and is small enough to be neglected in (4.3.1) and (4.3.2). There are only limited sources of data concerning α_L and α_T. Schmocker (S1) has suggested values for three geologic media.

Table 4.3.1 Values of α_L and α_T suggested by Schmocker (S1), for $v = 100$ m/yr

	α_L, m	α_T, m	α_L/α_T
Granite	30	10	3
Sandstone	10	3	3
Clay	0.2	0.07	3

Values of a_L and a_T are also seen in the numerical demonstrations of the SWIFT (D1) and IONMIG (R1) codes developed by the Sandia Laboratories for calculating the migration of radionuclides, as shown in Table 4.3.2.

Table 4.3.2 Values of α_L and α_T used by SWIFT (D1) and IONMIG (R1)

	v, m/yr	α_L, m	α_T, m	α_L/α_T
SWIFT	73	150	15	10
IONMIG	Very small	6.1	0.61	10

For the purpose of the parametric studies presented herein, we adopt the range of dispersivities shown in Table 4.3.3.

Table 4.3.3 The Range of Dispersivities Assumed in these Calculations

α_L, m	α_T, m	α_L/α_T
0.1 ~ 150	0.01 ~ 15	3 ~ 10

In these parametric studies, the longitudinal dispersion is first neglected so as to clarify the effect of transverse dispersion. The quantity $\Omega = zD_T/v = a_T z$ then becomes a key parameter in evaluating the effect of the transverse dispersion, as shown later in Section 4.3.2 and 4.3.3.

A configuration of a conceptual repository is shown in Figure 4.3.1.

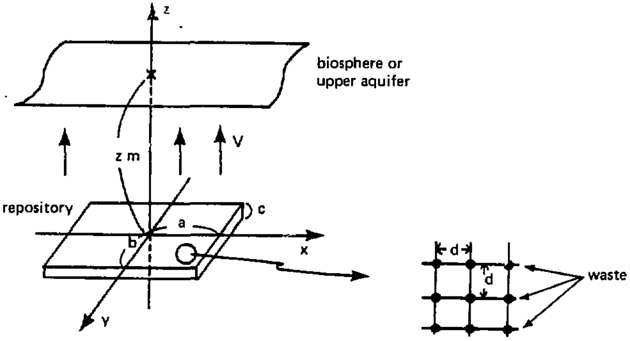

Figure 4.3.1 A conceptual configuration of a geologic respository

These assumptions are made:

1) Waste canisters are buried at a depth up to 1000 m.
2) The waste canisters are arranged in a square planar array (a=b) of overall dimensions a^2.
3) The thickness c of the waste layer is small enough to be neglected.
4) The waste is assumed to consist of 10-year old waste from a PWR, with heat generating rate of:
   Spent fuel: 550W/assembly, with one assembly per canister.
   High-level reprocessing waste: 1740 W/canister.
5) The areal loading of the repository is 20 W/m^2.
6) A maximum number of the waste canisters emplaced in the repository is assumed to be 8000.

Using assumption 1 and Table 4.3.3, the range of zD_T/v is defined as:

$$1 \leq \frac{zD_T}{v} \leq 1.5 \times 10^4 \quad m^2 \tag{4.3.3}$$

The pitch d of the waste canisters is given by assumption 4 and 5:

$$d_{\text{spent fuel}} = \sqrt{\frac{550}{20}} = 5.24 \text{ m} \tag{4.3.4}$$

$$d_{\text{reprocessing waste}} = \sqrt{\frac{1740}{20}} = 9.33 \text{ m} \tag{4.3.5}$$

We will adopt:

d = 5 m for spent fuel
d = 10 m for reprocessing waste

The array dimension a is given by assumption 6 and d = 10 m.

$$a = \sqrt{8000} \times 10 = 890 \text{ m} \tag{4.3.6}$$

We will adopt a = 900 m.

4.3.2 Comparison for a Finite Plane Source and for an Infinite Plane Source

We will assume here that the planar array of canisters can be approximated as a plane source of dimensions 2a × 2b. The solution of the transport equation for the concentration of a radionuclide with no precursor for a finite plane source of dimensions 2a x 2b, without longitudinal dispersion, and for a step release is given by applying Eq. (4.2.33):

$$ {}^{pl}N_1(x,y,z,t) = \frac{{}^{pl}N_1^o}{4}\, e^{-\lambda_1 t}\left[\mathrm{erf}\left(\frac{a+x}{2\sqrt{\frac{D_x z}{v}}}\right) + \mathrm{erf}\left(\frac{a-x}{2\sqrt{\frac{D_x z}{v}}}\right)\right] \cdot $$

$$ \mathrm{erf}\left[\left(\frac{b+y}{2\sqrt{\frac{D_y z}{v}}}\right) + \left(\frac{b-y}{2\sqrt{\frac{D_y z}{v}}}\right)\right] h\,(v_1 t - z) \qquad (4.3.7) $$

Where N_1^o is the concentration of 1^{st} nuclide at the source. The solution for an infinite plane source, without longitudinal dispersion and with the same dissolution rate per unit overall area of source, is (H1):

$$ {}^{\infty}N_1(z,t) = {}^{\infty}N_1^o\, e^{-\lambda_1 t}\, h(v_1 t - z) \qquad (4.3.8) $$

Dividing (4.3.7) by (4.3.8), and noting that ${}^{\infty}N_1^o = {}^{pl}N_1^o$, both λ_1 and t disappear, and there results:

$$ \frac{{}^{pl}N_1(x,y,z,t)}{{}^{\infty}N_1(z,t)} = \frac{1}{4}\left[\mathrm{erf}\left(\frac{a+x}{2\sqrt{\frac{D_x z}{v}}}\right) + \mathrm{erf}\left(\frac{a-x}{2\sqrt{\frac{D_x z}{v}}}\right)\right]\left[\mathrm{erf}\left(\frac{b+y}{2\sqrt{\frac{D_y z}{v}}}\right) + \mathrm{erf}\left(\frac{b-y}{2\sqrt{\frac{D_y z}{v}}}\right)\right] \qquad (4.3.9) $$

If $D_x = D_y = D_T$, Eq. (4.3.9) becomes:

$$ \frac{{}^{pl}N_1(x,y,z,t)}{{}^{\infty}N_1(z,t)} = \frac{1}{4}\left[\mathrm{erf}\left(\frac{a+x}{2\sqrt{\Omega}}\right) + \mathrm{erf}\left(\frac{a-x}{2\sqrt{\Omega}}\right)\right]\left[\mathrm{erf}\left(\frac{b+y}{2\sqrt{\Omega}}\right) + \mathrm{erf}\left(\frac{b-y}{2\sqrt{\Omega}}\right)\right] \qquad (4.3.10) $$

where $\Omega \equiv z D_T / v$.

In (4.3.10), a, b and Ω are the only parameters which affect the ratio of the two solutions. Since Eq. (4.3.10) does not include λ_1 and t, it holds for any nuclide and for any time $t \geq z/v$.

Figure 4.3.2 shows the relative concentration of the mother nuclide at $x = 0$, $y = 0$, as a function of the variable Ω for four sets of dimensions of the planar array. In the case of a relatively small array of 100m x 100m, the relative concentration at $x = 0$, $y = 0$ begins to decrease at $\Omega > 100\ m^2$. For example, if $v = 100$ m/yr and $D_T = 100\ m^2/yr$, the effect of the transverse dispersion becomes important at $z > 100$ m. For the larger array of 800m x 800m, the concentration profile along a normal through the center of the array (at x=y=o) is identical with the concentration profile predicted from no transverse dispersion, over the range of Ω shown here.

The concentration profiles in the direction of groundwater flow, along normals from the edges of the planar array, i.e., at x=a, y=b, will decrease more rapidly with Ω than shown in Figure 4.3.2.

4.3.3 Comparison for an Array of Point Sources and for an Infinite Plane Source

We will make a translation from an array of point sources to an equivalent infinite plane source in the water phase. First, we will translate from a single point source to an equivalent finite plane source of overall dimensions d^2, where d is the pitch between point sources on a square array. We can do this by writing a material balance which expresses the rate at which total waste dissolves into the water. For the point source of initial total atoms W^o, and for a total leach time T, the rate of dissolution of total atom is W^o/T. The point source strength for the nuclide is:

$$\frac{W^o}{T} n_1^o e^{-\lambda_1 t} \tag{4.3.11}$$

where n_1^o is the initial atom fraction of nuclide 1 in the waste.

This total rate of waste dissolution for the equivalent plane source must also equal W^o/T. The plane-source strength must be expressed as the rate of dissolution per unit cross-sectional area of water flow associated with the plane of overall dimension d^2. The initial total waste atoms W^o/S per unit cross-sectional area of water flow, as appearing in Eq. (2.1.20), is then:

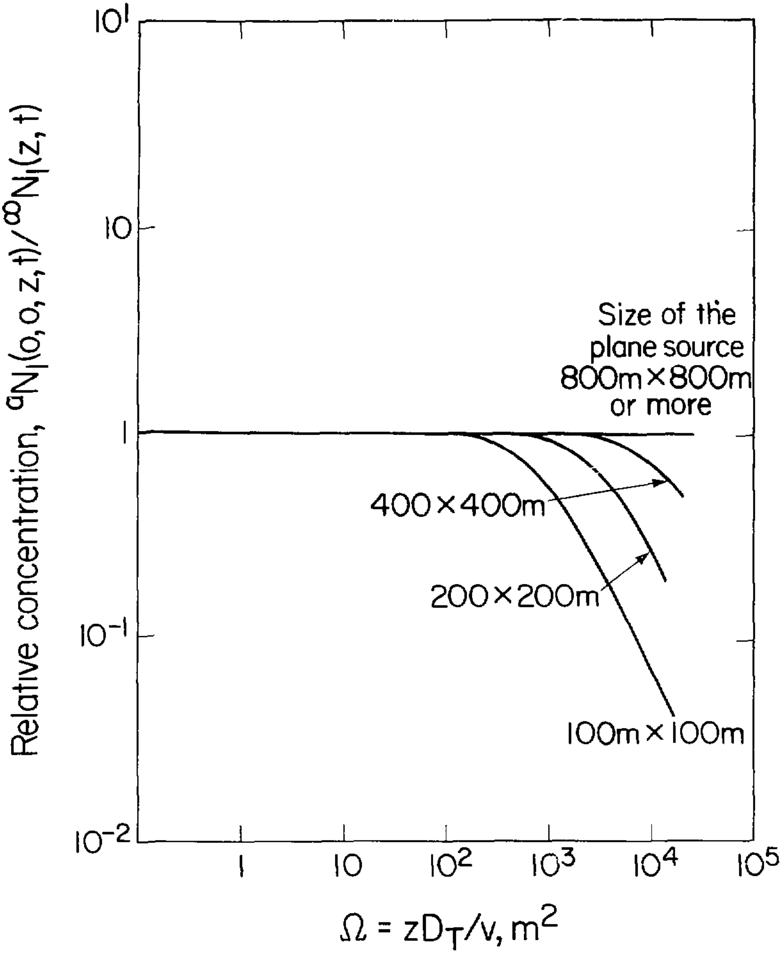

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Figure 4.3.2 Relative nuclide concentration (a finite plane source/an infinite plane source) for four dimensions of planar array as a function of the distance parameter Ω.

$$W_s^o = \frac{W^o}{S} = \frac{W^o}{\epsilon d^2} \tag{4.3.12}$$

where ϵ is the porosity. The strength of the plane source of the nuclide in the water phase is then:

$$\phi_1''(t) = \frac{W^o}{T\epsilon d^2} n_1^o e^{-\lambda_1 t} \tag{4.3.13}$$

We now assume that Eq. (4.3.13) also characterizes the source strength of nuclide $\underline{1}$ for an infinite plane source, so it now defines the plane source for calculating $N_1(z,t)$ from Eqs. (2.1.20) and (2.1.41). Alternatively, for the case of no axial dispersion, we could adopt the simpler form of the solution for the infinite plane source (H1):

$${}^{\infty}N_1(z,t) = {}^{pl}N_1^o e^{-\lambda_1 t} h(t - z/v_1) \tag{4.3.14}$$

Here, by a material balance at the source location, in the absence of axial dispersion, N_1^o is obtained from:

$$\frac{W^o n_1^o}{T} = \epsilon d^2 v\, {}^{pl}N_1^o \tag{4.3.15}$$

Which holds for either a finite or infinite plane source, in the absence of axial dispersion.

Substituting Eq. (4.3.15) into Eq. (4.3.14), and dividing the resulting equation into the point-source equation (4.1.66), we obtain the ratio of the concentration resulting from a single point source to the concentration resulting from an equivalent infinite plane source:

$$\frac{{}^{pt}N_1(x,y,z,t)}{{}^{\infty}N_1(z,t)} = \frac{\epsilon d^2}{4\pi\Omega} \exp\left(-\frac{x^2 + y^2}{4\Omega}\right) \tag{4.3.16}$$

where $\Omega \equiv \frac{zD_T}{v}$

As in the case of Eq. (4.3.10), the concentration ratio of Eq. (4.3.16) is independent of λ_1 and t. It is a function of the axial-distance parameter, Ω.

We can now use Eq. (4.3.16) to predict the concentration ratio for an array of identical point sources. For an array with canisters occupying M positions on the x coordinate and N positions on the y coordinate, the concentration ratio for the array is given by:

$$\frac{{}^{a}N_1(x,y,z,t)}{{}^{\infty}N_1(z,t)} = \frac{1}{{}^{\infty}N_1(z,t)} \sum_{m=1}^{M} \sum_{n=1}^{N} {}^{pt}N_1(x-x_m, y-y_n, z, t) \qquad (4.3.17)$$

where x_m, y_n is the coordinate for each canister location. Eq. (4.3.17) has been used to predict the concentration ratio shown herein.

Figure 4.3.3 shows the distribution of the relative nuclide concentration along the transverse coordinate x, normal to the direction of water flow, for values of the distance parameter Ω of 1, 4, 10, and 100 m^2. The radionuclide source consists of one hundred waste canisters emplaced in a square planar array in the x-y plane. The overall array dimensions are 100m x 100m, with a canister pitch of d = 10m. Sources are located at $\pm x = d/2$, $3d/2$, $5d/2$, etc.; $\pm y = o$, d, 2d, 3d, etc.

For Ω as large as $10m^2$ or greater, the array solution becomes almost identical with the solution for the infinite plane source. For Ω of $10m^2$ or less each concentration profile is periodic with respect to the transverse coordinate, with a period equal to the distance d between adjacent sources. The greatest deviations from the infinite-plane-source solution occur nearer the source, where Ω is small.

Figure 4.3.4 shows the relative nuclide concentration as a function of the distance parameter Ω for an array of point sources, with a source spacing of 10 m and for overall array dimensions varying from 10-canisters x 10-canisters to 80-canisters x 80-canisters. The near-field concentration ratios, for less than 10 m^2, are shown for the transverse peak ($x=d/2$, $y=o$) and for the minimum between adjacent sources ($x=o$, $y=o$). These two branches coincide at greater than about 10 m^2, where the array concentration becomes identical with that from an infinite plane source. The effect of the spacing of the point sources in the array on the convergence to the plane-source solution is shown in Figure 4.3.5, for pitches of 5 m and 10 m.

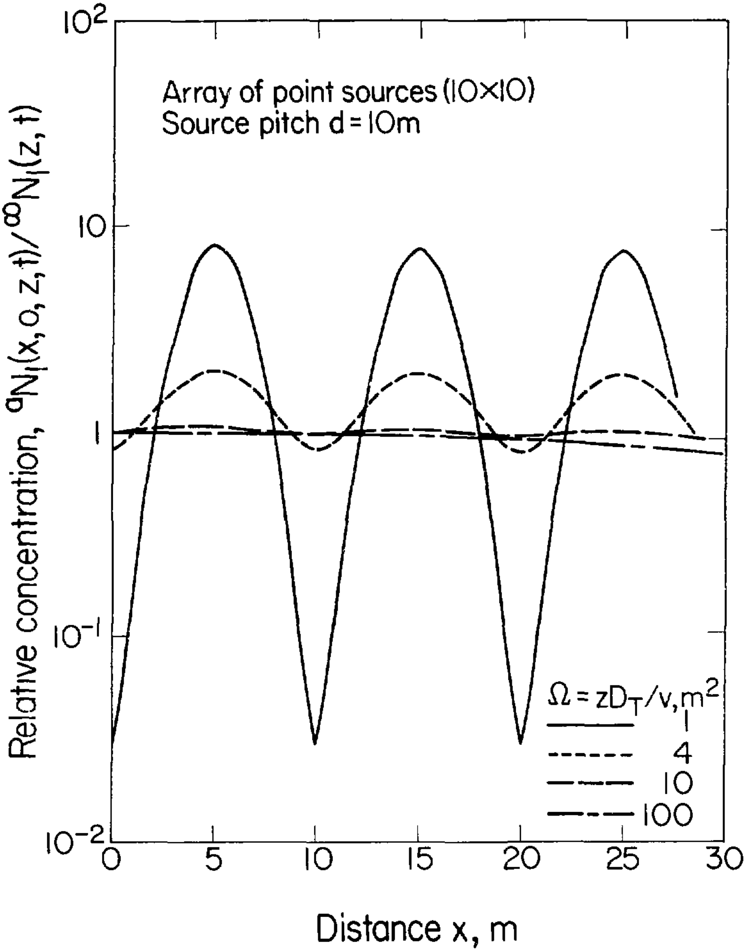

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Figure 4.3.3 Distribution of the relative nuclide concentration (an array of point sources/an infinite plance source) for four values, of along the transverse coordinate x, at y = 0.

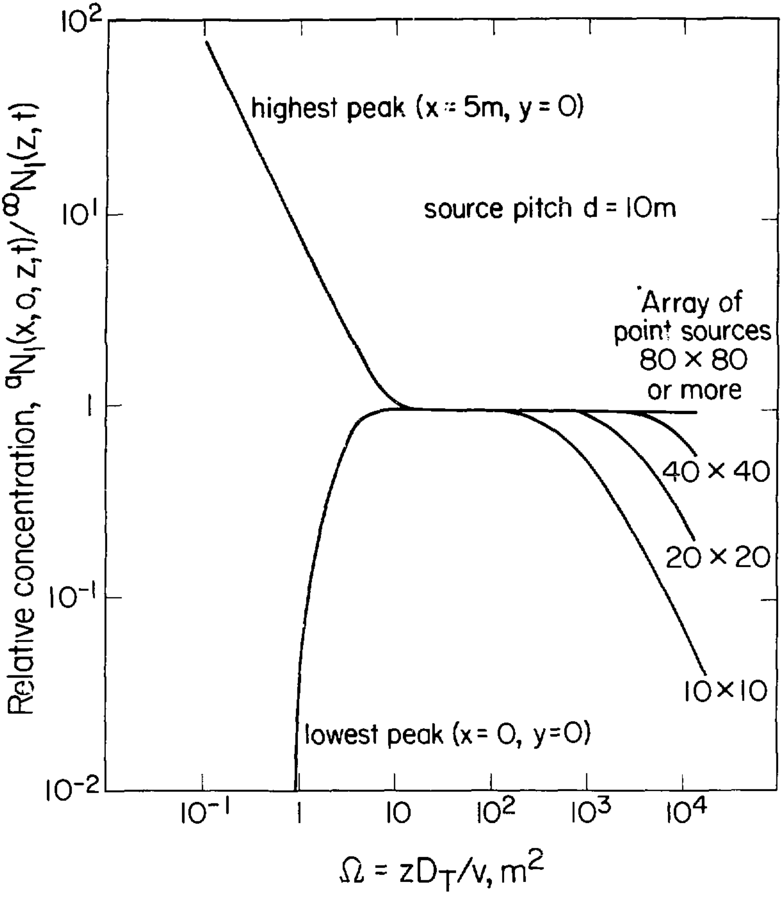

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Figure 4.3.4 Relative nuclide concentration as a function of the distance parameter Ω for an array of point sources at $y = 0$.

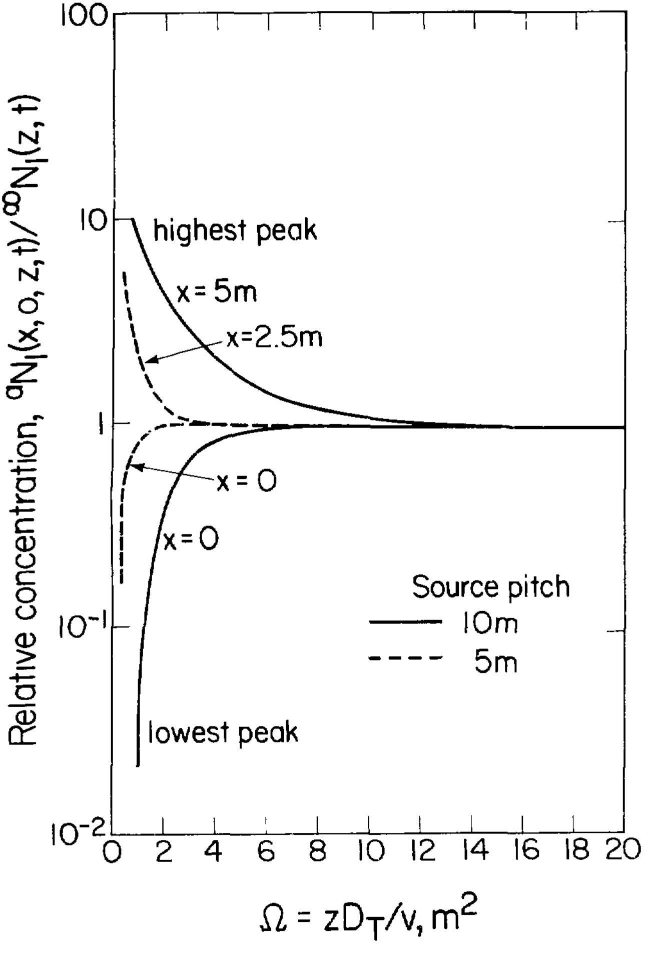

Figure 4.3.5 Effect of pitch of point sources on relative concentration profile (an array of point sources/an infinite plane source). The highest peaks are at $x = 2.5$ m and 5 m and $y = 0$. The lowest peaks are at $x = y = 0$.

Figure 4.3.4 shows that the concentration ratio remains unity until a larger value of the distance parameter is reached, depending upon the size of the array. The array concentration then becomes less than the infinite-plane-source concentration, because of overall transverse dispersion into the projected regions outside the array ($-a>x>a, -b>y>b$). Thus, comparing the concentrations in the porous medium, in a volume projected from the array surface in the direction of water flow, the array concentration will depart significantly from the infinite-plane-source concentration in the near field ($\Omega<\sim10$ m^2) due to discontinuities between discrete point sources in the source plane. These departures disappear at greater distances from the source plane, in the direction of water flow, because of local transverse dispersion, where the concentration field from the array of point sources becomes identical with that for a finite plane source. As has already been shown in Fig. 4.3.2, the concentration from the finite plane source is identical with that from the infinite plane source up to axial distance parameters Ω of about 100 m^2 or greater, for the array size considered herein. The concentration field of the array finally decreases below that of the infinite plane source because of transverse dispersion into the porous medium outside the projected region of the array.

4.3.4 Effect of Transverse Dispersion and Longitudinal Dispersion

When the longitudinal dispersion is taken into consideration, the solution for a point source is no longer the simple form of Eq. (4.3.18) which has a key parameter . Here the effects of longitudinal and transverse dispersion on the concentration profiles were examined by varying the ratio of the two dispersivities for an array of point sources. Parameters for the demonstrations and the array of the sources are shown in Table 4.3.4 and Figure 4.3.6.

Table 4.3.4 Parameters used in evaluating the effects of α_L and α_T

	Figure 4.3.7	Figure 4.3.8
Nuclide: ^{237}Np	Calculation for a fixed D_T and for varying D_L/D_T.	Calculation for a fixed D_L and for varying D_L/D_T
$T_{1/2} = 2.14\times10^6$ yr K_1=100, v = 100m/yr at t=10^4 yr z≈10^3 m	$D_T = 3\times10^2$ m^2/yr $D_L/D_T = 10^{-2}$, 10, 20	$D_L = 3\times10^3$ m^2/yr D_L/D_T = 3, 5, 10

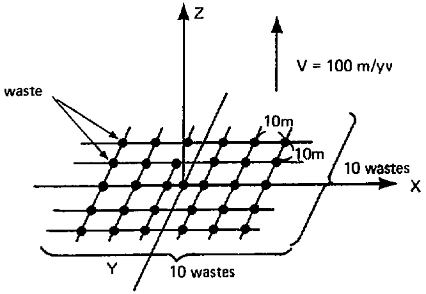

Figure 4.3.6 An array of point sources of 10 x 10.

Figure 4.3.7 shows the effect of varying D_L for a fixed D_T on the concentration profile of ^{237}Np along the x-axis at z=1000 m. In spite of the large variation in the ratio of D_L/D_T, from 10^{-2} to 20, there is little effect of D_L on the peak concentration and overall concentration profiles. However, as shown in Figure 4.3.8, the increasing D_T spreads the concentration in the transverse x direction and lowers the peak concentration at x=0, y=0.

We may conclude that it is not necessary to take into account longitudinal dispersion in evaluating the effect of transverse dispersion.

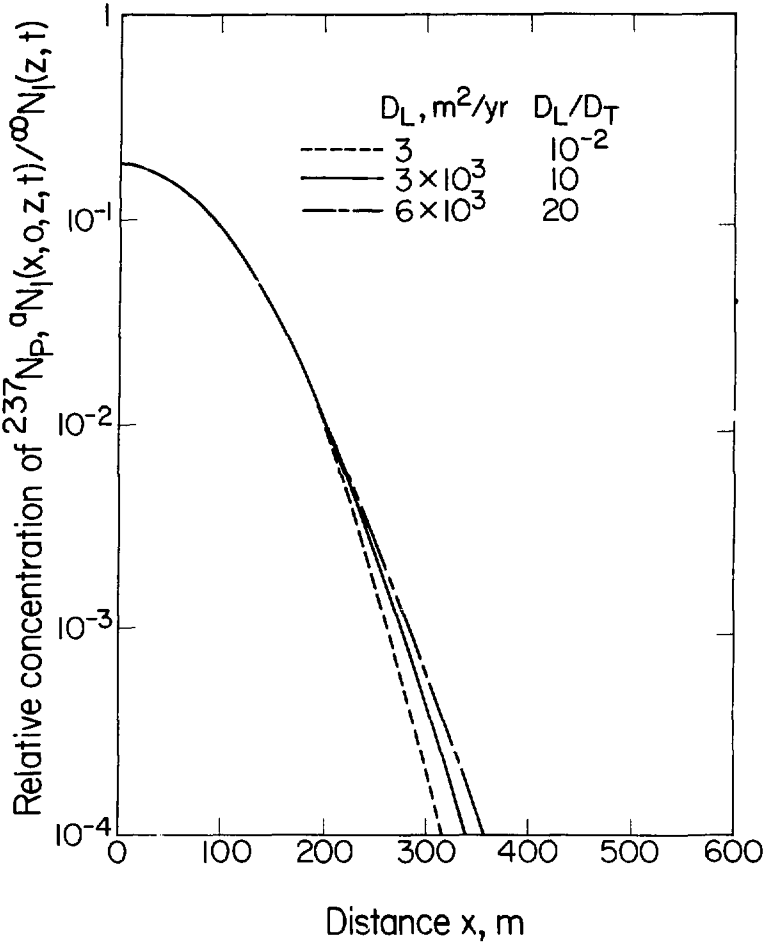

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Figure 4.3.7 Effect of axial dispersion on concentration profiles of ^{237}Np along the transverse coordinate x at $t = 10^4$ yr and $z = 10^3$ m. ($v = 100$ m/yr, $D_T = 3 \times 10^2$ m^2/yr, $K = 100$).

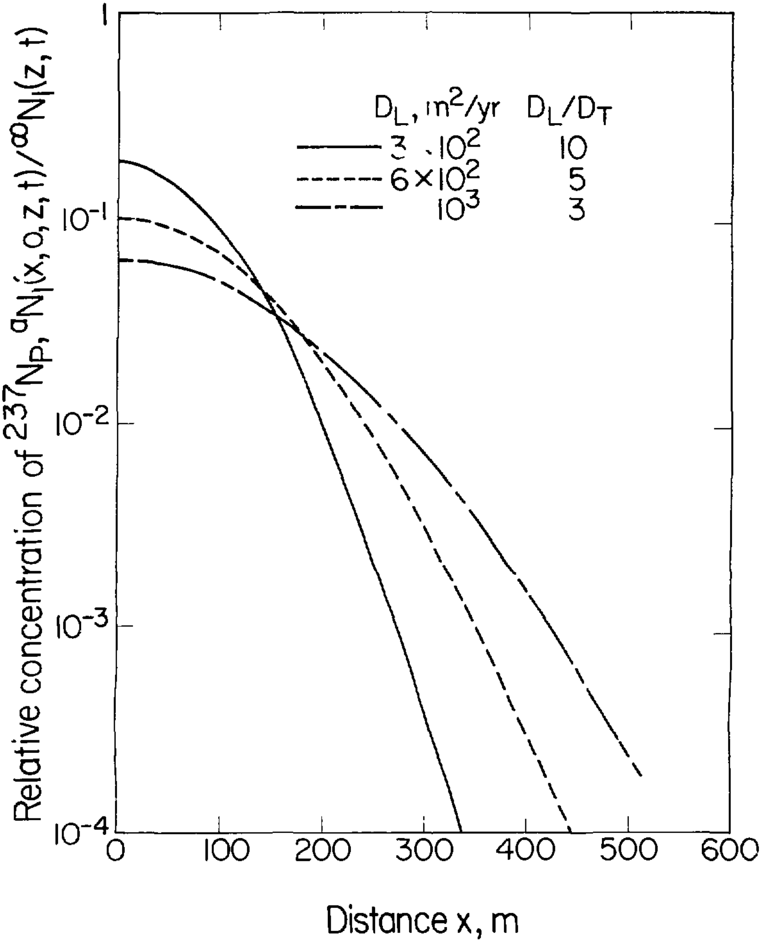

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Figure 4.3.8 Effect of transverse dispersion on concentration profiles of ^{237}Np along the transverse coordinate x at $t = 10^4$ yr and $z = 10^3$ m. ($v = 100$ m/yr, $D_L = 3 \times 10^3$ m^2/yr, $K = 100$).

4.5 Nomenclature

a : 2a x 2b is a size of the plane source

b : 2a x 2b is a size of the plane source

B_{ij} : coefficient of the Bateman equation, Eq. (4.1.25)

c : thickness of the repository

C : arbitrary constant, Eq. (4.1.53)

d : pitch of the emplaced wastes

D : dispersion coefficient, m^2/yr

D_L : Longitudinal dispersion coefficient, m^2/yr

D_m : Molecular diffusion coefficient, m^2/yr

D_T : Transverse dispersion coefficient, m^2/yr

D_x : dispersion coefficient in the x direction, m^2/yr

D_y : dispersion coefficient in the y direction, m^2/yr

D_z : dispersion coefficient in the z direction, m^2/yr

$\vec{\vec{D}}$: overall dispersion tensor

$\vec{\vec{D}}_L$: dispersion tensor in the liquid phase

$\vec{\vec{D}}_S$: dispersion tensor in the solid phase

$E(I,J,K,x,y,z,t)$: function defined by (4.1.48)

$F(u,a,b)$: function defined by (4.1.47)

$F^*(u,a,b)$: function defined by (4.1.78)

$G(\alpha,\mu)$: Gaussian function defined by (4.1.31)

$h(t)$: Heaviside step function

j : Dispersive current (atoms/m^2-sec)

J : Rate of dispersion across a boundary (atoms/sec)

K_{Di} : sorption equilibrium constant

K_i : Retardation coefficient

l : Characteristic dispersion length, m, Eq. (4.1.13)

L : Distance from the repository to a lateral boundary, m

m_i : Total amount of the i^{th} nuclide released in a medium, atoms

M_i^o : Atoms of isotope i at t=0

$\dot{M}_i^o$: Release rate, at t=0

M_{IJKL} : Constant defined by (4.1.56)

N_i^P : Concentration of the i^{th} nuclide released by its precursor, atoms/m^3 of liquid

N_i^R : Concentration of the i^{th} nuclide released from the waste, atoms/m^3 of liquid.

N_{il} : Concentration of the i^{th} nuclide in liquid phase, atoms/m^3 of liquid

N_{is} : Concentration of the i^{th} nuclide in solid phase, atoms/m^3 of solid

N_i^B : Concentration of the i^{th} nuclide resulting from a band release, atoms/m^3 of liquid

N_i^S : Concentration of the i^{th} nuclide resulting from a step release, atoms/m^3 of liquid

N_i^U : Concentration of the i^{th} nuclide in an unbounded medium, atms/m^3 of liquid

$N_1(z,t)$: Concentration of 1^{st} nuclide in water phase for infinite plane source (1-D solution)

${}^{pt}N_1(x,y,z,t)$: Concentration of 1^{st} nuclide in water phase for a point source

${}^{a}N_1(x,y,z,t)$: Concentration of 1^{st} nuclide in water phase for an array of point sources

${}^{pl}N_1(x,y,z,t)$: Concentration of 1^{st} nuclide in water phase for a plane source

N_1^0 : Concentration of 1^{st} nuclide at the plane source

Q : Flow rate of the ground water, m^3/yr

Q_i : Amount of the i^{th} nuclide released in an impulse release, atoms

S : Cross-sectional area of the repository, m^2

S_i : Volumetric source term for the i^{th} nuclide, atoms/m^3.s (cf section II.1)

$S_i(t)$: Source term for the i^{th} nuclide, for point source, atoms/s

S_{iL},S_{is} : Source terms for the i^{th} nuclide, respectively in liquid and solid phase (atoms/sec)

S_{ni} : Release rate at $x=y=z=0$ (Section 4.1.3)

$\mathcal{S}$: General source term defined by (4.1.17)

t : Time variable, yr

T : Duration of nuclide release, yr

v : Water velocity in z direction, m/yr

$\vec{v}$: Flow velocity vector in three dimensions, m/yr

v_i : Velocity of the i^{th} nuclide in the absence of dispersion, m/yr

$\tilde{x}$: Defined by (4.1.8)

$\tilde{y}$: Defined by (4.1.9)

W_0 or W^0 : Initial amount of waste in the repository, atoms

x,y,z : Space variables, m

$\vec{z}$: Unit vector in z direction

$\tilde{z}$: Defined by (4.1.12)

Greek Letters:

α_L : Longitudinal dispersivity

α_T : Transverse dispersivity

β : Space-domain of interest, section 4.1.4

β_{IJ} : Constant defined by (4.1.44)

γ_{IJK} : Constant defined by (4.1.45)

Γ_{ij} : Constant defined by (4.1.43)

δ : Delta function

ϵ : Porosity

θ : Time dummy variable

λ_i : Radioactive decay constant of the i^{th} nuclide, yr^{-1}

Λ_{ij} : Constant defined by (4.1.50)

χ_i : Atom fraction of the i^{th} nuclide in the waste

Ω : zD_T/v

4.6 Literature References

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5. The Transport of Radionuclides in Three-Dimensional Hydrological Flow Fields

5.1. The Formulation of the Governing Equations

This research is concerned with the analysis of the transport of a radionuclide chain in two and three dimensional flow fields in porous media. This analysis is an extension of our analytic work which has up to this point dealt with one-dimensional flow problems only. The results of the analysis are limited by the following

Assumptions: (1) The hydrological ground water flow is time independent, incompressible and is a Darcy type flow.

(2) Dispersion effects are neglected in Sections 5.1 through 5.5. Adsorption equilibrium is assumed and media properties characterizing soil, water and the radio-chemistry are treated as isotropic and constant unless otherwise noted.

Subject to assumption (2) one can write the conservation equation for the nuclide concentration N_n for the nth specie in the water as follows (Ref. H1, Eq. (2.52)):

$$\varepsilon_f K_n \frac{\partial N_n}{\partial t} + \varepsilon_f \nabla(\vec{V}_f N_n) = \varepsilon_f K_{n-1}\lambda_{n-1}N_{n-1} - \varepsilon_f K_n \lambda_n N_n \qquad (5.1.1)$$

$$n=1,2, \ldots , \lambda_0=0$$

In view of the incompressibility assumption, the ground-water velocity $\vec{V}_f$ satisfies the conservation of mass relation

$$\nabla\vec{V}_f = 0 \quad . \qquad (5.1.2)$$

On dropping the subscript f Eqs. (5.1.1) and (5.1.2) combine into

$$K_n \frac{\partial N_n}{\partial t} + \vec{V} \cdot \nabla N_n + K_n\lambda_n N_n = K_{n-1}\lambda_{n-1}N_{n-1} \quad . \qquad (5.1.3)$$

We shall solve this system of equations for $n = 1,2,\ldots$ with $\lambda_0 = 0$ so that $N_1(P,t)$ is the mother concentration. In general terms we shall seek the solutions for $N_n = N_n(P,t)$ in an open domain subject to the following side conditions, see Fig. 5.1.1

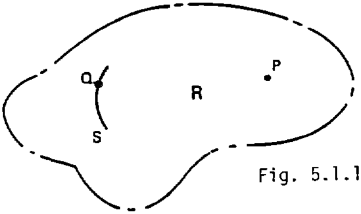

Fig. 5.1.1

The initial conditions at any point $P \epsilon R$ are

$$N_n(P,0) = 0 \tag{5.1.4}$$

and the boundary conditions on S

$$N_n(Q,t) = N_n^0 G_n(Q,t) \quad \text{where} \quad Q \epsilon S \ , \ t > 0 \tag{5.1.5}$$

with

$$G_n(P,t) \equiv 0 \quad , \quad t < 0 \quad , \quad P \epsilon R \cup S \quad . \tag{5.1.6}$$

The G_n functions are the concentration boundary condition functions (such as the Bateman functions) prescribed at the repository surface S and allow a position dependent release of nuclides on S. In view of Assumption (1) one can express the hydrological velocity field $\vec{V}$ as a Darcy flow with help of velocity potential function $\phi(P)$ as

$$\vec{V} = -k \nabla \phi \quad . \tag{5.1.7}$$

If the soil is homogeneous and isotropic. k is the permeability constant of the soil which is assumed constant. By defining the ground water potential function

$$\phi = k\,\Phi \tag{5.1.8}$$

Eq. (5.1.3) becomes in view of Eq. (5.1.7)

$$\frac{\partial C_n}{\partial t} - \frac{1}{K_n}(\nabla\phi)\cdot(\nabla C_n) + \lambda_n C_n = \lambda_{n-1}C_{n-1} \quad , \quad n = 1,2,\ldots \; ; \; \lambda_0 = 0 \tag{5.1.9}$$

where

$$C_n(P,t) \equiv K_n N_n(P,t) \quad . \tag{5.1.10}$$

For the new variables C_n there are the

$$\text{Initial conditions} \quad C_n(P,0) = 0 \quad , \quad P\epsilon R \tag{5.1.11}$$

and

$$\text{Boundary condition} \quad C_n(Q,t) = C_n^0 G_n(Q,t) \quad , \quad Q\epsilon S \quad , \quad t > 0 \tag{5.1.12}$$

and

$$G_n(P,t) \equiv 0 \quad , \quad t < 0 \quad , \quad P\epsilon RUS \tag{5.1.13}$$

$$C_n^0 = K_n N_n^0 \quad . \tag{5.1.14}$$

The Eqs. (5.1.9) through (5.1.14) form the system of governing equations to be solved.

Before proceeding with this task we note the following generalization. If the soil is anisotropic one can take the coordinate axes in the principal directions of permeability. Then Darcy's law states, in place of Eq. (5.1.7), that

$$\vec{V} = \left(-k_{xx}\frac{\partial\Phi}{\partial x} \, , \, -k_{yy}\frac{\partial\Phi}{\partial y} \, , \, -k_{zz}\frac{\partial\Phi}{\partial z}\right) \tag{5.1.15}$$

where $\Phi = \Phi(x,y,z)$ and k_{ii} are the three principal permeability coefficients. By introducing the new coordinates

$$\bar{x} = \frac{x}{\sqrt{k_{xx}}} \quad , \quad \bar{y} = \frac{y}{\sqrt{k_{yy}}} \quad , \quad \bar{z} = \frac{z}{\sqrt{k_{zz}}} \tag{5.1.16}$$

the convection term $(\nabla\phi)\cdot(\nabla C_n)$ in Eq. (5.1.9) remain invariant under the transformation Eq. (5.1.16). Furthermore ϕ is again a harmonic function in the $(\bar{x},\bar{y},\bar{z})$ space which facilitates the solution of Eq. (5.1.9) as we will show later.

In summary, our task is to obtain the general solution (non-recursive) for the system of Eqs. (5.1.9), i.e., the C_n function with the potential function either known through field measurements or to be found by analytical means.

5.2. Two-Dimensional Potential (Darcy) Flows

Suppose the open domain R is two-dimensional and S is one-dimensional. Suppose $\phi(x,y)$ is a two-dimensional potential function related to the stream function $\psi(x,y)$ by the Cauchy-Riemann Equations

$$\frac{\partial\phi}{\partial x} = \frac{\partial\psi}{\partial y} \quad , \quad \frac{\partial\phi}{\partial y} = -\frac{\partial\psi}{\partial x} \quad . \tag{5.2.1}$$

From Eqs. (5.1.7) and (5.1.8) we have then for the fluid velocity components

$$V_x = -\frac{\partial\phi}{\partial x} = -\frac{\partial\psi}{\partial y} \quad , \quad V_y = -\frac{\partial\phi}{\partial y} = \frac{\partial\psi}{\partial x} \tag{5.2.2}$$

With these the governing transport Eq. (5.1.9) reads

$$\frac{\partial C_n}{\partial t} - \frac{1}{K_n}\left(\frac{\partial\phi}{\partial x}\;\frac{\partial C_n}{\partial x} + \frac{\partial\phi}{\partial y}\;\frac{\partial C_n}{\partial y}\right) + \lambda_n C_n = \lambda_{n-1}C_{n-1} \tag{5.2.3}$$

Instead of the cartesian coordinate system (x,y) let us now introduce the new coordinates $\phi(x,y)$, $\psi(x,y)$. It is well known in hydrodynamics that these functions form an orthogonal coordinate system, i.e., they satisfy

$$(\nabla\phi)\cdot(\nabla\psi) = \frac{\partial\phi}{\partial x}\ \frac{\partial\psi}{\partial x} + \frac{\partial\phi}{\partial y}\ \frac{\partial\psi}{\partial y} = 0 \tag{5.2.4}$$

Furthermore $\phi(x,y)$, $\psi(x,y)$ are harmonic functions, i.e., they satisfy Laplaces equation. In transforming the convective term in Eq. (5.2.3) one obtains with help of Eq. (5.2.4)

$$\begin{aligned}\frac{\partial\phi}{\partial x}\ \frac{\partial C_n}{\partial x} + \frac{\partial\phi}{\partial y}\ \frac{\partial C_n}{\partial y} &= \frac{\partial\phi}{\partial x}\left(\frac{\partial C_n}{\partial\phi}\ \frac{\partial\phi}{\partial x} + \frac{\partial C_n}{\partial\psi}\ \frac{\partial\psi}{\partial x}\right)\\ &+ \frac{\partial\phi}{\partial y}\left(\frac{\partial C_n}{\partial\phi}\ \frac{\partial\phi}{\partial y} + \frac{\partial C_n}{\partial\psi}\ \frac{\partial\psi}{\partial y}\right) = \left(\left[\frac{\partial\phi}{\partial x}\right]^2 + \left[\frac{\partial\phi}{\partial y}\right]^2\right)\frac{\partial C_n}{\partial\phi}\end{aligned} \tag{5.2.5}$$

Hence there results for $C_n(\phi,\psi,t)$ the greatly simplified equation

$$\frac{\partial C_n}{\partial t} + \frac{1}{K_n}\ \frac{\partial C_n}{\partial\sigma} + \lambda_n C_n = \lambda_{n-1}C_{n-1}\quad,\quad n = 1,2,\ \ldots\quad,\quad \lambda_0 = 0 \tag{5.2.6}$$

where

$$\begin{aligned}d\sigma &= -\frac{d\phi}{\left(\left[\frac{\partial\phi}{\partial x}\right]^2 + \left[\frac{\partial\phi}{\partial y}\right]^2\right)}\\ &= -\frac{d\phi}{q^2}\quad,\quad q^2 = V_x^2 + V_y^2\end{aligned} \tag{5.2.7}$$

Since the velocity potential has the physical dimensions of a velocity times a length and the denominator is the square of the speed q of the fluid, σ has the dimensions of time. By Eq. (5.2.7), σ can of course be only a function of position $P(x,y)$. We will show that $\sigma(P)$ represents the time needed for a fluid particle to travel along a fixed streamline ψ = const, between two points lying on ψ. This flow is on account of Eq. (5.2.4) in the direction of the gradient of the velocity potential.

Since the gradient of ϕ and σ are thus parallel but of opposite sign one can assume without loss of generality that

$$\frac{\partial \phi}{\partial x}\ \frac{\partial \sigma}{\partial x} + \frac{\partial \phi}{\partial y}\ \frac{\partial \sigma}{\partial y} = -1 \tag{5.2.8}$$

We want to find the behavior of σ as the point (x,y) moves along a fixed streamline $\psi(x,y)$ = constant. Then

$$\begin{aligned} d\psi = 0 &= \frac{\partial \psi}{\partial x}\, dx + \frac{\partial \psi}{\partial y}\, dy \\ &= -\frac{\partial \phi}{\partial y}\, dx + \frac{\partial \phi}{\partial x}\, dy \end{aligned} \tag{5.2.9}$$

by Eq. (5.2.1). Therefore, for some parameter λ, Eq. (5.2.9) is satisfied if

$$dx = \frac{\partial \phi}{\partial x}\lambda \quad , \quad dy = \frac{\partial \phi}{\partial y}\lambda \tag{5.2.10}$$

Therefore

$$\begin{aligned} d\sigma &= \frac{\partial \sigma}{\partial x}\, dx + \frac{\partial \sigma}{\partial y}\, dy \\ &= \left(\frac{\partial \sigma}{\partial x}\ \frac{\partial \phi}{\partial x} + \frac{\partial \sigma}{\partial y}\ \frac{\partial \phi}{\partial y}\right)\lambda \end{aligned} \tag{5.2.11}$$

Comparing this with Eq. (5.2.8) we see that $\lambda = -d\sigma$ and by Eq. (5.2.10)

$$dx = -\frac{\partial \phi}{\partial x}\, d\sigma \quad , \quad dy = -\frac{\partial \phi}{\partial y}\, d\sigma \quad , \tag{5.2.12}$$

along a <u>fixed</u> streamline. But

$$d\phi = \frac{\partial \phi}{\partial x}\, dx + \frac{\partial \phi}{\partial y}\, dy \tag{5.2.13}$$

so along a <u>fixed</u> streamline we have on using Eq. (5.2.12)

$$d\phi = -\left(\left[\frac{\partial \phi}{\partial x}\right]^2 + \left[\frac{\partial \phi}{\partial y}\right]^2\right) d\sigma \tag{5.2.14}$$

which is precisely the relationship, Eq. (5.2.7).

Equation (5.2.7) (or Eq. (5.2.12)) may be used to determine the travel time σ as a function of x and y (or P) since the velocity potential $\phi(x,y)$ is known either by measurement or analysis.

The analytical process of constructing $\sigma(P)$ is simplified by representing the flow field in the complex $z(=x + iy)$ plane. It would then be convenient to use z and $\bar{z}$ as independent variables instead of x and y. Since

$$\frac{\partial}{\partial x} = \frac{\partial}{\partial z} + \frac{\partial}{\partial \bar{z}} \quad , \quad \frac{\partial}{\partial y} = i\left(\frac{\partial}{\partial z} - \frac{\partial}{\partial \bar{z}}\right)$$

one has

$$\frac{\partial}{\partial x} + i \frac{\partial}{\partial y} = 2 \frac{\partial}{\partial \bar{z}} \quad , \quad \frac{\partial}{\partial x} - i \frac{\partial}{\partial y} = 2 \frac{\partial}{\partial z} \tag{5.2.15}$$

On applying the first operator to the complex function $\phi + i\psi$

$$2 \frac{\partial}{\partial \bar{z}} (\phi + i\psi) = \left(\frac{\partial}{\partial x} + i \frac{\partial}{\partial y}\right)(\phi + i\psi)$$

$$= \frac{\partial \phi}{\partial x} + i \frac{\partial \psi}{\partial x} + i \frac{\partial \phi}{\partial y} - \frac{\partial \psi}{\partial y} = 0 \tag{5.2.16}$$

in view of the Cauchy-Riemann Eq. (5.2.1). From this one concludes that $(\phi + i\psi)$ is independent of $\bar{z}$ and a function of z only so that

$$\phi + i\psi = F(z) \tag{5.2.17}$$

F(z) is called the "complex potential." This holomorphic function F(z) allows one to effectively introduce complex variable techniques into the process of determining the details of the underground water flow field. The basic Eq. (5.2.7) can be cast as follows. From Eq. (5.2.17)

$$\frac{\partial \phi}{\partial x} + i \frac{\partial \psi}{\partial x} = F'(z) \tag{5.2.18}$$

so that by Eq. (5.2.1)

$$\frac{\partial\phi}{\partial x} - i\,\frac{\partial\phi}{\partial y} = F'(z) \qquad , \tag{5.2.19}$$

Hence

$$\left(\frac{\partial\phi}{\partial x}\right)^2 + \left(\frac{\partial\phi}{\partial y}\right)^2 = |F'(z)|^2 \tag{5.2.20}$$

which yields

$$d\sigma = -\frac{d\phi}{|F'(z)|^2} \tag{5.2.21}$$

along a fixed streamline.

Equally well one can re-express this as follows: For short hand notation set $W = \phi + i\psi$, so that

$$W = (\phi + i\psi) = F(z) \tag{5.2.22}$$

Consider this equation solved for z in terms of w

$$z = f(w) \tag{5.2.23}$$

Since

$$\frac{dz}{dw} = \frac{1}{\frac{dw}{dz}} = \frac{1}{F'(z)} \tag{5.2.24}$$

One has with Eqs. (5.2.19) and (5.2.2)

$$\frac{dz}{dw} = \frac{1}{V_x - iV_y} \quad , \quad \frac{d\bar{z}}{d\bar{w}} = \frac{1}{V_x + iV_y} \tag{5.2.25}$$

Hence

$$\frac{dz}{dw} \cdot \frac{d\bar{z}}{d\bar{w}} = \frac{1}{V_x^2 + V_y^2}$$

or

$$\left|\frac{dz}{dw}\right|^2 = \frac{1}{q^2(\phi,\psi)} \tag{5.2.26}$$

It is important to note that since z is expressed as a function of w by Eq. (5.2.23) that q is a function of ϕ and ψ. With this the form of Eqs. (5.2.7) or (5.2.21) changes to

$$d\sigma = - \left|\frac{dz}{dw}\right|^2 d\phi \tag{5.2.27}$$

In summary the key formulas for the computation of the time distance function $\sigma(P)$ are Eqs. (5.2.7) and (5.2.8), (the latter is not explicit in σ), (5.2.21) and (5.2.27). The use of these formulas will be illustrated in Section 5.4. The reader unfamiliar with the use of the complex potential in hydrodynamics may find the following remarks of use. Our original formulation of the two dimensional groundwater flow was represented in an (x,y) or (z) plane. the potential and stream function are harmonic functions in that plane, i.e., they satisfy Laplaces equation

$$\nabla^2\phi = 0 \quad , \quad \nabla^2\psi = 0 \quad \text{in} \quad R \tag{5.2.28}$$

All this has now been replaced by a formulation in terms of the single complex potential function $W = F(z)$ which is a holomorphic function of $z \epsilon R$. However, the solution of the flow field must not only satisfy the partial differential Eqs. (5.2.24) but also certain conditions along the boundary of R. Hence in terms of the complex potential, $W = F(z)$ must be holomorphic in R and in addition its real and imaginary parts must satisfy specific boundary conditions on S, the boundary of R.

In some cases of ground water flow, along a given part of the boundary S in the z plane, either the potential or the stream function assumes a constant value. Since lines of constant ϕ or ψ represent straight lines in the $W = F(z)$ plane, the original boundary conditions along S map into straight lines in the w plane defining a contour K. The geometric disposition of the mapping is then as shown below for a particular example

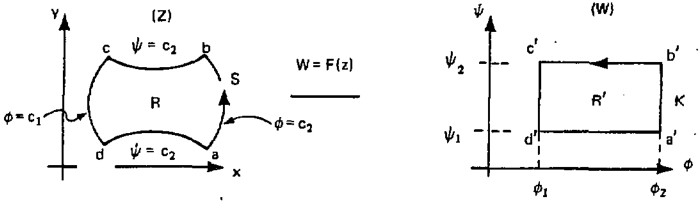

Fig. 5.2.1

Here R has mapped onto R' and the boundary points have their corresponding images. Now suppose that one has found that particular mapping function $w = F(z)$ that satisfies the required boundary conditions on K, then F(z) gives the complete solution to the problem because it yields by Eq. (5.2.17) both the potential function $\phi(x,y)$ and the stream function $\psi(x,y)$. This is a direct consequence of the following Theorem (Ref. C1, Section 9.4). Let D be an open set in the z-plane whose boundary B is a Jordan contour. Let D map by $w = w(z)$ where $w(z)$ is holomorphic, onto an open set E in the w-plane whose boundary C is a Jordan contour. Suppose

$$\frac{\partial^2\phi}{\partial x^2} + \frac{\partial^2\phi}{\partial y^2} = 0 \quad \text{in} \quad D$$

$$a(x,y)\ \phi(x,y) + b(x,y)\ \frac{\partial\phi(x,y)}{\partial n} = c(x,y) \quad \text{on} \quad B \quad ,$$

where n is the distance measured perpendicular to B. Then

$$\frac{\partial^2\phi(u,v)}{\partial u^2} + \frac{\partial^2\phi(u,v)}{\partial v^2} = 0 \quad \text{in} \quad E$$

$$a(u,v)\ \phi(u,v) + b[u,v]\left|\frac{dw}{dz}\right| \frac{\partial}{\partial N}\phi(u,v) = c(u,v) \quad \text{on} \quad C$$

where N is the distance measured perpendicular to C. The same theorem hold for the stream function ψ. The gist of the theorem is that the mapping $w = F(z)$ preserves the harmonic character of ϕ and ψ in going from the R into the R' plane. Furthermore for "simple" boundary conditions, where the coefficients b is equal to zero, the mapping assures the invariance of the boundary conditions. The solution for such "simple" boundary value problems can hence be obtained by determining the conformal mapping function $w = F(z)$ which maps the region R in the z-plane into the region R' in the w-plane. For this we have the extensive apparatus of conformal mapping techniques available to us. Some illustrations are given in Section 5.4.

Returning to the evaluation of $\sigma(P)$, the key equations for σ are integrated in the direction of decreasing potential ϕ. The constant of integration is chosen so that

$$\sigma(Q) = 0 \quad \text{for} \quad Q \epsilon S \tag{5.2.29}$$

$\sigma(P)$ then attains positive values downstream of the repository surface in its "shadow region"

$$\sigma(P) > 0 \quad . \tag{5.2.30}$$

These properties of the travel time function are used in its construction.

As a very simple example consider the case of a flow field induced by a sink located at $z = 0$. The zero potential line, i.e., the aquifer is located at $r = a$. Adopting polar coordinates and

$$\phi = -K \ \ln\,(a/r) \quad , \quad K\left(\frac{cm^2}{sec}\right) > 0 \tag{5.2.31}$$

and

$$\psi = K\theta \tag{5.2.32}$$

Eq. (5.2.17)

$$F(z) = -K \ \ln (a/z) \tag{5.2.33}$$

Then $F'(z) = K/z$ and so by Eq. (5.2.21)

$$d\sigma = -\frac{r^2}{K^2} \, d\phi \tag{5.2.34}$$

But by Eq. (5.2.31), $d\phi = (K/r)\, dr$, so $d\sigma = -\frac{r}{K}\, dr$. Therefore, with the initial condition Eq. (5.2.29)

$$\sigma(P) = \frac{1}{2K}(a^2 - r^2) \quad , \quad 0 \le r \le a \tag{5.2.35}$$

Fig. 5.2.2 shows the time-path history of a fluid element leaving the ring $r = a$ at $\sigma(a) = 0$,

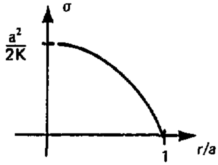

Fig. 5.2.2

5.3. The Solution of the Two Dimensional Transport Equation

We established in Section 5.2 that with the new orthogonal coordinate system (ϕ, ψ) the nuclide transport equation system takes on the form given by Eqs. (5.2.6) and (5.2.7)

$$\frac{\partial C_n}{\partial t} + \frac{1}{K_n} \frac{\partial C_n}{\partial \sigma} + \lambda_n C_n = \lambda_{n-1} C_{n-1} \qquad n = 1,2, \ldots ; \lambda_0 = 0 \tag{5.3.1}$$

$$d\sigma = -\frac{d\phi}{q^2} \tag{5.3.2}$$

for the functions $C_n(\phi,\psi,t)$. We established the fact that $\sigma(P)$ is a scalar function, with the properties that, see Eqs. (5.2.29) and (5.2.30),

$$\sigma(Q) = 0 \quad \text{for} \quad Q \epsilon S \tag{5.3.3}$$

$$\sigma(P) > 0 \quad \text{for} \quad P \epsilon R \quad , \tag{5.3.4}$$

and that the gradient of σ is parallel to the gradient of ϕ, see Eq. (5.2.8). This implies that $\nabla\sigma$ is parallel with the streamlines $\psi(x,y)$. These properties can be written

$$(\nabla\phi)\cdot(\nabla\sigma) = -1 \tag{5.3.5}$$

or as the orthogonality condition

$$(\nabla\psi)\cdot(\nabla\sigma) = 0 \tag{5.3.6}$$

Consider now the simple case that the boundary S is contracted to a single point which represents the repository as shown in Fig. 5.3.1.

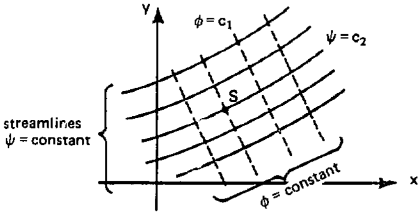

Fig. 5.3.1

According to the properties of σ, discussed in Section 5.2 the fluid moving along the streamline $\psi(x,y) = C_1$ which passes through S will be the carrier of the radionuclides and we can determine their travel time history by integration of Eqs. (5.3.1). However, these equations are precisely of the type for which recursive as well as general solutions were already derived by the author (Ref. H1). It was shown there, see Eq. (4.77), that

$$\frac{\partial N_i}{\partial t} + V_i \frac{\partial N_i}{\partial z} + \lambda_i N_i = \frac{K_{i-1}}{K_i} \lambda_{i-1} N_{i-1} \qquad i = 1,2, \ldots , \lambda_0 = 0 \tag{5.3.7}$$

with

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

$$N_i(z,0) = 0 \quad , \quad z > 0$$

has recursive solutions given by Eqs. (4.82) and (4.85) of Ref. H1 and the general solution given by Eq. (4.127). To make the comparison complete we need to append the side conditions (Eqs. (1.1.11) and (1.1.12)) to Eq. (5.3.1). The boundary points Q in the functions C_n and g_n are now described by the single point $\phi = c_1$, $\psi = c_2$, see Fig. 5.3.1, so the side conditions are

$$\text{I.C.} \quad C_n(P,0) = C_n(\phi,\psi,0) = 0 \quad , \quad \text{all } \phi,\psi \tag{5.3.8}$$

$$\text{B.C.} \quad C_n(Q,t) = C_n(c_1,c_2,t) = c_n^0 g_n(c_1,c_2,t) \quad , \quad t > 0$$

By recalling the scaling relation (Eq. (5.1.10)), the comparison between the equation system (Eqs. (5.3.11) and (5.3.8) with (5.3.7) is immediate and one can write down at once the solutions with help of the above mentioned equations.

Before doing this we first generalize the repository shape to a contour segment. We introduce at this point a mild but convenient restriction in that we assume that S coincides with a potential line as shown in Fig. 5.3.2 In this case the nuclide transport occurs along the streamlines intersecting S forming the shadow region R'. This restriction is mathematically described by the condition

$$(\nabla\phi)\cdot(\nabla g_n(Q,t)) = 0 \quad , \quad t \geq 0 \qquad (5.3.9)$$

But in view of the Lemma of Section 5.2 this means that $g_n(Q,t)$ is independent of ϕ and can thus only depend on ψ. Hence the boundary condition becomes

$$C_n(\psi,t) = C_n^0 g_n(\phi_0,\psi,t) \qquad (5.3.10)$$

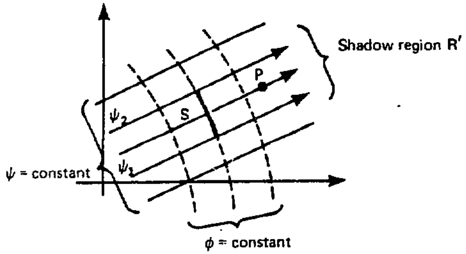

Fig. 5.3.2

As can be seen from Fig. 5.3.2, ψ is a function of the position P in the x-y coordinate system. P from now on is defined as a point lying on a trajectory (streamline) passing through the downstream side of the repository surface S. Thus one can write in place of Eq. (5.3.10)

$$C_n(\psi,t) = C_n^0 g_n(P,t) \quad , \quad P\epsilon R' \qquad (5.3.11)$$

For later use we generalize Eq. (5.3.9) for the scalar function $g_n(P,t)$ to

$$(\nabla\phi)\cdot(\nabla g_n(P,t)) = 0 \quad P\epsilon R' \ , \ t > 0 \qquad (5.3.12)$$

Equation (5.3.10) is a very general relation in that it specifies a concentration boundary condition which can vary on each streamline penetrating the repository surface S. Since the analytical nearfield solution to the nuclide migration problem is as yet unknown a reasonable assumption consists in considering a spatially constant release from S so that one can write the separable form of the boundary condition

$$C_n(\psi,t) = C_n^o\{h(\psi - \psi_1) - h(\psi - \psi_2)\}\ \tilde{g}_n(t) \quad , \quad \psi_1 < \psi_2 \tag{5.3.13}$$

The bracketed term represents a step function in ψ with ψ_1 and ψ_2 the bounding streamlines of the repository contour, see Fig. 5.3.2, and $\tilde{g}_n(t)$ the release rate.

We now set out the solutions for the above equations in terms of the original $N_i(P,t)$ variables, see Eq. (5.1.10).

Recursive Solution

We quote only one of these, i.e., Eq. (4.95), Ref. H1

$$\begin{aligned} N_n(P,t) = \sum_{r=1}^{n-1} \Big\{ & e^{-\lambda_r K_r \sigma(P)} N_{n,r}(P,t - K_r\sigma(P))h(t - K_r\sigma(P)) \\ & - e^{-\lambda_n K_n \sigma(P)} N_{n,r}(P,t - K_n\sigma(P))\ h(t - K_n\sigma(P))\Big\} \\ & + N_n^o e^{-\lambda_n K_n \sigma(P)} g_n(P,t - K_n\sigma(P)) \end{aligned} \tag{5.3.14}$$

where

$$N_{n,r}(P,\tau) = \frac{\lambda_{n-1}K_{n-1}}{K_n - K_r}\ e^{-\mu_{nr}\tau} \int_0^{\tau} N_{n-1,r}(P,\tau')\ e^{\mu_{nr}\tau'}\ d\tau'$$

General Solution

Here we make use of the authors result, Eq. (4.127), Ref. H1.

$$\begin{aligned} N_n(P,t) = {} & N_n^o e^{-\lambda_n K_n \sigma(P)}\ g_n(P,t - K_n\sigma(P)) \\ & + \sum_{j=1}^{n-1} A_n^{(j)} N_j^o \sum_{m=j}^{n} \frac{e^{-\lambda_m K_m \sigma(P)}}{B_m^{(j)}} \sum_{\substack{r=j \\ j\neq m}}^{n} D_{rm}^{(j)}(g_{rm}(t) * g_j(P,t)) \end{aligned} \tag{5.3.15}$$

The symbols have the same meaning as defined earlier except that with $\Delta_{rm} = \mu_{rm}$

$$g_{rm}(t) = \begin{cases} 0 \ , \ t < K_m\sigma(P) \\ e^{-\mu_{rm}(t-K_m\sigma(P))} \ , \ t > K_m\sigma(P) \end{cases} \quad . \tag{5.3.16}$$

The formula for the mother nuclide (n = 1) is

$$N_1(P,t) = N_1^0 \ e^{-\lambda_1 K_1 \sigma(P)} g_1(P, t - K_1\sigma(P)) \tag{5.3.17}$$

and that for the first daughter, n = 2, from Eq. (5.3.14)

$$\begin{aligned} N_2(P,t) = {} & e^{-\lambda_1 K_1 \sigma(P)} N_{2,1}(P, t - K_1\sigma(P)) \, h(t - K_1\sigma(P)) \\ & - e^{-\lambda_2 K_2 \sigma(P)} N_{2,1}(P, t - K_2\sigma(P)) \, h(t - K_2\sigma(P)) \\ & + N_2^0 \ e^{-\lambda_2 K_2 \sigma(P)} g_2(P, t - K_2\sigma(P)) \end{aligned} \tag{5.3.18}$$

where

$$N_{21}(P,\tau) = \frac{\lambda_1 K_1 N_1^0}{K_2 - K_1} \ e^{-\mu_{21}} \int_0^{\tau} g_1(P,\tau') \ e^{\mu_{21}\tau'} d\tau'$$

The corresponding expression from Eq. (5.3.15) is

$$\begin{aligned} N_2(P,t) = {} & N_2^0 \ e^{-\lambda_2 K_2 \sigma(P)} g_2(P, t - K_2\sigma(P)) \\ & + A_2^{(1)} N_1^0 \left\{ \frac{D_{21}^{(1)'}}{B_1^{(1)}} e^{-\lambda_1 K_1 \sigma(P)} \left[g_{21}(t) \circledast g_1(P,t) \right] \right. \\ & \left. + \frac{D_{12}^{(1)}}{B_2^{(1)}} e^{-\lambda_2 K_2 \sigma(P)} \left[g_{12}(t) \circledast g_2(P,t) \right] \right\} \end{aligned} \tag{5.3.19}$$

This can be shown to agree with the previous result. As a simple illustration consider again the nuclide transport in a flow field caused by a sink located at $z = 0$. The time-distance function was found in Eq. (5.2.35). The matriarchy can now be constructed from Eqs. (5.3.15). Since the $g_n(P,t)$ depend, in a two-dimensional flow field, only on the stream function, by Eqs. (5.2.29) and (5.3.10) we may consider these, functions of θ and t only, i.e., $g_n = g_n(\theta,t)$. By Eq. (5.3.17) the mother concentration is

$$N_1(P,t) = e^{-\frac{\lambda_1 K_1}{2K}(a^2 - r^2)} N_1^o \; g_1\left(\theta, t - \frac{K_1}{2K}(a^2 - r^2)\right) \tag{5.3.20}$$

If the repository is located between the rays $\theta_1 \leq \theta \leq \theta_2$ and the emission is independent of θ,

$$N_1(P,t) = e^{-\frac{\lambda_1 K_1}{2K}(a^2 - r^2)} N_1^o \left\{h(\theta-\theta_1)-h(\theta-\theta_2)\right\} g_1\left(\left(t-\frac{K_1}{2K}(a^2 - r^2)\right)\right) \tag{5.3.21}$$

5.4. <u>The Computation of the Time-Position Function in Two-Dimensional Flow Fields</u>

It is apparent from the previous discussion that the principal task in solving the governing equations is the determination of the time-position function $\sigma(P)$. Once this function has been found, its substitution into formulas (5.3.14) or (5.3.15) yields the analytical solution of nuclide transport along a given streamline.

In this section a number of illustrations are given for the determination of $\sigma(P)$. Both isotropic and non-isotropic media are considered.

A. <u>The Source and Sink Flow Field</u>

Consider a recharge and a pumping well in an infinite, homogeneous and isotropic medium. The flow field is assumed to be two-dimensional and the two wells are idealized by a line source and a line sink of equal strength μ.

With the source located at $(a,0)$ and the sink at $(-a,0)$ the complex potential function for this flow field is given by

$$W = \phi + i\psi = \mu \ln\left(\frac{a - z}{a + z}\right) , \quad \mu < 0 \tag{5.4.1}$$

The multiple valued logarithm is made single valued by cutting the complex z plane along the real axis for $\mathrm{Re}(z) < -a$, $\mathrm{Re}(z) > a$ as shown in Fig. 5.4.1.

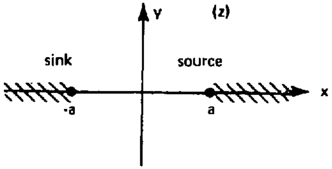

Fig. 5.4.1

Equation (5.4.1) allows one to find the inverse function $z = f(w)$ introduced in Eq. (5.2.23) so that one can employ the algorithm for computing $\sigma(P)$ from Eq. (5.2.27). It follows from Eq. (5.4.1) that

$$e^{w/\mu} = \frac{a - z}{a + z} \tag{5.4.2}$$

which yields

$$z = -a \tanh\left(\frac{w}{2\mu}\right) \quad , \qquad \bar{z} = -a \tanh\left(\frac{\bar{w}}{2\mu}\right) \quad .$$

With this

$$\begin{aligned} \left|\frac{dz}{dw}\right|^2 &= \left(\frac{a}{2\mu}\right)^2 \left[\mathrm{sech}\left(\frac{w}{2\mu}\right) \mathrm{sech}\left(\frac{\bar{w}}{2\mu}\right)\right]^2 \\ &= 4\left(\frac{a}{2\mu}\right)^2 \left\{\cosh\left(\frac{w + \bar{w}}{2\mu}\right) + \cosh\left(\frac{w - \bar{w}}{2\mu}\right)\right\}^{-2} \\ &= \left(\frac{a}{\mu}\right)^2 \left\{\cosh\left(\frac{\phi}{\mu}\right) + \cos\left(\frac{\psi}{\mu}\right)\right\}^{-2} \end{aligned} \tag{5.4.3}$$

Hence

$$d\sigma = -\left(\frac{a}{\mu}\right)^2 \frac{d\phi}{\left[\cosh\left(\frac{\phi}{\mu}\right) + \cos\left(\frac{\phi}{\mu}\right)\right]^2} \quad . \tag{5.4.4.}$$

This is a particularly useful form for integrating σ because, as explained in detail in Section 5.2, the stream function ψ is held constant. With the initial condition (Eq. (5.2.29)) taken on the potential line $\phi = \phi_0$

$$\sigma(P) = \frac{a^2}{\mu} \int_{\phi/\mu}^{\phi_c/\mu} \frac{dx}{[\cosh x + c]^2} \tag{5.4.5}$$

where

$$c = \cos(\psi/\mu) \tag{5.4.5a}$$

The quadrature can be expressed in terms of elementary function. For $c \neq \pm 1$

$$\sigma(P) = \frac{a^2}{\mu} \frac{1}{1 - c^2} \left\{ \frac{\sinh x}{c + \cosh x} - \frac{2c}{\sqrt{1 - c^2}} \tan^{-1} \left(\frac{1}{1} + \frac{c}{c} \tanh \frac{x}{2} \right) \right\} \Bigg|_{(\phi/\mu)}^{(\phi_0/\mu)} \tag{5.4.6}$$

For $c = \pm 1$ which corresponds to the stream lines $\psi = 0$ and $\psi = 2\pi$, the integral in Eq. (5.4.5) yields

$$\sigma(P) = \frac{a^2}{\mu} \left\{ \frac{1}{2} \tanh\left(\frac{x}{2}\right) - \frac{1}{6} \tanh^3\left(\frac{x}{2}\right) \right\} \Bigg|_{(\psi/\mu)}^{(\psi_0/\mu)} \quad . \tag{5.4.7}$$

This last result holds on the geodesic streamline connecting the source and sink. The streamline flow pattern is shown in Fig. 5.4.2. The circles with center on the x axis represent the lines of constant potential which varies over the range $-\infty < \phi < \infty$. The circles with center on the y axis represent the stream lines ψ for $0 \leq \psi \leq 2\pi$. For a source point located anywhere in the field, say at (ϕ_0, ψ_0), the Eqs. (5.4.6) or (5.4.7) yield the travel time along ψ_0 towards the sink.

B. The Source in a Uniform Flow Field

Suppose a source located at the origin is superposed on a uniform field of flow in the x direction. For this the complex potential is given by

$$w = \phi + i\psi = F(z) = \mu \ln z + Uz \quad . \tag{5.4.8}$$

μ and U are real constants, which characterize the source strength and the flow field velocity respectively.

The streamlines of this flow pattern are shown in Fig. 5.4.3. The streamlines are symmetrical about the x-axis. There is no flow across this axis. The dividing streamline BAC passes through the stagnation point A and separates the flow into two regions. One can suppose this dividing streamline to be replaced by an impenetrable boundary. The flow pattern

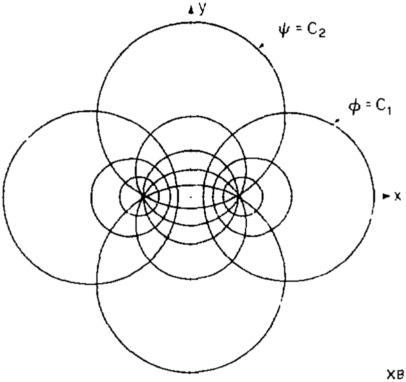

Figure 5.4.2 Potential and streamlines for the point source and point sink flow.

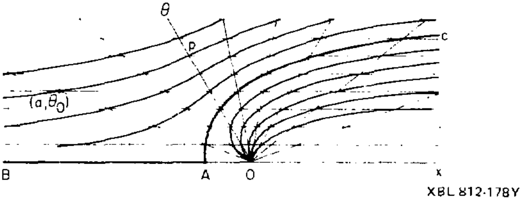

Figure 5.4.3 Streamlines for the uniform flow field with a point source.

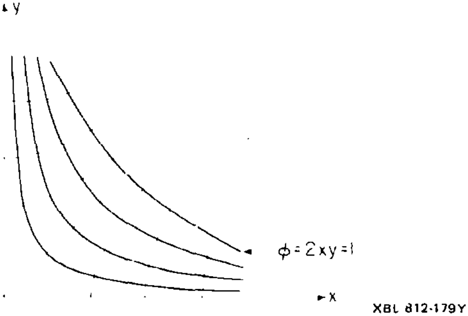

Figure 5.4.4 Potential and streamlines for the stagnation point flow.

above the streamline BAC offers several interesting physical interpretations. For example, one can regard Eq. (5.4.8) as giving the ground water flow in a homogeneous isotropic medium in the neighborhood of one end of a long bluff impenetrable substrate. The repository surface S could be situated as shown in Fig. 5.4.3.

Equation (5.4.8) cannot be solved readily for $z = f(w)$ so it is best in this case to consider $w = F(z)$ and employ Eq. (5.2.21) for the determination of $\sigma(P)$. From Eq. (5.4.8)

$$F'(z) = \frac{\mu}{z} + U \quad . \tag{5.4.9}$$

Therefore by Eq. (5.2.21)

$$d\sigma = -\frac{|z|^2}{|\mu + Uz|^2}\, d\phi \quad . \tag{5.4.10}$$

Now introduce the polar coordinates (r,θ) and set $z = re^{i\theta}$. By Eq. (5.4.8)

$$\phi = \mu \ln r + Ur \cos\theta \tag{5.4.11}$$

$$\psi = \mu\theta + Ur \sin\theta \tag{5.4.12}$$

Along a fixed streamline it follows from the last equation that

$$U \sin\theta\, dr = -(\mu + Ur \cos\theta)\, d\theta \tag{5.4.13}$$

Therefore by Eq. (5.4.11)

$$\begin{aligned} Ur \sin\theta\, d\phi &= U \sin\theta(\mu + Ur \cos\theta)\, dr - U^2r^2 \sin^2\theta\, d\theta \\ &= -[(\mu + Ur \cos\theta)^2 + U^2r^2\sin^2\theta]\, d\theta \\ &= -(\mu + Ure^{i\theta})(\mu + Ure^{-i\theta})\, d\theta \\ &= -|\mu + Uz|^2\, d\theta \end{aligned} \tag{5.4.14}$$

This expresses the change in potential in terms of the change in the polar ray θ. Combining this with Eq. (5.4.10), since $|z| = r$,

$$d\sigma = \frac{r}{U \sin\theta} d\theta \tag{5.4.15}$$

So that on substituting Eq. (5.4.12)

$$d\sigma = -\frac{\mu\theta - \psi}{(U \sin\theta)^2} d\theta \tag{5.4.16}$$

One can integrate this, starting at θ_0, to θ holding ψ constant in conformity with the discussion of Section 5.2.

$$\sigma(P) = \frac{1}{U^2} \int_{\theta}^{\theta_0} (\mu x - \psi) \csc^2 x \, dx$$

$$= \frac{1}{U^2} (\mu x - \psi) \cot x \Big|_{\theta_0}^{\theta} - \frac{\mu}{U^2} \ln(\sin x) \Big|_{\theta_0}^{\theta}$$

$$= \frac{1}{U^2} \left\{ (\mu\theta - \psi) \cot\theta - (\mu\theta_0 - \psi) \cot\theta_0 - \mu \ln\left|\frac{\sin\theta}{\sin\theta_0}\right| \right\} \tag{5.4.17}$$

The ray $\theta = \theta_0$ intersects the potential line $\phi = \phi_0$ at $r = a$. On ϕ_0 the boundary data is prescribed. The relation between ϕ_0 and ψ, θ_0 is obtained by combining Eqs. (5.4.11) and (5.4.12).

$$\phi_0 = \mu \ln\left(\frac{\psi - \mu\theta_0}{U \sin\theta_0}\right) + (\psi - \mu\theta_0) \cot\theta_0 \quad . \tag{5.4.18}$$

Equation (5.4.17) can be used as follows. Construct the $\phi - \psi$ flow net and locate the intersection of the potential line ϕ_0 and the streamline ψ where the boundary source point (a,θ_0) is located. (This is more convenient than trying to solve Eq. (5.4.18)). The point P moving along the streamline ψ is then characterized by the intersection of the variable ray θ with the streamline ψ = constant. See Fig. 5.4.3.

We now turn to an alternate analytical method for the determination of $\sigma(P)$. It is based on the derivative relation (Eq. (5.2.8)) which couples the gradient field of the velocity potential function $\phi(x,y)$ with that of the scalar function $\sigma(x,y)$

$$\left(\frac{\partial\phi}{\partial x}\right)\frac{\partial\sigma}{\partial x} + \left(\frac{\partial\phi}{\partial y}\right)\frac{\partial\sigma}{\partial y} = -1 \qquad (5.4.19)$$

Since ϕ and its derivatives are known in principle one can introduce the functions

$$\frac{\partial\phi}{\partial x} = \phi_1(x,y) \quad , \quad \frac{\partial\phi}{\partial y} = \phi_2(x,y) \qquad (5.4.20)$$

There results the first order linear partial differential equation

$$\phi_1(x,y)\,\frac{\partial\sigma}{\partial x} + \phi_2(x,y)\,\frac{\partial\sigma}{\partial y} = -1 \qquad (5.4.21)$$

The general solution can be constructed by the method of characteristics and is of the form

$$u_2(x,y) = g(u_1(x,y)) \qquad (5.4.22)$$

Here $u_1(x,y) = c_1$ and $u_2(x.y) = c_2$ are any two independent solutions of the associated ordinary differential equation system

$$\frac{dx}{\phi_1(x,y)} = \frac{dy}{\phi_2(x,y)} = -\,d\sigma \qquad (5.4.23)$$

The construction of this general solution proceeds as follows. One first solves the equation

$$\frac{dy}{dx} = \frac{\phi_2(x,y)}{\phi_1(x,y)} \qquad (5.4.24)$$

or its inverse. The solution can be represented in the form $u_1(x,y) = C_1$. This result allows one to express say y in terms of x and c_1. Then one considers the remaining equation in Eq. (5.4.23), say

$$\frac{d\sigma}{dx} = -\frac{1}{\phi_1(x,y)} \tag{5.4.25}$$

Before integrating one expresses y in terms of x and c_1. After integrating one replaces c_1 in terms of (x,y). The resulting solution is of the form

$$u_2 = \sigma + h(x,y) = c_2 \quad , \tag{5.4.26}$$

On substituting this into Eq. (5.4.22) there results the general solution

$$\sigma(x,y) = g(u_1(x,y)) - h(x,y) \tag{5.4.27}$$

Here h and u_1 are defined functions and g is an arbitrary function which must be determined from the boundary condition, Eq. (5.2.26)

$$\sigma(x_0,y_0) = 0 \tag{5.4.28}$$

where (x_0,y_0) is a point on the boundary curve which defines the repository position.

C. The Stagnation Point Flow

Consider the potential function $\phi(x,y) = \alpha(2xy)$ which describes the flow in the neighborhood of a stagnation point. The flow net is shown in Fig. 5.4.4. Equations (5.4.21) and (5.4.23) have the form

$$2y\,\frac{\partial\sigma}{\partial x} + 2x\,\frac{\partial\sigma}{\partial y} = -\frac{1}{\alpha} \tag{5.4.29}$$

$$\frac{dx}{2y} = \frac{dy}{2x} = -\alpha d\sigma$$

The first pair of equations yield

$$u_1(x,y) = x^2 - y^2 = C_1 \tag{5.4.30}$$

and with this the first and last term can be written

$$\frac{dx}{2\sqrt{x^2 - c_1}} = -\alpha d\sigma \tag{5.4.31}$$

This integrates into

$$u_2(x,y) \equiv \alpha\sigma + \frac{1}{2}\cosh^{-1}\left(\frac{x}{\sqrt{c_1}}\right) = c_2$$

$$= \alpha\sigma + \frac{1}{2}\cosh^{-1}\left(\frac{x}{\sqrt{x^2 - y^2}}\right) = c_2 \tag{5.4.32}$$

Thus the general solution is

$$\alpha\sigma(x,y) = g(x^2 - y^2) - \frac{1}{2}\cosh^{-1}\left(\frac{x}{\sqrt{x^2 - y^2}}\right) \tag{5.4.33}$$

in agreement with Eq. (5.4.27). Suppose that $\sigma = 0$ along part of the potential line $xy = 1$. Then

$$g(y^{-2} - y^2) = \frac{1}{2}\cosh^{-1}\left(\frac{y^{-1}}{\sqrt{y^{-2} - y^2}}\right) \tag{5.4.34}$$

which determines the arbitrary function g. To find its functional form set

$$u = y^{-2} - y^2 \tag{5.4.35}$$

Then for $y > 0$

$$y^{-1} = \left(\frac{2}{(u^2 + 4)^{1/2} - u}\right)^{1/2} \tag{5.4.36}$$

Both square roots are taken positive. Substituting for   in terms of u yields

$$g(u) = \frac{1}{2}\cosh^{-1}\left(\left[\frac{2}{u(u^2 + 4)^{1/2} - u^2}\right]^{1/2}\right) \tag{5.4.37}$$

With g determined, the time-distance function $\sigma(x,y)$ becomes with Eq. (5.4.33)

$$\sigma(x,y) = \frac{1}{\alpha}\left\{\frac{1}{2}\cosh^{-1}\left(\frac{2}{(x^2-y^2)[(x^2-y^2)^2+4]^{1/2}-(x^2-y^2)^2}\right)^{1/2} - \cosh^{-1}\left(\frac{x}{[x^2-y^2]^{1/2}}\right)\right\} \qquad (5.4.38)$$

If the repository surface S given by $xy = 1$, is restricted by $x_0 < x < x_1$ one must restrict the range in y to stay in the "shadow region."

The method just discussed is not restricted to rectangular Cartesian coordinates (x,y). In general curvilinear coordinates η_i Eq. (5.4.19) takes on the form.

$$\frac{1}{g_{11}}\frac{\partial\phi}{\partial\eta_1}\frac{\partial\sigma}{\partial\eta_1} + \frac{1}{g_{22}}\frac{\partial\phi}{\partial\eta_2}\frac{\partial\sigma}{\partial\eta_2} = -1 \qquad (5.4.39)$$

where the g_{ii} are the metric coefficients. The Eqs. (5.4.23) are replaced by

$$\frac{d\eta_1}{\frac{1}{g_{11}}\frac{\partial\phi}{\partial\eta_1}} = \frac{d\eta_2}{\frac{1}{g_{22}}\frac{\partial\phi}{\partial\eta_2}} = -d\sigma \qquad (5.4.40)$$

For cylindrical polar coordinates for example $g_{11} = 1$, $g_{22} = r^2$, $\eta_1 = r$, $\eta_2 = \theta$. In case the integrations in Eq. (5.4.40) are difficult to perform numerical integration method are obviously called for.

Before concluding this section we consider some generalizations mentioned in Section 5.1. The construction of the travel time distance function can be carried out even if the ground has an anisotropic permeability. This is very important in practical applications.

Consider a two-dimensional groundwater flow in a homogeneous anisotropic medium with principal axis permeabilities k_x, k_y. The combination of conservation of mass of the fluid and Darcy's law leads to the elliptic equation

$$\frac{\partial}{\partial x}\left(k_x\frac{\partial\Phi}{\partial x}\right) + \frac{\partial}{\partial y}\left(k_y\frac{\partial\Phi}{\partial y}\right) = 0 \qquad (5.4.41)$$

where Φ is the potential function first introduced in Eq. (5.1.7). On dividing by k_x this can be written

$$\frac{\partial^2 \Phi}{\partial x^2} + \sqrt{\frac{k_y}{k_x}}\ \frac{\partial}{\partial y}\left(\sqrt{\frac{k_y}{k_x}}\ \frac{\partial \Phi}{\partial y}\right) = 0 \tag{5.4.42}$$

With the new (scaled) Y coordinate

$$Y = y\sqrt{\frac{k_y}{k_x}} \tag{5.4.43}$$

and

$$X = x$$

there results the potential equation for an isotropic medium

$$\frac{\partial^2 \Phi}{\partial X^2} + \frac{\partial^2 \Phi}{\partial Y^2} = 0 \tag{5.4.44}$$

One can of course equally well rescale the x coordinate.

Suppose now that the boundary value problem for the harmonic function Φ has been solved from Eq. (5.4.44). Then the conjugate stream function $\psi(X,Y)$ and the couplex potential $F(Z)$ can be constructed as follows. In view of Eq. (5.2.17) the complex potential $F(Z)$ has the form

$$F(Z) = \Phi(X,Y) + i\psi(X,Y) \tag{5.4.45}$$

With

$$Z = X + iY \quad , \quad \bar{Z} = X - iY \quad , \tag{5.4.46}$$

$$F(Z) = \Phi\left(\frac{Z + \bar{Z}}{2}\ ,\ \frac{Z - \bar{Z}}{2i}\right) + i\psi\left(\frac{Z + \bar{Z}}{2}\ ,\ \frac{Z - \bar{Z}}{2i}\right) \quad . \tag{5.4.47}$$

Now set $\bar{Z} = Z$, then

$$F(Z) = \phi(Z,0) + i\psi(Z,0) \tag{5.4.47a}$$

To find F(Z), given the isotropic potential $\phi(X,Y)$ one computes from Eq. (5.4.45)

$$\begin{aligned} F'(Z) &= \frac{\partial\phi}{\partial X} + i\,\frac{\partial\psi}{\partial X} \\ &= \frac{\partial\phi}{\partial X} - i\,\frac{\partial\phi}{\partial y} \end{aligned} \tag{5.4.48}$$

by using the Cauchy-Riemann Eqs. (5.2.1). Since ϕ and its derivatives are in principle known function set.

$$\frac{\partial\phi}{\partial X} = \phi_1(X,Y) \tag{5.4.49}$$

$$\frac{\partial\phi}{\partial Y} = \phi_2(X,Y)$$

Thus

$$F'(Z) = \phi_1(X,Y) - i\phi_2(X,Y) \tag{5.4.50}$$

which with help of Eq. (5.4.47) becomes

$$F'(Z) = \phi_1(Z,0) - i\phi_2(Z,0) \tag{5.4.51}$$

On integrating there results the complex potential

$$F(Z) = \int^{Z} [\phi_1(Z',0) - i\phi_2(Z',0)]\, dZ' \tag{5.4.52}$$

from which one can readily isolate the stream function ψ.

As a simple illustration consider again the stagnation point potential $\phi(X,Y) = \alpha(2\,XY)$. Then

$$\phi_1(X,Y) = \alpha 2Y \quad ; \quad \phi_2(X,Y) = \alpha 2X \tag{5.4.53}$$

so that

$$F(Z) = \int^{Z} [-i\alpha 2Z'] \, dZ'$$
$$= -i\alpha Z^2 \quad . \tag{5.4.54}$$

Once $w = \phi(X,Y) + i\psi(X,Y) = F(Z)$ has been constructed $\sigma(P)$ can be determined by the methods discussed. Then one replaces the distorted coordinates (X,Y) or the distorted flow net $\phi - \psi$ with help of Eq. (5.4.43) in order to recover the original physical field variables in the anisotropic field.

5.5. The Solution of the Three-Dimensional Transport Equation

The construction of the solution to the transport equation in two dimensional flow fields depends seemingly on the existence and employment of the potential and stream functions describing the flow pattern. Unfortunately this conjugate pair of harmonic functions does not exist for three dimensional flows. Of course one has a ground water potential function for such flows but its mathematical coupling to the three-dimensional stream surface function is not as analytically connected as the Cauchy-Riemann equations in two-dimensional flow fields. Thus one must seek an alternate approach.

The proper suggestion comes from the properties of the functions which were required in the solution of the two dimensional problem. These are given by Eqs. (5.3.3), (5.3.4) and (5.3.5), i.e.,

$$(\nabla\phi)\cdot(\nabla\sigma) = -1 \quad P\epsilon R \tag{5.5.1}$$

$$\sigma(Q) = 0 \quad , \quad Q\epsilon S \tag{5.5.2}$$

$$\sigma(P) > 0 \quad , \quad P\epsilon R \tag{5.5.3}$$

It will be shown that if one postulates the existence of a travel time position relation $\sigma = \sigma(P)$ which satisfies the properties (Eqs. (5.5.1) and (5.5.3)) that one can then obtain the solution to the governing Eq. (5.1.9)

$$\frac{\partial C_n}{\partial t} - \frac{1}{K_n} (\nabla\phi)\cdot(\nabla C_n) + \lambda_n C_n = \lambda_{n-1} C_{n-1} \quad n = 1,2, \ldots , \lambda_o = 0 \tag{5.5.4}$$

The side conditions are (Eqs. (5.1.11) through (5.1.13))

$$C_n(P,0) = 0 \quad , \quad P \epsilon R \tag{5.5.5}$$

$$C_n(Q,t) = C_n^o g_n(Q,t) \quad , \quad Q \epsilon S \quad , \quad t \geq 0 \tag{5.5.6}$$

$$g_n(P,t) \equiv 0 \quad , \quad t < 0 \quad P \epsilon R U S \tag{5.5.7}$$

In view of Eq. (5.5.1) we now assume only the existence of a flow potential function $\phi(P)$ and the travel-position function $\sigma(P)$ which generates an orthogonal gradient field described by Eq. (5.5.1), with σ obeying the minor, non-negative properties of Eqs. (5.5.2) and (5.5.3). In addition we shall impose the geometrical restriction (Eq. (5.3.9)) on the position of the repository surface which leads to (see Eq. (5.3.12))

$$(\nabla \phi) \cdot (\nabla g_n(P,t)) = 0 \quad , \quad t \geq 0 \quad , \quad P \epsilon R U S \tag{5.5.8}$$

The above formulation has the interesting consequence that the solution to these governing equations presented below will also be the solution for the one, two and three dimensional flow fields. The reader must, however, keep in mind that it is now no longer possible to reduce the transport Eq. (5.5.4) to the single form given by Eq. (5.3.1) but Eq. (5.5.4) must be solved in the three dimensional space. We shall construct the general (non-recursive) analytical solution. The method employed is a generalization of the classical solution method described by the writer in Ref. (H1), p. 427.

The solution for the mother concentration (n=1) of Eq. (5.5.4) is

$$C_1(P,t) = C_1^o e^{-\lambda_1 K_1 \sigma(P)} g_1(P, t - K_1 \sigma(P)) \tag{5.5.9}$$

as already obtained for the two-dimensional case in Eq. (5.3.17). That this satisfies the three dimensional problem is verified by computing

$$\frac{\partial C_1}{\partial t} = C_1^o e^{-\lambda_1 K_1 \sigma(P)} \frac{\partial g_1}{\partial t} (P, t - K_1 \sigma(P)) \tag{5.5.10}$$

$$(\nabla\phi)\cdot(\nabla C_1) = C_1^0 e^{-\lambda_1 K_1 \sigma(P)} \Big\{ (\nabla\phi)\cdot(\nabla g_1(P,t - K_1\sigma(P))) - K_1(\nabla\phi)\cdot(\nabla\sigma)$$

$$\left[\frac{\partial g_1}{\partial t}(P,t - K_1\sigma(P)) + \lambda_1 g_1(P,t - K_1\sigma(P))\right]\Big\}$$

$$= C_1^0 K_1 e^{-\lambda_1 K_1 \sigma(P)} \left[\frac{\partial g_1}{\partial t}(P,t - K_1\sigma(P)) + \lambda_1 g_1(P,t - K_1\sigma(P))\right] \qquad (5.5.11)$$

by using Eqs. (5.5.1) and (5.5.8). Substitution of these expressions into Eq. (5.5.4) verifies the solution Eq. (5.5.9) for $n = 1$ ($\lambda_0 = 0$). Furthermore Eq. (5.5.9) satisfies the initial condition (Eq. (5.5.5)) because of Eq. (5.5.7). Finally, Eq. (5.5.9) obeys also the boundary condition (Eq. (5.5.6)) on account of Eq. (5.5.2).

The only remaining quantity to be constructed is $\sigma = \sigma(P)$. Once σ is known the mother concentration is completely determined. σ arises in the solution of every member of the radionuclide chain and the determination of this function can be obtained from the solution of the partial differential Eq. (5.5.1).

We now extend the result (Eq. (5.5.9)) and propose for the general solution of the governing Eq. (5.5.4),

$$C_n(P,t) = \sum_{p=1}^{n} \sum_{q=1}^{n} e^{-\lambda_p K_p \sigma(P)} f_{n,p,q}(P,t - K_p\sigma(P))$$

$$+ C_n^0 g_n(P,t - K_n\sigma(p)) e^{-\lambda_n K_n \sigma(P)} \quad , \quad n = 1,2, \ldots \qquad (5.5.12)$$

The functions $f_{n,p,q}$ are unknown at this point and are to be found subject to the imposed asymmetry condition

$$f_{n,p,q}(P,t) = - f_{n,q,p}(P,t) \quad , \quad t > 0 \qquad (5.5.13)$$

and where

$$f_{n,p,q}(P,t) = f_{n,p,q}(Q,t) = 0 \quad , \quad t < 0 \qquad (5.5.14)$$

The determination of the $f_{n,p,q}$ functions proceeds in a number of stages. Their determination rests on the fact that if $C_n(P,t)$ is the solution of Eq. (5.5.4) then the $f_{n,p,q}$ functions must satisfy certain constraining equations. If the reader is not interested in the mathematical details of the solution he may wish to turn to the final result, Eq. (5.5.50).

By using the asymmetry condition (Eq. (5.5.13)) one can re-write Eq. (5.5.12) as follows

$$C_n(P,t) = \frac{1}{2} \sum_{p=1}^{n} \sum_{q=1}^{n} e^{-\ K_p\sigma(P)} f_{n,p,q}(P,t-K_p\sigma(P)) - e^{-\lambda_q K_q\sigma(P)}$$

$$f_{n,p,q}(P,t-K_q\sigma(P))\Bigg\} + C_n^0 g_n(P,t - K_n\sigma(P))\, e^{-\lambda_n K_n\sigma(P)} \quad . \qquad (5.5.15)$$

From this one observes that the proposed solution form for $C_n(P,t)$ obeys the initial condition (Eq. (5.5.5)) by using Eqs. (5.5.3), (5.5.7) and (5.5.8). The boundary conditions (Eq. (5.5.6)) are also satisfied in view of Eq. (5.5.2). Therefore the only remaining condition to be met is to show that $C_n(P,t)$ obeys the governing partial differential equation. Substitution of Eq. (5.5.12) into Eq. (5.5.4) and using Eqs. (5.5.1) and (5.5.8) yields

$$\sum_{p=1}^{n} \sum_{q=1}^{n} \left[\left(1 - \frac{K_p}{K_n}\right) \frac{\partial}{\partial t} f_{n,p,q}(P,t - K_p\sigma(P)) + \left(\lambda_n - \lambda_p \frac{K_p}{K_n}\right) f_{n,p,q}(P,t - K_p\sigma(P))\right]$$

$$e^{-\lambda_p K_p\sigma(P)} = \lambda_{n-1} \sum_{p=1}^{n-1} \sum_{q=1}^{n-1} f_{n-1,p,q}(P,t - K_p\sigma(P))\, e^{-\lambda_p K_p\sigma(P)}$$

$$+ C_{n-1}^0 \lambda_{n-1} g_{n-1}(P,t - K_{n-1}\sigma(P))\, e^{-\lambda_{n-1}K_{n-1}\sigma(P)} \qquad (5.5.16)$$

Here use has been made of a property of the as yet unknown functions $f_{n,p,q}$, i.e.,

$$(\nabla\phi)\cdot(\nabla f_{n,p,q}(P,t)) = 0 \quad , \quad P\epsilon RUS \quad , \quad t \geq 0 \qquad (5.5.17)$$

This property will be verified subsequently.

It is Eq. (5.5.16), together with the various constraints such as Eqs. (5.5.13), (5.5.14) and (5.5.17), which when solved will yield the solution to the problem.

On equating coefficients of the different exponentials in Eq. (5.5.16) and slightly re-arranging the equations one obtains

$$\sum_{q=1}^{n}\left[\frac{\partial}{\partial t} f_{n,p,q}(P,t) + \mu_{n,p} f_{n,p,q}(P,t)\right] = \frac{\lambda_{n-1}K_{n-1}}{K_n - K_p}\left[\sum_{q=1}^{n-1} f_{n-1,p,q}(P,t) + \delta_{p,n-1} C^0_{n-1} g_{n-1}(P,t)\right] \quad , \quad p = 1,2, \ldots n-1 \tag{5.5.18}$$

where

$$\mu_{p,q} = \mu_{q,p} = \frac{\lambda_p K_p - \lambda_q K_q}{K_p - K_q} \quad , \quad \delta_{ij} = \begin{cases} 0 \ , & i \neq j \\ 1 \ , & i = j \end{cases} \tag{5.5.19}$$

The differential-difference equation system (Eq. (5.5.18)) for the functions $f_{n,p,q}$ can be reduced in several stages to a simpler difference equations system for a new set of unknown functions $h_{n,p,q}$. For this purpose introduce the integral transformation

$$f_{n,p,q}(P,t) = J(\mu_{p,q} \ , \ h_{n,p,q}(P,s) \ , \ t) \quad , \tag{5.5.20}$$

where the integral operator J is defined by

$$J(\mu, \ p(s) \ , \ t) = \begin{cases} e^{-\mu t}\int_0^t p(s)e^{\mu s}\, ds \ , & t>0 \\ 0 \ , & t<0 \end{cases} \tag{5.5.21}$$

One finds from Eqs. (5.5.20) and (5.5.21) that $f_{n,p,q}(P)$ is a solution of

$$\frac{df_{n,p,q}}{dt}(P,t) = -\mu_{p,q} f_{n,\mu,q}(P,t) + h_{n,p,q}(P,t) \quad . \tag{5.5.22}$$

Furthermore by Eqs. (5.5.13) and (5.5.22) there follows the asymmetry condition,

$$h_{n,p,q}(P,t) = -h_{n,q,p}(P,t) \quad . \tag{5.5.23}$$

If the last two equations are substituted into Eq. (5.5.18) there results with help of Eq. (5.5.19)

$$\sum_{q=1}^{n} (\mu_{n,p} - \mu_{p,q})\, f_{n,p,q}(P,t) + \sum_{q=1}^{n} h_{n,p,q}(P,t) = \frac{\lambda_{n-1}K_{n-1}}{K_n - K_p} \left\{ \sum_{q=1}^{n-1} f_{n-1,p,q}(P,t) + C^{o}_{n-1}\delta_{p,n-1}g_{n-1}(P,t) \right. \quad . \tag{5.5.24}$$

The key to the solution of this equation is the fact that $h_{n,p,q}$ and $f_{n,p,q}$ satisfy the recurrence relations

$$h_{n,p,q}(P,t) = \frac{\lambda_{n-1}K_n}{K_n - K_p} \frac{1}{\mu_{n,p} - \mu_{p,q}} h_{n-1,p,q}(P,t) \quad , \quad p,q = 1,2 \ldots (n-1) \tag{5.5.25}$$

$$f_{n,p,q}(P,t) = \frac{\lambda_{n-1}K_n}{K_n - K_p} \frac{1}{\mu_{n,p} - \mu_{p,q}} f_{n-1,p,q}(P,t) \quad , \quad p,q = 1,2 \ldots (n-1) \tag{5.5.26}$$

One shows that if Eq. (5.5.25) is assumed valid then Eq. (5.5.26) is a consequence of Eq. (5.5.20). Furtherfore, one establishes that if Eq. (5.5.23) is valid then Eq. (5.5.25) hold for the function pairs ($h_{n,p,q}$, $h_{n-1,p,q}$) and ($h_{n,q,p}$, $h_{n-1,q,p}$). One can now combine Eqs. (5.5.24) and (5.5.26). There results a cancellation of the two sums leaving only

$$\sum_{q=1}^{n} h_{n,p,q}(P,t) = \frac{\lambda_{n-1}K_n}{K_n - K_p} C^{o}_{n-1}\delta_{p,n-1}g_{n-1}(P,t) \quad p = 1,2, \ldots n-1 \tag{5.5.27}$$

Once <u>this</u> difference equation is solved, the $h_{n,p,q}$ functions when substituted into Eq. (5.5.20) yield the function $f_{n,p,q}$ which then determine the general solution given by Eq. (5.5.12). It is the solution of Eq. (5.5.27) which constitutes the major part of the following analysis. One observes from Eq. (5.5.23) that

$$h_{n,p,p}(P,t) = 0 \tag{5.5.28}$$

Now suppose $p > q$. It will be seen later that this condition allows one to satisfy the symmetry condition (Eq. (5.5.13)). One can then solve the Eq. (5.5.25) iteratively for n leading to

$$h_{n,p,q}(P,t) = \frac{L_{n,p}}{U_{n,p,q}} h_{p,q}(P,t) \quad , \quad p > q \quad . \tag{5.5.29}$$

where for short hand notation we have set

$$h_{p,q}(P,t) \equiv h_{p,p,q}(P,t) \quad . \tag{5.5.30}$$

$L_{n,p}$ is defined by

$$L_{n,p} = (K_n\lambda_{n-1})(K_{n-1}\lambda_{n-2}) \ldots . (K_{p+1}\lambda_p) \qquad p = 1,2, \ldots , n-1 \tag{5.5.31}$$

$L_{n,n} = 1$. $U_{n,p,q}$ is defined by

$$U_{n,p,q} = G_{n,p}(\mu_{p,q})\, G_{n-1,p}(\mu_{p,q}) \; . \; . \; . \; G_{p+1,p}(\mu_{p,q}) \, , n > p > q \tag{5.5.32}$$

$$U_{n,p,q} = 1 \qquad\qquad n = p > q$$

where

$$G_{p,q}(z) = (K_q - K_p)(z - \mu_{p,q}) \tag{5.5.33}$$

A number of properties of $G_{p,q}(z)$ are derived in Appendix G. If the solution (5.5.29) is now substituted into Eq. (5.5.27) and the asymmetry condition, Eq. (5.5.23), is used there results

$$L_{n,p} \sum_{q=1}^{p} \frac{1}{U_{n,p,q}} h_{p,q}(P,t) - \sum_{q=p}^{n} \frac{L_{n,q}}{U_{n,q,p}} h_{q,p}(P,t)$$

$$= \frac{\lambda_{n-1}K_n}{K_n - K_p} \delta_{p,n-1} C^{o}_{n-1} g_{n-1}(P,t) \quad , \quad p = 1,2, \ldots , n-1 \tag{5.5.34}$$

This system determines the functions $h_{p,q}$ successively beginning with $h_{2,1}$. These functions are linear combinations of the boundary condition functions $g_k(P,t)$ as can be seen from Eq. (5.5.34). For the solution of this equation set

$$h_{p,q}(P,t) = \sum_{k=1}^{p} A_{p,q,k} C_k^0 g_k(P,t) \quad p > q \tag{5.5.35}$$

The $A_{p,q,k}$ are unknown constant coefficients. It follows from Eq. (5.5.28) and (5.5.30) that

$$h_{p,p}(P,t) = 0 \tag{5.5.36}$$

and so in Eq. (5.5.35)

$$A_{p,p,k} = 0 \qquad k = 1,2, \ldots p \tag{5.5.37}$$

At this point one can verify the validity of Eq. (5.5.17). By Eqs. (5.5.8) and (5.5.35)

$$(\nabla\phi)\cdot(\nabla h_{p,q}(P,t)) = 0 \quad . \tag{5.5.38}$$

So that by Eq. (5.5.29)

$$(\nabla\phi)\cdot(\nabla h_{n,p,q}(P,t)) = 0 \quad . \tag{5.5.39}$$

Then in turn by Eq. (5.5.20)

$$(\nabla\phi)\cdot(\nabla f_{n,p,q}(P,t) = 0 \quad . \tag{5.5.40}$$

Thus, Eq. (5.5.17) is seen not to be an additional assumption but the natural outgrowth of conditions (Eq. (5.5.8)). One can now turn to the determination of the coefficients $A_{p,q,k}$.

Substituting Eq. (5.5.35) into (5.5.34) and interchanging the orders of the double sums

$$L_{n,p} \sum_{k=1}^{p} \sum_{q=k}^{p} \frac{1}{U_{n,p,q}} A_{p,q,k} C_k^0 g_k(P,t) - \tag{5.5.41}$$

$$- \sum_{k=1}^{p} \sum_{q=p}^{n} \frac{L_{n,q}}{U_{n,q,p}} A_{q,p,k} C_k^0 g_{n-1}(P,t) =$$

$$= \frac{\lambda_{n-1} K_n}{K_n - K_p} \delta_{p,n-1} C_{n-1}^0 g_{n-1}(P,t) \quad , \quad p = 1,2, \ldots (n-1)$$

Equating coefficient of the g_k functions gives for $k \leq n - 2$

$$L_{n,p} \sum_{q=k}^{p} \frac{1}{U_{n,p,q}} A_{p,q,k} - \sum_{q=p}^{n} \frac{L_{n,q}}{U_{n,q,p}} A_{q,p,k} = 0 \quad , \quad \begin{array}{l} p = 1,2, \ldots (n-1) \\ k = 1,2, \ldots (n-2) \end{array} \tag{5.5.42}$$

and for $k = n - 1$

$$A_{n,n-1,n-1} = \frac{L_{n,n-1}}{K_{n-1} - K_n} \quad . \tag{5.5.43}$$

If one separates out the $q = n$ term in Eq. (5.5.42)

$$A_{n,p,k} = \sum_{q=k}^{p} \frac{L_{n,p}}{U_{n,p,q}} A_{p,q,k} - \sum_{q=p}^{n-1} \frac{L_{n,q}}{U_{n,q,p}} A_{q,p,k} \quad ; \quad \begin{array}{l} p = 1,2, \ldots (n-1) \\ k = 1,2, \ldots (n-2) \end{array} \tag{5.5.44}$$

The problem has thus been finally reduced to the solution of this difference equation. It is shown in Appendix H that the required solution is given by

$$A_{p,q,k} = \frac{L_{p,k}}{k_q - k_p} \; \frac{1}{G_{k,q}(\mu_{p,q}) G_{k+1,q}(\mu_{p,q}) \ldots G_{p-1,q}(\mu_{p,q})} \, , \; p > q \geq k \tag{5.5.45}$$

Here it is to be understood that the factor $G_{p,q}(\mu_{p,q})$ is to be replaced by 1 when this term appears in the denominator.

We can now summarize the solution. Consider the expression for $C_n(P,t)$ in Eq. (5.5.12). By Eq. (5.5.13) there is no contribution when $p = q$. The terms corresponding to $q > p$ can be associated with those corresponding to $q < p$ by interchanging p and q and then using Eq. (5.5.13). There results

$$C_n(P,t) = \sum_{p=1}^{n} \sum_{q=1}^{p-1} \Big[e^{-\lambda_p k_p \sigma(P)} f_{n,p,q}(P,t - K_p\sigma(P)) - e^{-\lambda_q K_q \sigma(P)} f_{n,p,q}(P,t - K_q\sigma(P)) \Big] + C_n^0 g_n(P,t - K_n\sigma(P))\, e^{-\lambda_n K_n \sigma(P)} \tag{5.5.46}$$

By Eqs. (5.5.20) and (5.5.29) with $f_{p,q}(P,t) \equiv f_{p,p,q}(P,t)$

$$f_{n,p,q}(P,t) = \frac{L_{n,p}}{U_{n,p,q}} f_{p,q}(P,t) \quad . \tag{5.5.47}$$

In turn by equations (5.5.20) and (5.5.30)

$$f_{p,q}(P,t) = J(\mu_{p,q} , h_{p,q}(p,\tau) , t) \tag{5.5.48}$$

so that by equation (5.5.35)

$$f_{p,q}(P,t) = \sum_{k=1}^{q} A_{p,q,k} J(\mu_{p,q} , C_k^0 g_k(P,\tau) , t) \quad . \tag{5.5.49}$$

On combining Eqs. (5.5.46) and (5.5.47)

$$C_n(P,t) = \sum_{p=1}^{n} \sum_{q=1}^{p-1} \left[\frac{L_{n,p}}{U_{n,p,q}} F_{p,q}(P,t) \right] + e^{-\lambda_n K_n \sigma(P)} C_n^0 g_n(P , t - K_n\sigma(P)) \tag{5.5.50}$$

where

$$F_{p,q}(P,t) = e^{-\lambda_p K_p \sigma(P)} f_{p,q}(P,t - K_p\sigma(P)) - e^{-\lambda_q K_q \sigma(P)} f_{p,q}(P,t - K_q\sigma(P)) \tag{5.5.51}$$

This represents the final form of the general solution to our problem. The functions $f_{p,q}(P,t)$ are given in terms of the boundary data by Eq. (5.5.49) with the $A_{p,q,k}$'s fixed by Eq. (5.5.45). The solution holds for a radionuclide chain of arbitrary length. In the following are set out the first three members of the matriarchy.

The mother concentration (n = 1) is obtained from either Eqs. (5.5.9) or (5.5.50) with help of the scaling relation (Eq. (5.1.10))

$$N_1(P,t) = N_1^o e^{-\lambda_1 K_1 \sigma(P)} g_1(P,t - K_1\sigma(P)) \qquad (5.5.52)$$

The first daughter (n = 2) concentration by Eq. (5.5.50) is

$$C_2(P,t) = \frac{L_{22}}{U_{221}} F_{21}(P,t) + C_2^o e^{-\lambda_2 K_2 \sigma(P)} g_2(P,t - K_2\sigma(P)) \qquad (5.5.53)$$

But $U_{221} = 1$ by Eq. (5.5.32) and $L_{22} = 1$ by Eq. (5.5.31). Thus with Eq. (5.5.51)

$$\frac{L_{22}}{U_{221}} F_{21}(P,t) = e^{-\lambda_2 K_2 \sigma(P)} f_{21}(P,t - K_2\sigma(P)) - e^{-\lambda_1 K_1 \sigma(P)} f_{21}(P,t - K_1\sigma(P)) \qquad (5.5.54)$$

In turn by Eq. (5.5.49) and (5.5.21)

$$f_{21}(P,t) = A_{211} C_1^o \left\{ h(t)\ e^{-\mu_{21} t} \int_0^t g_1(P,\tau)\ e^{\mu_{21}\tau}\ d\tau \right\} \qquad (5.5.55)$$

where h(t) is the step function and by Eqs. (5.5.43) and (5.5.31)

$$A_{211} = \frac{L_{21}}{K_1 - K_2}$$

$$= \frac{K_2 \lambda_1}{K_1 - K_2} \qquad (5.5.56)$$

If these results are substituted into Eq. (5.5.53) and the scale relation (Eq. (5.1.10)) is employed one obtains

$$N_2(P,t) = e^{-\lambda_1 K_1 \sigma(P)} N_{2,1}(P,t - K_1\sigma(P))\, h(t - K_1\sigma(P)) \tag{5.5.57}$$

$$- e^{-\lambda_2 K_2 \sigma(P)} N_{2,1}(P,t - K_2\sigma(P))\; h(t - K_2\sigma(P)) + N_2^0 e^{-\lambda_2 K_2 \sigma(P)}\; g_2(P,t - K_2\sigma(P))$$

where

$$N_{1,2}(P,\tau) = \frac{\lambda_1 K_1 N_1^0}{K_2 - K_1}\, e^{-\mu_{21}\tau} \int_0^{\tau} g_1(P,\tau')\, e^{\mu_{21}\tau'}\, d\tau' \tag{5.5.58}$$

This expression formally agrees with the solution (5.3.18) except that σ(P) differs for these two flow fields.

Without filling in all the details of the solution for the second daughter (n = 3) one has from Eq. (5.5.50)

$$C_3(P,t) = \left[\frac{L_{32}}{G_{31}(\mu_{21})} F_{21}(P,t) + F_{31}(P,t) + F_{32}(P,t)\right]$$

$$+ e^{-\lambda_3 K_3 \sigma(P)} C_3^0 g_3(P,t - K_3\sigma(P)) \tag{5.5.59}$$

By Eq. (5.5.35)

$$\begin{aligned} h_{31}(P,t) &= A_{311} C_1^0 g_1(P,t) \\ h_{32}(P,t) &= A_{321} C_1^0 g_1(P,t) + A_{322} C_2^0 g_2(P,t) \end{aligned} \tag{5.5.60}$$

These two functions are used in the construction of the $f_{p,q}$ functions in Eq. (5.5.48). In turn, the $f_{p,q}$'s are used in forming the $F_{i,j}$ function with help of Eq. (5.5.51). Finally, the coefficients $A_{i,j,k}$ are obtained from Eqs. (5.5.43) and (5.5.45)

$$A_{311} = \frac{L_{31}}{K_1 - K_3} \frac{1}{G_{21}(\mu_{21})} \quad ; \quad A_{321} = \frac{L_{31}}{K_2 - K_3} \frac{1}{G_{12}(\mu_{32})} \qquad (5.5.61)$$

$$A_{322} = \frac{L_{32}}{K_2 - K_3}$$

The L and G coefficients are given in Eqs. (5.5.31) and (5.5.33) respectively. The construction of the analytical solution for additional members of the radio nuclide chain proceeds then in the same manner. The reader should note that with the appropriate form of $\sigma(P)$ Eq. (5.5.50) represents the general (non-recursive) solution of the nuclide transport in one, two and three-dimensional flow fields.

The time position function $\sigma(P)$ for a three-dimensional field is as previously the trajectory of a particle of fixed identity. The governing equation is Eq. (5.5.1), which in arbitrary curvilinear, orthogonal coordinates η_i with metric coefficients g_{ii} can be written

$$\frac{1}{g_{11}} \frac{\partial \phi}{\partial \eta_1} \frac{\partial \sigma}{\partial \eta_1} + \frac{1}{g_{22}} \frac{\partial \phi}{\partial \eta_2} \frac{\partial \sigma}{\partial \eta_2} + \frac{1}{g_{33}} \frac{\partial \phi}{\partial \eta_3} \frac{\partial \sigma}{\partial \eta_3} = -1 \qquad (5.5.62)$$

This is an extension of Eq. (5.4.39). The path line of a fluid particle which is the locus of its position in space as σ increase is described by the ordinary differential equations system

$$\frac{d\eta_1}{\frac{1}{g_{11}}\left(\frac{\partial \phi}{\partial \eta_1}\right)} = \frac{d\eta_2}{\frac{1}{g_{22}}\left(\frac{\partial \phi}{\partial \eta_2}\right)} = \frac{d\eta_3}{\frac{1}{g_{33}}\left(\frac{\partial \phi}{\partial \eta_3}\right)} = -d\sigma \qquad (5.5.63)$$

For shorthand notation set

$$\frac{1}{g_{ii}} \frac{\partial \phi}{\partial \eta_i} = F_i(\eta_1, \eta_2, \eta_3) \qquad i = 1,2,3 \qquad (5.5.64)$$

These functions are known in principle because the potential ϕ is assumed known. Then

$$\frac{d\eta_1}{F_1} = \frac{d\eta_2}{F_2} = \frac{d\eta_3}{F_3} = -d\sigma \qquad (5.5.65)$$

To find σ it is necessary to solve the system

$$\frac{d\eta_1}{F_1} = \frac{d\eta_2}{F_2} \quad , \quad \frac{d\eta_2}{F_2} = \frac{d\eta_3}{F_3} \quad , \quad \frac{d\eta_1}{F_1} = \frac{d\eta_3}{F_3} \tag{5.5.66}$$

These equation in turn are equivalent to the ordinary differential equation system

$$\begin{aligned} \frac{d\eta_2}{d\eta_1} &= \frac{F_2}{F_1} = g_1(\eta_1 , \eta_2 , \eta_3) \\ \frac{d\eta_3}{d\eta_1} &= \frac{F_3}{F_2} = g_2(\eta_1 , \eta_2 , \eta_3) \end{aligned} \tag{5.5.67}$$

If f and g are analytic in the neighborhood of $(\eta_1^0 , \eta_2^0 , \eta_3^0)$ the Cauchy Majorant method assures the existence and uniqueness of the solution to the system (5.5.66). The solution has the general form

$$\begin{aligned} f_1(\eta_1 , \eta_2 , \eta_3 ; c_1 , c_2) &= 0 \\ f_2(\eta_1 , \eta_2 , \eta_3 ; c_1 , c_2) &= 0 \end{aligned} \tag{5.5.68}$$

In order to actually construct the solutions by quadratures a number of methods may be employed.

(a) It may happen that one of the equations in Eq. (5.5.66) can be integrated directly due to the absence of one of the variables. Let it be the first equation and suppose $u_1(\eta_1 , \eta_2) = c_1$ is its integral. With help of this one can eliminate η_1 from the second equation or η_2 from the third equation. One obtains respectively $u_2(\eta_2 , \eta_3) = c_2$ or $v_2(\eta_1 , \eta_3) = c_3$.

(b) It may happen that two of the equations (Eqs. (5.5.66)) are of such a form that they contain only the variables whose differentials enter the equations. Then one can integrate these two to obtain two solutions, say of the form

$$u_1(\eta_1,\eta_2) = c_1 \quad ; \quad u_2(\eta_1,\eta_3) = c_2 \tag{5.5.69}$$

(c) One can re-state the Eq. (5.5.66) with help of the fraction as follows

$$\frac{d\eta_1}{F_1} = \frac{d\eta_2}{F_2} = \frac{d\eta_3}{F_3} = \frac{k_1 d\eta_1 + k_2 d\eta_2 + k_3 d\eta_3}{k_1F_1 + k_2F_2 + k_3F_3} \qquad (5.5.70)$$

where k_1, k_2 and k_3 are any multipliers, not necessarily constants, which may be chosen as any functions of η_1, η_2, η_3. The choice can be made so that

i) The last expression in Eq. (5.5.70) taken with one of the first three terms yields an equation which can be solved by quadrature. With this integral one can proceed as done under (a).

ii) Two different choices of (k_1,k_2,k_3) are made so as to give two integrals

iii) Choose the multipliers so that

$$k_1F_1 + k_2F_2 + k_3F_3 = 0 \qquad (5.5.71)$$

Then the numerator in the last term in Eq. (5.5.70) must vanish also which gives rise to the Pfaffian equation

$$k_1 d\eta_1 + k_2 d\eta_2 + k_3 d\eta_3 = 0 \qquad (5.5.72)$$

If one defines the vector $\bar{k}$ with help of the unit vectors $\bar{a}_i$

$$\bar{k} = \bar{a}_1 k_1 + \bar{a}_2 k_2 + \bar{a}_3 k_3 \qquad (5.5.73)$$

the condition of integrability of Eq. (5.5.72) is that

$$\bar{k} \times \text{curl}\, \bar{k} = 0 \qquad (5.5.74)$$

If this is met, Eq. (5.5.72) can be integrated by standard means.

In summary the above method give rise to integrals of the form of Eq. (5.5.69). These equations can be used to construct the scalar function $\sigma(P)$ as follows. Consider that Eq. (5.5.65) is solved say for the last pair

$$\frac{d\sigma}{d\eta_3} = -F_1^{-1}(\eta_1,\eta_2,\eta_3) \qquad (5.5.75)$$

Now eliminate η_1 and η_2 from the right hand side with help of Eqs. (5.5.69). The resultant integral will be of the form

$$u_3 \equiv \sigma + h_o(\eta_3,c_1,c_2) = c_3 \qquad (5.5.76)$$

If c_1 and c_2 are replaced again with Eq. (5.5.69)

$$\mu_3 = \sigma + h(\eta_1,\eta_2,\eta_3) = c_3 \qquad (5.5.77)$$

where h is a specific function.

Now the general solution of the original partial differential Eq. (5.5.62) is of the form

$$u_3(\eta_1,\eta_2,\eta_3) = g(u_1(\eta_1,\eta_2)\ ,\ u_2(\eta_1,\eta_3)) \qquad (5.5.78)$$

where g is an arbitrary function. Thus one has finally

$$\sigma(\eta_1,\eta_2,\eta_3) = g(u_1(\eta_1,\eta_2)\ ,\ u_2(\eta_1,\eta_3) - h(\eta_1,\eta_2,\eta_3)) \qquad (5.5.79)$$

The arbitrary function g is determined from the boundary condition (Eq. 5.5.2) where Q is a point on the potential surface which coincides with the repository surface. As a very simple illustration consider the three-dimensional flow field into a sink. The potential function is given by

$$\phi(r) = -\frac{\lambda}{r} \quad . \qquad (5.5.80)$$

Equation (5.5.62) takes on the form with $\eta_1 = r$, $\eta_2 = \theta$, $\eta_3 = \psi$, $g_{11} = 1$, $g_{22} = r^2$, $g_{33} = (r\sin\theta)^2$ (spherical coordinate system)

$$\frac{\lambda}{r^2}\ \frac{\partial\sigma}{\partial r} + 0\ \frac{\partial\sigma}{\partial\theta} + 0\ \frac{\partial\sigma}{\partial\psi} = -1 \quad . \qquad (5.5.81)$$

The associated ordinary differential equations system (5.5.63) is

$$r^2 dr = \frac{d\theta}{0} = \frac{d\psi}{0} = -d\sigma \qquad (5.5.82)$$

so that θ and ψ are constant and

$$\lambda\sigma(r) = c - \frac{r^3}{3} \quad . \tag{5.5.83}$$

If $\sigma = 0$ at $r = R$ then

$$\sigma(P) = \frac{1}{3\lambda}(R^3 - r^3) \quad , \quad 0 \leq r \leq R \quad . \tag{5.5.84}$$

We will discuss the travel time position function $\sigma(P)$ for a number of three-dimensional flow fields of interest to use in a forthcoming report.

5.6. The Nuclide Transport in Presence of Dispersion

In this last section we introduce the effects of dispersion on the nuclide transport in two-dimensional hydrological flow fields. With dispersion taken into account Eq. (5.2.23) reads

$$\frac{\partial C_n}{\partial t} + \frac{1}{K_n}\left(V_x \frac{\partial C_n}{\partial x} + V_y \frac{\partial C_n}{\partial y}\right) + \lambda_n C_n = \frac{D}{K_n}\left(\frac{\partial^2 C_n}{\partial x^2} + \frac{\partial^2 C_n}{\partial y^2}\right) + \lambda_{n-1} C_{n-1} \tag{5.6.1}$$

The velocity components are determined with help of either the potential function $\phi(x,y)$ or the stream function $\psi(x,y)$ by using Eq. (5.2.2)

$$V_x = \frac{\partial\phi}{\partial_x} = \frac{\partial\psi}{\partial y} \, , \; V_y = \frac{\partial\phi}{\partial y} = -\frac{\partial\psi}{\partial x} \quad . \tag{5.6.2}$$

Recall that ϕ and ψ are harmonic functions

$$\nabla^2\phi = 0 \quad , \quad \nabla^2\psi = 0 \quad , \tag{5.6.3}$$

and that they form an orthogonal coordinate system

$$(\nabla\phi)\cdot(\nabla\psi) = 0 \quad . \tag{5.6.4}$$

As in Section 5.2 one introduces instead of the Cartesian coordinate system (x,y) the new coordinates $\phi(x,y)$, $\psi(x,y)$. The convective terms in Eq. (5.6.1) becomes with help of Eqs. (5.6.2) (see also Eq. (5.2.5)),

$$V_x \frac{\partial C_n}{\partial x} + V_y \frac{\partial C_n}{\partial y} = q^2 \frac{\partial C_n}{\partial \phi} \tag{5.6.5}$$

where

$$q^2 = V_x^2 + V_y^2 \tag{5.6.6}$$

is the square of the fluid velocity vector. The Laplacian operator transforms as follows

$$\frac{\partial^2 C_n}{\partial x^2} + \frac{\partial^2 C_n}{\partial y^2} = (\nabla^2 \phi) \frac{\partial C_n}{\partial \phi} + q^2 \frac{\partial^2 C_n}{\partial \phi^2} + 2(\nabla \phi \cdot \nabla \psi) \frac{\partial^2 C_n}{\partial \phi \partial \psi}$$
$$+ (\nabla^2 \psi) \frac{\partial C_n}{\partial \psi} + q^2 \frac{\partial^2 C_n}{\partial \psi^2} \tag{5.6.7}$$

In view of Eqns. (5.6.3) and (5.6.4) this simplifies to (Lamme, 1835)

$$\frac{\partial^2 Cn}{\partial x^2} + \frac{\partial^2 Cn}{\partial y^2} = q^2 \left(\frac{\partial^2 Cn}{\partial \phi^2} + \frac{\partial^2 Cn}{\partial \psi^2} \right) \tag{5.6.8}$$

When Eqs. (5.6.5) and (5.6.8) are combined with Eq. (5.6.1) there results

$$\frac{\partial C_n}{\partial t} + \frac{q^2(\phi,\psi)}{K_n} \frac{\partial C_n}{\partial \phi} + \lambda_n C_n = D \frac{q^2(\phi,\psi)}{K_n} \left(\frac{\partial^2 C_n}{\partial \phi^2} + \frac{\partial^2 C_n}{\partial \psi^2} \right) + \lambda_{n-1} C_{n-1} \,, \quad n=1,2 \ldots \lambda_0=0 \tag{5.6.9}$$

In this equation one must treat q^2 as a function of the independent variables ϕ,ψ. This relation has already been derived in Eq. (5.2.26)

$$q^2(\phi,\psi) = \left| \frac{dz}{dw} \right|^{-2} \quad . \tag{5.6.10}$$

Recall that one can always construct in principle the inverse function $z = f(w)$ to the complex potential $w = F(z)$. With $f(w)$ the derivative on the right hand side is then known. The side conditions for Eq. (5.6.9) are, see Eqs. (5.3.8) through (5.3.12)

$$C_n(\phi,\psi,0) = 0 \tag{5.6.11}$$

$$C_n(\phi_o,\psi, t) = C_n^o g_n(\psi, t) \tag{5.6.12}$$

Equation (5.6.9) is a variable coefficient linear, parabolic partial differential equations system. The solution to this set of governing equations will be discussed in our next report.

I wish to acknowledge many helpful discussions with E. J. Pinney about this work.

5.7. Literature References

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H1. M. Harada, P. L. Chambre', M. Foglia, K. Higash; F. Iwamoto, D. Leung, T. H. Pigford, D. Ting, "Migration of Radionuclides through Sorbing Media, Analytical Solutions--I," LBL-10500, 1980.

6. Finite Element Analysis of Radionuclide Migration

6.1 Introduction

The prediction of the space and time dependent concentration of radionuclides in geologic media from various analytical and/or numerical methods may give some important implications in the underground storage and disposal of high level nuclear wastes. Analytical solutions of the one dimensional transport equation for aqueous concentrations of nuclides in sorbing media, with and without dispersion, have been published (H1, H2, L1). Although these analytical solutions give exact expressions and valuable insight, they can be applied only to the limiting cases where the transport parameters are assumed to be constant or to the transport in geometrically simplified space. In order to solve the more general problem we must eventually rely on some numerical technique. The numerical solution to the radionuclide migration problem based on the finite difference method has been carried out by R. T. Dillon, R. B. Lantz, and S. B. Pahwa (D1) The finite difference method is a useful method to solve the partial differential equation directly, but this method is not well matched to the transport problem with complicated geometrical boundaries. A more effective method is that of finite element matrix analysis, which allows us to solve a more generalized problem (Y1).

We here present two matrix transport equations; one is for transport with constant water velocity and the other is for the general case. The matrix equations are applied to compute the aqueous concentrations of a three-member decay chain in a one dimensional geological medium with constant parameters. The finite element solutions for the concentration boundary condition are compared with the analytical solutions. It is found that the finite element approach gives a sufficiently good approximation to the exact analytical solution.

The finite-element solutions for a current boundary condition and those for transport with a dissolution source term are presented, and the differences resulting from these three different boundary situations are discussed.

The application of the finite element matrix equation to the transport in one dimensional, multi-layered geometric media and to the transport in multi-dimensional geologic media are also discussed.

6.2 Theoretical Approach

6.2.1 Basic Model Equations

The basic equation which governs the water flow in the porous medium can be derived in part from the equation of continuity

$$\frac{\partial}{\partial t}(\varepsilon\rho_w) + \nabla\cdot(\rho_w v) = q \qquad (6.2.1)$$

where v (v_x, v_y, v_z) is the velocity of water, ρ_w is the density of the water, $q(x,y,z,t)$ is the source of the water, ε is the porosity of the porous medium. The water velocity v is given by Darcy's law

$$v = -\frac{k}{\mu}\nabla\phi \qquad (6.2.2)$$

where k is the Darcy's constant $(k=(k_x, k_y k_z))$, μ is the viscosity of the water, and ϕ is the pressure and gravitational potentials

$$\phi = p - \rho_w\Omega \qquad (6.2.3)$$

For noncompressible fluids, Eq. (6.2.1) can be written as

$$\nabla\cdot(\rho_w v) = q \qquad (6.2.4)$$

Substitution of Eq (6.2.2) with Eq. (6.2.3) into Eq. (6.2.4) gives the basic equation which governs the static pressure

$$\frac{\partial}{\partial x}(k_x \frac{\partial p}{\partial x}) + \frac{\partial}{\partial y}(k_y \frac{\partial p}{\partial y}) + \frac{\partial}{\partial z}(k_z \frac{\partial p}{\partial z})$$

$$-\rho_w \frac{\partial}{\partial x}(k_x \frac{\partial\Omega}{\partial x}) - \rho_w \frac{\partial}{\partial y}(k_y \frac{\partial\Omega}{\partial y}) - \rho_w \frac{\partial}{\partial z}(k_z \frac{\partial\Omega}{\partial z}) = \frac{\mu}{\rho_w} q\ (x,y,z,t) \qquad (6.2.5)$$

where the Ω and q are known functions. When the pressure field is solved as a function of the space, we can immediately calculate the profiles of the water velocity from Darcy's law.

The transport equation for the aqueous concentration N_i of nuclide i in ground water flowing at velocity v through a porous medium results from the mass balance

$$\frac{\partial}{\partial t}(K_iN_i) + \frac{\partial}{\partial x}(v_xN_i) + \frac{\partial}{\partial y}(v_yN_i) + \frac{\partial}{\partial z}(v_zN_i) - \frac{\partial}{\partial x}(D_x \frac{\partial N_i}{\partial x}) - \frac{\partial}{\partial y}(D_y\frac{\partial N_i}{\partial y})$$

$$- \frac{\partial}{\partial z}(D_z \frac{\partial N_i}{\partial z}) + \lambda_iK_iN_i - \lambda_{i-1}K_{i-1}N_{i-1} = 0, \ \lambda_0 = 0, \ i=1,2,3,\ldots \qquad (6.2.6)$$

where K_i is the sorption constant defined by $K_i = 1+(1-\varepsilon)K_{Di}/\varepsilon$, K_{Di} is the distribution coefficient, D_x,D_y, and D_z are components of the dispersion coefficient D, and λ_i is the decay constant of nuclide i. Generally the sorption constant, dispersion coefficient, and water velocity are given by space-dependent functions.

Before the initial release of nuclides begins the concentration of each nuclide is considered to be zero.

$$N_i(x,y,z,0) = 0 \qquad (6.2.7)$$

These systematic equations should be solved simultaneously under the appropriate boundary conditions.

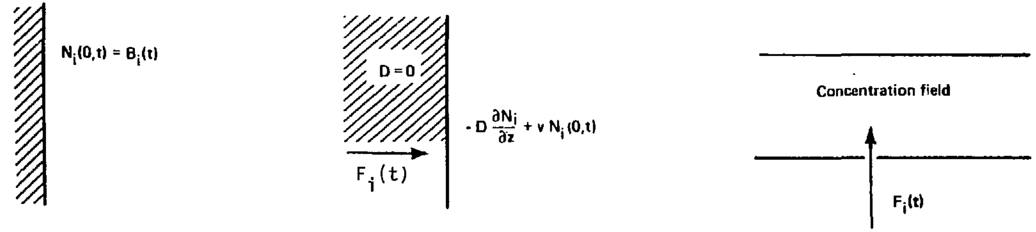
6.2.2 Boundary Condition

For the hydrogeological transport equation, three different types of source boundary conditions are considered. One is the concentration boundary condition, the boundary problem of the first kind. Another is the current expression, which includes convective and dispersive transport at the boundary, the boundary problem of the third kind. The analytical solutions for the first kind have been obtained by D. H. Lester, G. Jansen, and H. C. Burkholder (L1) and the analytical solution for the third kind has been presented in recursive form in our previous work (H1). Another important and realistic boundary condition is the source condition, which can be directly inserted in the basic transport equation (H1). The physical schemes of these three different boundary conditions are shown in Fig. 6.2.1.

The infinite medium boundary condition is

$$N_i(x,y,z,t) = 0 \ , \qquad x,y,z \in S_I \qquad (6.2.8)$$

where S_I denotes the boundary at infinite distance from the repository.



(a) Concentration boundary condition

(b) Current boundary condition

(c) Transport with source

Fig. 6.2.1 Physical scheme of three different boundary conditions

For a step release, the concentration boundary condition at the repository is given by

$$N_i(x,y,z,t) = B_i(t) \qquad x,y,z \in S_R \tag{6.2.9}$$

where S_R denotes the surface of repository. The $B_i(t)$ is given by the Bateman equation

$$B_i(t) = \sum_{j=1}^{i} b_{ji} \exp(-\lambda_j t) \tag{6.2.10}$$

with the Bateman coefficient

$$b_{ji} = \sum_{m=1}^{j} N_m^0 \Big(\prod_{l=m}^{i} \lambda_l \Big) \lambda_i / \prod_{\substack{l=m \\ (l \neq j)}}^{i} (\lambda_l - \lambda_j) \tag{6.2.11}$$

If we assume that the nuclear waste matrix and its contained radionuclides dissolve at a constant leach rate over the time period T and neglect the dispersive transport at the repository, the initial concentration of i-th nuclide can be written by

$$N_i^0 = M_i^0 / QT \tag{6.2.12}$$

where M_i^0 is the initial amount of nuclide i per unit initial amount of waste, and Q is the volume flow rate of water per unit time, per unit initial amount of waste.

For a band release, the concentration boundary condition is given by

$$N_i(x,y,z,t) = B_i(t)\,[h(t) - h(t-T)], \quad x,y,z \in S_R \tag{6.2.13}$$

Another important boundary condition is that of the third kind which results from the consideration that the leach rate of nuclide must be equal to a sum of the rates of convective and diffusional transport, thus

$$\frac{M_i^0}{T\,S}\,[h(t) - h(t-T)] = -D\nabla N_i + v\, N_i(x,y,z,t)\ , \quad x,y,z \in S_R \tag{6.2.14}$$

where S_R is the area of waste surface in the water phase.

In the special case without diffusional transport at the repository, Eq. (6.2.14) becomes

$$\frac{M_i^0}{T\ S}\,[h(t) - h(t-T)] = v\ N_i(x,y,z,t) \tag{6.2.15}$$

This is equivalent to Eq. (6.2.13).

6.3 Finite Element Approach

We apply the finite element method of weighted residuals to solve the system of equations for water flow and hydrogeological transport. We present two matrix equations. One is convenient for solving the problem with constant migration velocity and with concentration boundary condition. The other is convenient for solving the problem with the current boundary condition.

6.3.1 Matrix Equation of Darcy Flow

Consider the pressure, graviational potential, and source fields in a given arbitrary element. We assume these fields to be approximated as

$$P^{(e)} = [H^P]\{P\} = \sum_i^M H_i^P(x,y,z)P_i \tag{6.3.1}$$

$$\Omega^{(e)} = [H^\Omega]\{\Omega\} = \sum_i^M H_i^\Omega(x,y,z)\Omega_i \tag{6.3.2}$$

$$q^{(e)} = [H^q]\ \{q\} = \sum_i^M H_i^q(x,y,z)q_i \tag{6.3.3}$$

where H^P, H^Ω, H^q are the interpolation shape functions for the pressure, gravitational potential, and water source fields, P_i, Ω_i, and q_i are the numerical values of the pressure, potential, and of the flux of the water at i-th node of the element, and m is the number of nodes in the element considered. The notations [ ] and { } denote the row and column matrixes, respectively.

Generally the Darcy constant is space dependent, and we express the components as

$$k_x^{(e)} = [H^k]\,\{k_x\} \tag{6.3.4}$$

$$k_y^{(e)} = [H^k]\,\{k_y\} \tag{6.3.5}$$

$$k_z^{(e)} = [H^k]\,\{k_z\} \tag{6.3.6}$$

In Eqs. (6.3.1) ~ (6.3.6), different shape functions are used, but use of a single shape function will not introduce significant errors. To simplify the mathematical treatment, we put

$$[H^p] = [H\ ] = [H^q] = [H^k] = [H^v] = [H] \tag{6.3.7}$$

According to the Galerkin's method of weighted residuals, multiplying the both sides of Eq. (6.2.5) by the weight function H_i and integrating the resultant equation over the volume of the element, we have

$$\begin{aligned}
&\int_{Ve} H_i\,\Big[\frac{\partial}{\partial x}\Big(k_x\,\frac{\partial p^{(e)}}{\partial x}\Big) + \frac{\partial}{\partial y}\Big(k_y\,\frac{\partial p^{(e)}}{\partial y}\Big) + \frac{\partial}{\partial z}\Big(k_z\,\frac{\partial p^{(e)}}{\partial z}\Big)\Big]\,dV_e \\
&\quad -\rho_w\int_{Ve} H_i\Big[\frac{\partial}{\partial x}\Big(k_x\,\frac{\partial \Omega}{\partial x}\Big)^{(e)} + \frac{\partial}{\partial y}\Big(k_y\,\frac{\partial \Omega}{\partial y}\Big)^{(e)} + \frac{\partial}{\partial z}\Big(k_z\,\frac{\partial \Omega}{\partial z}\Big)^{(e)}\Big]\,dVe \\
&\quad = \frac{\mu}{\rho_w}\int_{Ve} H_i\,q(x,y,z,t)\,dVe
\end{aligned} \tag{6.3.8}$$

After integrating the first and second terms by parts, we have

$$\begin{aligned}
&\int_{Se}\Big(H_i k\,\frac{\partial p}{\partial n}\Big)dS_e - \ Ve\Big[\sum_{q=x,y,z} k_q\,\frac{\partial H_i\,\partial p^{(e)}}{\partial q\ \partial q}\Big]\,dVe \\
&\quad -\rho_w\ \ Se\Big(H_i\ k\,\frac{\partial p}{\partial n}\Big)dS_e + \rho_w\int_{Ve}\Big[\sum_{q=x,y,z} k_q\,\frac{\partial H_i}{\partial q}\,\frac{\partial \Omega^{(e)}}{\partial q}\Big]dVe \\
&\quad = \frac{\mu}{\rho_w}\int_{Ve} H_i q\ dVe
\end{aligned} \tag{6.3.9}$$

Substitution of Eq. (6.3.1) ~ (6.3.6) into Eq. (6.3.9) gives the finite element equation for Darcy flow in porous media

$$\int_{Ve} [\sum_{q=x,y,z} (\frac{\partial H_i}{\partial q}[H]\{k_q\} \frac{\partial [H]}{\partial q})]dV_e]\ dVe\ \{P_i\}$$

$$= \rho_w \int_{Ve} [\sum_{q=x,y,z} (\frac{\partial H_i}{\partial q}[H]\{k_q\}\frac{\partial [H]}{\partial q})]dVe\{\Omega_i\} + \frac{\mu}{\rho_w} \int_{Ve}[H]\{q\}dVe$$

$$- \int_{Se}(H_i k\ \frac{\partial p^{(e)}}{\partial n})\ dSe + \rho w \int_{Se}(H_i k\ \frac{\partial \Omega^{(e)}}{\partial n})\ dSe \tag{6.3.10}$$

or in matrix form:

$$[k_V^e]\{p\} = \rho_w[k_V^e]\{\Omega\} + \{S_w^e\} - \{s_p^e\} + \rho_w\ \{s_\Omega^e\} \tag{6.3.11}$$

where

$$[k_V^e] = \int_{Ve}(\frac{\partial [H]}{\partial x}[H]\{k_x\}\frac{\partial [H]}{\partial x} + \frac{\partial [H]}{\partial y}[H]\{k_y\}\frac{\partial [H]}{\partial y}$$

$$+ \frac{\partial [H]}{\partial z}[H]\{k_z\}\frac{\partial [H]}{\partial z})dVe \tag{6.3.12}$$

$$\{S_w^e\} = \frac{\mu}{\rho_w} \int_{Ve}[H]\{q\}\ dVe \tag{6.3.13}$$

$$\{s_p^e\} = \int_{Se}(H_i\ k\frac{\partial p^{(e)}}{\partial n})dSe \tag{6.3.14}$$

$$\{s_\Omega^e\} = \int_{Se}(H_i k\ \frac{\partial\ ^{(e)}}{\partial n})dSe. \tag{6.3.15}$$

This is an inhomogeneous algebraic equation for $p_1, p_2, \ldots p_m$ and should be solved if given values of Ω_i and q_i. After construction of the global matrix, the pressure gradient and gravitational potential gradient surface-integrals will vanish from a force balance at the interface area of adjacent elements.

6.3.2 Matrix Transport Equation with Concentration Boundary Condition

We approximate the concentration field in a given element as

$$N_i^{(e)} = [H^N]\, N_i \quad = \sum_k^m H_k^N N_{i,k} \tag{6.3.16}$$

The H_k^N (k=1,2,...m) are shape functions, $N_{i,k}$ is a value of N_i at k-th node of the element, and m is the number of nodes. As stated in Section 6.2, the dispersion coefficient, sorption constant, and the water velocity are generally given by the functions of space

$$D_q^{(e)} = [H^D]\,\{D_q\} = \sum_k^m H_k^D\, D_{q,k}\ ,\ q = x,y,z \tag{6.3.17}$$

$$K_i^{(e)} = [H^K]\,\{K_i\} = \sum_k^m H_k^K K_{i,k} \tag{6.3.18}$$

$$v_q^{(e)} = [H^V]\,\{v_q\} = \sum_k^m H_k^V\, v_{q,k}\ ,\ q = x,y,z \tag{6.3.19}$$

From the similar consideration used in above section, we put

$$H^N = H^D = H^K = H^V = H \tag{6.3.20}$$

In accordance with Galerkin's weighted residual method, multiplying the both sides of Eq. (6.2.6) by the shape function H_k and integrating the resultant equation over the volume of the element, we have the finite matrix representation

$$\begin{aligned}\int_{Ve} H_k \Big[&\frac{\partial}{\partial t}(K_i N_i^{(e)}) + \frac{\partial}{\partial x}(v_x N_i^{(e)}) + \frac{\partial}{\partial y}(v_y N_i^{(e)}) + \frac{\partial}{\partial z}(v_z N_i^{(e)}) \\ &- \frac{\partial}{\partial x}\Big(D_x \frac{\partial N_i^{(e)}}{\partial x}\Big) - \frac{\partial}{\partial y}\Big(D_y \frac{\partial N_i^{(e)}}{\partial y}\Big) - \frac{\partial}{\partial z}\Big(D_z \frac{\partial N_i^{(e)}}{\partial z}\Big) \\ &+ \lambda_i K_i N_i^{(e)} - \lambda_{i-1} K_{i-1} N_{i-1}^{(e)}\Big]\, dVe = 0\end{aligned} \tag{6.3.21}$$

The diffusive terms can be reduced to much a simpler expression, which involves only the space derivatives of the first order, by integrating by parts:

$$\int_{Ve} H_k\Big[\sum_{q=x,y,z} \frac{\partial}{\partial q}\Big(D_q \frac{\partial N_i^{(e)}}{\partial q}\Big)\Big]\, dVe = -\int_{Ve}\Big[\sum_{q=x,y,z} D_q \frac{\partial H_k}{\partial q}\frac{\partial N_i^{(e)}}{\partial q}\Big]\, dVe$$

$$-\int_{Se} H_k\, D \frac{\partial N_i^{(e)}}{\partial n}\, dSe \qquad (6.3.22)$$

where we used the Gauss theorem to replace the divergence terms by the surface integral terms.

At this stage, we approximate the nonsteady term by a finite difference and write as

$$\frac{\partial(K_i N_i^{(e)})}{\partial t} = \frac{K_i N_i^{(e)}(t) - K_i N_i^{'(e)}(t-\Delta t)}{\Delta t} \quad . \qquad (6.3.23)$$

Substituting Eqs. (6.3.22) and (6.3.23) into Eq. (6.3.21) and arranging the result, we have

$$\int_{Ve} H_k \Big[K_i\Big(\frac{1}{\Delta t} + \lambda_i\Big)[H] + \sum_{q=x,y,z} [H]\{v_q\}\frac{\partial[H]}{\partial q}$$

$$+ \sum_{q=x,y,z} \frac{\partial[H]}{\partial q}\{v_q\}[H] + \sum_{q=x,y,,,z} D_q \frac{\partial H_k}{\partial q}\frac{\partial[H]}{\partial q}\Big]\, dv_e\, \{N_i\}$$

$$= \int_{Ve} H_k[H]\ \frac{K_i}{\Delta t}\{N_i'\} + \lambda_{i-1} K_{i-1}\{N_{i-1}\})dVe + \int_{Se} H_k\, D \frac{\partial N_i^{(e)}}{\partial n}\, dSe \qquad (6.3.24)$$

Equation (6.3.24) can be reduced to the algebraic equation after evaluation of the volume and surface integrals

$$[k^e]\,\{N_i\} = \{r^e\} + \{s^e\} \qquad (6.3.25)$$

where

$$[k^e] = \int_{Ve} \{H\}\Big[\Big(K_i\Big(\frac{1}{\Delta t} + \lambda_i\Big)[H] + \sum_{q=x,y,z} [H]\{v_q\}\frac{\partial[H]}{\partial q}$$

$$+ \sum_{q=x,y,z} \frac{\partial[H]}{\partial q}\{v_q\}[H] + \sum_{q=x,y,z} D_q \frac{\partial H_k}{\partial q}\frac{\partial[H]}{\partial q}\Big]\, dVe \qquad (6.3.26)$$

$$\{r^e\} = \int_{Ve} \{H\}\,[H]\,\left(\frac{K_i}{\Delta t}\{N_i'\} + \lambda_{i-1}\,K_{i-1}\,\{N_{i-1}\}\right) dVe \qquad (6.3.27)$$

$$\{s^e\} = \int_{Se} \{H\}\,D\,\frac{\partial N_i^{(e)}}{\partial n}\,dSe \qquad (6.3.28)$$

At the boundary between two adjacent elements, the continuity of mass is

$$\left[N_i^{(e)}\,v - D\,\frac{\partial N_i^{(e)}}{\partial n}\right]_{jth} = \left[N_i^{(e)}v - D\,\frac{\partial N_i^{(e)}}{\partial n}\right]_{(j+1)th} \qquad (6.3.29)$$

In the usual case where the concentration profiles and water velocity are continuous functions, Eq. (6.3.29) can be rewritten as

$$\left[-D\,\frac{\partial N_i^{(e)}}{\partial n}\right]_{j\,th} = \left[-D\,\frac{\partial N_i^{(e)}}{\partial n}\right]_{(j+1)th} \qquad (6.3.30)$$

With aid of this equation, the surface-integral terms in all elements, with the exception of the boundary element, cancel after assembling the matrix element equations into a single global equation. The global equation then takes the form

$$[k^g]\,\{N_i\} = \{r^g\} \qquad (6.3.31)$$

or

$$\sum_{j=1} k^g_{kj}\,N_{i,j} = r^g_k\,,\quad k = 1,2,3,\ \ldots\ n \qquad (6.3.31)$$

where n is the total number of nodes in the space of interest. Our problem is now reduced to that of solving the n linear, nonhomogeneous algebraic equations with the appropriate boundary condition.

This equation cannot be applied to the problem with the boundary condition of third kind, because the equation does not include the current term given by Eq. (6.2.14) in explicit form. But use of this equation is convenient for the concentration boundary condition, in which the diffusional transport at the boundary is strictly zero.

6.3.3 Matrix Transport Equation with Current Boundary Condition

It is necessary to extend the matrix equation in 6.3.2 to include the problem with a discontinuous concentration field. In order to derive the matrix element equation in more general form, we split the convective transport term in Eq. (6.3.21) into two terms. Thus,

$$\int_{Ve} \sum_{q=x,y,z} \frac{\partial}{\partial q} (v_q N_i^{(e)}) dV_e = \int_{Ve} \sum_{q=x,y,z} v_q \frac{\partial N_i^{(e)}}{\partial q} dVe$$

$$+ \int \sum_{q=x,y,z} (\frac{\partial v_q}{\partial q} N_i^{(e)}) dVe \qquad (6.3.32)$$

Substituting Eqs. (6.3.23) and (6.3.29) into Eq. (6.3.21) and arranging the resulting equation by use of the similar way in 6.3.2, we have

$$[k^e] \{N_i\} = \{r^e\} + \{s^e\} \qquad (6.3.33)$$

but with

$$[k^e] = \int_{Ve} \{H\} [(\frac{K_i}{\Delta t} + \lambda_i K_i)[H] - \sum_{q=x,y,z} [H] \{v_q\} [H] \frac{\partial [H]}{\partial q}$$

$$+ \sum_{q=x,y,z} D_q \frac{\partial \{H\}}{\partial q} \frac{\partial [H]}{\partial q}] dVe \qquad (6.3.34)$$

$$\{r^e\} = \int_{Ve} \{H\} [H] (\frac{K_i}{\Delta t} \{N_i'\} + \lambda_{i-1} K_{i-1} \{N_{i-1}\}) dVe \qquad (6.3.35)$$

$$\{s^e\} = \int_{Se} \{H\} (D \frac{\partial N_i^{(e)}}{\partial n} - v N_i^{(e)}) dSe \qquad (6.3.36)$$

Assembly of the element matrix equations with help of the relation given by Eq. (6.3.29) yields the global equation

$$[k^g] \{N_i\} = \{r^g\} + \{s^g\} \qquad (6.3.37)$$

or

$$\sum_{j=1}^{n} k_{kj}^{g} N_{i,j} = r_{k}^{g} \quad , \quad k = 1,2,3, \ldots$$
$$\sum_{j=1}^{n} k_{kj}^{g} N_{i,j} = r_{k}^{g} + s_{k}^{g} \ , \ k = \alpha,\beta,\gamma,\ldots \tag{6.3.37}$$

where the integers $\alpha,\beta,\gamma,\ldots$ denote the number of nodes at which the nuclide sources are located. Equation (6.3.37) involves the current surface integrations, which account for convective and dispersive transports. This matrix equation can be applied directly to solve the transport equation subject to the current boundary condition.

6.4 One Dimensional Transport with Concentration Boundary Condition-Constant Parameters

The purpose of this section is to show the applicability of the finite element approach to the migration of an arbitrary radionuclide chain in geologic media. For this purpose we consider the three-member radionuclide chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ in a one dimensional medium with constant physicochemical parameters, and we present a comparison of the finite element solutions with the analytical solutions (H1).

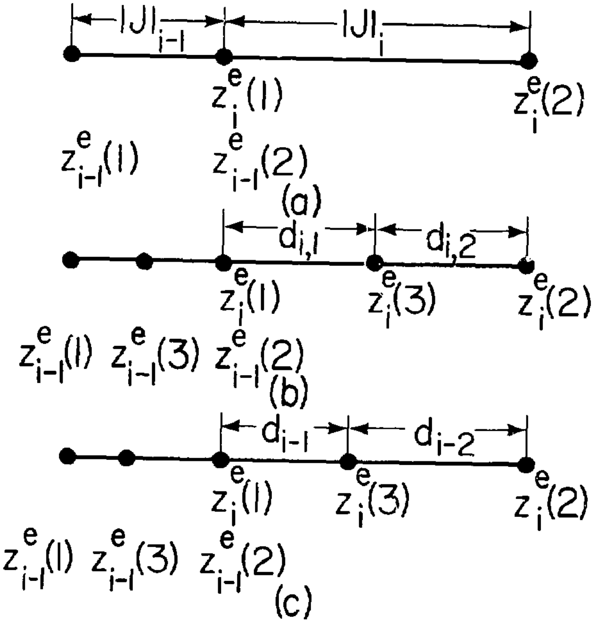
6.4.1 Numerical Method

We first check the error due to different ways of subdivision of the migration space. For this purpose we select three different elements. They are a) a two-nodal linear element, b) a three-nodal quadratic element with two rods of equal size, and c) a three-nodal quadratic element with two rods of different size as shown in Fig. 6.4.1. The linear and quadratic interpolation functions used in the computations are, respectively,

$$[H] = [L_1 \quad L_2] \tag{6.4.1}$$

$$[H] = [(2L_1-1)L_1 \quad (2L_2-1)L_2 \quad 4L_1L_2] \tag{6.4.2}$$

where $L_i (i=1,2)$ denotes the normalized, natural coordinate system. L_i takes the value from zero to unity. When the element a) or b) is used to subdivide the space, all memebers of the volume integrations in the matrix $[k^e]$ and r^e can be integrated analytically.



XBL 812-261

Fig. 6.4.1 Finite elements used in this computation, a) two-nodal linear element, b) three-nodal quadratic element with two rods of equal size, and c) three-nodal quadratic element with two rods of different size.

Let z_1 and z_2 be positions of two nodal points which are measured in the original z-scale, then we can write L_1 and L_2 as

$$L_1 = (z-z_1)/(z_2-z_1) = \xi \tag{6.4.3}$$

$$L_2 = (z_2-z)/(z_2-z_1) = 1 - \xi \tag{6.4.4}$$

where ξ is the relative coordinate measured from $z= z_1$. From Eqs. (6.4.3) and (6.4.4), we have the derivative of shape function

$$\frac{\partial[H]}{\partial z} = \frac{1}{|J|}\frac{\partial H}{\partial \xi} = \frac{1}{|J|}\begin{bmatrix} 1 & -1 \end{bmatrix} \tag{6.4.5}$$

where $|J|$ is the Jacobian $|J| = z_2-z_1$. Introducing Eqs. (6.4.1) and (6.4.5) into the integration terms which are included in the element matrices given by Eqs. (6.3.26) and (6.3.27) and using the integrations:

$$\int_{Ve}\{H\}\ [H]\ \ dVe = |J|\begin{bmatrix} 1/3 & 1/6 \\ 1/6 & 1/3 \end{bmatrix} \tag{6.4.6}$$

$$\int_{Ve}\{H\}\ \frac{\partial[H]}{\partial z}\ dVe = -\begin{bmatrix} 1/2 & -1/2 \\ 1/2 & -1/2 \end{bmatrix} \tag{6.4.7}$$

$$\int_{Ve}\frac{\partial\{H\}}{\partial z}\frac{\partial[H]}{\partial z}\ dVe = \frac{1}{|J|}\begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix} \tag{6.4.8}$$

we have

$$[k^e] = \left(\frac{K_i}{\Delta t} + \lambda_i K_i\right)|J|\begin{bmatrix} 1/3 & 1/6 \\ 1/6 & 1/3 \end{bmatrix} - v\begin{bmatrix} 1/2 & -1/2 \\ 1/2 & -1/2 \end{bmatrix} + \frac{D}{|J|}\begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix} \tag{6.4.9}$$

$$\{r^e\} = |J|\begin{bmatrix} 1/3 & 1/6 \\ 1/6 & 1/3 \end{bmatrix}\begin{bmatrix} (K_i/\Delta t)N'_{i,1} + \lambda_{i-1}K_{i-1}N_{i-1,1} \\ (K_i/\Delta t)N'_{i,2} + \lambda_{i-1}K_{i-1}N_{i-1,2} \end{bmatrix} \tag{6.4.10}$$

For element b) similar mathematical treatment gives

$$[k^e] = \left(\frac{K_i}{\Delta t} + \lambda_i K_i\right)|J| \begin{bmatrix} 2/12 & -1/30 & 1/15 \\ -1/30 & 2/15 & 1/15 \\ 1/15 & 1/15 & 8/15 \end{bmatrix}$$

$$- v \begin{bmatrix} 1/2 & 1/6 & -2/3 \\ -1/6 & -1/2 & 2/3 \\ 2/3 & -2/3 & 0 \end{bmatrix} + \frac{D}{|J|} \begin{bmatrix} 7/3 & 1/3 & -8/3 \\ 1/3 & 7/3 & -8/3 \\ -8/3 & -8/3 & 16/3 \end{bmatrix} \qquad (6.4.11)$$

$$\{r^e\} = |J| \begin{bmatrix} 2/15 & -1/30 & 1/15 \\ -1/30 & 2/15 & 1/15 \\ 1/15 & 1/15 & 8/15 \end{bmatrix} \begin{bmatrix} (K_i/\Delta t)N'_{i,1}+\lambda_{i-1}K_{i-1}N_{i-1,1} \\ (K_i/\Delta t)N'_{i,2}+\lambda_{i-1}K_{i-1}N_{i-1,2} \\ (K_i/\Delta t)N'_{i,3}+\lambda_{i-1}K_{i-1}N_{i-1,3} \end{bmatrix} \qquad (6.4.12)$$

where $|J| = z_2 - z_1$.

For the quadratic element c), the integrations of all members in the element matrix must be evaluated by some numerical technique. When the method of Gauss quadrature is employed, the elements k^e_{ij} and r^e_i can be calculated directly from

$$k_{ij} = \sum_{l=1}^{L} \left[\left(\frac{K_i}{\Delta t} + \lambda_i K_i\right)(H_i)_l (H_j)_l - v(H_i)_l \left(\frac{\partial H_j}{\partial z}\right)_l\right.$$

$$\left. + D_z \left(\frac{\partial H_i}{\partial z}\right)\left(\frac{\partial H_j}{\partial z}\right)_l\right] w_l \qquad (6.4.13)$$

$$r_i = \sum_{l=1}^{L} (H_i)_l \left[\left(\sum_{j=1}^{m} H_j N_{i,j}\right)_l + \lambda_{i-1} K_{i-1} \left(\sum_{j=1}^{m} H_j N_{i-1,j}\right)_l\right] w_l \qquad (6.4.14)$$

where

$$\frac{\partial H}{\partial z} = \frac{1}{|J|} [4L_1 - 1 \quad -4L_2 + 1 \quad 4L_2 - 4L_1] \qquad (6.4.15)$$

$$J = (4L_1-1)z_1 + (4L_2-4L_1)z_3 + (-4L_2+1)z_2 \qquad (6.4.16)$$

The $(H_i)_\ell$ denotes the value of the shape function at a given point $L_2=(L_2)_\ell$, and w_ℓ is the numerical weight factor. In this computation the 5-point approximation is used

ℓ	L_2	w_1
1	0.0469100770	0.1184634425
2	0.2307653449	0.2393143353
3	0.5	0.2844444444
4	0.7692346551	0.2393143353
5	0.9530899230	0.1184634425

In computing the finite element solution, the semi-infinite space was replaced by a finite space ranging from z=0 to z=10,436 m. This model space is sufficiently large enough to be regarded here as an infinite space, since radionuclides do not reach such a distance within a time scale of tens of thousands of years. The model space was divided into 50 elements and the size of each element was determined by a series of equal ratio, namely $|J_j| = a\ b^{j-1}$, with a=1 m and b=1.16 (see first column in Table 6.1).

The time step Δt=1000 year was used from t=0 to t=10,000 year. After this Δt=2000 year was used. The computations were continued until the time reached 52,000 year.

6.4.2 Numerical Results -Error Due to Subdivision

The finite element solutions were computed for the concentration profiles of the three-member decay chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ in a geologic medium with constant parameters. As an illustration of the results, the concentrations of ^{234}U, ^{230}Th, and of ^{226}Ra at time t=10,000 year, computed by use of three different elements, are compared in Tables 6.4.1 ~ 6.4.6. The assumed constant physicochemical parameters are listed at the bottom of the tables. As seen from a comparison among these three finite-element solutions, the errors resulting from the use of different elements are very small. The maximum error is less than ten percent, with the exception of the results at greater distance. Therefore, the two-nodal linear element which yields the most simple matrix transport equation is most preferable for use of computations.

Table 6.4.1 Finite-element solutions for $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ chain resulting from three different subdivisions, concentrations of ^{234}U at t=10,000 year, assumed parameters are $T=3.33 \times 10^4$ year, v=110 m/yr, $K_U=1.43 \times 10^4$, $K_{Th}=5.00 \times 10^4$, $K_{Ra}=5.00 \times 10^2$, $D=1.76 \times 10^3$ m^2/yr (For element b the distance should be replaced by $z_{2i}=(z_{2i-1}+z_{2i+1})/2$.)

Node	Distance	Element a) $N_1/N_1^0 \times 10$	Element b) $N_1/N_1^0 \times 10$	Element c) $N_1/N_1^0 \times 10$
1	0.000	9.7229	9.7229	9.7229
2	1.00	9.7087	9.7075	9.7088
3	2.16	9.6911	9.6910	9.6909
4	3.51	9.6691	9.6671	9.6692
5	5.06	9.6414	9.6412	9.6410
6	6.88	9.6064	9.6031	9.6066
7	8.98	9.5615	9.5611	9.5607
8	11.4	9.5035	9.4981	9.5042
9	14.2	9.4281	9.4274	9.4267
10	17.5	9.3290	9.3198	9.3305
11	21.3	9.1982	9.1972	9.1957
12	25.7	9.0243	9.0082	9.0272
13	30.9	8.7925	8.7914	8.7881
14	36.8	8.4839	8.4565	8.4889
15	43.7	8.0757	8.0752	8.0687
16	51.7	7.5424	7.4974	7.5471
17	60.9	6.8609	6.8628	6.8531
18	71.7	6.0183	5.9528	6.0119
19	84.1	5.0244	5.0323	5.0288
20	98.6	3.9251	3.8521	3.8973
21	115	2.8033	2.8234	2.8473
22	135	1.7909	1.7458	1.7718
23	157	0.98452	1.0015	1.0475
24	184	0.44724	0.44356	0.46438
25	214	0.15891	0.17070	0.20105
26	249	0.040882	0.046582	0.056257

Table 6.4.2 Finite-element solutions for $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ chain resulting from three different subdivisions, concentrations of ^{234}U at t=50,000 year, assumed parameters are $T=3.33 \times 10^4$ year, v=110 m/yr, $K_U=1.43 \times 10^4$, $K_{Th}=5.00 \times 10^4$, $K_{Ra}=5.00 \times 10^2$, $D=1.76 \times 10^3$ m^2/yr (For element b, the distance should be replaced by $z_{2i}=(z_{2i-1} + z_{2i+1})/2$.)

Node	Distance Z (m)	Element a) $N_1/N_1^0 \times 10$	Element b) $N_1/N_1^0 \times 10$	Element c) $N_1/N_1^0 \times 10$
1	0.00	0.0000000	0.0000000	0.0000000
2	1.00	0.0033915	0.0036980	0.0033665
3	2.16	0.0075926	0.0076464	0.0076566
4	3.51	0.012839	0.013370	0.012843
5	5.06	0.019450	0.019585	0.019616
6	6.88	0.027860	0.028662	0.027885
7	8.98	0.038666	0.038925	0.038997
8	11.4	0.052702	0.054249	0.052717
9	14.2	0.071139	0.071591	0.071753
10	17.5	0.095634	0.098338	0.095534
11	21.3	0.12855	0.12930	0.12966
12	35.7	0.17328	0.17813	0.17279
13	30.9	0.23469	0.235866	0.23671
14	36.8	0.31973	0.32863	0.31820
15	43.7	0.43825	0.43993	0.44195
16	51.7	0.60394	0.62041	0.60038
17	60.9	0.83515	0.83701	0.84176
18	71.7	1.1551	1.1847	1.1496
19	84.1	1.5907	1.5910	1.6005
20	98.6	2.1674	2.2155	2.1673
21	115	2.8997	2.8935	2.9035
22	135	3.7754	3.8365	3.7996
23	157	4.7367	4.7183	4.7071
24	184	5.6669	5.7047	5.7107
25	214	6.3949	6.3690	6.3174
26	249	6.7268	6.6866	6.7263
27	290	6.5001	6.4874	6.4268
28	338	5.6492	5.5336	5.5613
29	392	4.2720	4.2824	4.2781
30	456	2.6730	2.5986	2.6289
31	530	1.2877	1.2993	1.3601
32	616	0.43136	0.44459	0.48806

Table 6.4.3 Finite element solutions for $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ chain resulting from three different subdivisions, concentrations of ^{230}Th at t=10,000 year, assumed parameters are $T=3.33 \times 10^4$ year, v=110 m/yr, $K_U=1.43 \times 10^4$, $K_{Th}=5.00 \times 10^4$, $K_{Ra}=5.00 \times 10^2$, $D=1.76 \times 10^3$ m^2/yr, (For element b, the distance should be replaced by $z_{2i}=(z_{2i-1} + z_{2i+1})/2$.)

Node	Distance Z (m)	Element a) $N_2/N_1^0 \times 10$	Element b) $N_2/N_1^0 \times 10$	Element c) $N_2/N_1^0 \times 10$
1	0.00	2.6537	2.6537	2.6537
2	1.00	2.5837	2.5785	2.5837
3	2.16	2.5047	2.5046	2.5047
4	3.51	2.4141	2.4075	2.4141
5	5.06	2.3132	2.3130	2.3132
6	6.88	2.1982	2.1901	2.1982
7	8.98	2.0724	2.0720	2.0724
8	11.4	1.9305	1.9210	1.9305
9	14.2	1.7798	1.7789	1.7798
10	17.5	1.6121	1.6019	1.6121
11	21.3	1.4423	1.4406	1.4423
12	25.7	1.2575	1.2476	1.2575
13	30.9	1.0832	1.0802	1.0831
14	36.8	0.89970	0.89132	0.89970
15	43.7	0.74237	0.73799	0.74237
16	51.7	0.58393	0.57733	0.58393
17	60.9	0.46034	0.45559	0.46034
18	71.7	0.34066	0.33571	0.34066
19	84.1	0.25245	0.24857	0.25245
20	98.6	0.16863	0.16573	0.16863
21	115	0.11159	0.10837	0.11159
22	135	0.060985	0.059464	0.060985
23	157	0.033132	0.030691	0.033132
24	184	0.013104	0.012221	0.013104
25	214	0.0053444	0.0042892	0.0053444
26	249	0.0013560	0.0010674	0.0013560

Table 6.4.4 Finite element solutions for $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ chain resulting from three different subdivisions, concentrations of ^{230}Th at t=50,000 year, assumed parameters are $T=3.33 \times 10^4$ year, v=110 m/yr, $K_U=1.43 \times 10^4$, $K_{Th}=5.00 \times 10^4$, $K_{Ra}=5.00 \times 10^2$, $D=1.76 \times 10^3$ m^2/yr (For element b, the distance should be replaced by $z_{2i}=(z_{2i-1} + z_{2i+1})/2$.)

Node	Distance Z (m)	Element a) $N_2/N_1^0 \times 10^2$	Element b) $N_2/N_1^0 \times 10^2$	Element c) $N_2/N_1^0 \times 10^2$
1	0.00	0.000000	0.000000	0.000000
2	1.00	0.024066	0.026056	0.023738
3	2.16	0.053832	0.053831	0.053922
4	3.51	0.090870	0.093942	0.090344
5	5.06	0.13723	0.13721	0.13746
6	6.88	0.19557	0.20040	0.19468
7	8.98	0.26933	0.26924	0.26980
8	11.4	0.36291	0.37059	0.36144
9	14.2	0.48177	0.48149	0.48259
10	17.5	0.63250	0.64466	0.63042
11	21.3	0.82251	0.82179	0.82371
12	25.7	1.0593	1.0777	1.0576
13	30.9	1.3485	1.3468	1.3493
14	36.8	1.6910	1.7161	1.6935
15	43.7	2.0787	2.0751	2.0752
16	51.7	2.4886	2.5141	2.5002
17	60.9	2.8801	2.8738	2.8630
18	71.7	3.1975	3.2076	3.2110
19	84.1	3.3816	3.3736	3.3494
20	98.6	3.3926	3.3750	3.3852
21	115	3.2307	3.2256	3.2083
22	135	2.9419	2.9140	2.9262
23	157	2.5955	2.5956	2.5983
24	184	2.2474	2.2274	2.2479
25	214	1.9152	1.9192	1.9258
26	249	1.5845	1.5637	1.5815
27	290	1.2379	1.2430	1.2489
28	338	0.88099	0.86186	0.87227
29	392	0.54744	0.55200	0.56406
30	456	0.28151	0.27722	0.28646
31	530	0.11141	0.11448	0.12611
32	616	0.030530	0.033315	0.038836

Table 6.4.5 Finite element solutions for $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ chain resulting from three different subdivisions, concentrations of ^{230}Th at t=50,000 year, assumed parameters are $T=3.33 \times 10^4$ year, v=110 m/yr, $K_U=1.43 \times 10^4$, $K_{Th}=5.00 \times 10^4$, $K_{Ra}=5.00 \times 10^2$, $D=1.76 \times 10^3$ m^2/yr (For element b, the distance should be replaced by $z_{2i}=(z_{2i-1} + z_{2i+1})/2$.)

Node	Distance Z (m)	Element a) $N_3/N_1^0 \times 10^3$	Element b) $N_3/N_1^0 \times 10^3$	Element c) $N_3/N_1^0 \times 10^3$
1	0.00	0.41373	0.41373	0.41373
2	1.00	0.48170	0.48698	0.48211
3	2.16	0.55792	0.55778	0.55765
4	3.51	0.64289	0.64334	0.64335
5	5.06	0.73698	0.73663	0.73631
6	6.88	0.84031	0.84783	0.83920
7	8.98	0.95272	0.95210	0.95146
8	11.4	1.0736	1.0821	1.0744
9	14.2	1.2019	1.2010	1.1998
10	17.5	1.3360	1.3448	1.3370
11	21.3	1.4733	1.4719	1.4701
12	25.7	1.6109	1.6193	1.6122
13	30.9	1.7451	1.7433	1.7406
14	36.8	1.8718	1.8786	1.8731
15	43.7	1.9865	1.9844	1.9808
16	51.7	2.0851	2.0891	2.0862
17	60.9	2.1631	2.1609	2.1568
18	71.7	2.2169	2.2174	2.2177
19	84.1	2.2428	2.2408	2.2366
20	98.6	2.2378	2.2340	2.2377
21	115	2.1999	2.1986	2.1950
22	135	2.1295	2.1218	2.1280
23	157	2.0291	2.0288	2.0271
24	184	1.8936	1.8935	1.9016
25	214	1.7584	1.7594	1.7601
26	249	1.5999	1.5889	1.5981
27	290	1.4313	1.4326	1.4351
28	338	1.2566	1.2457	1.2548
29	392	1.0794	1.0810	1.0849
30	456	0.90369	0.89394	0.90207
31	530	0.73393	0.73547	0.74033
32	616	0.57497	0.56767	0.57447
33	716	0.43152	0.43277	0.43770
34	831	0.30764	0.30353	0.30875
35	965	0.20615	0.20705	0.21101
36	1120	0.12814	0.12682	0.13027
37	1300	0.072662	0.073242	0.075683
38	1510	0.036794	0.036910	0.038668
39	1750	0.016942	0.016509	0.017598
40	2030	0.0059845	0.0062648	0.0068668
41	2360	0.0017754	0.0018957	0.0022125

Table 6.4.6 Finite element solutions for $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ chain resulting from three different subdivisions, concentrations of ^{230}Th at t=50,000 year, assumed parameters are $T=3.33 \times 10^4$ year, v=110 m/yr, $K_U=1.43 \times 10^4$, $K_{Th}=5.00 \times 10^4$, $K_{Ra}=5.00 \times 10^2$, $D=1.76 \times 10^3\ m^2/yr$ (For element b, the distance should be replaced by $z_{2i}=(z_{2i-1} + z_{2i+1})/2$.)

Node	Distance Z (m)	Element a) $N_3/N_1^0 \times 10^2$	Element b) $N_3/N_1^0 \times 10^2$	Element c) $N_3/N_1^0 \times 10^2$
1	0.00	0.0000000	0.0000000	0.0000000
2	1.00	0.0024290	0.0026370	0.0024016
3	2.16	0.0054360	0.0054507	0.0054596
4	3.51	0.0091861	0.0095234	0.0091536
5	5.06	0.013899	0.013934	0.013960
6	6.88	0.019869	0.020420	0.019817
7	8.98	0.027493	0.027557	0.027616
8	11.4	0.037306	0.038220	0.037201
9	14.2	0.050031	0.050133	0.050256
10	17.5	0.066640	0.068180	0.066423
11	21.3	0.088429	0.088573	0.088827
12	25.7	0.11710	0.11971	0.11669
13	30.5	0.15482	0.15499	0.15549
14	36.8	0.20424	0.20861	0.20366
15	43.7	0.26840	0.26852	0.26939
16	51.7	0.35042	0.35734	0.35007
17	60.9	0.45290	0.45274	0.45381
18	71.7	0.57708	0.58679	0.57793
19	84.1	0721894	0.72100	0.72115
20	98.6	0.88312	0.89426	0.88532
21	115	1.0541	1.0523	1.0502
22	135	1.2264	1.2364	1.2285
23	157	1.3921	1.3893	1.3852
24	184	1.5438	1.5506	1.5461
25	214	1.6746	1.6712	1.6655
26	249	1.7753	1.7766	1.7777
27	290	1.8335	1.8304	1.8234
28	338	1.8352	1.8273	1.8323
29	392	1.7702	1.7687	1.7636
30	456	1.6386	1.6238	1.6308
31	530	1.4535	1.4536	1.4545
32	616	1.2380	1.2242	1.2333
33	716	1.0159	1.0169	1.0226
34	831	0.80416	0.79461	0.80371
35	965	0.61245	0.61351	0.61988
36	1120	0.44632	0.44092	0.44816
37	1300	0.30899	0.30977	0.31477
38	1510	0.20151	0.19951	0.20435
39	1750	0.12257	0.12314	0.12621
40	2030	0.068737	0.068595	0.071158
41	2360	0.035045	0.035397	0.036858

6.4.3 Comparison with Analytical Solutions

The analytical solutions for concentrations of a three member decay chain for band release have been presented in our previous report (H1).

$$N_i(z,t) = N_i^S(z,t) - N_i^S[z,\ t-T,\ b_{ji}\exp(-\lambda_j T)]h(t-T) \tag{6.4.17}$$

where the N_i^S is the solution for step release

$$N_1^S(z,t) = b_{11}E(1,1;1) \tag{6.4.18}$$

$$N_2^S(z,t) = \sum_{j=1}^{2} b_{j2}E(j,j;2) + \frac{b_{11}\lambda_1}{v_1(\Lambda_{12}-\lambda_1\Gamma_{12})}[E(1,1;2) - E(1,1;1)$$

$$+ E(1,2;1) - E(1,2;2)] \tag{6.4.19}$$

$$N_3(z,t) = \sum_{j=1}^{3} b_{j3}E(j,j;3) + \frac{\lambda_2}{v_2} \sum_{j=1}^{2} \frac{b_{j2}}{(\Lambda_{23}-\lambda_j\Gamma_{23})}[E(j,j;3)$$

$$- E(j,j;2) + E(2,3;2) - E(2,3;3)]$$

$$+ \frac{\lambda_1\lambda_2 b_{11}}{v_1 v_2} \sum_{\substack{j=1\\k\neq j\\ \ell\neq j}}^{3} \frac{E(1,1;j)}{[(\Lambda_{kj}-\lambda_1\Gamma_{kj})(\Lambda_{lj}-\lambda_1\Gamma_{lj})}$$

$$+ \frac{\Gamma_{kj}E(k,j;j)}{(\Lambda_{kj}-\lambda_1\Gamma_{kj})(\Gamma_{lj}\Lambda_{kj}-\Gamma_{kj}\Lambda_{lj})} + \frac{\Gamma_{lj}E(l,j;j)}{(\Lambda_{lj}-\lambda_1\Gamma_{lj})(\Gamma_{kj}\Lambda_{lj}-\Gamma_{lj}\Lambda_{kj})}] \tag{6.4.20}$$

with

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

$$E(i,j;k) = e^{(z/2\alpha-\ \beta_{ij}t)} \frac{2}{\sqrt{\pi}} \int_{\frac{z}{\sqrt{4\alpha v_k t}}}^{\infty} \exp -[y^2+\gamma(z/4\alpha y)^2]\, dy \tag{6.4.21}$$

$$\beta_{ij} = \begin{cases} \lambda_i & i=j \\ \Lambda_{ij}/\Gamma_{ij} & i\neq j \end{cases}$$

$$\gamma = 1 + 4\alpha(\lambda_k-\beta_{ij})/v_k$$

$$\alpha = D/v, \qquad v_i=v/K_i$$

The finite element solutions for the concentration profiles of three-member decay chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ at t=10,000 year and at t=50,000 year, which are computed by use of the linear element are compared with the analytical solutions in Figs. 6.4.2 and 6.4.3. The assumed parameters used in calculations are included in these figures. The concentration profiles of ^{234}U and of ^{230}Th at t=10,000 year ($t<T$) computed by the finite-element method agree well with those given by the analytical solutions.

Although the finite-element solution for the concentration of ^{226}Ra gives slightly higher values than those given by the analytical solution, its concentration curve is closely related to that from the analytical solution. At the leach time T, the concentration at the repository becomes zero discontinuously, subject to the band-release boundary condition. This discontinuous concentration with respect to time t at the repository can introduce error in numerical results at time t greater than T. In analytical treatments the superposition theorem is easily applied (H1), but in numerical analysis there is no effective method corresponding to superposition. In spite of this difficulty at $t>T$, the finite element solution gives a good approximation to the exact solution, as shown in Fig. 6.4.3. In this computation the time step used is every 1000 year at $t<10,000$ year and every 2000 year at $t\geq 10,000$ year, whereas the leach time is T=33,300 year. If one selects the time step such that the leach time T is included in the commutation time series $t=(n-1)\Delta t$ $(n=1,2,..)$, better results are obtained.

Comparisons of finite-element solutions with the analytical solutions given in Figs. 6.4.2 and 6.4.3 demonstrate that the finite element method is a useful numerical technique to predict the migration behavior of radionuclide chains in geologic media.

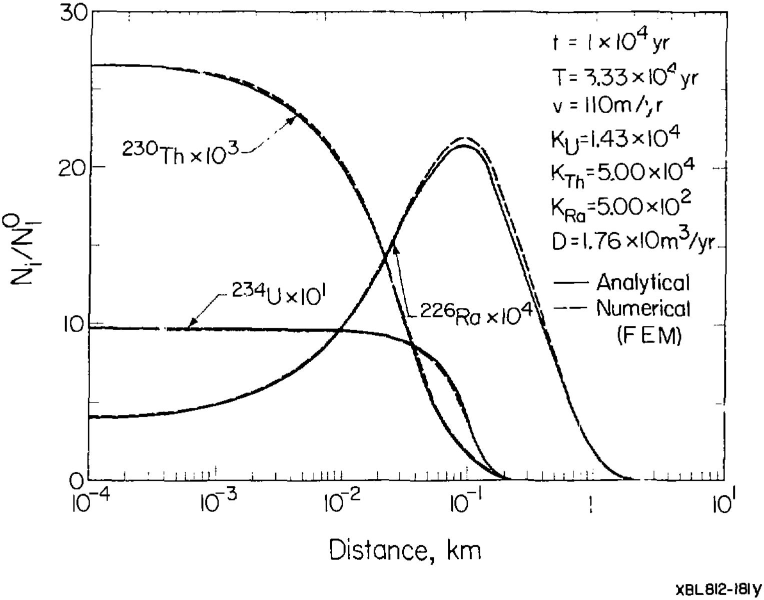


Fig. 6.4.2 Comparison of finite element solution with analytical solution, concentration profiles of the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain at t = 1 x 10^4 year, concentration boundary condition.

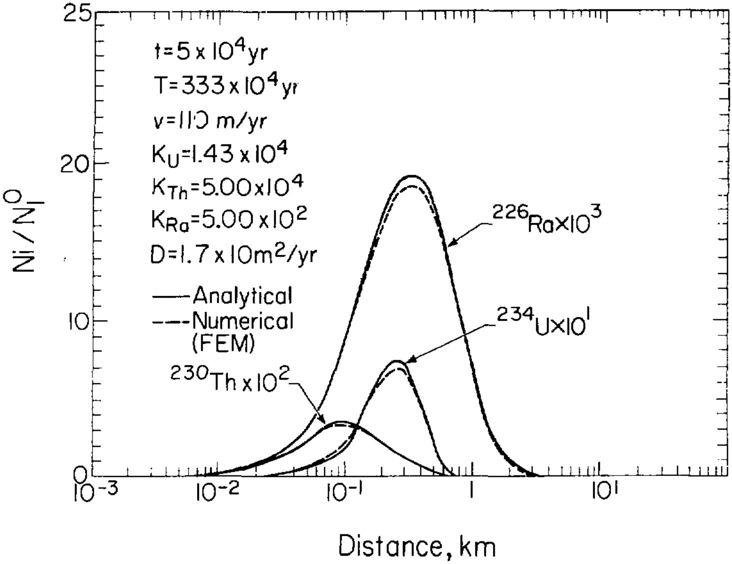


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Fig. 6.4.3 Comparison of finite element solution with analytical solution, concentration profiles of the $^{234}U \rightarrow$ $^{230}Th \rightarrow$ ^{226}Ra decay chain at t = 5 x 10^4 year, concentration boundary condition.

6.5. One Dimensional Transport with Current Boundary Condition

We describe here the application of the general matrix transport equation given by Eq. (6.3.33) to the one-dimensional problem with constant parameters.

6.5.1 Numerical Method

According to the similar mathematical treatment in Sec. 6.4.11, we can reduce the matrix equation into the simpler algebraic equation. The members included in the element matrices given by Eqs. (6.3.34) and (6.3.35) are for element a)

$$[k^e] = \left(\frac{K_i}{\Delta t} + \lambda_i K_i\right) |J| \begin{bmatrix} 1/3 & 1/6 \\ 1/6 & 1/3 \end{bmatrix} + v \begin{bmatrix} 1/2 & 1/2 \\ -1/2 & -1/2 \end{bmatrix} + \frac{D_z}{|J|} \begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix} \tag{6.5.1}$$

$$r^e = |J| \begin{bmatrix} 1/3 & 1/6 \\ 1/6 & 1/3 \end{bmatrix} \begin{bmatrix} (K_i/\Delta t)N^{\prime}_{i,1} + \lambda_{i-1}K_{i-1}N_{i-1,1} \\ (K_i/\Delta t)N^{\prime}_{i,2} + \lambda_{i-1}K_{i-1}N_{i-1,2} \end{bmatrix} \tag{6.5.2}$$

for element b)

$$[k^e] = \left(\frac{K_i}{\Delta t} + \lambda_i K_i\right) |J| \begin{bmatrix} 2/15 & -1/30 & 1/15 \\ -1/30 & 2/15 & 1/15 \\ 1/15 & 1/15 & 8/15 \end{bmatrix} + v \begin{bmatrix} 1/2 & -1/6 & 2/3 \\ 1/6 & -1/2 & -2/3 \\ -2/3 & 2/3 & 0 \end{bmatrix} + \frac{D_z}{|J|} \begin{bmatrix} 7/3 & 1/3 & -8/3 \\ 1/3 & 7/3 & -8/3 \\ -8/3 & -8/3 & 16/3 \end{bmatrix} \tag{6.5.3}$$

$$\{r^e\} = |J| \begin{bmatrix} 2/15 & -1/30 & 1/15 \\ -1/30 & 2/15 & 1/15 \\ 1/15 & 1/15 & 8/15 \end{bmatrix} \begin{bmatrix} (K_1/\Delta t)N^{\prime}_{i,1} + \lambda_{i-1}K_{i-1}N_{i-1,1} \\ (K_i/\Delta t)N^{\prime}_{i,2} + \lambda_{i-1}K_{i-1}N_{i-1,2} \\ (K_i/\Delta t)N^{\prime}_{i,3} + \lambda_{i-1}K_{i-1}N_{i-1,3} \end{bmatrix} \tag{6.5.4}$$

The model space and the subdivision of the space are the same as those described in Sec. 6.4.1.

6.5.2 Numerical Results

The concentration profiles of the three-member decay chain ^{234}U > ^{230}Th > ^{226}Ra at t=10,000 year for the current boundary condition are shown in Fig. 6.4.4. The assumed constant parameters are listed on the figure. The solid curves in the figure show the finite-element solution for the concentration boundary condition. As has already been discussed in Sec. 6.3.2 the concentration boundary condition covers only the convective transport of nuclides at the repository and is not physically consistent with the basic transport equation when the concentration of each nuclide at the repository is specified by the source term. Use of the concentration boundary condition can introduce a significant error in the prediction of the behaviour of nuclide migration near the repository, especially when the diffusional transport is relatively large. In fact, as seen from Fig. 6.4.4, these solutions for two different boundary conditions give appreciably different curves for the concentration profiles of the nuclides near the repository. The concentrations of ^{234}U and ^{230}Th for the current boundary condition are lower than those for the concentration boundary condition, whereas the concentration of ^{226}Ra for the current boundary condition is higher than that for the concentration boundary condition. This physical meaning is clear. Since ^{226}Ra has a relatively higher mobility compared with those of precursors nuclides, there arises a peak in concentration (reconcentration) at a relatively early time, less than the leach time. The peak concentration occurs near the leading edge of the ^{230}Th precursor. The positive gradient of ^{226}Ra concentration at the waste causes dispersive transport in the negative direction. Consequently, the concentration of ^{226}Ra for the current boundary condition can reach higher values than for the concentration boundary condition. The gradients of concentration of ^{234}U and of ^{230}Th, on the other hand, have negative values at the waste, and dispersive transport in the positive direction causes ^{234}U and ^{230}Th concentrations to decrease with distance.

The finite element solutions for the concentration profiles at t=50,000 year, about 17,000 year after the assumed leach time, for the two different boundary condition are also compared in Fig. 6.4.5. The solution for the current boundary condition, in contrast to that for the concentration

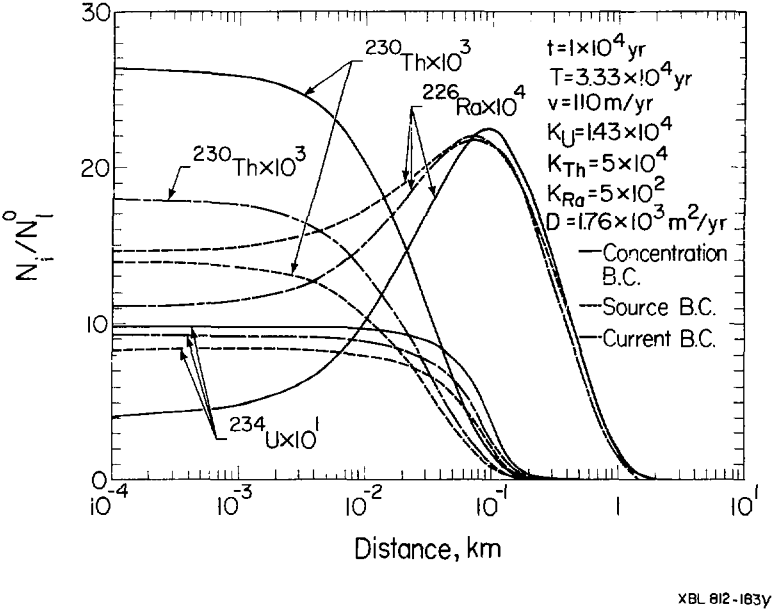

Fig. 6.4.4 Finite element solutions for three different boundary conditions, concentration profiles of the three-member decay chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ for band release, at t = 10^4 yr.

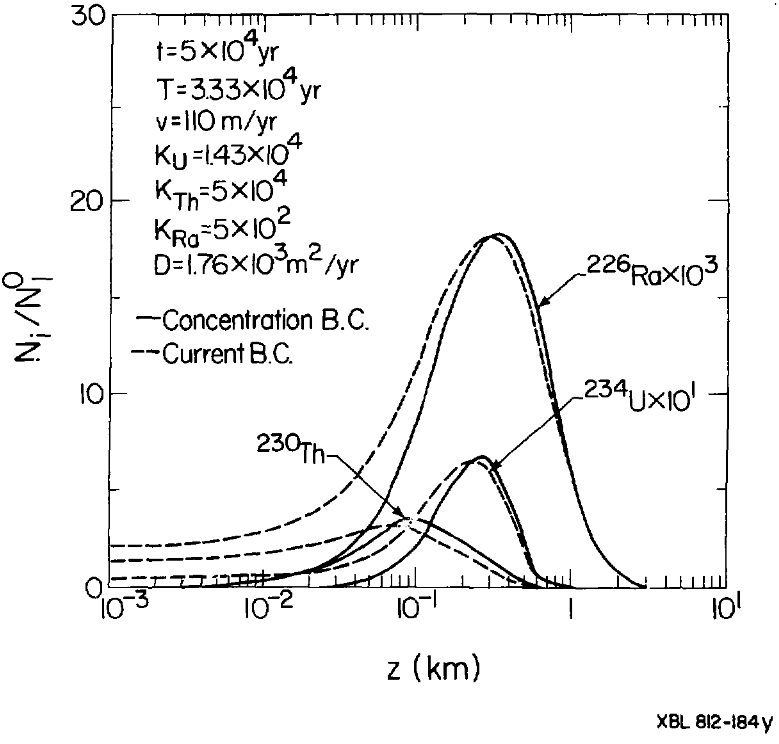

Fig. 6.4.5 Finite element solutions for two different boundary conditions; concentration profiles of the three-member decay chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ for a band release at $t = 5 \times 10^4$ yr.

three members do not disappear at the repository. At t greater than T all members have maxima in their concentration profiles, and the nuclides in the region between the waste and the peak undergo dispersive transport towards the waste. At distance greater than the locus of maximum concentration there is no appreciable difference between these two results. The concentration profiles for the concentration boundary condition and those for the current boundary condition are almost identical near the leading edges of the bands. We can conclude that the effects of dispersive transport near the waste source on concentration profiles of nuclide chain are important near the repository, but are less significant at greater distances.

6.5.3 Finite-Element Solution of the Transport Equation with a Dissolution Source Term

The transport equation which includes the source term can be expressed by

$$\frac{\partial}{\partial t}(K_i N_i) + \nabla\cdot(vN_i) - \nabla\cdot(D\nabla N_i) + \lambda_i K_i N_i - \lambda_{i-1} K_{i-1} N_{i-1} = \phi_i(z,t) \quad (6.5.5)$$

where $\phi_i(z,t)$ is the source term.

The matrix transport equation becomes

$$[k^e]\{N_i\} = \{r^e\} + \{s^e\} + \{s'^{(e)}\} \quad (6.5.6)$$

where the matrices $[k^e]$, $\{r^e\}$, and $\{s^e\}$ are given by Eqs. (6.3.34), (6.3.35), and (6.3.36). The $\{s'^{(e)}\}$ is given by

$$\{s'^{(e)}\} = \int_{Ve}\{H\}\,\phi_i(z,t)\,dVe \quad (6.5.7)$$

Since all members in the surface-integral column matrix are canceled out by use of Eq. (6.3.29), the global matrix equation reduces to

$$[k^g]\{N_i\} = \{r^g\} + \{s'^g\}$$

or

$$\sum_{j=1}^{n} k^g_{kj} N_{i,j} = r^g_k + S'^g_k\ ,\ k = 1,2,3\ldots \quad (6.5.8)$$

This equation has the same form as that given by Eq. (6.3.37), with the exception that the surface-integral column matrix is replaced by the source-term column matrix. If the source term is given by a plane source at $z=z_1$, the column matrix $\{s'^e\}$ becomes

$$\{s'^{\,e}\} = \int_{Ve} \{H\} \frac{M_i^0}{TS} \delta(z-z_\ell) dVe,$$

$$= \begin{cases} \frac{M_i^0}{TS} H & k=\ell \\ 0 & k\neq 1 \end{cases} \qquad (6.5.9)$$

where k=ℓ denotes the position of the plane source.

6.5.4 One Dimensional Transport with Plane Source

Consider a plane source located at $z=z_\ell$, the interface between l-th and (ℓ+1)-th elements. When the nuclides from the source are transported away by convective and dispersive transports, the following mass balance equation should hold

$$\int_S \left[\left(-D \frac{\partial N_i}{\partial z} + v\, N_i\right)_{z=z_\ell-0} - \left(-D \frac{\partial N_i}{\partial z} + v\, N_i\right)_{z=z_\ell+0}\right] dS$$

$$= \int_S \int_{z_\ell-0}^{z_\ell+0} \phi_i(t) \delta(z-z_\ell) dz\ dS \qquad (6.5.10)$$

Thus, in the global matrix equation, the plane source volume-integration term can be replaced by the current surface integration. Therefore, we can apply the matrix transport equation given by Eq. (6.3.37) to nuclide transport with a plane source by regarding the current surface-integration column matrix as the source-volume-integration column matrix. In this problem of one dimensional flow with infinite plane source in an infinite medium in x and y, a nuclide can migrate in the positive and negative z-directions, but there is no transverse dispersion.

If we apply the matrix equation to the concentrations of nuclides in infinite medium but not in semi-infinite medium, the matrix equation will lead us directly to the solution for the transport equation with a plane source.

In this section, instead of transforming the modeled space, we adopt the iteration procedure for a semi-infinite medium. We first compute the solution for the current boundary condition. Then we can know the dispersive current in the negative direction and calculate a new correct current in the positive direction, for the case of dispersive transport in both directions. After setting the new current as an input quantity and solving the matrix transport equation, we have another solution for the aqueous concentrations of nuclides. Successive iterations yield the convergent solution, which is identical to the solution of the transport equation with a source term.

In Fig. 6.5.1 the finite element solutions or the transport problem with a source term, which are computed by use of the linear element, are compared with the analytical solutions given by Eq. (6.4.18) ~ (6.4.20), with the function E(i,j;k) defined by

$$E(i,j;k) = e^{(z/2\alpha-\beta_{ij}t)} \frac{2}{\sqrt{\pi}} \int_0^{\sqrt{v_k t/4\alpha}} \exp\{-[\gamma y^2+(z/4\alpha y)^2]\}\, dy \quad (6.5.11)$$

Although the finite-element solutions give slightly lower values for the concentration of ^{234}*U and slightly higher values for the concentrations of* ^{230}Th and of ^{226}Ra, they give good approximations to the concentration profiles given by the exact analytical solutions.

In Fig. 6.4.4 the finite-element solutions for the problem with a source term are also shown with the dashed curves. The concentrations of ^{234}U and ^{230}Th take the lowest values, and that of ^{226}Ra takes the highest value near the waste source, when compared with the concentrations given by the solutions for the concentration and current boundary conditions.

At greater distance these three solutions show almost identical concentration profiles and the effects of dispersive transport as the repository becomes less important.

6.6 Application to Transport to Multi-Layered Media

There are many instances where the prediction of the migration behavior of radionuclides in geologic media composed of several layers is important. As has already been discussed in Section 5.5.4 in the previous work (H1), the sorption equilibrium constant is one of the most important parameters which have significant effects on the concentration profiles of nuclides. We here apply the matrix transport equation to the problem of migration of nuclide in multi-layered geologic media of which constituent layers have different sorption equilibrium constants.

6.6.1 One Dimensional Transport in Two-Layered Media

The concentration profiles of ^{234}U, ^{230}Th, and ^{226}Ra in geologic media composed of two layers which have different sorption constants are

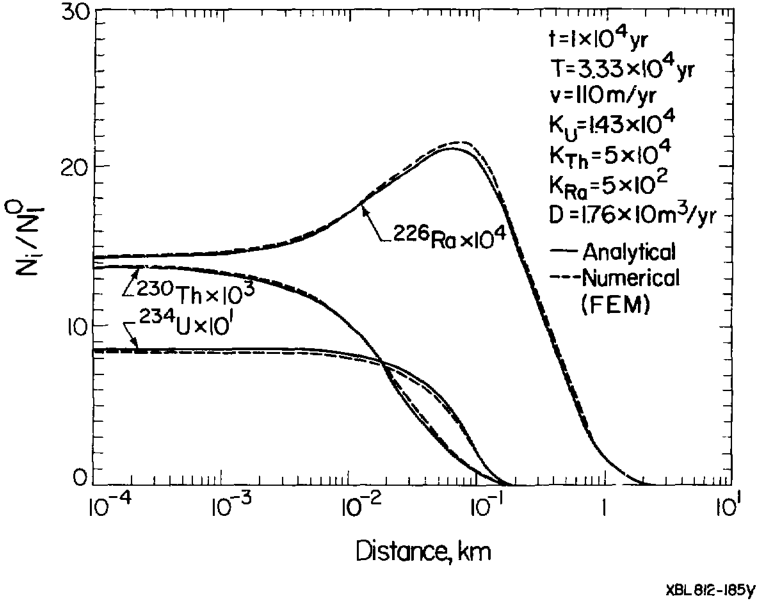

Fig. 6.5.1 Comparison of finite element solution with analytical solution, concentration profiles of the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain at $t = 1 \times 10^4$ year, source boundary conditions.

computed for the concentration boundary condition. The numerical results at t=10,000 year are shown in Fig. 6.6.1. In this situation, we selected the two layer media which are in contact with each other at z=98.6 m from the dissolving waste. The interface between the two layers is perpendicular to the direction of groundwater flow. The assumed parameters used in computations are included in the figure. The solid curves show the concentration profiles in a single infinite medium, with K_u=1.43 x 10^4, K_{Th}=5.00 x 10^4, and K_{Ra}=5.00 x 10^2. The dashed curves show the concentration profiles of nuclides in a two-layer medium, with reduced sorption constants in the outer medium. For the assumed time of 10,000 year, the leading edges of the ^{234}U and ^{230}Th bands locate in the outer medium. The presence of the second layer has no significant effect on the concentration profiles in the inner medium, but it causes a local maximum in the concentration of ^{230}Th in the outer layer. This local peak results because of the higher mobility of ^{230}Th in the second layer. For ^{226}Ra we find no appreciable change in concentration in the inner layer, but its lower sorption constant in the outer layer results in an increase in the maximum concentration and a shift in the locus of the maximum to a greater distance. The concentration profiles of ^{234}U, ^{230}Th, and ^{226}Ra at t=50,000 year are shown in Fig. 6.6.2. Here the nuclides have penetrated farther into the outer layer with lower assumed sorption constants, resulting in increased local maximum concentrations of each nuclide. The locus of each maximum is shifted to greater distance from the waste source.

6.7 Application to Multi-Dimensional Problem

In this section, we discuss briefly the application of the finite-element method to the multi-dimensional transport of radionuclides through geological media.

6.7.1 Two Dimensional Dispersion with Constant Parameters

As an illustration we present the finite-element solutions for the concentration profiles of the three member decay chain ^{234}U > ^{230}Th > ^{226}Ra in two dimensional geologic media. We adopt the concentration boundary condition. We select the two dimensional space in y-z plane, the rectangular area of $0 \leq y \leq 160$ m, $-40\text{m} \leq z \leq 240$ m for the domains of ^{234}U and ^{230}Th and the other rectangular area of $0 \leq y \leq 320$ m, $-160 \text{ m} \leq z \leq 960$ m for the domain of ^{226}Ra and split these domains into 52 triangular elements of different

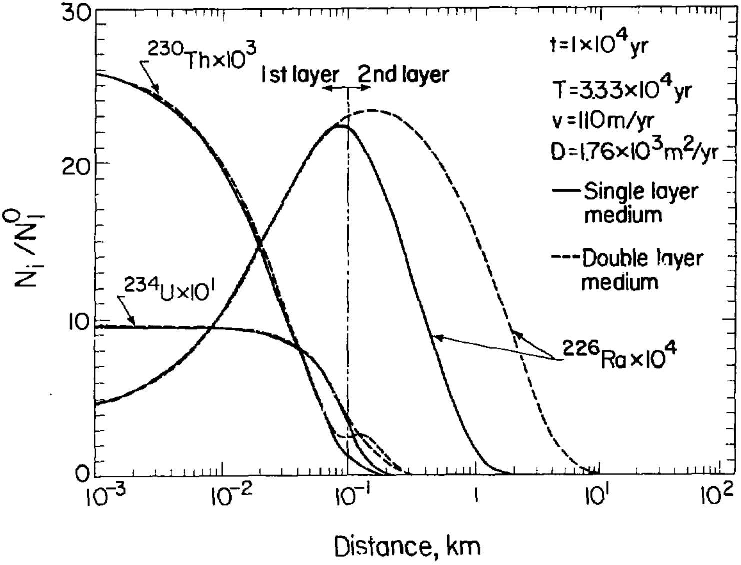

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Fig. 6.6.1 Concentration profiles of the ^{234}U → ^{230}Th → ^{226}Ra decay chain migrating through two layer geologic medium at t = 1 x 10^4 year, concentration boundary condition.

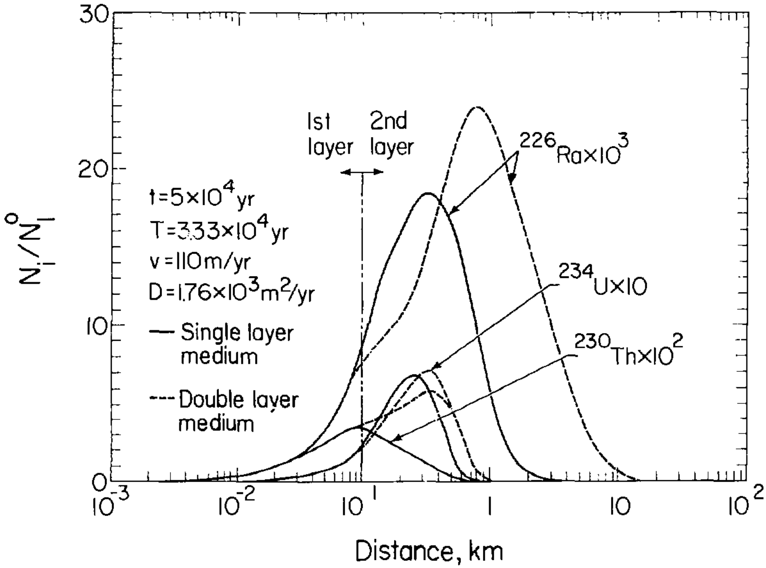

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Fig. 6.6.2 Concentration profiles of the $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ decay chain migrating through a two-layer geologic medium at $t = 5 \times 10^4$ year, concentration boundary condition.

sizes. The discretization of the model space is shown in Fig. 6.7.1. The concentration field is approximated as

$$N_i^{(e)} = \lfloor H \rfloor \{N_i\} = \sum_{j=1}^{6} H_j N_{ij} \tag{6.7.1}$$

where N_{ij} is the value of concentration at j-th node of the element and H is the shape function. The shape function used here is

$$\lfloor H \rfloor = \left[(2L_1 - 1)L_1 \ (2L_2 - 1)L_2 \ (2L_3 - 1)L_3 \ 4L_1L_2 \ 4L_2L_3 \ 4_3L_1 \right] \tag{6.7.2}$$

where L_i's are natural coordinates (or surface coordinates) for the triangle element, among which the following relation holds

$$L_1 + L_2 + L_3 = 1 \tag{6.7.3}$$

The coordinates y and z in the model space are approximated by the shape functions as

$$q = \lfloor H \rfloor \{q\} = \sum_{q=1}^{6} H_j g_j \ , \ q = y,z \tag{6.7.4}$$

From Eq. (6.7.3) we can put

$$L_1 = \zeta, \quad L_2 = \eta, \quad L_3 = 1 - \zeta - \eta \tag{6.7.5}$$

Then we have

$$\begin{bmatrix} \dfrac{\partial \lfloor H \rfloor}{\partial \zeta} \\ \\ \dfrac{\partial \lfloor H \rfloor}{\partial \eta} \end{bmatrix} = \begin{bmatrix} 4L_1 - 1 & 0 & 1 - 4L_3 & 4L_2 & -4L_2 & 4L_3 - 4L_1 \\ \\ 0 & 4L_2 - 1 & 1 - 4L_3 & 4L_1 & 4L_3 - 4L_3 & -4L_1 \end{bmatrix} \tag{6.7.6}$$

but

$$\begin{bmatrix} \dfrac{\partial \lfloor H \rfloor}{\partial \zeta} \\ \\ \dfrac{\partial \lfloor H \rfloor}{\partial \eta} \end{bmatrix} = J \begin{bmatrix} \dfrac{\partial \lfloor H \rfloor}{\partial z} \\ \\ \dfrac{\partial \lfloor H \rfloor}{\partial y} \end{bmatrix} \tag{6.7.7}$$

where

$$J = \begin{bmatrix} J_{11} & J_{12} \\ \\ J_{21} & J_{22} \end{bmatrix} = \begin{bmatrix} \dfrac{\partial z}{\partial \zeta} & \dfrac{\partial y}{\partial \zeta} \\ \\ \dfrac{\partial z}{\partial \eta} & \dfrac{\partial y}{\partial \eta} \end{bmatrix} \tag{6.7.8}$$

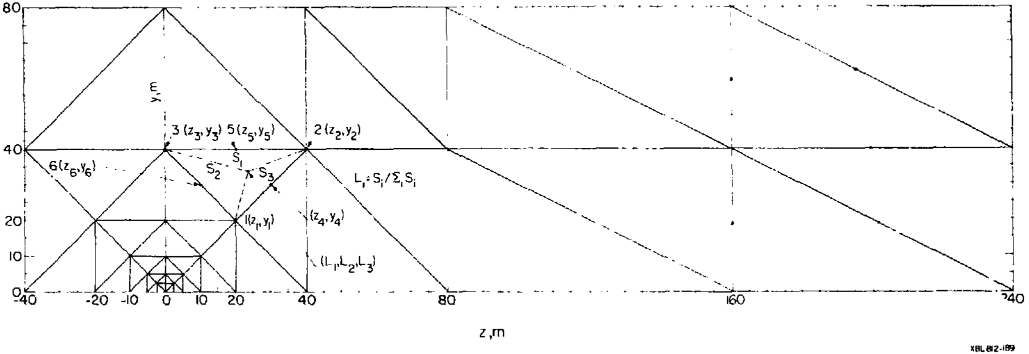

Fig. 6.7.1 Discretization of two dimensional model space.

From Eq. (6.7.7) the derivatives of shape functions can be written as

$$\begin{bmatrix} \dfrac{\partial [H]}{\partial z} \\ \dfrac{\partial [H]}{\partial y} \end{bmatrix} = J^{-1} \begin{bmatrix} \dfrac{\partial [H]}{\partial \zeta} \\ \dfrac{\partial [H]}{\partial \eta} \end{bmatrix} \tag{6.7.9}$$

The Jacobian J is given by

$$J = \begin{bmatrix} 4L_1-1 & 0 & 1-4L_3 & 4L_2 & -4L_2 & 4L_3-4L_1 \\ 0 & 4L_2-1 & 1-4L_3 & 4L_1 & 4L_3-4L_2 & -4L_1 \end{bmatrix} \begin{bmatrix} z_1 & y_z \\ z_2 & y_2 \\ z_3 & y_3 \\ z_4 & y_z \\ z_5 & y_5 \\ z_6 & z_6 \end{bmatrix} \tag{6.7.10}$$

where (z_i, y_i) is the position of i^{th} node of the triangular element which is measured on the z-y scale. If we wish to compute the solution for the space which has curved linear boundaries, we can select the points arbitrarily to fit the boundaries.

Although the matrix transport equation given by Eq. (6.3.31) with Eq. (6.7.2) and (6.7.9) can be directly applied to solve the two dimensional dispersion with two dimensional water flow, we here present the results for the simple case of two dimensional dispersion with one dimensional water flow in z-direction. In computations, the time dependent concentration given by Eq. (6.2.13) was applied at z=0 and y=0 for the boundary condtion.

The concentration profiles of the ^{234}U, ^{230}Th, and ^{226}Ra decay chain in the z-direction at y=0 at time t=10,000 year are shown in Fig. 6.7.2. As easily seen from a comparison of these curves with those for the one dimensional which is given in Fig. 6.4.2, the concentration of each member takes much smaller values than those given by the one dimensional solution. The maximum concentration of ^{226}Ra is one fourth less than that given by the one dimensional solution. This lower maximum concentration is explained by the effects of transverse dispersion which smoothes the concentration peak. The figure suggests that the effect of transverse dispersion on the concentration profiles is very important for the exact prediction of radionuclide migration behavior.

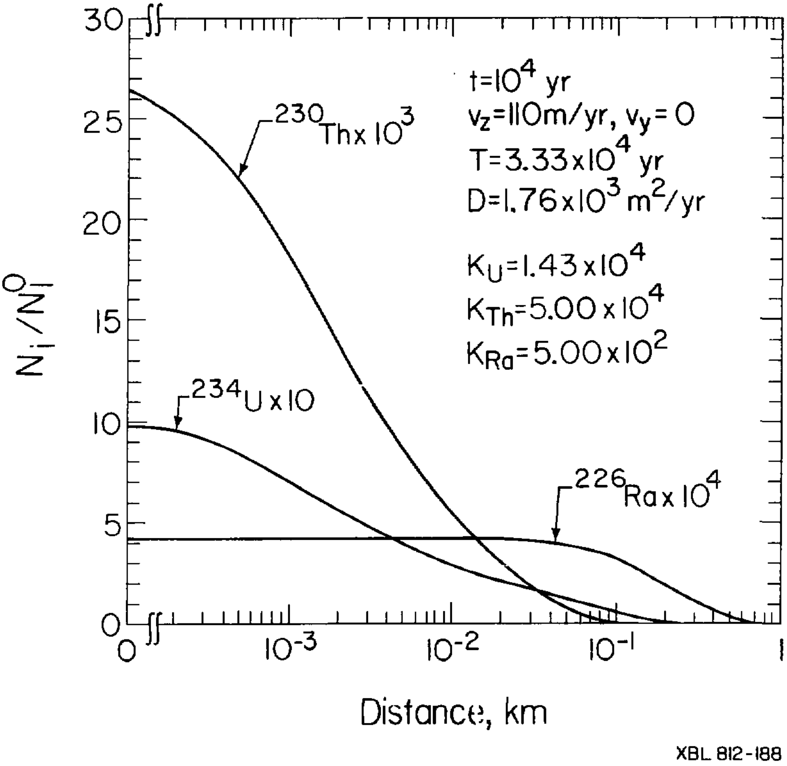

Fig. 6.7.2 Concentration profiles of the three-member decay chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra$ in a two-dimensional medium in longitudinal direction.

6.8 Proposed Future Study

The Darcy flow matrix equation and the general matrix transport equation have been derived here. Although these flow and mass-transport equations can be applied to the general problem of the transport of radionuclides in sorbing media, only the solutions for some limiting cases (one- and two-dimensional problems with constant parameters) have been demonstrated. The solutions of the Darcy flow problem have not been presented. There remain some important problems to be solved for future study.

They are

1. Solutions of one-dimensional transport with space-dependent physicochemical parameters.
2. Solutions of multidimensional transport with space-dependent physicochemical parameters and of multidimensional transport with geometrically complicated boundaries for the three different boundary conditions.
3. Solutions of coupled Darcy flow and mass-transport equations in multidimensional geologic media, including three-dimensional transport with Darcy flow.
4. Introduction of the concept of secular equilibrium between precursor and daughter nuclides into the matrix transport equation and into the solutions for concentration profiles of multi-member nuclide chains.
5. Derivation and solutions of the general matrix equation for the transport of radionuclides without local chemical equilibria.
6. Formulation of the matrix equation for transport with space- time-dependent physicochemical parameters.
7. Formulation of the energy matrix transport equation which describes thermal effects on mass transport.

6.9 Conclusion

In this chapter we have presented some applications of the finite element method to the prediction of behaviour of radionuclides which migrate through geologic media. Based on the discussion above we can conclude:

1. From comparisons of the one-dimensional finite element solutions with the analytical solutions the finite element method gives sufficiently good approximations to the exact solutions. The finite-element solution is, therefore, a useful numerical technique to evaluate radionuclide migration.

2. The finite-element matrix equations presented here can be applied to problems with concentration and current boundary conditions and also to solving the transport equation with a source term. The finite-element solutions for the problem with a current boundary condition of third kind, and also with a source boundary condition, agree well with the physical phenomena of near-field radionuclide migration.

3. The finite-element matrix equations can be applied directly to the migration of the radionuclides through multi-layer geologic media. The effects of the second layer in two-layer media are discussed in Sec. 6.6.1. Even when the physicochemical parameters and the water velocity are space dependent, the matrix transport equations can be applied simply by changing the input data for these parameters.

4. The finite-element method can be applied to the multidimensional radionuclide transport problem by finding a set of suitable discretizations. Although demonstrations of the solution for the problem with curved linear boundaries have not been given here, the finite-element matrix equations can be easily extended to include the migration problem with such complicated boundary geometries.

5. The matrix transport equations are not restricted in terms of the number of members in a nuclide chain. They can easily be applied to calculate the concentration profiles of multimember nuclide chains.

6. The finite-element analysis presented here is a more effective approach to the solution of the basic transport equation than is the direct numerical solution of the partial differential equation.

6.10 Nomenclature

b_{ji} : Bateman coefficient

$B_i(t)$: Bateman equation

D : Dispersion coefficient

H : Shape function

$k(k_x, k_y, k_z)$: Darcy's constant

$[k]$: Matrix for mass transport

$[k_V]$: Matrix for flow

K_{Di} : Distribution coefficient

K_i : Retardation coefficient

L : Order of Gauss quadrature approximation

L_i : Natural coordinate

m : Number of nodes in element

M_i^0 : Initial amount of nuclide i

n : Total number of nodes

n : Unit vector normal to surface

N_i : Concentration of radionuclides in aquifer

N_i^0 : Initial concentration of nuclide i

P : Pressure

q : Source of water

Q : Volume flow rate of water per initial amount of wastes

r : Column matrix for nonsteady and decay generation terms

s : Column matrix for surface integration of flux

s' : Column matrix for source term

s_p : Column matrix for surface integration of pressure

s_Ω : Column matrix for surface integration of gravitational potential

s_w : Column matrix for source of water

S_R : Area of surface of repository

Se : Area of surface of element

S_I : Area of surface of infinite medium

t : Time

T : Leach time

$v(v_x,v_y,v_z)$: Velocity of water

Ve : Volume of element

x,y,z : Cartesian coordinate

α : Longitudinal dispersivity factor $\equiv D/v_z$

ε : Porosity of sorbing medium

λ_i : Decay constant of nuclide i

μ : Viscosity of water

ρ_w : Density of water

$\phi_i\ (t)$: Plane source term in the water phase

$\phi_i(z,t)$: General source term in the water phase

Ω : Gravitational potential

Subscripts

e : Element

g : Global

i : Nuclide

q : x,y,z

6.11 References

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Appendix A. The Semiequilibrium Transport Equation with Irreversible Mineralization

With Eq. (4.2) in our previous report (H1) we presented a one dimensional differential equation, the transport equation, which includes the assumption that the atoms dissolved in water, N_i, and the ones adsorbed on the solid, M_i, are at anytime in local chemical equilibrium:

$$N_i(z,t) = K_{D,i} M_i(z,t) \qquad \text{(A.1)}$$

$$K_{D,i} = \text{sorption equilibrium constant} = \frac{\text{concentration of nuclide i adsorbed on solid}}{\text{concentration of nuclide i in water}}$$

Here we show how it is possible to include into the transport equation the effect of a possible irreversible mineralization of the sorbed species into a different chemical species, which contributes no longer to the transport process.

We assume that an isotope i can be found only in three different states:

(1) N_i, dissolved in water;

(2) M_i, sorbed on the solid, and in equilibrium with N_i according to Eq. (A.1);

(3) M_i, mineralized on the solid.

The sorbed nuclide undergoes an irreversible chemical reaction at a rate R:

$$\dot{R}_i = \vec{k}_i M_i \ , \ \text{atoms/(yr)}\ m^3_{\text{solid}} \qquad \text{(A.2)}$$

where $\vec{k}_i$ = first-order reaction rate constant for irreversible mineralization of species i, sorbed in the solid, 1/yr. We also assume that a mineralized species m_i decays to a daughter m_{i+1} which is also mineralized.

If we note by $\dot{r}_i$ the sorption rate, i.e., the net rate, per unit volume of solid, of which the nuclide transfers from the liquid to the sorbed phase, the previous assumptions can be characterized by:

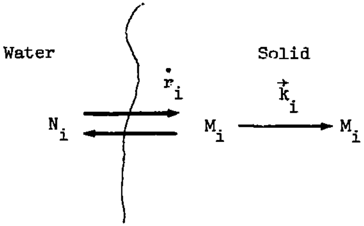


Figure A.1: Irreversible mineralization reaction schematic.

A mass balance on nuclide i becomes (H1):

$$\frac{\partial N_i}{\partial t} = D\frac{\partial^2 N_i}{\partial z^2} - v\frac{\partial N_i}{\partial z} - \lambda_i N_i + \lambda_{i-1}N_{i-1} - \left(\frac{1-\varepsilon}{\varepsilon}\right)\dot{r}_i + \phi_i'''(z,t) \tag{A.3}$$

$$\frac{\partial M_i}{\partial t} = -\lambda_i M_i + \lambda_{i-1}M_{i-1} + \dot{r}_i - \vec{k}_i M_i \tag{A.4}$$

$$\frac{\partial m_i}{\partial t} = -\lambda_i m_i + \lambda_{i-1}m_{i-1} + \vec{k}_i M_i \tag{A.5}$$

We multiply now Eq. (A.4) by $\left(\frac{1-\varepsilon}{\varepsilon}\right)$, add it to Eq. (A.3), use K_i defined by Eq. (2.1.3) and rearrange, and we obtain the one-dimensional semi-equilibrium transport equation for $N_i(z,t)$:

$$\hat{L}_i N_i(z,t) = \lambda_{i-1}K_{i-1}N_{i-1}(z,t) + \phi_i'''(z,t) \tag{A.6}$$

where

$$\hat{L}_i \equiv -D \frac{\partial^2}{\partial z^2} + v \frac{\partial}{\partial z} + K_i \frac{\partial}{\partial t} + \eta_i K_i \qquad (A.7)$$

η_i being given by:

$$\eta_i = \lambda_i + \left(\frac{1 - \varepsilon}{\varepsilon}\right) \frac{\vec{k}_i K_{D,i}}{K_i} \quad . \qquad (A.8)$$

Comparing Eq. (A.6) and Eq. (A.7) with Eq. (4.2) in Ref. (H1), it is apparent that we have indeed the same differential equation, the only difference being the constant λ_i in the previous transport equation (H1) is now replaced by η_i.

Appendix B. Non Recursive Solution of the One-Dimensional Equilibrium Transport Equation

A) With dispersion

We must solve Eq. (2.1.1), with the conditions of Eqs. (2.1.7) and (2.1.8). Let $\tilde{N}_i(z,p)$ be the Laplace Transform of $N_i(z,t)$:

$$\tilde{N}_i(z,p) = L[N_i(z,t)] = \int_0^\infty e^{-pt} N_i(z,t)\, dt \tag{B.1}$$

Define also:

$$\tilde{y}_i(z,p) = \tilde{N}_i(z,p)\, e^{-vz/2D} \tag{B.2}$$

If we use Eq. (B.1) and the initial condition Eq. (2.1.8) in Eq. (2.1.1), and then Eq. (B.2) in the resulting expression, we get:

$$\frac{d^2\tilde{y}_i(z,p)}{dz^2} - g_i\tilde{y}_i = -\left(\tilde{\beta}_{i-1}\tilde{y}_{i-1} + \frac{1}{D}\,\tilde{\phi}_i'''(z,p)\, e^{-vs/2D}\right) \tag{B.3}$$

where:

$$\tilde{\phi}_i'''(z,p) = L[\phi_i'''(z,t)] \tag{B.4}$$

$$g_i = \frac{1}{D}\left[\frac{v^2}{4D} + \gamma_i\right] \tag{B.5}$$

$$\tilde{\beta}_i = \frac{K_i\lambda_i}{D} \tag{B.6}$$

$$\gamma_i = K_i p + K_i \eta_i \tag{B.7}$$

Due to the condition of Eq. (2.1.7), we can use the Fourier Transform F for $\tilde{y}_i$, and for $\tilde{\phi}_i'''$ too, because we can expect, for physical reasons, the general source term ϕ_i''' to behave as Eq. (2.1.7). Accordingly we define:

$$\hat{y}_i(s,p) = F[\tilde{y}_i(z,p)] = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-isz}\, \tilde{y}_i(z,p)\, dz \qquad \text{(B.8)}$$

where $i \equiv \sqrt{-1}$. Equations (B.3) and (B.8) give:

$$s^2\hat{y}_i + g_i\hat{y}_i = \hat{F}_i(s,p) + \tilde{\beta}_{i-1}\hat{y}_{i-1} \qquad \text{(B.9)}$$

where:

$$\hat{F}_i(s,p) = \frac{1}{D} F[\tilde{\phi}_i'''(z,p)\, e^{-vz/2D}] \qquad \text{(B.10)}$$

From Eq. (B.10) we get the recursive formula:

$$\hat{y}_i = \frac{1}{S^2 + g_i} (\hat{F}_i + \tilde{\beta}_{i-1}\hat{y}_{i-1}) \quad . \qquad \text{(B.11)}$$

Then:

$$\hat{y}_0 = 0 \qquad \text{(B.12)}$$

$$\hat{y}_1 = \frac{\hat{F}_1}{s^2 + g_1} \qquad \text{(B.13)}$$

$$\hat{y}_2 = \frac{\hat{F}_2}{s^2 + g_2} + \tilde{\beta}_1 \frac{\hat{F}_1}{(s^2 + g_1)(s^2 + g_2)} \qquad \text{(B.14)}$$

$$\hat{y}_3 = \frac{\hat{F}_3}{s^2 + g_3} + \tilde{\beta}_2 \frac{\hat{F}_2}{(s^2 + g_2)(s^2 + g_3)} + \tilde{\beta}_1\tilde{\beta}_2 \frac{\hat{F}_1}{(s^2 + g_1)(s^2 + g_2)(s^2 + g_3)} \qquad \text{(B.15)}$$

and so on, which suggests:

$$\hat{y}_i(s,p) = \sum_{j=1}^{i} A_i^j \ \alpha \left[\prod_{l=j}^{i} \frac{1}{\alpha}\right] \frac{\hat{F}_j(s,p)}{\prod\limits_{m=j}^{i} (s^2 + g_m)} \tag{B.16}$$

where

$$A_i^j = \frac{v_i}{\lambda_i}\left[\prod_{l=j}^{i} \frac{\lambda_l}{v_l}\right] \tag{B.17}$$

$$\alpha = D/V \tag{B.18}$$

Equation (B.16) is a non recursive formula. We can take its inverse Fourier and Laplace transforms by the use of tables (e.g., Ref. (C1)) and the use of the convolution properties of these transforms (see Ref. (H3), pages 67 and 263). After use of Eq. (B.2), the solution given in Section 2.1 follows. Notice: In order to find the inverse transforms in the tables, we must rewrite such fractions as:

$$\left[\prod_{m=j}^{i} (s^2 + g_m)\right]^{-1}$$

as:

$$\sum_{m=j}^{i} \frac{c_m^-}{\sqrt{g_m} - is} + \frac{c_m^+}{\sqrt{g_m} + is}$$

where C_m^- and C_m^+ are constants which can be found by standard mathematical methods. These operations impose the following restrictions on the solution:

$$\beta_{r,m} \neq \beta_{l,k} \quad \text{if} \quad r,m \neq l,k \tag{B.19}$$

where $\beta_{r,m}$ is defined by Eq. (2.1.26), and

$$v_r \neq v_m \quad \text{if} \quad r \neq m \tag{B.20}$$

The general time-dependent plane source $\phi_i''(t)$ of nuclide i at $z = 0$, for the step release, is given by Eq. (2.1.17):

$$\phi_i''(t) = \frac{W_s^0}{T} B_i(t)\ h(t) \tag{B.21}$$

where $B_i(t)$ is defined by Eq. (2.1.12).

The step-release functions $F_{i,i}^i(z,t)$ and $F_{r,m}^j(z,t)$, given by Eq. (2.1.35) and (2.1.36) respectively, are obtained by substituting Eq. (B.21) into Eq. (2.1.33) and (2.1.34): the following formulas, derived from Eq. (5.33) in Ref. (H1) by a change of variable and integration by parts, are used to perform the integrations:

$$e^{-\lambda_q t} \int_0^t e^{\theta(\lambda_q - \eta_i)}\ v_i\ \frac{e^{-(z-v_i\theta)^2/4v_i\alpha\theta}}{\sqrt{4\pi\theta\alpha v_i}}\ d\theta = E_{q,q,i}(z,t) \tag{B.22}$$

and

$$e^{-\lambda_q t} \int_0^t e^{\lambda_q \theta} E_{r,m,m}(z,\theta)\ d\theta \tag{B.23}$$

$$= \frac{1}{(\lambda_q - \beta_{rm})} [E_{r,m,m}(z,t) - E_{q,q,m}(z,t)] \quad , \quad \text{for } r \neq m$$

where $E_{q,q,i}$ and $E_{r,m,m}$ are given by Eq. (2.1.32).
Notice: The preceding mathematical operations introduce the factor v at the denominator in Eqs. (2.1.35) and (2 1.36).

B) Without dispersion

We set $D = 0$ in Eq. (2.1.2). The procedure is then the same as the one for the case with dispersion. Equation (B.2) is no longer needed.

Appendix C. The Superposition Equation for the Time-Dependent Source of Section 2.3

When the release rate at the repository depends on t or t^2, as in Section 2.3, the superposition presented by Eq. (2.1.18) is no longer valid. Such a release can be expressed as:

$$\phi_i''(t) = \frac{\dot{R}\rho}{S\epsilon} B_i(t)[c_0 + c_1 t + c_2 t^2][h(t) - h(t - T)] \qquad \text{(C.1)}$$

The correct superposition is given by Eq. (C.11), which is derived here. For simplicity sake, we consider a single-member chain, and a release rate such as:

$$\phi_1''(t) = \frac{\dot{R}\rho}{S\epsilon}\,{}^1B_{1,1} t e^{-\lambda t} \qquad \text{(C.2)}$$

where:

$${}^1B_{1,1} \equiv n_1^0 c_1 \qquad \text{(C.3)}$$

We seek a function $N_i^b(z,t)$ which satisfies:

$$\hat{L}_1 N_i^b(z,t) = \frac{\dot{R}\rho}{S\epsilon}\,{}^1B_{1,1} t e^{-\lambda t}[h(t) - h(t - T)]\;\delta(z) \qquad \text{(C.4)}$$

with conditions of Eqs. (2.1.7) and (2.1.8). $N_i^b(z,t)$ is given by:

$$\begin{aligned} N_1^b &= {}^1n_1^S(z,t;{}^1B_{1,1}) \\ &\quad - h(t - T)[{}^1n_1^S(z,t - T\,;\,{}^1B_{1,1}') + T\,{}^0n_1^S(z,t - T;\,{}^1B_{1,1}')] \end{aligned} \qquad \text{(C.5)}$$

where:

$$\hat{L}_1\,{}^1n_1^S(z,t\,;\,{}^1B_{1,1}) = \frac{\dot{R}\rho}{S\epsilon}\,{}^1B_{1,1} t e^{-\lambda t} h(t)\;\delta(z) \qquad \text{(C.6)}$$

$$\hat{L}_1\,{}^0n_1^S(z,t\;;\;{}^1B_{1,1}) = \frac{\dot{R}\rho}{s\epsilon}\,{}^1B_{1,1}e^{-\lambda t}h(t)\;\delta(z) \tag{C.7}$$

${}^1n_1^S$ and ${}^0n_1^S$ satisfy Eqs. (2.1.7) and (2.1.8); and:

$${}^1B'_{1,1} = {}^1B_{1,1}e^{-\lambda T} \tag{C.8}$$

Proof: When T < 0 Eq. (C.5), clearly satisfies Eq. (C.4). When t > T, by substituting Eqs. (C.5) to Eq. (C.8), into Eq. (C.4) we get:

$$\hat{L}_1N_1^b = \frac{\dot{R}\rho}{S\epsilon}\,{}^1B_{1,1}te^{-\lambda t}\delta(z) \tag{C.9}$$

$$- \frac{\dot{R}\rho}{s\epsilon}\left[{}^1B_{1,1}e^{-\lambda T}(t - T)\;e^{-\lambda(t-T)} + Tv\,{}^1B_{1,1}e^{-\lambda T}e^{-\lambda(t-T)}\right]\delta(z) = 0$$

(q.e.d.)

Furthermore, Eq. (C.5) satisfies Eqs. (2.1.7) and (2.1.8) because ${}^1n_1^s$ and ${}^0n_1^s$ satisfy such equations.

Notice: Although Eqs. (C.4) and (C.6) do not represent a band or a step release, because the release rate is time dependent, we still used the superscripts b (band) and s(step) as a convenient way to characterize the step functions in Eqs. (C.4) and C.6).

By the same way we can show that the solution to:

$$\hat{L}_iN_i^b(z,t) = \lambda_{i-1}K_{i-1}N_{i-1}^b(z,t) + \frac{\dot{R}\rho}{s\epsilon}\,B_i(t)$$

$$\cdot[c_0 + c_1t + c_2t^2][h(t) - h(t - T)]\delta(z) \quad -\infty < z < \infty\;,\; t > 0 \tag{C.10}$$

with the conditions Eqs. (2.1.7) and (2.1.8), is given by the following superposition:

$$N_{i,b}(z,t) = {}^0n_i^b(z,t) + {}^1n_i^b(z,t) + {}^2n_i^b(z,t) \tag{C.11}$$

where

$$ {}^0n_i^b(z,t) \equiv {}^0n_i^s(z,t\ ;\ {}^0B_{i,q}) - h(t - T)\ {}^0n_i^s(z,t - T\ ;\ {}^0B'_{i,q}) \qquad (C.12) $$

$$ {}^1n_i^b(z,t) \equiv {}^1n_i^s(z,t\ ;\ {}^1B_{i,q}) - h(t - T) $$
$$ \cdot\left[{}^1n^s(z,t - T\ ;\ {}^1B'_{i,q}) + T\ {}^0n_1^s(z,t - T\ ;\ {}^1B'_{i,q})\right] \qquad (C.13) $$

$$ {}^2n_i^b(z,t) \equiv {}^2n_i^s(z,t\ ;\ {}^2B_{i,q}) - h(t - T) $$
$$ \cdot\Big[{}^2n_i^s(z,t - T\ ;\ {}^2B'_{i,q}) + 2T\ {}^1n_i^s(z,t - T\ ;\ {}^2B'_{i,q}) $$
$$ + T^2\ {}^0n_i^s(z,t - T\ ;\ {}^2B'_{i,q})\Big] \qquad (C.14) $$

$$ {}^xB_{i,q} \equiv \sum_{m=1}^{q} n_m^o X_{i,q}^m C_x \qquad x = 0,1,2 \qquad (C.15) $$

$$ {}^xB'_{i,q} \equiv {}^xB_{i,q} e^{-\lambda_q T} \qquad x = 0,1,2 \qquad (C.16) $$

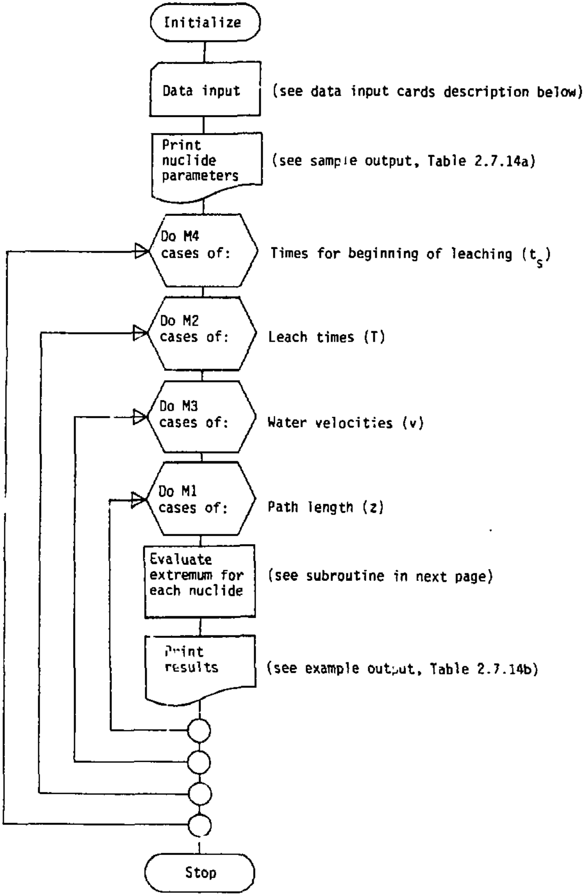
$X_{i,q}^m$ is defined by Eq. (2.1.14) and ${}^xn_i^s(z,t\ ;\ {}^yB_{i,q})$ is a solution of:

$$ L_i\ {}^xn_i^s(z,t\ ;\ {}^yB_{i,q}) = \lambda_{i-1}K_{i-1}\ {}^xn_{i-1}^s(z,t\ ;\ {}^yB_{i,q}) $$
$$ + \frac{R\rho}{S\varepsilon}\ t^x \sum_{q=1}^{i} {}^yB_{i,q} e^{-\lambda_q t}\ h(t) \qquad (C.17) $$

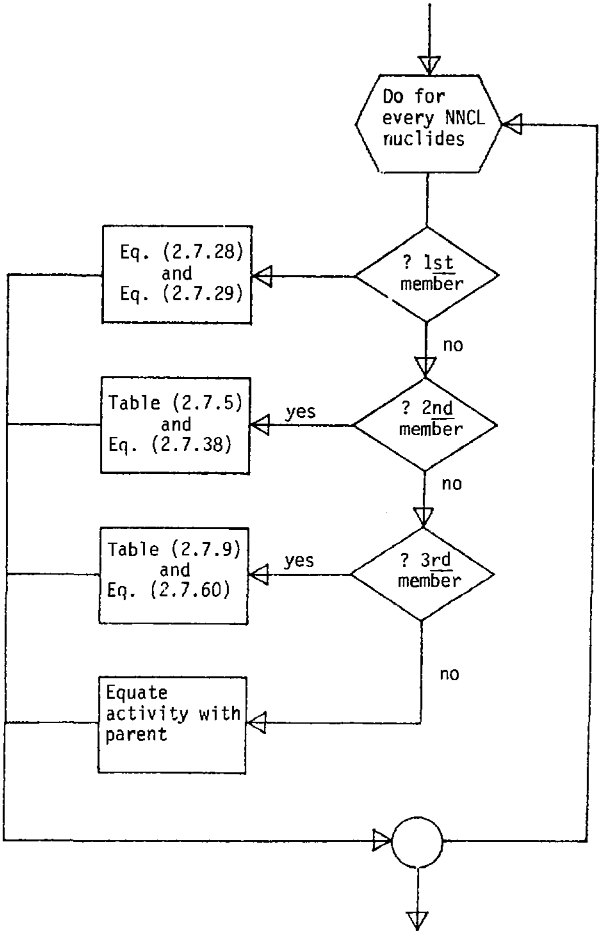
with conditions Eqs. (2.1.4) and (2.1.8).

Appendix D: Description of the Computer Program UCBNE25

D-1) The following flowchart describes the structure of the program UCBNE25:



Subroutine to evaluate the extremum for each nuclide



D-2) Description of the data cards for UCBNE25

The following cards are read by UCBNE25:

1) 1st card: Initial system description:

VARIABLE	FORMAT	DESCRIPTION
NNUCL	I2	total number of nuclides being considered
z0	E12.4	first value of path length to be used (meters); ($z0 \neq 0$)
T0	E12.4	first value of leach time to be used (yrs); ($T0 \neq 0$)
v0	E12.4	first value of water velocity to be used (m/yr); ($v0 \neq 0$)
TBL0	E12.4	first value of time for beginning of leach (yr); ($TBL0 \neq 0$)

2) 2nd card: System parameters variation description

VARIABLE	FORMAT	DESCRIPTION
M1	I2	total number of different path length cases to be evaluated M1 = 1,2 ...
DEL1	E10.4	multiplying factor for different path length cases: $z = z0 \times (DEL1)^n$; n = 1,2 ... M1
M2	I2	total number of different leach time cases to be evaluated M2 = 1,2 ...
DEL2	E10.4	multiplying factor for different leach time cases: $T = T0 \times (DEL2)^n$; n = 1,2 ... M2
M3	I2	total number of different water velocity cases to be evaluated, M3 = 1,2 ...
DEL3	E10.4	multiplying factor for different water velocity cases $vw = v0 \times (DEL3)^n$; n = 1,2 ... M3
M4	I2	total number of different time for beginning of leaching' cases to be evaluated, M4 = 1,2,3 ...
DEL4	E10.4	multiplying factor for different time for beginning of leaching cases $TBL = TBL0 \times (DEL4)^n$; n = 1,2 ... M4

3) 3rd through (NNUCL + 2)th card: Nuclides description

It is required one card for each nuclide being considered, for a total of NNUCL cards. Each card has the following inputs:

VARIABLE	FORMAT	DESCRIPTION
NN	I2	Nuclide identification number
NUCL	A6	Nuclide name (e.g., TH-229)
NH	I2	Nuclide hierarchy: NH = 1 : if it is a first member of a chain NH = 2 : if it is a second member of a chain NH = 3 : if it is a third member of a chain NH = 4 : if it is in secular equilibrium with the nearest long lived precursor
NP1	I2	NP1 = identification number of the 1st precursor if NH = 2 or 3. NP1 = identification number of the nearest long lived precursor if NH = 4. NP1 = 0 if NH = 1
NP2	I2	If NH = 3, NP2 is the identification number of the 2nd precursor. NP2 = 0 otherwise
HFLV	E12.4	Nuclide half-life (yr)
KD	E12.4	Nuclide sorption coefficient, $KD = 1 + \frac{(1 - \varepsilon)}{\varepsilon} \rho_s k_D$
TOX	E12.4	Nuclide maximum permissible concentration (MPC); (Ci/m^3)
C	E12.4	Initial activity at the time of emplacement, i.e., $t_s = 0$, (Ci)

Appendix E. Radioactive Transmutation of a Mineralized Species.

The equations of Section 2.1 are applicable to completely irreversible mineralization ($\tilde{b} = \gamma = 0$). However, it is reasonable to expect that radioactive transmutation of a mineralized radionuclide may result in a radioactive daughter which, because it is a different chemical element, reverts back to the non-mineralized sorbed species. The equations of Sections 3.3.1-2 are applicable to this case by setting $\tilde{k} = 0$ and retaining γ as a finite quantity. The explicit solutions for a three-member decay chain, assuming a step release with the source term given by Eq. (2.1.17), are as follows:

$$N_1^s(z,t) = \frac{e^{zv_1/2\tilde{D}_1}}{2K_1\sqrt{\pi\tilde{D}_1}} \int_0^t {}^{\prime\prime}_1(t-\tau)\, e^{-\lambda_1\tau}\, \sigma_1(z,\tau)\, d\tau \tag{E.1}$$

$$\begin{aligned} N_2^s(z,t) = {} & \frac{e^{zv_2/2\tilde{D}_2}}{2K_2\sqrt{\pi\tilde{D}_2}} \int_0^t \phi_2''(t-\tau)\, e^{-\lambda_2\tau}\, \sigma_2(z,\tau)\, d\tau \\ & + \frac{e^{zv_2/2\tilde{D}_2}}{2\sqrt{\pi\tilde{D}_2}} \int_{-\infty}^{\infty} e^{-z'v_2/2\tilde{D}_2} \left\{ \int_0^t f_2^p(z',t-\tau) \right. \\ & \left. \cdot\, e^{-\lambda_2}\, \sigma_2(z-z',\tau)\, d\tau \right\} dz' \end{aligned} \tag{E.2}$$

$$\begin{aligned} N_3^s(z,t) = {} & \frac{e^{zv_3/2\tilde{D}_3}}{2K_3\sqrt{\pi\tilde{D}_3}} \int_0^t \phi_3''(t-\tau)\, e^{-\lambda_3\tau}\, \sigma_3(z,\tau)\, d\tau \\ & + \frac{e^{zv_3/2\tilde{D}_3}}{2\sqrt{\pi\tilde{D}_3}} \int_{-\infty}^{\infty} e^{-z'v_3/2\tilde{D}_3} \left\{ \int_0^t f_3^p(z',t-\tau) \right. \\ & \left. \cdot\, e^{-\lambda_3\tau}\, \sigma_3(z-z',\tau)\, d\tau \right\} dz' \end{aligned} \tag{E.3}$$

where;

$$f_i^p(z,t) = \frac{1}{K_i}\left[K_{i-1}\lambda_{i-1}N_{i-1}(z,t) + \gamma_{i-1}\lambda_{i-1}\left(\frac{1-\varepsilon}{\varepsilon}\right) m_{i-1}(z,t)\right] \tag{E.4}$$

and the other symbols have been defined in the previous sections.

Equations (E.1) through (E.3) yield the following non-recursive formulas:

$$N_1^S(z,t) = N_{1,1}(z,t) \tag{E.5}$$

$$N_2^S(z,t) = \sum_{j=1}^{3} N_{2,j}(z,t) \tag{E.6}$$

$$N_3^S(z,t) = \sum_{j=1}^{8} N_{3,j}(z,t) \tag{E.7}$$

The functions $N_{i,j}(z,t)$ $(i = 1,2,3,\ j = 1,2,\ldots)$ are given by

$$N_{1,1}(z,t) = B_{11}E(1,1;1) \tag{E.8}$$

$$N_{2,1}(z,t) = B_{22}E(2,2,2)+B_{21}E(1,1;2) \tag{E.9}$$

$$N_{2,2}(z,t) = \frac{B_{11}K_1\lambda_1 v_1}{K_2(v_1-v_2)(\beta_{12}-\lambda_1)}\,[E(1,1;1)-E(1,2;1)-E(1,1;2)+E(1,2;2)] \tag{E.10}$$

$$\begin{aligned} N_{2,3}(z,t) = \frac{B_{11}\gamma_1\lambda_1(\eta_1-\lambda_1)\ v_2}{(-\lambda_1+\beta_{12})^2(v_1-v_2)} \Big\{&[E(1,2;1)-E(1,2;2)] - \\ &- [1+(\lambda_1-\beta_{12})t][E(1,1;1)-E(1,1;2)]\Big\} \end{aligned} \tag{E.11}$$

$$N_{3,1}(z,t) = B_{33}E(3,3,3)+B_{32}E(2,2,3)+B_{31}E(1,1,3) \tag{E.12}$$

$$\begin{aligned} N_{3,2}(z,t) = {} & \frac{K_2\lambda_2 v_2}{K_3(v_2-v_3)}\ \frac{B_{22}}{\beta_{23}-\lambda_2}\,[E(2,2;2)-E(2,2;3)-E(2,3;2)+E(2,3;3)] \\ & + \frac{K_2\lambda_2 v_2}{K_3(v_2-v_3)}\ \frac{B_{21}}{\beta_{23}-\lambda_1}\,[E(1,1;2)-E(1,1;3)-E(2,3;2)+E(2,3;3)] \end{aligned} \tag{E.13}$$

$$N_{3,3}(z,t) = -\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{13}\Gamma_{23}}\ \frac{1}{(-\lambda_1+\beta_{13})(-\beta_{23}+\lambda_1)}\ E(1,1;3)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{13}\Gamma_{23}}\ \frac{1}{(-\lambda_1+\beta_{13})(-\beta_{13}+\beta_{23})}\ E(1,3;3)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{13}\Gamma_{23}}\ \frac{1}{(-\beta_{13}+\beta_{23})(-\beta_{23}+\lambda_1)}\ E(2,3;3)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{32}\Gamma_{12}}\ \frac{1}{(-\lambda_1+\beta_{32})(-\beta_{12}+\lambda_1)}\ E(1,1;2)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{32}\Gamma_{12}}\ \frac{1}{(-\lambda_1+\beta_{32})(-\beta_{32}+\beta_{12})}\ E(2,3;2)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{32}\Gamma_{12}}\ \frac{1}{(-\beta_{32}+\beta_{12})(-\beta_{12}+\lambda_1)}\ E(1,2;2)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{21}\Gamma_{31}}\ \frac{1}{(-\lambda_1+\beta_{21})\ (\beta_{31}+\lambda_1)}\ E(1,1;1)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{21}\Gamma_{31}}\ \frac{1}{(-\lambda_1+\beta_{23})(-\beta_{21}+\beta_{31})}\ E(1,2:1)$$

$$-\frac{\lambda_1\lambda_2B_{11}}{v_1v_2\Gamma_{21}\Gamma_{31}}\ \frac{1}{(\beta_{21}+\beta_{31})(-\beta_{31}+\lambda_1)}\ E(1,3;1) \qquad (E.14)$$

$$N_{3,4}(z,t) = \frac{\gamma_1\lambda_1\lambda_2(\eta_1-\lambda_1)\ B_{11}}{v_1v_2\Gamma_{13}\Gamma_{23}}$$

$$\times\left[\left[\frac{1}{(-\lambda_1+\beta_{13})(-\lambda_1+\beta_{23})}\,t-\frac{(-\lambda_1+\beta_{13})+(-\lambda_1+\beta_{23})}{(-\lambda_1+\beta_{13})^2(-\lambda_1+\beta_{23})^2}\right]E(1,1;3)\right.$$
$$+\frac{1}{(-\beta_{13}+\lambda_1)^2(-\beta_{13}+\beta_{23})}\ E(1,3;3)$$
$$\left.+\frac{1}{(-\beta_{23}+\lambda_1)^2(-\beta_{23}+\beta_{13})}\ E(2,3;3)\right]$$

(Continued on next page)

$$+ \frac{\gamma_1 \lambda_1 \lambda_2 (\eta_1 - \lambda_1) B_{11}}{\nu_1 \nu_2 \Gamma_{32} \Gamma_{12}} \times \left[\begin{array}{l} \left[\frac{1}{(-\lambda_1 + \beta_{32})(-\lambda_1 + \beta_{12})} t - \frac{(-\lambda_1 + \beta_{32}) + (-\lambda_1 + \beta_{12})}{(-\lambda_1 + \beta_{32})^2 (-\lambda_1 + \beta_{12})^2} \right] E(1,1;2) \\ + \frac{1}{(-\beta_{32} + \lambda_1)^2 (-\beta_{32} + \beta_{12})} E(2,3;2) \\ + \frac{1}{(-\beta_{12} + \lambda_1)^2 (-\beta_{12} + \beta_{32})} E(1,2;2) \end{array} \right]$$

$$+ \frac{\gamma_1 \lambda_1 \lambda_2 (\eta_1 - \lambda_1) B_{11}}{\nu_1 \nu_2 \Gamma_{21} \Gamma_{31}} \times \left[\begin{array}{l} \left[\frac{1}{(-\lambda_1 + \beta_{21})(-\lambda_1 + \beta_{31})} t - \frac{(-\lambda_1 + \beta_{21}) + (-\lambda_1 + \beta_{21})}{(-\lambda_1 + \beta_{21})^2 (-\lambda_1 + \beta_{31})^2} \right] E(1,1;1) \\ + \frac{1}{(-\beta_{21} + \lambda_1)^2 (-\beta_{21} + \beta_{31})} E(1,2;1) \\ + \frac{1}{(-\beta_{31} + \lambda_1)^2 (-\beta_{31} + \beta_{21})} E(1,3;1) \end{array} \right] \tag{E.15}$$

$$N_{3,5}(z,t) = \frac{\gamma_2 (1-\gamma_1) \lambda_1 \lambda_2 (\eta_1 - \lambda_1) \nu_3 B_{11}}{\nu_1 - \nu_3}$$

$$\times \left[\begin{array}{l} \left[\frac{1}{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{31})} t - \frac{(-\lambda_1 + \lambda_2) + (-\lambda_1 + \beta_{31})}{(-\lambda_1 + \lambda_2)^2 (-\lambda_1 + \beta_{31})^2} \right] \times \left[E(1,1;1) - E(1,1;3) \right] \\ + \frac{1}{(-\lambda_2 + \lambda_1)^2 (-\lambda_2 + \beta_{31})} \left[E(2,2;1) - E(2,2;3) \right] \\ \frac{1}{(-\beta_{31} + \lambda_1)^2 (-\beta_{31} + \lambda_2)} \left[E(1,3;1) - E(1,3;3) \right] \end{array} \right] \tag{E.16}$$

$$N_{3,6}(z,t) = \frac{\gamma_2\lambda_2(\eta_2-\lambda_2)\ v_3}{v_2-v_3} B_{22} \frac{1}{(-\lambda_2+\beta_{23})^2}$$

$$\begin{bmatrix} [E(2,3;2) - E(2,3;3)] \\ - [1 + (\lambda_2-\beta_{23})\ t] \times [E(2,2;2) - E(2,2;3)] \end{bmatrix}$$

$$- \frac{\gamma_2\lambda_2(\eta_2-\lambda_2)\ v_3}{v_2-v_3} B_{21} \times \begin{bmatrix} \dfrac{1}{(-\lambda_2+\lambda_1)(-\beta_{23}+\lambda_2)} \quad E(2,2;2) - E(2,2;3) \\ + \dfrac{1}{(-\lambda_2+\lambda_1)(-\lambda_1+\beta_{23})} \quad E(1,1;2) - E(1,1;3) \\ + \dfrac{1}{(-\lambda_1+\beta_{23})(-\beta_{23}+\lambda_1)} \quad E(2,3;2) - E(2,3;3) \end{bmatrix} \qquad (E.17)$$

$$N_{3,7}(z,t) = \frac{\gamma_2\lambda_1\lambda_2(\eta_2-\lambda_2)\ B_{11}}{v_1 v_2 \Gamma_{13}\Gamma_{23}}$$

$$\times \begin{bmatrix} \dfrac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{13})(-\lambda_1+\beta_{23})} E(1,1;3) \\ + \dfrac{1}{(-\lambda_2+\lambda_1)(-\lambda_2+\beta_{13})(-\lambda_2+\beta_{23})} E(2,2;3) \\ + \dfrac{1}{(-\beta_{13}+\lambda_1)(-\beta_{13}+\lambda_2)(-\beta_{13}+\beta_{23})} E(1,3;3) \\ + \dfrac{1}{(-\beta_{23}+\lambda_1)(-\beta_{23}+\lambda_2)(-\beta_{23}+\beta_{13})} E(2,3;3) \end{bmatrix}$$

(Continued on next page)

$$
+ \frac{\gamma_2\lambda_1\lambda_2(\eta_2-\lambda_2)\ B_{11}}{\nu_1\nu_2\Gamma_{32}\Gamma_{12}}
$$

$$
\times \left[\begin{aligned}
&\frac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{32})(-\lambda_1+\beta_{12})} E(1,1;2) \\
&+ \frac{1}{(-\lambda_2+\lambda_1)(-\lambda_2+\beta_{32})(-\lambda_2+\beta_{12})} E(2,2;2) \\
&+ \frac{1}{(-\beta_{32}+\lambda_1)(-\beta_{32}+\lambda_2)(-\beta_{32}+\beta_{12})} E(2,3;2) \\
&+ \frac{1}{(-\beta_{12}+\lambda_1)(-\beta_{12}+\lambda_2)(-\beta_{12}+\beta_{32})} E(1,2;2)
\end{aligned} \right]
$$

$$
+ \frac{\gamma_2\lambda_1\lambda_2(\eta_2-\lambda_2)\ B_{11}}{\nu_1\nu_2\Gamma_{21}\Gamma_{31}}
$$

$$
\times \left[\begin{aligned}
&\frac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{21})(-\lambda_1+\beta_{31})} E(1,1;1) \\
&+ \frac{1}{(-\lambda_2+\lambda_1)(-\lambda_2+\beta_{21})(-\lambda_2+\beta_{31})} E(2,2;1) \\
&+ \frac{1}{(-\beta_{21}+\lambda_1)(-\beta_{21}+\lambda_2)(-\beta_{21}+\beta_{31})} E(1,2;1) \\
&+ \frac{1}{(-\beta_{31}+\lambda_1)(-\beta_{31}+\lambda_2)(-\beta_{31}+\beta_{21})} E(1,3:1)
\end{aligned} \right] \tag{E.18}
$$

$$
N_{3,8}(z,t) = \frac{\gamma_1\gamma_2\lambda_1\lambda_2(\eta_1-\lambda_1)(\eta_2-\lambda_2)\ B_{11}}{\nu_1\nu_2}
$$

$$
\times \left[\begin{aligned}
&\frac{1}{\Gamma_{13}\Gamma_{23}} \left[\frac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{13})(-\lambda_1+\beta_{23})}\, t \right. \\
&\qquad \left. - \frac{(-\lambda_1+\lambda_2)(\lambda_1+\beta_{13})+(-\lambda_1+\beta_{13})(-\lambda_1+\beta_{23})+(-\lambda_1+\beta_{23})(-\lambda_1+\lambda_2)}{(-\lambda_1+\lambda_2)^2(-\lambda_1+\beta_{13})^2(-\lambda_1+\beta_{23})^2} \right] E(1,1;3) \\
&+ \frac{1}{\Gamma_{13}\Gamma_{23}} \quad \frac{1}{(-\lambda_2+\lambda_1)^2(-\lambda_2+\beta_{13})(-\lambda_2+\beta_{23})} E(2,2;3)
\end{aligned} \right]
$$

(Continued on next page)

$$
\times\left[\begin{array}{l}
+\dfrac{1}{\Gamma_{13}\Gamma_{23}}\quad\dfrac{1}{(-\beta_{13}+\lambda_1)^2(-\beta_{13}+\lambda_2)(-\beta_{13}+\beta_{23})}\,E(1,3;3)\\[2ex]
+\dfrac{1}{\Gamma_{13}\Gamma_{23}}\quad\dfrac{1}{(-\beta_{23}+\lambda_1)^2(-\beta_{23}+\lambda_2)(-\beta_{23}+\beta_{13})}\,E(2,3;3)\\[2ex]
+\dfrac{1}{\Gamma_{32}\Gamma_{12}}\Bigg[\dfrac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{32})(-\lambda_1+\beta_{12})}\,t\\[2ex]
\qquad-\dfrac{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{32})+(-\lambda_1+\beta_{32})(-\lambda_1+\beta_{12})+(-\lambda_1+\beta_{12})(-\lambda_1+\lambda_2)}{(-\lambda_1+\lambda_2)^2(-\lambda_1+\beta_{32})^2(-\lambda_1+\beta_{12})^2}\Bigg]\,E(1,1;2)\\[2ex]
+\dfrac{1}{\Gamma_{32}\Gamma_{12}}\quad\dfrac{1}{(-\lambda_2+\lambda_1)^2(-\lambda_2+\beta_{32})(-\lambda_2+\beta_{12})}\,E(2,2;2)\\[2ex]
+\dfrac{1}{\Gamma_{32}\Gamma_{12}}\quad\dfrac{1}{(-\beta_{32}+\lambda_1)^2(-\beta_{32}+\lambda_2)(-\beta_{32}+\beta_{12})}\,E(2,3:2)\\[2ex]
+\dfrac{1}{\Gamma_{32}\Gamma_{12}}\quad\dfrac{1}{(-\beta_{12}+\lambda_1)^2(-\beta_{12}+\lambda_2)(-\beta_{12}+\beta_{32})}\,E(1,2;2)\\[2ex]
+\dfrac{1}{\Gamma_{21}\Gamma_{31}}\Bigg[\dfrac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{21})(-\lambda_1+\beta_{31})}\,t\\[2ex]
\qquad-\dfrac{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{21})+(-\lambda_1+\beta_{21})(-\lambda_1+\beta_{31})+(-\lambda_1+\beta_{31})(-\lambda_1+\lambda_2)}{(-\lambda_1+\lambda_2)^2(-\lambda_1+\beta_{21})^2(-\lambda_1+\beta_{31})^2}\Bigg]\,E(1,1;1)\\[2ex]
+\dfrac{1}{\Gamma_{21}\Gamma_{31}}\quad\dfrac{1}{(-\lambda_2+\lambda_1)^2(-\lambda_2+\beta_{21})(-\lambda_2+\beta_{31})}\,E(2,2;1)\\[2ex]
+\dfrac{1}{\Gamma_{21}\Gamma_{31}}\quad\dfrac{1}{(-\beta_{21}+\lambda_1)^2(-\beta_{21}+\lambda_2)(-\beta_{21}+\beta_{31})}\,E(1,2;1)\\[2ex]
+\dfrac{1}{\Gamma_{21}\Gamma_{31}}\quad\dfrac{1}{(-\beta_{31}+\lambda_1)^2(-\beta_{31}+\lambda_2)(-\beta_{31}+\beta_{21})}\,E(1,3;1)
\end{array}\right]
\tag{E.19}
$$

where the function $E(i,j;k)$ is related to the function $E_{i,j,k}(z,t)$ defined by Eq. (2.1.32) as

$$E(i,j;k) = \frac{w_s^0}{Tv} E_{i,j,k}(z,t) \tag{E.20}$$

The explicit solutions for the band release can be obtained from the superposition Eq. (2.1.18).

Appendix F. Consistency of Point-Source and Plane-Source Solutions

When we compare Figs. 4.3.2 and 4.3.4 in Section 4.3, four lines in each figure, which show the size of the repository and the corresponding array of point sources, agree with each other where $10 < \Omega$ for $d = 10\text{m}$, and where $4 < \Omega$ for $d = 5\text{m}$. As a result, the solution to the finite plane source gives a good approximation to that to the array of point sources where Ω is relatively large.

Here we will consider the relationship between Eqs. (4.2.33) and (4.1.65). Equation (4.1.65) can be extended to the solution for the finite plane source by using a superposition method with respect to x and y.

$$ {}^{pl}N_1(x,y,z,t) = {}^{pl}N_1^o e^{-\lambda_1 t} \int_{-b}^{b}\int_{-a}^{a} \frac{\exp\left(-\frac{(x-\eta)^2}{4zD_x/v}\right)}{\sqrt{\frac{4\pi z D_x}{v}}} \; \frac{\exp\left(-\frac{(y-\xi)^2}{4zD_y/v}\right)}{\sqrt{\frac{4\pi z D_y}{v}}} \, d\eta \, d\xi \tag{F.1} $$

where the double integral can be calculated separately.

$$ \int_{-a}^{a} \exp\left(-\frac{(x-\eta)^2}{4zD_x/v}\right) d\eta = \frac{\sqrt{\pi}}{2} \sqrt{\frac{4zD_x}{v}} \left[\operatorname{erf}\left(\frac{a+x}{\sqrt{\frac{4zD_x}{v}}}\right) + \operatorname{erf}\left(\frac{a-x}{\sqrt{\frac{4zD_x}{v}}}\right) \right] \tag{F.2} $$

$$ \int_{-b}^{b} \exp\left(-\frac{(y-\xi)^2}{4zD_y/v}\right) d\xi = \frac{\sqrt{\pi}}{2} \sqrt{\frac{4zD_y}{v}} \left[\operatorname{erf}\left(\frac{b+y}{\sqrt{\frac{4zD_y}{v}}}\right) + \operatorname{erf}\left(\frac{b-x}{\sqrt{\frac{4zD_y}{v}}}\right) \right] \tag{F.3} $$

Finally Eq. (F.1) can be written by substituting Eqs. (F.2) and (F.3).

$$ {}^{pl}N_1(x,y,z,t) = \frac{{}^{pl}N_1^o}{4} e^{-\lambda_1 t} \left[\operatorname{erf}\left(\frac{a+x}{2\sqrt{\frac{D_x z}{v}}}\right) + \operatorname{erf}\left(\frac{a-x}{2\sqrt{\frac{D_x z}{v}}}\right) \right] \times \left[\operatorname{erf}\left(\frac{b+y}{2\sqrt{\frac{D_y z}{v}}}\right) + \operatorname{erf}\left(\frac{b-y}{2\sqrt{\frac{D_y z}{v}}}\right) \right] \tag{F.4} $$

Equation (F.4) is identical to Eq. (4.3.7), which means that the solution for the plane source by superimposing the injected point sources is identical to the solution for the plane source with the concentration boundary condition, if longitudinal dispersion is neglected.

Appendix G. Properties of the Function $G_{p,q}(z)$ for the Solution of the Three-Dimensional Transport Equation

Property 1

$$\frac{\mu_{p,r} - \mu_{q,r}}{K_p - K_q} = \frac{\mu_{p,q} - \mu_{q,r}}{K_p - K_r} \tag{G.1}$$

From Equation (5.5.19)

$$\begin{aligned}(K_p - K_r)(\mu_{p,r} - \mu_{q,r}) &= (K_p - K_r)\,\mu_{p,r} - (K_p - K_q + K_q - K_r)\,\mu_{q,r} \\ &= \lambda_p K_p - \lambda_r K_r - (K_p - K_q)\,\mu_{q,r} - \lambda_q K_q + \lambda_r K_r \\ &= \lambda_p K_p - \lambda_q K_q - (K_p - K_q)\,\mu_{q,r} \\ &= (K_p - K_q)\,\mu_{p,q} - (K_p - K_q)\,\mu_{q,r} \\ &= (K_p - K_q)(\mu_{p,q} - \mu_{q,r})\ .\end{aligned}$$

On dividing by $(K_p - K_q)(K_p - K_r)$ there results Eq. (G.1). Note that Eq. (G.1) can be expressed also in terms of the function $G_{p,q}(z)$ defined by $G_{p,q}(z) = (K_q - K_p)(z - \mu_{p,q})$.

Property 2

$$G_{n,p}(\mu_{p,q}) = G_{n,q}(\mu_{p,q}) \tag{G.2}$$

Property 3

$$G_{p,q}(z) = -G_{q,p}(z) \tag{G.3}$$

Property 4

$$G_{p,q}(\mu_{p,q}) = 0 \tag{G.4}$$

Property 5

$$G'_{p,q}(z) = (K_q - K_p) = -G'_{q,p}(z) \quad . \tag{G.5}$$

These properties are an immediate consequence of the definition of $G_{p,q}(z)$. Let

$$G_{n,p,k}(z) \equiv G_{k,p}(z)\, G_{k+1,p}(z) \,\dots\, G_{p-1,p}(z)\, G_{p+1,p}(z) \,\dots\, G_{n,p}(z) \tag{G.6}$$

Property 6

$$G_{n,p,k}(\mu_{p,q}) = 0 \quad \text{for} \quad k \leq q < p \tag{G.7}$$

because one of the terms in the product is $G_{q,p}(\mu_{p,q})$ which vanishes by Eq. (G.4)

Property 7

$$G'_{n,p,k}(\mu_{p,q}) = -(K_q - K_p)\, G_{n,p,k}(\mu_{p,q}) \tag{G.8}$$

This result is obtained by termwise differentiation of $G_{n,p,k}(z)$ and making use of Eqs. (G.4) and (G.5).

Appendix H. Verification of the Solution for $A_{p,q,k}$ in the Three-Dimensional Difference Eq. (5.5.45)

To verify Eq. (5.5.45) note that when $p = 2$ and $q = k = 1$ (Eq. (5.5.45)) is satisfied in view of the comment below this equation. By the method of induction we now assume the equation to be valid for $p = 3,4, \ldots (n - 1)$ and prove it to be valid for $p = n$. For this one substitutes Eq. (5.5.45) for $p = (n - 1)$ into the right-hand side of Eq. (5.5.44). We denote the right-hand side of this equation by R and use the shorthand notation introduced in Eq. (G.6)

$$R = \sum_{q=k}^{p-1} \frac{L_{n,p}L_{p,k}}{(K_q - K_p)} \frac{1}{G_{n,p,k}(\mu_{p,q})} - \sum_{q=p+1}^{n-1} \frac{L_{n,q}L_{q,k}}{K_p - K_q} \frac{1}{G_{n,p,k}(\mu_{p,q})} \tag{H.1}$$

by Eq. (G.8) and with

$$L_{n,p}L_{p,k} = L_{n,k} \quad , \quad L_{n,q}L_{q,k} = L_{n,k}$$

can be shown to transform into

$$R = - L_{n,k} \sum_{\substack{q=k \\ q \neq p}}^{n-1} \frac{1}{G'_{n,p,k}(\mu_{p,q})} \tag{H.2}$$

To study this sum consider the integral

$$I = \frac{1}{2\pi i} \int_C \frac{dz}{G_{n,p,k}(z)} \tag{H.3}$$

where C is a Jordan curve enclosing $\mu_{k,p}, \mu_{k+1,p} \cdots \mu_{p-1,p}, \mu_{p+1,p} \cdots \mu_{n,p}$. By the residue theorem

$$I = \sum_{q=k}^{n} \frac{1}{G'_{n,p,k}(\mu_{p,q})} \quad . \tag{H.4}$$

Thus Eq. (H.2)

$$R = -L_{n,k}\left[I - \frac{1}{G'_{n,p,k}(\mu_{p,n})}\right] \tag{H.5}$$

But $G_{n,p,k}(z)$ is a polynomial of degree $n - k > n - p \geq 1$. So $G_{n,p,k}(z)$ is a polynominal of degree at least 2. Therefore the residue of $G^{-1}_{n,p,k}(z)$ at infinity is zero. Hence by Eq. (H.3) $I = 0$ and therefore by Eq. (H.5)

$$R = \frac{L_{n,k}}{G'_{n,p,k}(\mu_{p,n})} \quad . \tag{H.6}$$

But by Eq. (G.8)

$$G'_{n,p,k}(\mu_{p,n}) = -(K_n - K_p)\, G_{n,p,k}(\mu_{p,n}) \tag{H.7}$$

So that

$$R = \frac{L_{n,k}}{K_p - K_n} \; \frac{1}{G_{k,p}(\mu_{n,p})\, G_{k+1,p}(\mu_{n,p}) \cdots G_{n-1,p}(\mu_{n,p})} \tag{H.8}$$

Here the factor $G_{p,p}(\mu_{n,p})$ has been replaced by 1 by our convention. This equation is precisely of the form of the right-hand side of Eq. (5.5.45) when p is replaced by n and q is replaced by p. This establishes the validity of Eq. (5.5.45) as a solution of Eq. (5.5.44) for arbitrary values of n.