AECL EACL

AECL-11321

Source-Term Model for the SYVAC3-NSURE Performance Assessment Code

Modèle de terme-source pour le code de calcul SYVAC3-NSURE d'évaluation du comportement

J.H. Rowat, D.S. Rattan, G.M. Dolinar

November 1996 novembre

AECL

Source-Term Model for the SYVAC3-NSURE Performance Assessment Code

by

J.H. Rowat, D.S. Rattan and G.M. Dolinar

Waste Management and Decommissioning
Waste Management Operations
Chalk River Laboratories
Chalk River, Ontario K0J 1J0

1996 November

AECL-11321

EACL

Modèle de terme-source pour le code de calcul SYVAC3-NSURE d'évaluation du comportement

par

J.H. Rowat, D.S. Rattan et G.M. Dolinar

RÉSUMÉ

Les radionucléides qui se trouvent dans les déchets mis en place dans des installations de stockage permanent ne resteront pas indéfiniment dans ces installations. Les barrières artificielles finiront par se détériorer et laisseront des matières radioactives s'échapper de l'installation. La vitesse de libération des radionucléides issus d'une installation de stockage permanent de déchets de faible activité, le terme-source, constitue un élément clé de l'évaluation du comportement du système de stockage permanent. Dans le présent rapport, on donne une description du modèle de terme-source qui a été utilisé dans la version 1.03 du code de calcul SYVAC3-NSURE[1]. NSURE est un programme d'évaluation du comportement qui estime les effets du stockage permanent de déchets de faible activité à faible profondeur en tenant compte du transport par les eaux souterraines. Le modèle de terme-source dont on fait ici la description a été élaboré pour l'installation de stockage permanent appelée IRUS (construction souterraine anti-intrusion), qui sera située dans les morts-terrains non saturés aux Laboratoires de Chalk River d'EACL. Les processus qui sont compris dans le modèle de l'installation sont le comportement du toit et du conditionnement des déchets, et la diffusion, l'advection et la sorption des radionucléides dans le remblai de l'installation. Le modèle qui est présenté ici a été mis au point pour l'IRUS; toutefois, il s'applique à d'autres installations de stockage permanent à faible profondeur.

Gestion des déchets et Déclassement
Activités de gestion des déchets
Laboratoires de Chalk River
Chalk River (Ontario) K0J 1J0
1996 Novembre

[1] SYVAC3-NSURE = Systems Variability Analysis Code generation 3-Near Surface Repository.

AECL-11321

AECL

Source-Term Model for the SYVAC3-NSURE Performance Assessment Code

by

J.H. Rowat, D.S. Rattan and G.M. Dolinar

ABSTRACT

Radionuclide contaminants in wastes emplaced in disposal facilities will not remain in those facilities indefinitely. Engineered barriers will eventually degrade, allowing radioactivity to escape from the vault. The radionuclide release rate from a low-level radioactive waste (LLRW) disposal facility, the source term, is a key component in the performance assessment of the disposal system. This report describes the source-term model that has been implemented in Ver. 1.03 of the SYVAC3-NSURE[1] code. NSURE is a performance assessment code that evaluates the impact of near-surface disposal of LLRW through the groundwater pathway. The source-term model described here was developed for the Intrusion Resistant Underground Structure (IRUS) disposal facility, which is a vault that is to be located in the unsaturated overburden at AECL's Chalk River Laboratories. The processes included in the vault model are roof and waste package performance, and diffusion, advection and sorption of radionuclides in the vault backfill. The model presented here was developed for the IRUS vault; however, it is applicable to other near-surface disposal facilities.

Waste Management and Decommissioning
Waste Management Operations
Chalk River Laboratories
Chalk River, Ontario K0J 1J0

1996 November

[1] SYVAC3-NSURE = Systems Variability Analysis Code generation 3–Near Surface Repository.

AECL-11321

CONTENTS

LIST OF FIGURES

1.0 INTRODUCTION

AECL is planning to construct a low-level radioactive waste (LLRW) disposal facility called the Intrusion Resistant Underground Structure (IRUS) at Chalk River Laboratories (CRL) in Chalk River, Ontario (Dolinar et al. 1994). The IRUS repository is a near-surface concrete vault that will be built in a sand ridge (i.e., in the unsaturated zone). Radionuclide contaminants in wastes emplaced in disposal vaults such as IRUS will not remain in these vaults indefinitely. With time, waste containers will fail, allowing nuclides to diffuse out of waste packages and subsequently out of the vault. The vault's concrete roof will also fail, and admit infiltration that will transport contaminants out of the vault by advection. The radionuclide release rate an LLRW disposal facility, the source term, is a key component in the performance assessment of the disposal system. This report describes the source-term model that was developed for the IRUS facility. The model has been implemented in Version 1.03 of the SYVAC3-NSURE[2] performance assessment code (Rattan et al. 1996a).

An earlier version of the source-term model for the IRUS facility was documented in two reports: Selander et al. (1991) and Rowat et al. (1991). Vault design changes and a reassessment of available data necessitated modification of the previous model. As a mathematical convenience, it was assumed in the previous model that the vault backfill was nonsorbing; the present model includes sorption. Reliance on waste form performance for IRUS performance assessment calculations (particularly for baled wastes) has been decreased because sufficient experimental data are not available. However, waste form performance can be modeled if the radionuclide fractional release rates for the waste package are known. Additional support is provided for assumptions concerning mass transfer in the unsaturated zone.

Section 2 of this report provides a description of the IRUS facility in its post-closure phase. Section 3 describes the conceptual model for the disposal system, and the equation for nuclide mass transfer in the vault. Particular solutions of the mass-transfer equation in the zero- and steady-infiltration regions of the vault are derived in Section 4. In Section 5, these solutions are integrated with performance functions for the roof and waste packages to derive the three components of the IRUS source-term model. Section 6 discusses the interpretation of the source-term components and provides a summary of results.

2.0 SYSTEM DESCRIPTION

The IRUS vault is to be located in the sand ridge southwest of Lake 233 in the Outer Area of AECL's Chalk River Laboratories. Figure 1 shows a cross-section of the IRUS vault. The vault will have 60 cm thick scalloped outer concrete walls, a 1 m thick sloped concrete roof, and a permeable bottom that permits it to be free-draining (to prevent flooding). A multilayer soil cover (about 2 m thick) will be placed above the concrete roof to prevent frost penetration, divert infiltration and prevent biointrusion (plant and animal).

---

[2] SYVAC3-NSURE = Systems Variability Analysis Code generation 3–Near Surface Repository.

The vault will be filled with waste packages embedded in a free-draining backfill in about a 1:1 volumetric ratio. The backfill around the packages in the vault will be a 90:10 mixture of sand:clinoptilolite by weight (Woods 1996). Waste packages consist of bales of miscellaneous trash, and 0.205 m^3 drums of bitumenized wastes. Bales have a volume of about 0.4 m^3 and are made by compacting about 50 to 60 bags of laboratory trash. The miscellaneous trash in bales comprises mainly cellulose, plastics and rubber (e.g., paper towels, mop heads, platics bags, rubber gloves). Two different types of bituminized waste will be emplaced in IRUS: bituminized ash from the incineration of bales, and bituminized liquid waste.

Below the vault are the buffer layers, which consist of a 30 cm sand/clay layer over a 30 cm sand/clinoptilolite layer. These two engineered barriers provide additional cation sorption capacity for nuclides that migrate out of the backfill (Woods 1996). The base of the vault will be about 1 m above the level of the highest recorded water table. Contaminants that escape from the vault will descend through this unsaturated zone to the water table and enter the groundwater in the underlying sandy aquifer.

The permeable backfill and buffer layers allow the vault to drain freely, which in turn reduces contact between infiltrating water and waste packages. Water could accumulate in the vault only if the infiltration rate exceeded the saturated hydraulic conductivity of the backfill (Rubin and Steinhardt 1963); that is, if:

$$q > K_{sat} \tag{1}$$

where q ($m \cdot a^{-1}$) is the infiltration rate and K_{sat} ($m \cdot a^{-1}$) is the saturated hydraulic conductivity of the backfill. For the IRUS backfill material a representative value for K_{sat} is 4000 $m \cdot a^{-1}$, based on the hydraulic conductivity of the main sandy unit in the Lake 233 aquifer as reported by Klukas and Moltyaner (1995). Infiltration rates would therefore have to be about 10^4 times the seasonally-averaged infiltration rate of 0.3 $m \cdot a^{-1}$ before water could accumulate within the vault—an unlikely event. Accumulation of water has occurred in trenches at the West Valley and Maxey Flats LLRW disposal sites in the United States (Meyer 1976); those trenches had permeable caps and were situated in low-permeability media.

3.0 CONCEPTUAL MODEL AND GOVERNING EQUATION

The vault source term is obtained from a multicomponent mass-transfer model, each component having its own characteristic rate. Together, these rates determine the kinetics of the system as a whole. The processes included in the IRUS source-term model are advection and diffusion, nuclide sorption in the backfill, roof performance and waste package performance. Radioactive decay, which is an important process for attenuating nuclide release, is not included in the equations presented here. It is inserted when all of the mass-transfer calculations are finished, which is permissible because the solution is separable into a spatial and time component; this is no longer true when decay chains are included in the model. A comprehensive discussion of general issues pertaining to LLRW source terms has been given elsewhere (Sullivan 1991).

The interior of the IRUS vault is modelled as a parallelepiped of length 28.7 m, width 18.3 m, and height 8.34 m. As Figure 2 shows, the vault is divided into two regions: a zero- and a steady-infiltration region. The zero-infiltration region lies below the part of the roof not admitting infiltration; the steady-infiltration region lies below the part of the roof admitting infiltration. Mass transfer in the zero-infiltration region is governed by diffusion; mass transfer in the steady-infiltration region is assumed to be advection-dominated. The fraction of the horizontal area of the vault that is subject to infiltration is represented by the roof performance function, which is a time series specified by the user. Container performance is quantified in a similar manner by a time series that gives the fraction of the containers that have failed. Nuclide sorption in the backfill is modeled using the conventional K_d approach.

Nuclides released from the waste packages diffuse into the surrounding backfill and migrate out of the vault, either avoiding or passing through other waste forms. Mathematically, however, it is much easier to model mass transfer in the vault if it is treated as a single homogeneous medium, rather than as a heterogeneous system of packages and backfill. The approach taken here is to treat the vault as a single homogeneous medium.

Mass transfer in the vault is modeled using the one-dimensional advection-dispersion equation (see Appendix A):

$$R\frac{\partial C}{\partial t} + U\frac{\partial C}{\partial x} - D\frac{\partial^2 C}{\partial x^2} = 0 \tag{2}$$

where D ($m^2 \cdot a^{-1}$) is the diffusivity, U ($m \cdot a^{-1}$) is the average pore-water velocity, and R is the retardation factor of the backfill. The diffusivity and average pore-water velocity are represented by "effective" values to account for the heterogeneous nature of the real system, and are calculated as follows.

The effective diffusivity of the homogeneous vault is calculated using Maxwell's formula assuming that the backfill diffusivity is much higher than that of the waste packages. The effective diffusivity is:

$$D = \frac{1-p}{1+0.5p} D_b \,; \qquad D_b = D_0/\tau_b^2 \tag{3}$$

where:

D_b = the backfill diffusivity
p = the volume fraction of the waste packages
D_0 = the free-water diffusivity
τ_b = the backfill tortuosity.

Using Maxwell's formula, D is 2.5 times less than D_b when $p = 0.5$. Crank (1975, Chapter 12) discusses several other formulae by which to calculate effective diffusivities for two-phase systems; for comparison, the value of D derived from Cheng and Vachon's formula is 3.2 times less than D_b. Given the inherent uncertainties associated with factors such as distribution coefficients, and given that diffusive mass-transfer rates are proportional to $D^{1/2}$, mass-transfer rates are not sensitive to the correction for vault heterogeneity.

The average pore-water velocity in the vault is calculated assuming that waste packages are impermeable to flow, which leads to the following expression:

$$U = \frac{q}{\theta(1-p)} \tag{4}$$

Because $0 < p < 1$, the average pore-water velocity is higher inside the vault than in the unsaturated zone surrounding the vault.

4.0 NUCLIDE RELEASE RATES IN ZERO- AND STEADY-INFILTRATION REGIONS

The source-term model derived here is a two-region model that is constructed from solutions of the governing equation in the zero- and steady-infiltration regions. The particular solutions of the mass-transfer equation that are used for the source-term model are discussed in Sections 4.1 to 4.3.

4.1 Concentration and Release Rate: Zero-Infiltration Region

When the vault roof is intact, mass transfer inside the vault is governed by diffusion, and the mass-transfer equation is

$$D\frac{\partial^2 C_1}{\partial x^2} - R\frac{\partial C_1}{\partial t} = 0 \tag{5}$$

The subscript "1" denotes the zero-infiltration region (i.e., the region where $U = 0$). Assuming that the total nuclide inventory, N (Bq), is uniformly distributed throughout the vault, the initial nuclide concentration is equal to:

$$C_1(x,t) = \frac{N}{R\theta V(1-p)} = C_0 \quad \text{at} \quad t = 0 \tag{6}$$

where V (m^3) is the volume of the vault, $\theta V(1\text{-}p)$ is the total volume of pore water in the backfill and $1/R$ is the fraction of nuclides in the pore water. The solution for Eq. (5) derived in this section assumes that the entire nuclide inventory is present in the backfill at $t = 0$.

The boundary conditions for the zero-infiltration region are:

$$C_1(x,t) = 0 \quad \text{at} \quad x = L, \ t > 0 \tag{7}$$

and

$$\frac{\partial C_1}{\partial x} = 0 \quad \text{at} \quad x = 0, \ t > 0 \tag{8}$$

where $x = L$ corresponds to the bottom of the vault, that is, the vault/buffer interface. The boundary condition at the bottom of the vault, Eq. (7), represents a zero-concentration boundary; the boundary condition for the top of the vault, Eq. (8), represents a zero-flux boundary. The IRUS structure is constructed from concrete that is much less permeable to diffusion than the sand/clinoptilolite backfill, hence upward mass transfer at $x = 0$ is expected to be negligible. The zero-concentration boundary at $x = L$ would correspond physically to having a fast-flowing aquifer located at the base of the vault. This boundary condition is conservative because the water table is expected to be at least 1 m below the base of the vault. Nuclide release rates are not very sensitive to the choice of boundary condition at the bottom of the vault (Rowat and Lane 1994).

Solving Eqs. (5-8) yields (Crank 1975, Eq. (4.17)):

$$C_1(x,t) = C_0 \sum_{n=0}^{\infty} B_n \cos(\omega_n x) \exp\left\{-\frac{D}{R}\omega_n^2 t\right\} \tag{9}$$

where:

$$B_n = \frac{4(-1)^n}{(2n+1)\pi} \quad \text{and} \quad \omega_n = \frac{(2n+1)\pi}{2L} \tag{10}$$

The flux per unit horizontal area of the vault at $x = L$ is equal to:

$$J_1(t) = -\theta(1-p)D\left.\frac{\partial C_1}{\partial x}\right|_{x=L} \tag{11}$$

Multiplying $J_1(t)$ ($Bq{\cdot}m^{-2}{\cdot}a^{-1}$) by V/L, the cross-sectional area of the vault, yields the total release rate:

$$\Phi_V^1(t) = \frac{2ND}{L^2R} \sum_{n=0}^{\infty} \exp\left\{-\frac{D}{R}\omega_n^2 t\right\} \tag{12}$$

Equation (12) can also be written as (Carslaw and Jaeger 1959, pg. 97):

$$\Phi_V^1(t) = N\sqrt{\frac{D}{RL^2\pi t}}\left\{1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left\{-\frac{n^2RL^2}{Dt}\right\}\right\} \tag{13}$$

and can be derived from Eq. (12) using Poisson's summation formula (Papoulis 1962).

The cumulative fraction of the nuclide inventory released (CFR) by time t is:

$$\chi_1(t) = \frac{1}{N}\int_0^t \Phi_V^1(\tau)\,d\tau = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\left[1 - \exp\left(-\frac{D}{R}\omega_n^2 t\right)\right](2n+1)^{-2} \tag{14}$$

The release rate $\Phi_V^1(t)$ is mass conserving, because $\chi_1(\infty) \to 1$ as $t \to \infty$.

4.2 Closed-Form Expressions for $\Phi_V^1(t)$ and $\chi_1(t)$

From Eq. (12) it is clear that the release rate at large times is represented asymptotically by a single exponential, the term $n = 0$. For short times, the release rate condenses to a singularity of the form $t^{-1/2}$, as shown by Eq. (13). To simplify computations, Eq. (12) is replaced with the closed form expression:

$$\Phi_V^1(t) = N\left\{\frac{2D}{L^2R}\exp\left(-\frac{D}{R}\omega_0^2 t\right) + \frac{1}{\sqrt{t}}\sqrt{\frac{D}{\pi R L^2}}\exp\left(-\beta\frac{D}{R}\omega_0^2 t\right)\right\} \tag{15}$$

where:

$$\beta = \left(\frac{2}{\pi(1 - 8/\pi^2)}\right)^2 \approx 11.294 \tag{16}$$

Equation (15) is a good approximation for Eq. (12) because it is asymptotically exact for both limits, $t \to 0$ and $t \to \infty$, and by the choice of the dimensionless parameter β it also satisfies the requirement that total mass is conserved, that is $\chi_1(t) \to 1.0$ as $t \to \infty$. Detailed calculations comparing Eqs. (12) and (15) have shown the two expressions to be numerically close, uniformly over all values of t (see Appendix B). The corresponding two-term approximation for the CFR is:

$$\chi_1(t) = \frac{8}{\pi^2}\left[1 - \exp\left(-\frac{D\omega_0^2 t}{R}\right)\right] + \frac{2}{\pi}\sqrt{\frac{1}{\beta}}\,\mathrm{erf}\left(\sqrt{\frac{\beta D\omega_0^2 t}{R}}\right) \tag{17}$$

which is obtained by integrating Eq. (15) with respect to t.

This singularity in Φ_V^1 at $t = 0$ is a mathematical artifact of the initial condition, and occurs in any model which prescribes an infinite initial concentration gradient. In NSURE, the singularity at $t = 0$ is avoided by using linear interpolation to calculate the release rate near the time origin. As Figure 3 shows, the release rate near $t = 0$ is calculated as follows:

1) Choose a small time interval δt.
2) Calculate $\chi_1(\delta t)$, which is the total mass released between $t = 0$ and $t = \delta t$.
3) Calculate $\Phi_V^1(0)$ by linear interpolation from $\Phi_V^1(\delta t)$ by requiring mass conservation in the interval $0 \le t < \delta t$. The result is that

$$\Phi_V^1(0) = \frac{2N}{\delta t}\chi_1(\delta t) - \Phi_V^1(\delta t) \tag{18}$$

4) In the short interval $0 \le t < \delta t$, calculate $\Phi_V^1(t)$ by linear interpolation from:

$$\Phi_V^1(t) = \left[\frac{\Phi_V^1(\delta t) - \Phi_V^1(0)}{\delta t}\right]t + \Phi_V^1(0) \qquad \text{for} \quad 0 \le t < \delta t \tag{19}$$

For times $t \geq \delta t$, $\Phi^1_V(t)$ is calculated using Eq. (15). The use of linear interpolation near the time origin is justified because δt can always be chosen small enough to ensure that the total mass released between $t = 0$ and $t = \delta t$ is negligible.

The closed-form expressions for $\Phi^1_V(t)$ and $\chi_1(t)$ implemented in NSURE differ slightly from Eqs. (15) and (17); the expressions for $\Phi^1_V(t)$ and $\chi_1(t)$ that have been implemented in NSURE are described in Appendix B.

4.3 Concentration and Release Rates: Steady-Infiltration Region

The onset of infiltration following failure of a section of the roof is assumed to be instantaneous, and mass transfer in the part of the vault exposed to infiltration is assumed to occur by advection only (i.e., slug flow). As before, the system starts from a uniform initial concentration and the governing equation is

$$R \frac{\partial C_2}{\partial t} + U \frac{\partial C_2}{\partial x} = 0 \qquad (20)$$

with

$$C_2(x, t) = \frac{N}{[R\,\theta\,V(1-p)]} = C_0 \qquad \text{at} \quad t=0 \quad \text{and} \quad 0 \leq x \leq L \qquad (21)$$

The subscript "2" denotes the steady-infiltration region. The only boundary condition is a zero-flux condition at the top of the vault ($x = 0$), which is equivalent to a zero-concentration boundary because mass transfer is purely advective. Therefore

$$C_2(x,t) = 0 \qquad \text{at} \quad x=0 \quad \text{and} \quad t \geq 0 \qquad (22)$$

The solution for Eqs. (20) and (22), obtained by Laplace transformation, is

$$C_2(x,t) = \begin{cases} C_0\,, & t < \dfrac{xR}{U} \\ 0\,, & t > \dfrac{xR}{U} \end{cases} \qquad (23)$$

and the nuclide release rate at $x = L$ is

$$\Phi^2_V(t) = (V/L)\ \theta U(1-p)\ C_2(L, t) = \begin{cases} \dfrac{UN}{LR} \quad \ldots\ldots & t < \dfrac{LR}{U} \\ 0 \quad \ldots\ldots & t \geq \dfrac{LR}{U} \end{cases} \qquad (24)$$

This solution corresponds to a slug-flow release of solute following the onset of infiltration.

5.0 NUCLIDE RELEASE RATES INCLUDING CONTAINER AND ROOF PERFORMANCE

The vault roof and waste package containers are expected to fail gradually with time. The functions derived in the previous section do not account for roof and container failure events (i.e., the gradual failure of the roof and containers). Sections 5.1 to 5.6 describe how roof and waste package performance have been integrated with the release rates derived in Sections 4.2 and 4.3.

5.1 Roof Performance

Conceptually, the IRUS vault is partitioned into two regions: a zero- and a steady-infiltration region. The zero-infiltration region lies below the part of the roof not admitting infiltration; the steady-infiltration region lies below the part of the roof admitting infiltration. In the zero-infiltration region, mass transfer is assumed to occur by diffusion only; in the steady-infiltration region, mass transfer is assumed to be purely advective, with the infiltration rate given by Eq. (4). The roof performance function, $F_r(t)$, represents the cumulative fraction of the roof surface area that is leaking at time t, and hence determines the fraction of the vault that is subject to infiltration. The roof failure rate, $f_r(t)$, is the time derivative of $F_r(t)$.

5.2 Waste Package Performance

Containers affect radionuclide release by delaying the onset of waste form leaching. The container performance function, $F_c(t)$, represents the cumulative fraction of containers that have failed as a function of time. The mass-transfer model assumes that a failed container is one that has been stripped away from the waste form.

No credit is taken for nuclide retention by the waste forms. Once a container has failed, it is assumed that the nuclide inventory in the package is instantaneously released and uniformly distributed throughout the vault backfill. For example, when the value of $F_c = 0.5$, the nuclide inventory in half of the waste packages has been released into the backfill. The container failure rate, $f_c(t)$, is the time derivative of $F_c(t)$.

For any given type of waste package (e.g., bitumen in steel drums), $F_c(t)$ effectively represents the cumulative fraction released (CFR) for nuclides from that type of waste package. It is the CFR for the collection of waste packages of that type, not the CFR of a single package. Because nuclide retention by waste forms is assumed to be negligible, all nuclides are released at the same rate. This is a very conservative assumption, particularly for conditioned wastes.

The dose impact of nuclide retention by waste forms can be assessed in a limited fashion using the present model. This is done on a nuclide-by-nuclide basis by prescribing, as NSURE input, a "container" performance function $F_c(t)$ that combines waste form and container performance. Mathematically, the user specifies an array $F_c(t)$ that is the convolution of a true container failure function with a nuclide-specific CFR for the waste form.

5.3 Vertical Partitioning of the Repository

To derive expressions for nuclide release rates that include roof and container failure processes, it is convenient to discretize the variables. The time variable is discretized in uniform increments Δt. The increments in the roof performance function, labelled $\Delta F_r(t_i)$, are defined as:

$$\Delta F_r(t_k) = \begin{cases} F_r(t_1) \quad \dots\dots\dots & \text{if } k=1 \\ F_r(t_k) - F_r(t_{k-1}) & \text{for } k=2,3,\dots \end{cases} \tag{25}$$

where $t_k = t_{k-1}+\Delta t$, and the point t_1 corresponds to $t = 0$. Increments for the container performance function, ΔF_c, are defined similarly.

Each element of the ΔF_r array corresponds to a vertical section of the vault. Each vertical section is characterized by two parameters: a fractional width ΔF_r, and a time that specifies the onset of infiltration, which is labelled T. The transition within a vertical section from zero to steady infiltration (at $t = T$) is assumed to be instantaneous. Mathematically, this abrupt change is reflected by a switch in the governing equation from the diffusion to the advection equation.

The vault source term is made up of three components:

1) The diffusive component, φ_1, which is the nuclide release rate from the zero-infiltration region of the vault (i.e., nuclides that migrate out of the vault by diffusion).

2) The advective component, φ_2. This component is generated by waste packages for which the containers fail after the onset of infiltration.

3) The wash-out component, φ_3. When the switch from zero to steady infiltration occurs, nuclides may be present in the backfill as a result of container failures that happened prior to the onset of infiltration. With the onset of infiltration these nuclides get "washed out" of the vault generating the φ_3 component.

The function $\varphi_1(t)$ is the total nuclide release rate for the zero-infiltration region; the sum of $\varphi_2(t)$ and $\varphi_3(t)$ is equal to the total nuclide release rate for the steady-infiltration region.

5.4 The Diffusive Component, φ_1

Before roof failure occurs, mass transfer in the vault is diffusion controlled. The expression for the nuclide release rate from a single vertical section, prior to the onset of infiltration at time $t = T$, is:

$$
\begin{aligned}
\varphi_1(0) &= \Delta F_r(T)\ \{\Delta F_c(0)\Phi_V^1(0)\} \\
\varphi_1(\Delta) &= \Delta F_r(T)\ \{\Delta F_c(0)\Phi_V^1(\Delta) + \Delta F_c(\Delta)\Phi_V^1(0)\} \\
&\ \vdots \\
\varphi_1(n\Delta) &= \Delta F_r(T) \sum_{m=0}^{n} \Delta F_c(m\Delta)\, \Phi_V^1[(n-m)\Delta] \qquad \text{for} \quad n\Delta < T
\end{aligned}
\tag{26}
$$

where Φ_V^1 is given by Eq. (15) and Eq. (19). Note that $\varphi_1 = 0$ at times $t \geq T$. The ΔF_c increments in Eq. (26) account for the time-distributed failure of containers and subsequent nuclide releases into the vault backfill. The time shifts in Φ_V^1 reflect the delay in the onset of leaching associated with each container failure increment. The sum in Eq. (26) is multiplied by $\Delta F_r(T)$ to account for the fraction of the vault occupied by the vertical section.

The discrete convolution of ΔF_c with Φ_V^1 in Eq. (26) can be written as:

$$
\varphi_1(t) = H(T-t)\ \Delta F_r(T) \int_0^t f_c(\tau)\, \Phi_V^1(t-\tau)\, d\tau \tag{27}
$$

where H is the Heaviside step function. Allowing for time-distributed roof failure by summing over all intact vertical sections of the vault (i.e., the zero-infiltration region) gives

$$
\varphi_1(t) = [1 - F_r(t)] \int_0^t f_c(\tau)\, \Phi_V^1(t-\tau)\, d\tau \tag{28}
$$

5.5 The Advective Component, φ_2

The derivation of the expression for $\varphi_2(t_k)$ is analogous to that for $\varphi_1(t_k)$. Nuclide release begins with the onset of infiltration at $t = T$, instead of at $t = 0$, and the release rate is given by Φ_V^2 instead of Φ_V^1. Focussing on a single vertical section, the expression for φ_2 at times $t \geq T$ is:

$$
\begin{aligned}
\varphi_2(T) &= \Delta F_r(T)\ \{\Delta F_c(T)\Phi_V^2(0)\} \\
\varphi_2(T+\Delta) &= \Delta F_r(T)\ \{\Delta F_c(T)\Phi_V^2(\Delta) + \Delta F_c(T+\Delta)\Phi_V^2(0)\} \\
&\ \vdots \\
\varphi_2(T+n\Delta) &= \Delta F_r(T) \sum_{m=0}^{n} \Delta F_c(T+m\Delta)\, \Phi_V^2((n-m)\Delta)
\end{aligned}
\tag{29}
$$

Prior to $t = T$ the φ_2 term is equal to zero. The continuous representation for Eq. (29) is:

$$
\varphi_2(t) = \Delta F_r(T) \int_0^t H(\tau - T)\, f_c(\tau)\, \Phi_V^2(t-\tau)\, d\tau \tag{30}
$$

Allowing for time-distributed roof failure, by summing over all vertical sections in the steady-infiltration region, yields:

$$\varphi_2(t) = \int_0^t f_r(T) \left\{ \int_0^t H(\tau - T) f_c(\tau)\, \Phi_V^2(t-\tau)\, d\tau \right\} dT \tag{31}$$

Because both integrals in Eq. (31) have the same limits, the expression simplifies to

$$\varphi_2(t) = \int_0^t F_r(\tau)\, f_c(\tau)\, \Phi_V^2(t-\tau)\, d\tau \tag{32}$$

5.6 The Wash-Out Component, φ_3

The nuclide concentration at time T (i.e., at the onset of infiltration) is obtained by convolution of the concentration profile, $C_1(x,T)$, with the container failure rate, and is:

$$C_c(x,T) = \int_0^T f_c(\tau)\, C_1(x, T-\tau)\, d\tau \tag{33}$$

If the entire vault roof were to fail suddenly at time T, then the wash-out component at times $t \geq T$ would be

$$\Phi_V^3(t,T) = \theta U(1-p) \frac{V}{L}\, C_c\left(L - \frac{U}{R}(t-T),\, T \right) \tag{34}$$

Prior to $t = T$ this term is zero. If the roof fails gradually, then

$$\varphi_3(t) = \int_0^t f_r(T)\, \Phi_V^3(t,T)\, dT \tag{35}$$

Equation (35) is a nested integral over both space and time variables. The present version of the SYVAC code does not have a module to evaluate this type of integral; however, this difficulty can be circumvented by replacing $C_1(x,t)$ with its spatial average. The spatial average of $C_1(x,t)$ is

$$\bar{C}_1(t) = \frac{N}{\theta R V(1-p)} \left(1 - \chi_1(t) \right) \tag{36}$$

Figure 4 shows how $\bar{C}_1(t)$ and $C_1(x,t)$ evolve with time. The leading and trailing edges of Φ_V^3 are proportional to the concentration at the points at $x = L$ and $x = 0$, respectively. Release rates derived using $\bar{C}_1$ rather than C_1 will overpredict the magnitude of Φ_V^3 soon after wash-out starts, and underpredict it somewhat at long times. As Figure 4 shows, the peak value of $C_1(x,t)$ may exceed $\bar{C}_1(t)$ by at most a factor of two. Hence, by employing $\bar{C}_1$ rather than C_1, the peak value of φ_3, may be underestimated; the discrepancy, however, will be at most a factor of two.

Recalculating C_c from Eq. (33) using $\bar{C}_1$ rather than C_1 yields:

$$\bar{C}_c(T) = \frac{N}{\theta R V(1-p)} \int_0^T f_c(\tau)\left(1 - \chi_1(T-\tau)\right) d\tau \tag{37}$$

As in Eq. (34), if the entire roof were to fail suddenly at time T, then the wash-out component at times $t \geq T$ would be

$$\Phi_V^3(t,T) = \left[H(L - \frac{U}{R}(t-T)) - H(\frac{U}{R}(T-t))\right] \phi(T) \tag{38}$$

where

$$\phi(T) = \frac{UN}{RL} \int_0^T f_c(\tau)\left[1 - \chi_1(T-\tau)\right] d\tau \tag{39}$$

Rearranging the step functions in Eq. (38) leads to the following equivalent form for Φ_V^3:

$$\Phi_V^3(t,T) = \left\{H(t-T) - H\left[t - \left(T + \frac{RL}{U}\right)\right]\right\} \phi(T) \tag{40}$$

Substituting Eq. (40) into Eq. (35) to obtain the expression for the wash-out component when roof failure is distributed over time yields:

$$\varphi_3(t) = \int_0^t H\left(T - \left[t - \frac{RL}{U}\right]\right) f_r(T)\, \phi(T)\, dT \tag{41}$$

The pathway for subsurface radionuclide migration from the IRUS facility is: the vault, the unsaturated zone beneath the vault and the underlying Lake 233 Aquifer. The horizontal extent of the vault (20 m by 30 m) is much greater than the depth of the unsaturated zone beneath the vault (1-2 m). Hence, it is assumed that the zero- and steady-infiltration regions extend through the vault and unsaturated zone below it, as shown in Figure 2. Mass transfer in the unsaturated zone below the vault is not part of the source term, however, it is described by a set of components analogous to the vault components. For this reason the equations for mass transfer in the unsaturated zone below the vault are included in this report, and are described in Appendix C.

6.0 SUMMARY

A source-term model has been developed to calculate radionuclide release rates from the IRUS vault. The major processes incorporated into the model are:

- Time-distributed failure of the infiltration barrier.
- Time-distributed release of radionuclides from waste packages.
- Advective and diffusive mass transfer in a sorbing backfill.

The vault is treated as a homogeneous porous medium. Mass transfer is modelled using the 1D advection-dispersion equation, assuming a uniform moisture content and either zero- or steady-infiltration conditions. Contaminant sorption is modelled using a linear sorption isotherm (i.e., the K_d model). A roof performance function determines the fraction of the vault contents that are exposed to infiltration (i.e., steady-infiltration conditions). The change from zero- to steady-infiltration conditions within the vault is assumed to be instantaneous. For IRUS safety assessment calculations, radionuclide releases from waste forms are assumed to be instantaneous; when a waste container fails, the nuclide inventory in the package is assumed to be instantaneously and uniformly distributed throughout the backfill. Hence, the container failure function is *de facto* the CFR for nuclide release from the waste package.

The complete source term for the repository is the sum of three components:

$$\varphi(t) = \varphi_1(t) + \varphi_2(t) + \varphi_3(t) \tag{42}$$

where φ_1, φ_2 and φ_3 represent the diffusive, advective and wash-out components, and are given by Eqs. (28), (32) and (41), respectively. The source-term model is semi-analytical, which in contrast with purely numerical models, provides better insight into facility performance and is computationally efficient. The diffusive component, φ_1, is usually only significant for mobile nuclides. The migration of strongly-sorbed species is dominated by advection. The total release rate, φ, of mobile long-lived nuclides, such as ^{14}C, is often dominated by the wash-out component, φ_3. If the time-scale for nuclide release from waste packages is comparable to, or exceeds that of, the period during which roof failure occurs, then φ is dominated by the φ_2 component; conditioned wastes tend to be in this category.

The source-term model derived here is of sufficient flexibility to represent a variety of a near-surface disposal systems. The IRUS repository is intended for radioactive wastes with low actinide contents, therefore, decay chains have not been included in the model. The model has been implemented in SYVAC3-NSURE, and can be run in a deterministic or probabilistic mode. The complexity of the model is believed to be commensurate with the inherent variability of the processes that will occur in a near-surface vault such as IRUS. Solubility-limited mass transfer, for example, has not been incorporated into the present model, because the chemistry of the vault is not well enough defined to take credit for nuclide solubility constraints. Omitting solubilities is one of several approximations in the model that may cause it to overestimate rates of nuclide release.

Results generated using the COSMOS code, an earlier version of NSURE (see Rattan et al. 1996b), were used in an IAEA intercode comparison study involving seventeen participants (IAEA 1995). For the test cases considered in the IAEA study, the results obtained using COSMOS (the NSURE predecessor) were generally within the range of values produced by other participants (i.e., not an outlier). This provides some validation for the NSURE models and indicates that NSURE results will be representative of results that would be generated using other performance assessment codes.

ACKNOWLEDGEMENT

The authors would like to thank Dr. T.M. Sullivan and Dr. W.N. Selander for their technical review of this report.

REFERENCES

IAEA 1995. "Safety assessment of near surface radioactive waste disposal facilities: model intercomparison using simple hypothetical data (Test Case 1)", IAEA-TECDOC-846, International Atomic Energy Agency, Vienna.

Carslaw, H.S., and J.C. Jaeger 1959. *Conduction of Heat in Solids*, Second Editionn, Clarendon Press, Oxford.

Crank, J. 1975. *The Mathematics of Diffusion*, Second Edition, Clarendon Press, Oxford.

Dolinar, G.M., D.S. Rattan and J.H. Rowat 1994. "AECL IRUS Near-Surface Low-Level Waste Repository", 16th Annual U.S. Department of Energy Low-Level Radioactive Waste Management Conference (1994 December 13-15, Phoenix, Arizona).

Klukas, M. and G.M. Moltyaner 1995. "Numerical Simulations of Groundwater Flow and Solute Transport in the Lake 233 Aquifer", Atomic Energy of Canada Limited Report AECL-11330.

Meyer, G.L. 1976. "Recent Experience with the Land Burial of Solid-Low-Level Radioactive Wastes", pp. 383-395 in: Management of Radioactive Wastes from the Nuclear Fuel Cycle Vol. II, International Atomic Energy Agency, Vienna (IAEA-SM-207/64).

Papoulis, A. 1962. *The Fourier Integral and its Applications*, McGraw-Hill, New York.

Rattan, D.S., J.H. Rowat, S.R. Wilkinson and F.E. Lane 1996a. "NSURE Code: Theory, Documentation and User's Guide", Atomic Energy of Canada Limited Report AECL-11675.

Rattan, D.S., F.E. Lane and L. Wojciechowski 1996b. "NSURE Code Verification", Atomic Energy of Canada Limited Technical Record TR-604.*

Rowat, J.H. and F.E. Lane 1994. "Vault Source Term for the IRUS Repository: Sensitivity to Mathematical Boundary Conditions", Atomic Energy of Canada Limited Technical Record TR-602.*

Rowat, J.H., W.N. Selander and S.R. Wilkinson 1991. "Analytical Source Term For A Low-Level Radioactive Waste Repository", Atomic Energy of Canada Limited Technical Record TR-510.*

Rubin, J., and R. Steinhardt 1963. "Soil Water Relations During Rain Infiltration: I. Theory", Soil Science Society of America Proceedings, **27**, pp. 246-251.

Selander, W.N., S.R. Wilkinson and J.H. Rowat 1991. "A Vault Mass Transfer Model for LLRW", Atomic Energy of Canada Limited Technical Record TR-509.*

Sullivan, T.M. 1991. "Selection of Models to Calculate the LLW Source Term", U.S. Nuclear Regulatory Commission Report NUREG/CR-5773.

Woods, B.L. 1996. "Chemical and Physical Properties of IRUS Buffer and Backfill Materials", Atomic Energy of Canada Limited Technical Record TR-661.*

---

* Unpublished report available from SDDO, Atomic Energy of Canada Limited, Chalk River Laboratories, Chalk River, Ontario K0J 1J0.

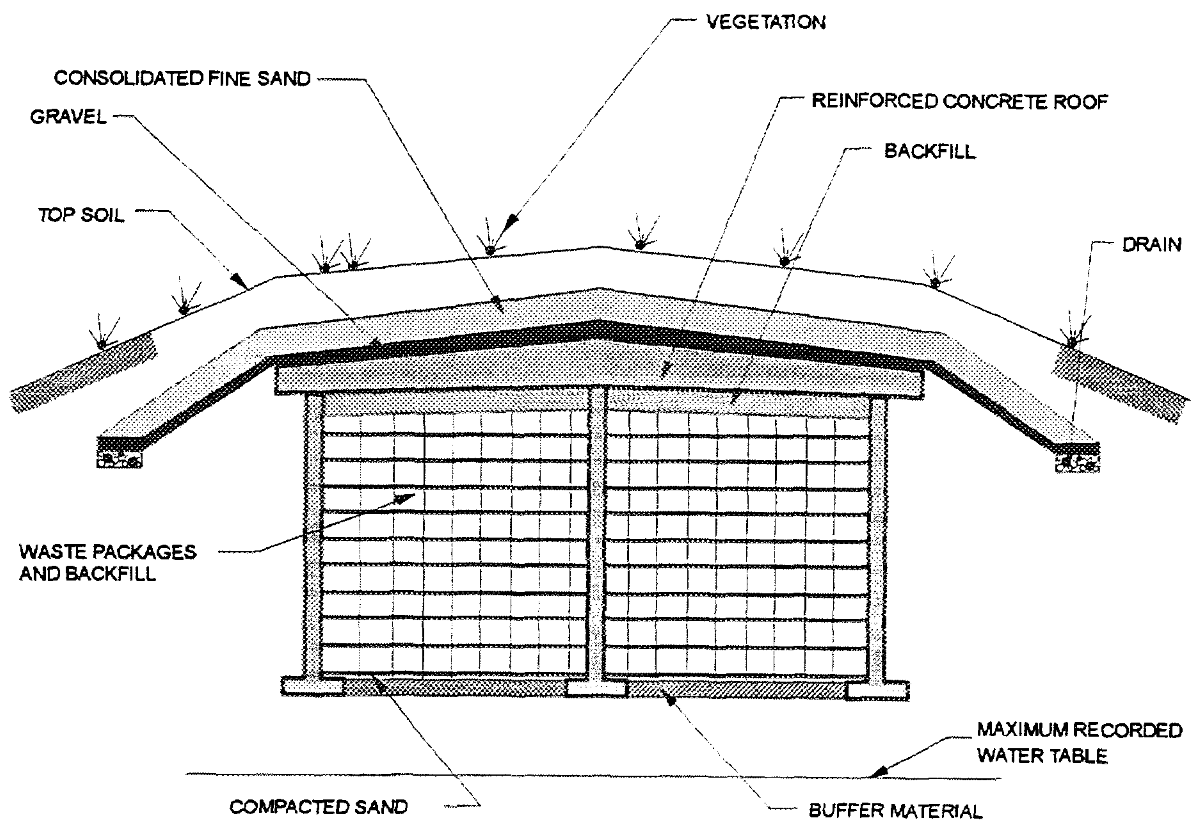

Figure 1: IRUS - transverse section of unit after closure.

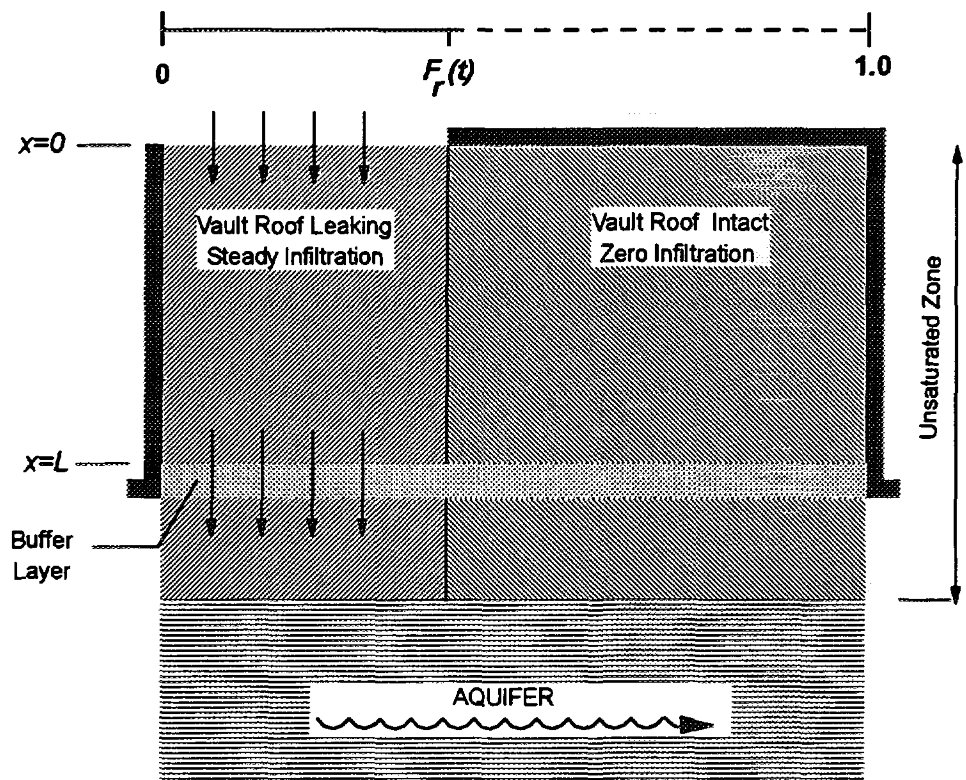

Figure 2: Vertical partitioning of the vault and unsaturated layers into zero- and steady-infiltration regions

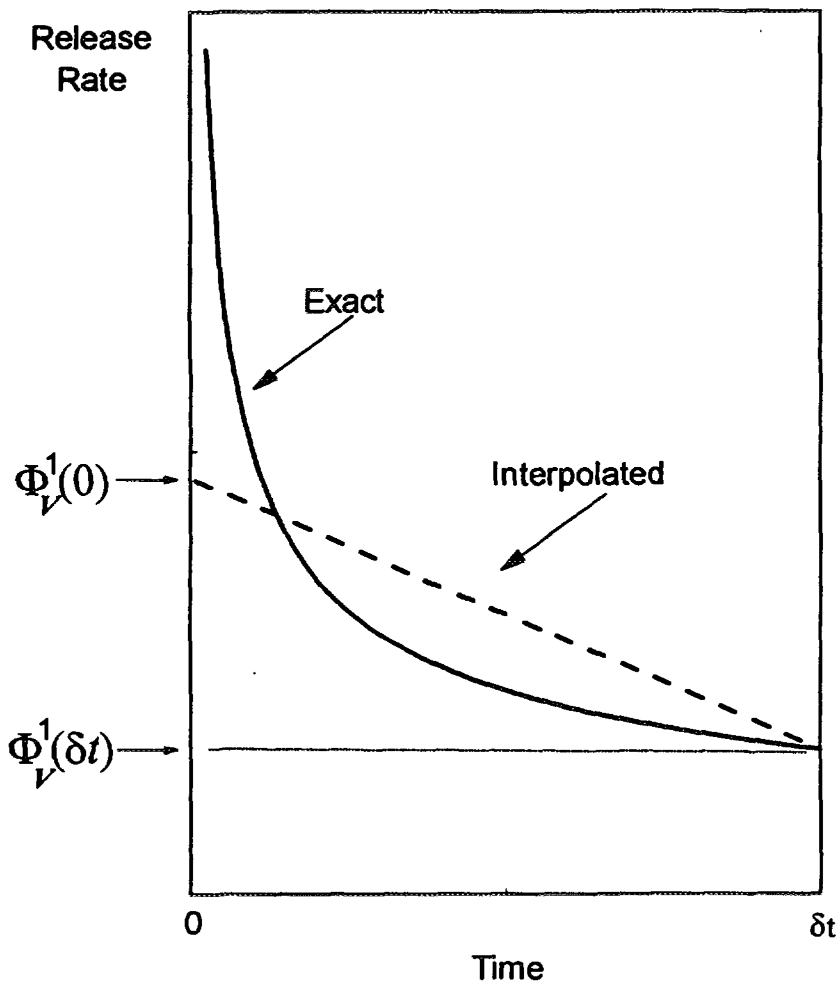

Figure 3: Method of interpolation for treating singularity in Φ_V^1 at $t = 0$.

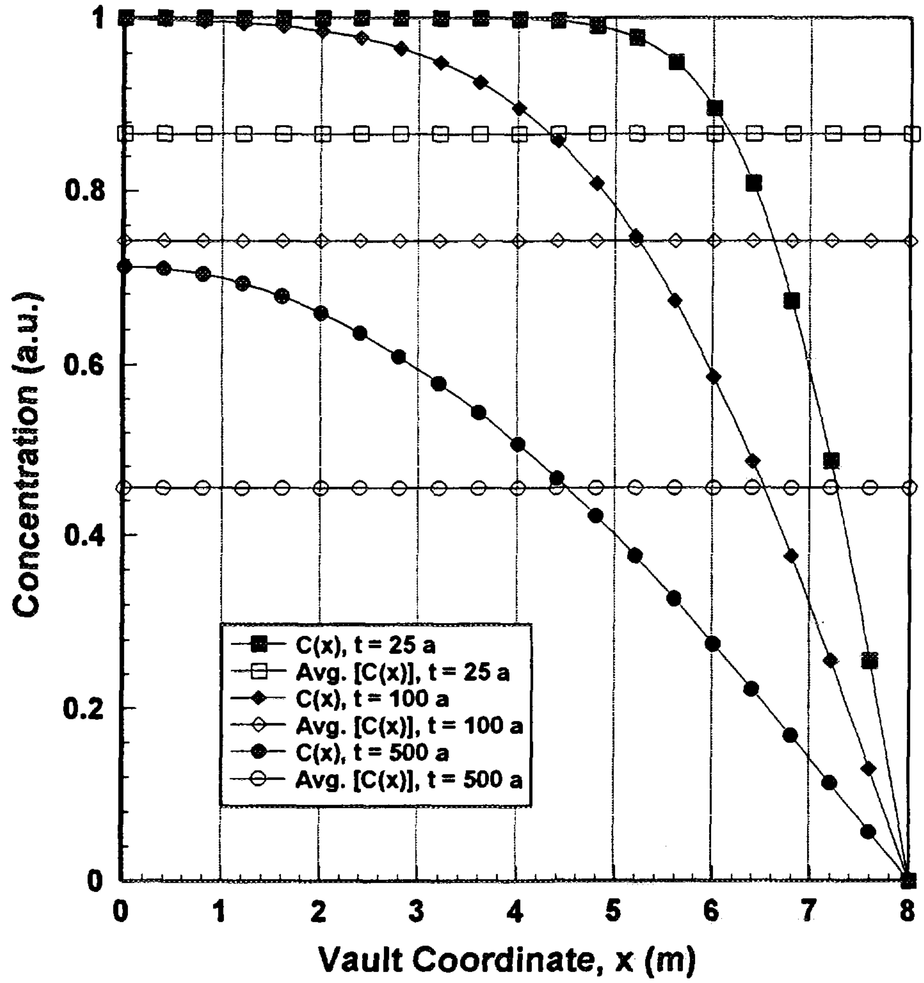

Figure 4: Spatially-averaged nuclide concentration profiles in a vault of depth of 8 m (arbitrary concentration units).

APPENDIX A

THE ADVECTION-DISPERSION EQUATION

A.1 INTRODUCTION

The purpose of this appendix is to define the terminology and notation used in the present report, and to describe how mass transfer is modeled in the unsaturated zone and in the aquifer beneath the vault.

The IRUS vault is to be located above the water table, hence, nuclide migration within the vault occurs in the unsaturated zone. A rigorous treatment of nuclide migration in the unsaturated zone requires simultaneous solution of two second-order partial differential equations: the flow equation, and the equation for nuclide mass transfer (Bear and Verruijt 1987). The flow equation is difficult to solve because it is nonlinear, and because it is necessary to determine soil-specific functions (the moisture characteristic and the unsaturated hydraulic conductivity). The flow calculation is further complicated because the configuration of the engineered system varies with time (i.e., the vault roof and waste containers are subject to time-distributed failure). Rather than solving the flow equation, the approach taken here for mass-transfer modeling, and one commonly used in field studies of contaminant migration, is to employ the advection-dispersion equation assuming a seasonally-averaged infiltration rate and a spatially-uniform moisture content, θ.

The time scales for nuclide migration are not days or weeks, rather years; therefore, it is reasonable to use a seasonally-averaged infiltration rate for mass-transfer calculations, rather than transient infiltration rates derived from detailed flow simulations. Wierenga (1977), Duguid and Reeves (1977) and Gelbard (1989) have shown this to be a good approximation. Regarding the assumption of uniform moisture content, Cassel et al. (1975) have compared numerical solutions of the advection-dispersion equation using depth-averaged and depth-varying moisture content. Good agreement was found between experimental data and the solutions based on both the depth-averaged (i.e., spatially uniform) and depth-varying moisture content. The choice of a particular value for the average moisture content is somewhat arbitrary; however, it is usually set equal to the field capacity of the soil (Till and Meyer 1983, Wierenga 1977). Other methods for selecting an average moisture content have been discussed by Enfield et al. (1982) and Hamlen and Kachanoski (1992).

A.2 SORPTION IN UNSATURATED MEDIA

There are few experimental data that pertain specifically to sorption in the unsaturated zone; batch equilibration or flow-through column measurements using water-saturated soil samples are the norm. All other things being equal, sorption in the saturated and unsaturated zones differs by the specific contact area between the soil particles and the pore water (i.e., the surface area of the solid "seen" by a unit volume of pore water); the specific contact area will depend on the pore structure of the medium and the moisture content. Because sorption occurs at the interface of the aqueous and solid phases, larger interfacial areas result in a greater sorption capacity, and may result in larger distribution coefficients.

Most authors assume that the degree of saturation has no effect on sorption processes (Nielsen et al., 1986). This is the conventional assumption, and is the one adopted here. Furthermore, it is assumed that sorption follows a linear equilibrium isotherm, that is:

$$s = K_d C \tag{A-1}$$

where K_d is the partition coefficient ($m^3 \cdot kg^{-1}$), C is the pore-water concentration ($Bq \cdot m^{-3}$) and s is the sorbed concentration ($Bq \cdot kg^{-1}$). The advantages and limitations of the K_d model for performance assessment applications have been discussed by Serne (1992).

A.3 THE ADVECTION-DISPERSION EQUATION

The average pore-water velocity U ($m \cdot a^{-1}$) in the unsaturated zone is calculated as:

$$U = q/\theta \tag{A-2}$$

where q ($m \cdot a^{-1}$) is the specific discharge. Because the moisture content is assumed to be uniform, the specific discharge is also uniform and is equal to the seasonally-averaged infiltration rate (Hillel 1980).

In the literature, two pore-water velocities are discussed frequently: the average pore-water velocity U as defined by Eq. (A-2), and the solute (or apparent) velocity, U_s. The solute velocity is the velocity at which a nonsorbing solute front would move through the medium. It is assumed here that U and U_s are identical. Although they are usually identical, U and U_s can sometimes differ. In certain soil types (e.g., clays) anions can migrate faster than the average pore-water velocity, so that $U_s > U$. This phenomena, called anion-exclusion, occurs because repulsion between anions and negatively charged pore surfaces tends to force anions into pore centers where the pore-water velocity is higher. In sands, anion exclusion has been found to be negligible (Gvirtzman and Gorelick 1991), and hence is not expected to affect nuclide migration rates in the sand overburden at CRL. Gelhar et al. (1985) have reported a "solute-lag" effect, which has $U > U_s$; this effect, however, has not been extensively studied.

Solute dispersion in unsaturated media follows Fick's law, which is

$$J = -\theta D(\theta) \, \partial C/\partial x \tag{A-3}$$

where J is the solute flux ($Bq \cdot m^{-2} \cdot a^{-1}$) and D is the dispersion coefficient ($m^2 \cdot a^{-1}$). Studies of dispersion in unsaturated media show that the dispersion coefficient has the form (Yule and Gardner 1978, Gelhar et al. 1985):

$$D(\theta) = D_p(\theta) + \alpha U \tag{A-4}$$

where $D_p(\theta)$ is the coefficient of molecular diffusion and the product αU is the coefficient of mechanical mixing. The dispersion length, α (m), is an experimental constant that depends on the scale of the system, the pore structure of the medium and the moisture content. For the IRUS vault model, no credit will be taken for mechanical mixing, hence, the parameter α is not required for the source-term calculation.

For saturated porous media the coefficient of molecular diffusion is often written as:

$$D_p = D_0 / \tau^2 \tag{A-5}$$

where D_0 is the "free-water" diffusion coefficient ($m^2 \cdot a^{-1}$) and τ is the tortuosity. The free-water diffusion coefficient varies from about 0.03 to 0.07 $m^2 \cdot a^{-1}$, and the tortuosity for saturated sand is about 1.2 (Marsily 1986, pg. 233). The same expression for D_p is sometimes used for unsaturated media; however, the tortuosity factor is adjusted to reduce the diffusion coefficient in the unsaturated zone compared with the saturated zone (e.g., Olague and Price 1991, McCarthy and Johnson 1995). Expressions that relate the diffusion coefficient to the moisture content have been developed; one expression that is frequently used is the Millington and Quirk relation (Jury 1988, Mott and Weber 1991):

$$1/\tau^2 = \theta^{7/3} / \varepsilon^2 \tag{A-6}$$

where ε is the porosity. Generally, $D_p(\theta)$ exhibits a gradual decline as the moisture content decreases. This is followed by a steep drop-off at very low moisture contents because of the loss of continuous water pathways.

The total mass flux in the unsaturated zone is (Bear and Verruijt 1987, Eq. (6.2.19):

$$J = -\theta \left(D(\theta) \frac{\partial C}{\partial x} + U(\theta) C \right) \tag{A-7}$$

Mass conservation requires that:

$$\frac{\partial}{\partial t} (\rho_b s + \theta C) = - \frac{\partial}{\partial x} J \tag{A-8}$$

Assuming a steady infiltration rate and a uniform moisture content, the governing equation for mass transfer in the unsaturated zone is:

$$R \frac{\partial C}{\partial t} = D(\theta) \frac{\partial^2 C}{\partial x^2} - U(\theta) \frac{\partial C}{\partial x} \tag{A-9}$$

where

$$R = 1 + \frac{\rho_b K_d}{\theta} \tag{A-10}$$

Equation (A-9) is the advection-dispersion equation for solute transport in the unsaturated zone.

A.4 MASS TRANSFER IN SATURATED MEDIA

Equation (A-9) is identical in form to the advection-dispersion equation for solute transport in saturated media. The coefficients of the advection-dispersion equation for saturated media are obtained by replacing the moisture content by the porosity; that is:

$$R = 1 + \frac{\rho_b K_d}{\varepsilon}$$

$$U = \frac{q}{\varepsilon} \tag{A-11}$$

The dispersion coefficient for saturated media is identical to Eq. (A-4) with D_p given by Eq (A-5) and U as in Eq. (A-11).

REFERENCES

Bear, J. and A. Verruijt 1987. *Modelling Groundwater Flow and Pollution*, D. Reidel Pub. Co., Dordrecht, Holland.

Cassel, D.K., M.Th. Van Genuchten and P.J. Wierenga 1975. "Predicting Anion Movement in Disturbed and Undisturbed Soils", Soil Sci. Soc. Am. J., **39**, pp. 1015-1019.

Duguid, J.O. and M. Reeves 1977. "A Comparison of Mass Transport Using Average and Transient Rainfall Boundary Conditions", in: Finite Elements in Water Resources, Eds. W.G. Gray et al., Lentech Press, London.

Enfield, C.G., R.F. Carsel, S.Z. Cohen, T. Phan, and D.M. Walters 1982. "Approximating Pollutant Transport to Ground Water", Ground Water, **20**, pp. 711-722.

Gelbard, F. 1989. "Modeling One-Dimensional Radionuclide Transport Under Time-Varying Fluid-Flow Conditions", U.S. Nuclear Regulatory Commission Report NUREG/CR-5412.

Gelhar, L.W., A. Mantoglou, C. Welty and K.R. Rehfeldt 1985. "A Review of Field-Scale Physical Solute Transport Processes in Saturated and Unsaturated Porous Media", Electric Power Research Institute Report EPRI EA-4190.

Gvirtzman, H. and S.M. Gorelick 1991. "Dispersion and advection in unsaturated porous media enhanced by anion exclusion", Nature, **352**, pp. 793-795.

Hamlen, C.J. and R.G. Kachanoski 1992. "Field Solute Transport Across a Soil Horizon Boundary", Soil Sci. Soc. Am. J., **56**, pp. 1716-1720.

Hillel, D. 1980. *Applications of Soil Physics*, Academic Press, New York.

Jury, W.A. 1988. "Behavior of Organic Contaminants in Soil", pp. 75-81 in: *Contaminated Soil '88*, Eds. K. Wolf, W.J. van den Brink and F.J. Colon, Kluwer Academic Publishers, Dordrecht, Holland.

Marsily, G. de 1986. *Quantitative Hydrogeology*, Academic Press, San Diego, California.

McCarthy, K.A. and R.L. Johnson 1995. "Measurement of Trichloroethylene Diffusion as a Function of Moisture Content in Sections of Gravity-Drained Soil Columns", J. Environ. Qual., **24**, pp. 49-55.

Mott, H.V. and W.J. Weber Jr. 1991. "Factors Influencing Organic Contaminant Diffusivities in Soil Bentonite Cutoff Barriers", Environmental Science and Technology, **25**, pp. 1708-15.

Nielsen, D.R., M.Th. Van Genuchten and J.W. Biggar 1986. "Water Flow and Solute Transport Processes in the Unsaturated Zone", Water Resources Research, **22**, pp. 89S-108S.

Olague, N.E. and L.L. Price 1991. "A First Approximation for Modelling the Liquid Diffusion Pathway at the Greater Confinement Disposal Facilities", Waste Management'91 Vol. 2, R.G. Post (Ed.), pp. 525-531.

Serne, R.J. 1992. "Conceptual Adsorption Models and Open Issues Pertaining to Performance Assessment", pp. 237-282 in: *Radionuclide Sorption from the Safety Evaluation Perspective*, OECD Nuclear Energy Agency, Paris.

Till, J.E. and H.R. Meyer 1983. *Radiological Assessment, A Textbook on Environmental Dose Analysis*, U.S. Nuclear Regulatory Commission Report NUREG/CR-3332.

Wierenga, P.J. 1977. "Solute Distribution Profiles Computed With Steady-State and Transient Water Movement Models", Soil Sci. Soc. Am. J., **41**, pp. 1050-1055.

Yule, D.F and W.R. Gardner 1978. "Longitudinal and Transverse Dispersion Coefficients in Unsaturated Plainfield Sand", Water Resources Research, **14**, pp. 582-588.

APPENDIX B

CLOSED-FORM EXPRESSIONS FOR $\Phi_V^1(t)$

The expression for $\Phi_V^1(t)$ that is implemented in SYVAC3-NSURE Ver. 1.03 is slightly different from Eq. (15). The two-term approximation for $\Phi_V^1(t)$ that has been implemented in NSURE is:

$$\Phi_V^1(t) = N\left\{\frac{2D}{L^2R}\exp\left(-\frac{D}{R}\omega_0^2 t\right) + \frac{1}{\sqrt{t}}\left(1-\frac{8}{\pi^2}\right)\left[\frac{D\omega_1^2}{R\pi}\right]^{1/2}\exp\left(-\frac{D}{R}\omega_1^2 t\right)\right\} \qquad \text{(B-1)}$$

or

$$\Phi_V^1(t) = N\left\{\frac{2D}{L^2R}\exp\left(-\frac{D}{R}\omega_0^2 t\right) + \frac{0.893}{\sqrt{t}}\sqrt{\frac{D}{\pi RL^2}}\exp\left(-9\frac{D}{R}\omega_0^2 t\right)\right\} \qquad \text{(B-2)}$$

The approximate CFR corresponding to Eq. (B-1) is:

$$\chi_1(t) = \frac{8}{\pi^2}\left[1-\exp\left(-\frac{D}{R}\omega_0^2 t\right)\right] + \left(1-\frac{8}{\pi^2}\right)\operatorname{erf}\left(\sqrt{\frac{D}{R}\omega_1^2 t}\right) \qquad \text{(B-3)}$$

The vault roof is assumed to fail and admit infiltration between about 500 and 2000 years after closure. Hence, for practical purposes, the time series representing Φ_V^1 and χ_1 are only calculated for times less than about 2000 years. Figures B-1 and B-2 show plots of the two-term and exact time series for Φ_V^1 and χ_1 out to 2000 years for a range of R values, assuming $D = 0.03$ $m^2 \cdot a^{-1}$ and $L = 8$ m. The two-term approximations match the exact solution closely for all values of R. When R is large, the differences would be of no consequence, because only a small fraction of the total inventory would be released by diffusion.

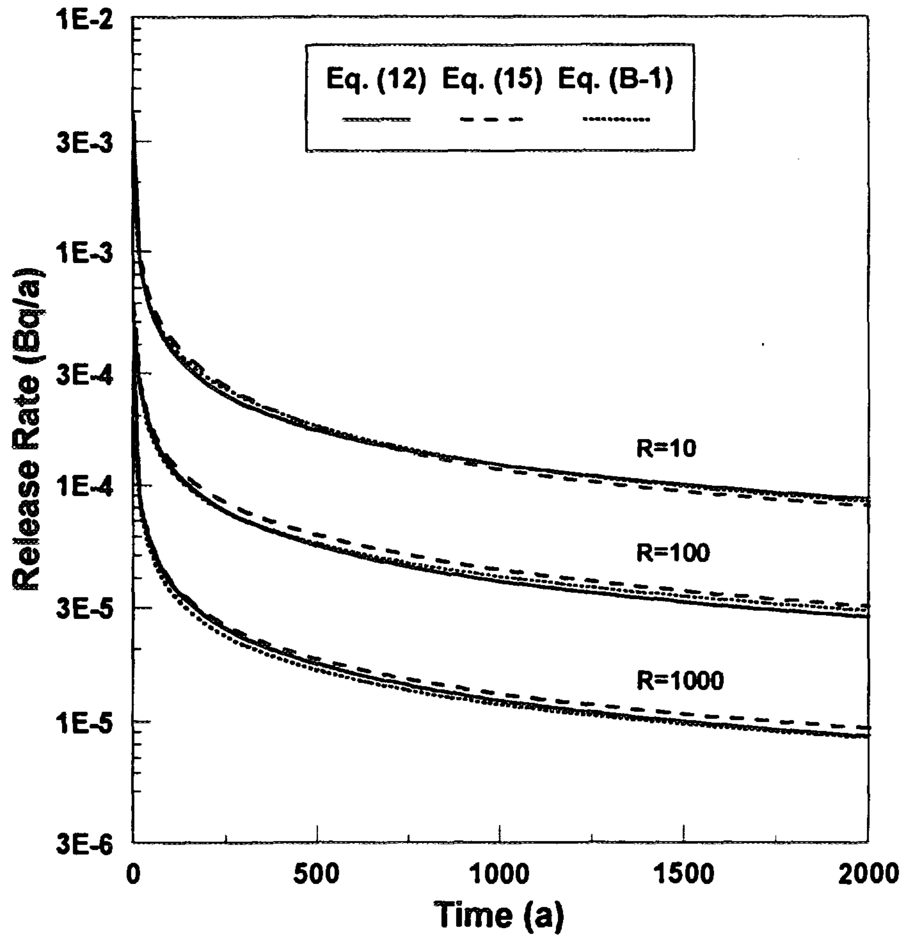

Figure B-1: Plots of exact formula (Eq. (12)) and two-term approximations for Φ_V^1.

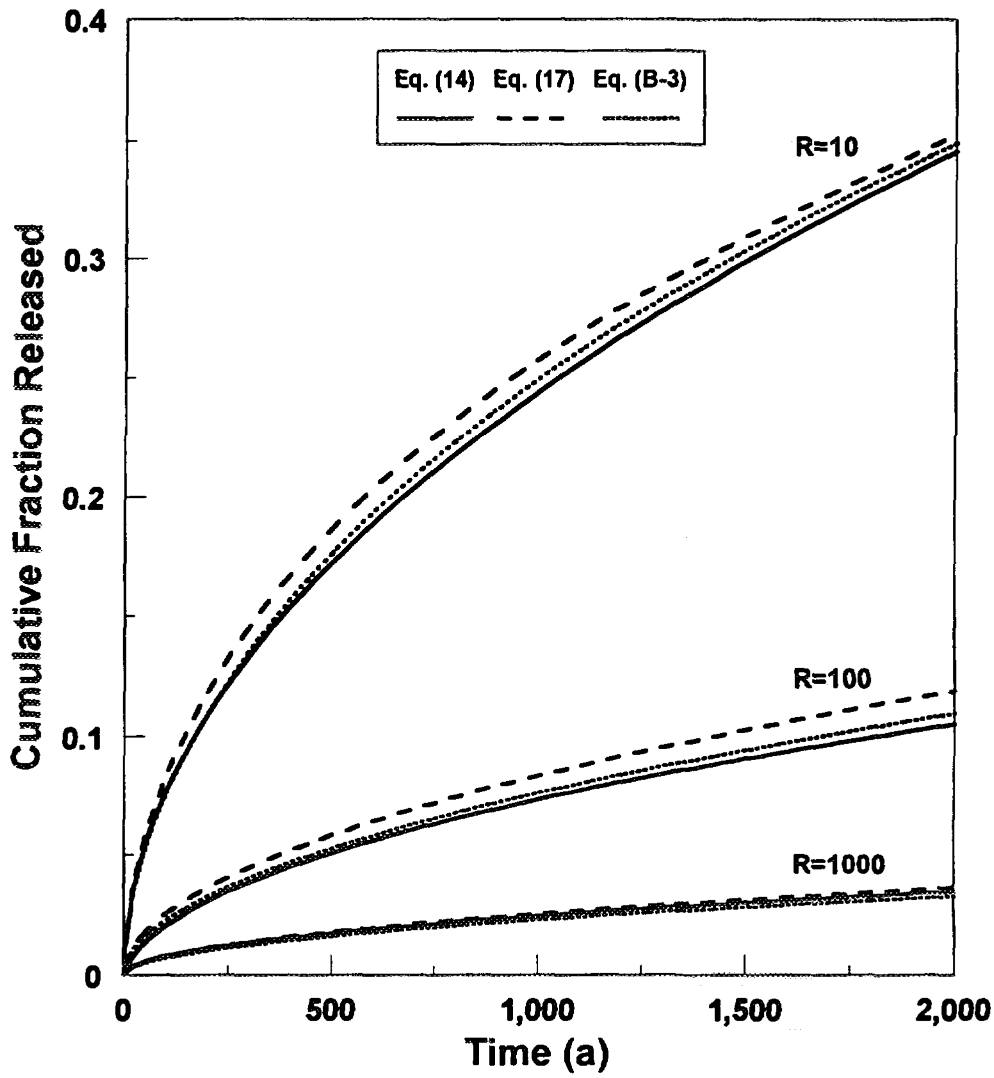

Figure B-2: Plots of exact formula (Eq. (14)) and two-term approximations for $\chi_1(t)$.

APPENDIX C

MULTILAYER MASS TRANSFER IN THE UNSATURATED ZONE AND LAKE 233 AQUIFER

C.1 MULTILAYER MASS TRANSFER

This appendix describes the model for mass transfer in the downstream links in the IRUS groundwater pathway, which are the unsaturated zone and Lake 233 aquifer beneath the IRUS vault.

Each link in the multilayer system of vault, unsaturated layers and aquifer is modelled using the 1D advection-dispersion equation. In principle, the mathematical model for mass transfer through a multilayer system requires solutions for a coupled set of equations. To avoid the mathematical and numerical complications associated with the coupled system, it is assumed that mass transfer in any given layer is not correlated with downstream layers, and that each layer is of semi-infinite extent. This amounts to the assumption that in a given layer nuclide travel times are not influenced by downstream layers. This approximation was introduced by Shamir and Harleman (1967), and has been found to provide a satisfactory description of multilayer mass transfer (Barry and Parker 1987, Dyson and White 1989).

The different layers are assumed to be characterized by piecewise constant parameters (i.e., each layer is assigned a set of parameters l_i, D_i, U_i and R_i). The expression for the release rate from the i^{th} layer is

$$\Phi_i(l_i,t) = \int_0^t \Phi_i(0,\tau)\, G(l_i, t-\tau)\, d\tau \qquad \text{where} \qquad \Phi_i(0,t) = \Phi_{i-1}(l_{i-1},t) \tag{C-1}$$

where:

$\Phi_i(0,t)$ = rate at which nuclides enter into the i^{th} layer at $x=0$ (Bq/a).

$\Phi_i(l_i,t)$ = rate at which nuclides are released from the i^{th} layer at $x=l_i$ (Bq/a).

$G(x,t)$ = Green's function for mass transfer in a semi-infinite medium.

l_i = length of i^{th} layer.

The Green's function for a semi-infinite medium, $G(x,t)$, is (Moltyaner 1983):

$$G(x,t) = \frac{x R_i^{1/2}}{\sqrt{4\pi D_i t^3}} \exp\left[-\frac{(R_i x - U_i t)^2}{4 D_i R_i t}\right] \tag{C-2}$$

where:

U_i = average pore-water velocity.

D_i = dispersion coefficient for i^{th} layer.
R_i = nuclide retardation factor for i^{th} layer.

The multilayer mass-transfer calculation represented by Eq. (C-1) is semianalytic because the convolution integral in Eq. (C-1) is evaluated numerically.

C.2 MASS TRANSFER IN THE UNSATURATED LAYERS BELOW THE VAULT

The unsaturated layers below the vault are treated as an extension of the vault. They are also modelled using the 1D advection dispersion equation, because edge effects should be small, given that the horizontal extent of the unsaturated layers (about 20 × 30 m) is much greater than their depth (1-2 m). Like the vault, the unsaturated layers are partitioned into a zero-flow region and a steady-flow region. Sections of the unsaturated layers that are below intact sections of the vault roof are assumed to have zero-flow conditions ($U_i = 0$); the sections below failed sections of the vault roof (i.e., roof sections admitting infiltration) are assumed to have steady-flow conditions.

Similar to the vault, the release rate for each of the unsaturated layers has three components. The nuclide migration rate in the zero-flow region is given by the diffusive component Φ_i^1. In the steady-flow region, the nuclide migration rate is the sum of an advective component Φ_i^2 and a wash-out component Φ_i^3.

The diffusive component, Φ_i^1, is calculated directly from Eq. (C-1) according to:

$$\Phi_i^1(l_i,t) = \int_0^t \Phi_i^1(0,\tau)\, G(l_i, t-\tau)\, d\tau \tag{C-3}$$

where

$$\Phi_1^1(0,t) = \varphi_1(t) \quad \text{and} \quad \Phi_i^1(0,t) = \Phi_{i-1}^1(l_{i-1},t) \text{ for } i > 1 \tag{C-4}$$

The advective component, Φ_i^2, is also calculated directly from Eq. (C-1), according to:

$$\Phi_i^2(l_i,t) = \int_0^t \Phi_i^2(0,\tau)\, G(l_i, t-\tau)\, d\tau \tag{C-5}$$

where

$$\Phi_1^2(0,t) = \varphi_2(t) + \varphi_3(t) \quad \text{and} \quad \Phi_i^2(0,t) = \Phi_{i-1}^2(l_{i-1},t) + \Phi_{i-1}^3(t) \text{ for } i > 1 \tag{C-6}$$

At the onset of infiltration, nuclides stored in the unsaturated layers will give rise to a wash-out component. The calculation for the wash-out component, Φ_i^3, requires an expression for the average concentration in the zero-flow region, which is

$$\bar{C}_i(t) = \frac{1}{l_i} \int_0^{l_i} C_i(\xi,t)\, d\xi = \frac{1}{l_i} \int_0^{l_i} \left\{ \int_0^t J_i(0,\tau)\, G_{fc}(\xi,t-\tau)\, d\tau \right\} d\xi \qquad \text{(C-7)}$$

where G_{fc} is the flux-to-concentration Green's function for the zero-flow region (Černík et al. 1994), and $J_i(x,t)$ is the radionuclide flux in the i^{th} layer. It has the form

$$G_{fc}(x,t) = \frac{1}{\theta_i} \frac{1}{\sqrt{\pi D_i R_i t}} \exp\left\{ \frac{-x^2 R_i}{4 D_i t} \right\} \qquad \text{(C-8)}$$

After some minor rearrangements it can be shown that

$$\bar{C}_i(t) = \frac{1}{l_i \theta_i R_i} \int_0^t J_i(0,t-\tau)\, \text{erf}\left(\frac{l_i}{\sqrt{4 D_i \tau / R_i}} \right) d\tau \qquad \text{(C-9)}$$

The diffusive flux out of the vault into the first of the unsaturated layers is needed to calculate $\bar{C}_i$ for each of the unsaturated layers. It is:

$$J_1(0,t) = \frac{L}{V} \int_0^t f_c(\xi)\, \Phi_V^1(t-\xi)\, d\xi \qquad \text{(C-10)}$$

The radionuclide flux in subsequent layers is calculated from Eq. (C-1), that is:

$$J_{i+1}(0,t) = J_i(l_i,t) = \int_0^t J_i(0,\tau)\, G(l_i,t-\tau)\, d\tau \qquad \text{for } i = 1, 2 \cdots \qquad \text{(C-11)}$$

If the entire roof were to fail at time T, then the wash-out release rate for layer i would be

$$\Phi_i^3(t) = \theta_i U_i \frac{V}{L} \bar{C}_i(T) \left[H(t-T) - H(t-T-\frac{L_i R_i}{U_i}) \right] \qquad \text{(C-12)}$$

When roof failure is gradual, then

$$\Phi_i^3(t) = \theta_i U_i \frac{V}{L} \int_0^t f_r(\tau)\, \bar{C}_i(\tau) \left[H(t-\tau) - H(t-\tau-\frac{L_i R_i}{U_i}) \right] d\tau = \theta_i U_i \frac{V}{L} \int_0^t f_r(t-\tau)\, \bar{C}_i(t-\tau)\, H(\frac{L_i R_i}{U_i}-\tau)\, d\tau \qquad \text{(C-13)}$$

C.3 MASS TRANSFER IN LAKE 233 AQUIFER

Klukas and Moltyaner (1995) have carried out detailed flow and mass-transfer modelling for the Lake 233 aquifer; they have shown that there is little seasonal variability in the groundwater flow velocity and practically no seasonal variability in the groundwater flow direction. Because of the relative simplicity of the local hydrology, it was found that the 1D advection-dispersion equation adequately describes mass transfer in the Lake 233 aquifer for performance assessment purposes.

The width of the contaminant plume in the aquifer is assumed to be the same as the effective inside width of the vault (18.3 m). The plume is assumed to be uniformly mixed over a cross-sectional area equal to the width times the thickness of the plume. No lateral dispersion of the aquifer flow is assumed in the model. The expression for mass transfer through the aquifer, as per Eq. (C-1), is

$$\Phi_A(l_A, t) = \int_0^t \Phi_A(0, \tau)\, G(l_A, t-\tau)\, d\tau \qquad \text{(C-14)}$$

where:

$$\Phi_A(0, t) = \Phi_M^1(t) + \Phi_M^2(t) + \Phi_M^3(t) \qquad \text{(C-15)}$$

and where l_A is the aquifer length and M is the index for the unsaturated layer adjacent to the aquifer.

REFERENCES

Barry, D.A. and J.C. Parker 1987. "Approximations for Solute Transport Through Porous Media with Flow Transverse to Layering", Transport in Porous Media, **2**, 65-82.

Černík, M., P. Federer, M. Borkovec and H. Sticher 1994. "Modeling of Heavy Metal Transport in a Contaminated Soil", Journal of Environmental Quality, **23**, pp. 1239-1248.

Dyson, J.S. and R.E. White 1989. "A Simple Predictive Approach to Solute Transport in Layered Soils", Journal of Soil Science, **40**, pp. 525-542.

Klukas, M.H. and G.L. Moltyaner 1995. "Numerical Simulations of Groundwater Flow and Solute Transport in the Lake 233 Aquifer", Atomic Energy of Canada Limited Report AECL-11330.

Moltyaner, G.L. 1983. "Migration of Radionuclides in Unconsolidated Materials: Equilibrium Transport Models", Atomic Energy of Canada Limited Report AECL-8253.

Shamir, U.Y., and D.R.F. Harleman 1967. "Dispersion in Layered Porous Media", Proc. Am. Soc. Civil Eng. Hydr. Div., **93**, 236-260.

Cat. No. /No de cat.: CC2-11321E
ISBN 0-660-16703-4
ISSN 0067-0367

To identify individual documents in the series, we have assigned an AECL- number to each.

Please refer to the AECL- number when requesting additional copies of this document from:

Scientific Document Distribution Office (SDDO)
AECL
Chalk River, Ontario
Canada K0J 1J0

Fax: (613) 584-1745 Tel.: (613) 584-3311 ext. 4623

Price: A

Pour identifier les rapports individuels faisant partie de cette serie, nous avons affecté un numéro AECL- à chacun d'eux.

Veuillez indiquer le numéro AECL- lorsque vous demandez d'autres exemplaires de ce rapport au:

Service de Distribution des Documents Officiels
EACL
Chalk River (Ontario)
Canada K0J 1J0

Fax: (613) 584-1745 Tél.: (613) 584-3311 poste 4623

Prix: A