A MATHEMATICAL MODEL FOR EVALUATING THE SUITABILITY OF A LOW-LEVEL RADIOACTIVE WASTE SITE

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ABSTRACT

A mathematical model intended to study the one-dimensional transport of radionuclides in a non-homogeneous soil system under saturated-unsaturated and isothermal conditions is presented. The model is composed of two modules: The first to calculate the pressure distribution, enabling one to compute velocities and soil moisture; the second to calculate the migration of species by considering the major processes associated with the transport of a dissolved substance in porous media, i.e., advection, mechanical dispersion, molecular diffusion, radioactive decay, and sorption, assuming a linear equilibrium isotherm. The numerical method of solving both flow and solute equations used here is the finite-element method based on the weighted residual technique. The flow equation is solved by the Bubnov-Galerkin method. The solute equation is solved by a Petrov-Galerkin type method. The model allows for a variety of boundary conditions; e.g., infiltration, drainage and/or evaporation. A test case involving the movement of a non-reacting ionic species is used to validate the model. Use of the model is illustrated by the analysis of the movement Sr-90 and Cs-137 and H-3 (as water) from a low-level solid-waste disposal trench subject to a steady rate of rainfall.

INTRODUCTION

Shallow land burial has been generally accepted for isolating solid low-level radioactive waste (LLW). This type of isolation concept consists of placing the waste in trenches above the water table of unconfined aquifers and covering it with a few meters of excavated fill and/or compacted clay. Engineering techniques used

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in the covering and sealing of the trenches are being improved. These practices vary according to the hydrogeological and climatological conditions at the disposal site location.

When surface water or groundwater breaches a repository, radionuclides will be released through leaching, thus posing a potential threat to the quality of groundwater. To assess the long-term containment integrity of a burial site, one must select from among the engineering alternatives the ones that will be most successful in minimizing the threat of groundwater contamination.

One reliable approach for solving such a complex problem is the application of a mathematical model for various release scenarios. There are two main points to consider. First, the physics of water movement in the saturated-unsaturated domain is well understood. Reasonably accurate numerical predictions may be obtained under these flow regimes if geohydrologic data are available. Second, the prediction of radionuclide migration requires an understanding of geochemical processes, such as ion-exchange and adsorption characteristics. The corrosion of the storage canisters must also be considered. Some of these problems are still the subjects of research, and thus several simplifications of the sorption, dilution and corrosion mechanisms are necessary. In this paper we assess the potential impact on groundwater quality resulting from the release of the fission products Sr-90, Cs-137 and H-3 (as water) from a low-level solid-waste disposal trench.

A typical release scenario related to the influence of precipitation on the leaching of solid waste is investigated at a hypothetical site where the annual precipitation rate exceeds the evaporation rate. The calculations were based on a normalized concentration since the dissolution rates of the source are not known.

GOVERNING EQUATIONS

The continuity equation describing transient saturated-unsaturated vertical flow of water, assumed compressible, in the vadose zone of a slightly deformable unconfined aquifer, in the absence of sources and sinks and under isothermal conditions, may be written as

$$\Lambda(h) = (c_m + S_w S_s) \frac{\partial h}{\partial t} - \frac{\partial}{\partial z} [K \frac{\partial h}{\partial z} + K] = 0, \qquad (1)$$

where Λ is the differential operator, z is the vertical Cartesian coordinate (L), t is time (T), c_m is soil moisture capacity defined as ϕ $(\partial S_w/\partial h)(L^{-1})$, S_w is soil-water saturation θ/ϕ $(0 \leq S_w \leq 1)$, θ is volumetric moisture content, ϕ is porosity, S_s is specific storage coefficient (L^{-1}), $K = K_r K^s$ is hydraulic conductivity (LT^{-1}), K_r is relative hydraulic conductivity $(0 \leq K_r \leq 1)$, K^s is the saturated hydraulic conductivity (LT^{-1}), and h is soil-water pressure head (L).

In the derivation of Eq. (1), the specific discharge relative to the solid particles is assumed to be given by Darcy's law. The solution of Eq. (1) will permit computation of the soil-water content and specific discharge parameters needed to solve the solute-transport equation.

From the mass conservation requirement, the hydrodynamic dispersion equation describing the migration of a solute γ in a liquid phase α through a sorbing saturated-unsaturated porous medium s may be written as

$$\Lambda(C) = \frac{\partial}{\partial t}(\theta C + \theta_s \rho_s S) - \frac{\partial}{\partial z}\left[\theta D \frac{\partial C}{\partial z} - C\bar{q}\right] - Q' = 0, \qquad (2)$$

where C is concentration (ML^{-3}) of solute γ in the liquid phase, S is concentration of the solute γ in the absorbed phase (M^0), ρ_s is soil bulk density (ML^{-3}), $\bar{q}$ is the specific discharge (LT^{-1}), θ_s is volumetric fraction of the solid phase $(1 - \phi)$, D is coefficient of hydrodynamic dispersion (L^2T^{-1}), and Q' is the rate of production or removal of solute due to radioactive decay ($ML^{-3}T^{-1}$).

The general form of D for an isotropic media may be written after Bear[1] as

$$D = a_1 |V| + D_d \tau , \qquad (3)$$

where V is the average apparent interstitial fluid velocity (q/θ), a_1 is the longitudinal dispersivity of the medium (L), D_d is the molecular diffusion coefficient (L^2T^{-1}), and τ is tortuosity (L^0).

The mechanism of adsorption may be adequately described by a linear equilibrium isotherm, $S = K_d C$, where K_d is the distribution coefficient (L^3M^{-1}) of solute γ.

The instantaneous rate of removal Q' of a typical radionuclide in a sorbing porous medium in both dissolved and solid states may be written as:

$$Q' = -\lambda(\theta C + \theta_s \rho_s S), \qquad (4)$$

where λ is the radioactive decay constant (T^{-1}).

With the above considerations, the differential equation describing the movement of a radionuclide, i, leaching from a trench at a solid-radioactive-waste disposal site is given by:

$$\Lambda(C_i) = \frac{\partial}{\partial t}(\theta R_{d_i} C_i) - \frac{\partial}{\partial z}\left(\theta D \frac{\partial C_i}{\partial z} - C_i \bar{q}\right) + \lambda_i \theta R_{d_i} C_i = 0, \qquad (5)$$

where $R_d = 1 + [(1 - \phi)/\theta]\rho_s K_d$, is the retardation factor embodying the overall effect of the sorption/desorption reactions.

INITIAL AND BOUNDARY CONDITIONS

A general set of initial and boundary conditions for the system of Eqs. (1) and (5) is given by

$$h(z,0) = h_o \qquad ; 0 \leq z \leq L \qquad (6a)$$

$$h(z,t) = \tilde{h} \qquad ; z = 0 \quad \text{or} \quad z = L \qquad (6b)$$

$$-K\left(\frac{\partial h}{\partial z} + 1\right) + V_i = 0 \qquad ; z = 0 \qquad (6c)$$

$$C(z,0) = C_o \qquad ; 0 \leq z \leq L \qquad (7a)$$

$$C(0,t) = \tilde{C} \qquad (7b)$$

$$\theta D \frac{\partial C}{\partial z} = 0 \qquad ; z = 0 \quad \text{or} \quad z = L \qquad (7c)$$

$$\left(-\theta D \frac{\partial C}{\partial z} + C\bar{q}\right)_+ = (\tilde{C}\bar{q})_- \qquad ; z = 0 \qquad (7d)$$

where V_i corresponds to the external value of the imposed fluxes (precipitation, evaporation), and L is the thickness of the vadose zone. Subscripts + and - in Eq. (7d) denote the two sides of the boundary between the flow region and the external domain respectively. In the above equations, h_o, $\tilde{h}$, V, C_o, $\tilde{C}$ and $\bar{q}$ are all known functions of position and/or time. If no boundary conditions are specified at Z = 0, then an impermeable surface condition is automatically assumed, i.e., V_i=0. Note that at the boundary in contact with the atmosphere, conditions may change in time Eq. (6b) to Eq. (6c) (i.e., Dirichlet to Neumann, or vice versa).

Equations (6b) and (7b) correspond to Dirichlet (or first type) boundary conditions, Eqs. (6c) and (7c) to Neumann (or second type) boundary conditions and Eq. (7d) to Cauchy (or third type) boundary conditions.

FINITE ELEMENT FORMULATION

The spatial discretization of Eqs. (1) and (5) in the solution domain R is obtained through a finite element technique[2] based on the method of weighted residuals.

An approximate solution is sought through an orthogonalization process[3] written as

$$\int_R w_i L(\hat{\psi})dR = 0 \qquad i = 1, 2, \ldots, \Omega \ . \qquad (8)$$

where w_i are the weighting (or test) functions, ψ is the unknown function, and Ω is the total number of nodes.

The weighted and integrated residuals resulting from the substitution of Eqs. (1) and (5) in Eq. (8) and application of Green's theorem yield two sets of simultaneous equations. An approximation for the coefficients of h and C has also been considered to accommodate the spatial variation of these parameters within an element.

Flow Equation

The matrix form of the flow equation (Eq. 1) may be written as

$$[M]\{h\}+[P]\ \{\frac{dh}{dt}\} + \{G\} = 0 \quad , \qquad (9)$$

in which typical matrix elements are

$$M^e_{mn} = \int_{R^e} \sum_{I=1}^{\Omega_k} N_I K_{rI}\ K^s\ \frac{\partial N_m}{\partial z} \frac{\partial N_n}{\partial z}\, dR^e \quad , \qquad (10a)$$

$$P^e_{mn} = \int_{R^e} \sum_{I=1}^{\Omega_k} N_I (c_m + S_w S_s)_I\ N_m N_n dR^e \quad , \text{ and} \qquad (10b)$$

$$G^e_m = \int_{R^e} \sum_{I=1}^{\Omega_k} N_I K_{rI}\ K^s\ \frac{\partial Nm}{\partial z}\, dR^e$$

$$- \ N_m \sum_{I=1}^{\Omega_k} N_I K_{rI} \left(K^s\ \frac{\partial N_n}{\partial z}\ h_n + 1\right) \qquad (10c)$$

where Ω_k is the number of nodes in a particular element e. In this instance, the weighting (or test) functions (see Eq. 8) have been taken to correspond to the shape (or trial) functions ($w_i = N_i$), and the standard Galerkin (or Bubnov-Galerkin) type finite element method (FEM) is obtained.

A mass lumping procedure of the coefficients of the time derivative term, as suggested by Hinton et al.,[4] was used to improve the accuracy of the solution of Eq. (9).

Solute Equation

The matrix equation related to a typical species (Eq. 5) may be written as

$$[B]\{C\} + [E]\ \{\frac{dC}{dt}\} + \{F\} = 0 \quad , \qquad (11)$$

in which typical matrix elements are

$$B^e_{mn} = \int_{R^e} \left[\sum_{I=1}^{\Omega_k} N_I(\theta D)_I \frac{\partial W_m}{\partial z} \frac{\partial N_n}{\partial z} - \sum_{I=1}^{\Omega_k} (N_I q_I) \frac{\partial W_m}{\partial z} N_n + \sum_{I=1}^{\Omega_k} N_I (\lambda \theta R_d)_I N_m N_n \right] dR^e \, , \qquad (12a)$$

$$E^e_{mn} = \int_{R^e} \sum_{I=1}^{\Omega_k} N_I(\theta R_d)_I N_m N_n dR^e \quad \text{, and} \qquad (12b)$$

$$F^e_m = - W_m \left(\theta D \frac{\partial C}{\partial z} - C\bar{q}\right) \, . \qquad (12c)$$

With the exception of the second member of Eq. (5), described as the dispersive-convective component, which is weighted with asymmetric quadratic test functions (i.e., $w_i = W_i$), all of the terms of this equation are weighted with the usual basis functions. In this instance a Petrov-Galerkin type finite element method[5] yielding an optimally 'upwind' convective term is obtained.

Specific Discharge

The velocity at each node of the flow region is computed in a manner similar to that in Oden's conjugate function theory[6] using the same orthogonalization procedure as before. The matrix equation in this case may be written as

$$[D] \{q_x\} + \{Q\} = 0 \, . \qquad (13)$$

where

$$D^e_{mn} = \int_{R^e} N_m N_n dR^e \quad \text{, and} \qquad (13a)$$

$$Q^e_m = \int_{R^e} \sum_{I=1}^{\Omega_k} N_I K_{rI} K^s N_m \frac{\partial N_n}{\partial z} H_n dR^e \, . \qquad (13b)$$

where H is the hydraulic potential (i.e., h + z). Note that q is evaluated at each step of the computation once the values of h are computed.

ELEMENT FORMULATION

Isoparametric linear elements were used to discretize the flow region.

The shape (trial) function[2] for the ith node of a linear element with corner nodes $\xi_i = \pm 1$, is given by

$$N_i(\xi) = \frac{1}{2}(1+\xi\xi_i) \quad , \qquad \xi_i = \pm 1 \quad . \tag{14}$$

The weighting functions, which are used in the solution of the solute equation, are quadratic and have a non-symmetric form.[7,8] These may be written as

$$W_i(\xi) = N_i(\xi) + \alpha_{ij}\,\frac{3\xi_i(1-\xi^2)}{4} \quad , \quad \xi_i = \pm 1 \; , \tag{15}$$

and α_{ij} is the upstream parameter (scalar quantities associated with nodes i and j of a typical element), the sign of which is dependent on the direction of the average velocity along the element. Note that the test and trial functions coincide when $\alpha = 0$.

Details regarding the numerical mapping from the local coordinates to the global coordinates may be found in Reference 2.

The optimal value of the upstream parameter α as suggested by Huyakorn and Nilkuha[9] is approximated[10] by the following expressions:

$$\alpha = 0 \quad , \qquad Cr < 0.2 \quad , \tag{16a}$$

$$\alpha = 0.0545 \exp 2.797(1 - e^{-1.056Cr}), \; 0.2 \le Cr \le 1.6, \text{ and} \tag{16b}$$

$$\alpha = 1 \quad , \qquad Cr > 1.6 \quad , \tag{16c}$$

in which Cr is the cell Courant number, a critical non-dimensional parameter defined as

$$Cr = \left| \frac{V'\Delta t}{\Delta z} \right| \; , \tag{17}$$

where Δz is the length of a typical element, Δt is the time increment, and V' is the average velocity along the element written as

$$V'_{ij} = (V_i + V_j)/2 \; . \tag{18}$$

In this work the positive sign convention of V' was one in the direction of the vertical axis.

Note that when a typical species reacts with the soil matrix, V' in Eq. (17) will be reduced to $V^{*'}$ where $V^{*'} = V'/R_d$.

SOLUTION SCHEMES

Flow Equation

Equation (9) defines a set of Ω differential equations with non-linear coefficients. Subject to its boundary conditions Eq. (6), Eq. (9) is then linearized by under-relaxing the coefficient matrices [M] and [P], which are evaluated at half the time step $(t + \Delta t/2)$ and solved by an iterative technique using an implicit backward difference scheme which guarantees unconditional stability.[11,12]

In the steady-state case, Eq. (9) becomes elliptical in form. An iterative technique is used after linearizing and solving Eq. (9) subject to its boundary conditions (Eqs. 6b-c). The procedure is continued until the convergency criterion is met. This is written as

$$\left|h_k - h_{k-1}\right| = \varepsilon_1\left|h_{k-1}\right| + \varepsilon_2 \quad , \quad k > 1 \quad , \tag{19}$$

where k is the iteration index and ε_1 and ε_2 correspond to the relative and absolute error, respectively.

Empirical fitted equations,[13] mathematically defined as hyperbolic functions, are adopted in this study; to simulate the soil characteristic curves these may be expressed as

$$K_r = \frac{a}{b + |h|^m} \, , \; S_w = \frac{c}{d + |h|^n} \quad , \tag{20}$$

where a, b, c, d, m and n are constants.[b] The constants in Eq. (20) are conveniently obtained through a least-squares fit performed on the available set of experimental data.[14]

Solute Equation

Equation (11) defines a set of Ω linear differential equations. In this instance the Crank-Nicolson scheme centered in time was used to solve this equation subject to its initial and boundary conditions, Eqs. (7a-d).

A Gaussian elimination algorithm operating only on the band portion of the coefficient of h and C was used to solve Eqs. (9) and (11), respectively.

RESULTS AND DISCUSSIONS

The accuracy of the mathematical model was tested on a typical problem for which experimental data were available.

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[b]Dimensions of a and b are L^m; dimensions of c and d are L^n.

problem for which experimental data were available.

The performance of the model in prediction of water and solute transport of a conservative ion (Cl^-) in an unsaturated homogeneous soil was evaluated by comparing our results with the experimental ones reported by Warrick et al.[15] In that experiment, water and chloride were allowed to infiltrate an unsaturated homogeneous soil with the water table at a great depth. Figures 1a and 1b show a comparison of the moisture and concentration profiles for nine hours of infiltration. In both cases, the results show good agreement with the experimental values. In the solute case, a third type of boundary condition yields superior results.

Application of the Model to the Transport of Radionuclides

To evaluate the long-term performance of waste repositories by means of a mathematical model, an estimate of the concentrations of radionuclides at the source is desirable. In this work, the following assumptions were made: (a) the buried waste was simulated by soil with properties of a homogeneous coarse sand and a retardation factor of unity; (b) the activity per unit volume of waste for each fission product is known; (c) corrosion of the containers occurs at a uniform rate, $\beta(T^{-1})$, and total exposure of the waste to water is achieved within 50 years after burial; (d) the dissolution rate of the nuclides is proportional to the exposure of the solid to the water in contact with it; (e) solubility was assumed independent of pH and Eh. Thus $1m^3$ of soil of porosity ϕ and specific gravity ρ_b ($2.6\ g/cm^3$) corresponds to $\rho_b(1 - \phi) \times 10^6$ g of soil.

If d (mg/L) is the dissolution rate of an exposed representative sample of the waste subjected to a flow rate Q, and if we assume that the total activity of a particular nuclide in a unit volume of soil is $A(Ci/m^3)$ or $A\ 10^{-6}/(1 - \phi)\rho_b$ (Ci/g), the concentration of that nuclide at the source under unsaturated flow conditions is

$$C_{max} = \tilde{C}(t) = \frac{|q|}{\bar{\theta}} \frac{Ad}{Q} \frac{(1 - e^{-\beta t})}{\rho_b(1-\phi)} e^{-\lambda t}\ \text{pCi/mL} \qquad (21)$$

where $\bar{\theta}$ is the average volumetric moisture content in the repository. Note that the term Ad/Q was taken as unity, allowing the results to be interpreted in a flexible fashion. Maximum exposure of the waste was supposed to occur in a period of 50 years, in which case $\beta = 0.133\ year^{-1}$.

A schematic cross-section of the hypothetical site is shown in Figure 2. A layer of clay 0.5-m thick overlays the 6-m-deep trench, which is then capped with 2 m of loam. The characteristic curves of the three soils simulating the repository are shown in Figures 3.a and 3.b. The precipitation is assumed to be 1.25 mm per day. The approximate steady-state moisture profile is shown in Fig. 4. Note the abrupt changes in the shape of this profile at the interface of the different soil layers and the low moisture content prevailing in the trench.

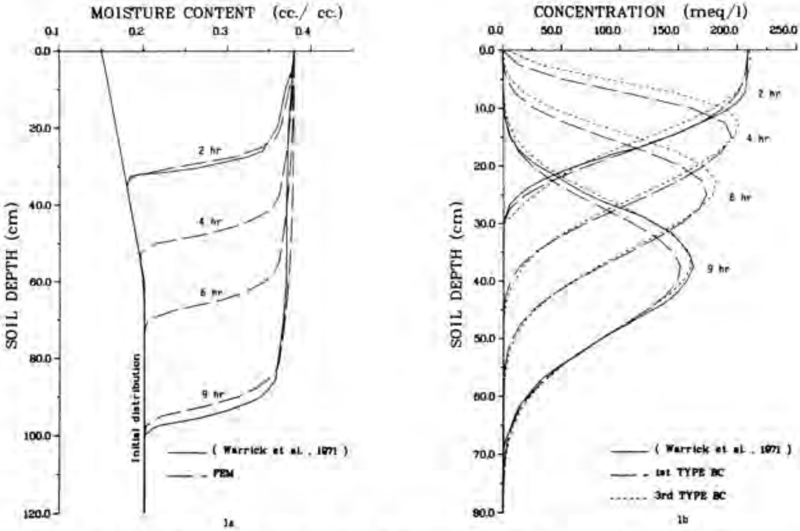

Fig. 1. Profiles of (Fig. 1a) Moisture Content and (Fig. 1b) Concentration for Infiltration Times of 2, 4, 6 and 9 hours.

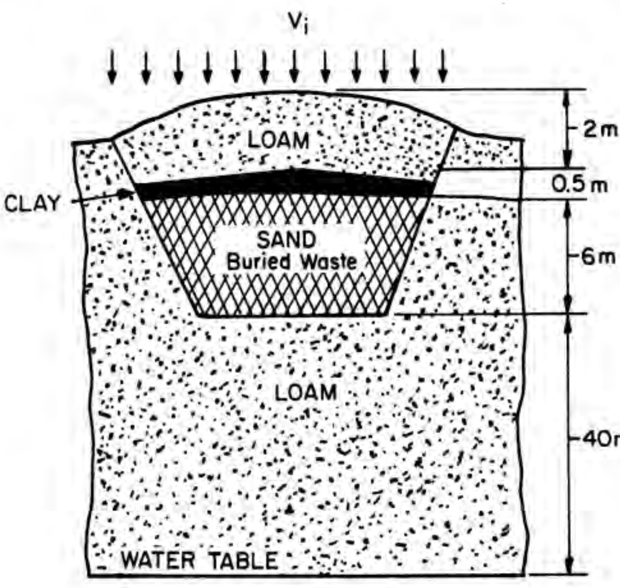

Fig. 2. Vertical Cross Section of a Shallow Waste Disposal Site.

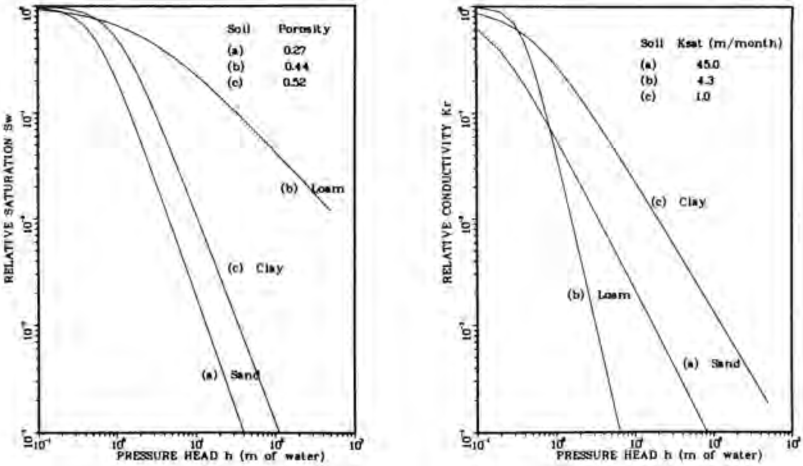

Fig. 3. Relationships between the Soil-Water Pressure Head and (Fig. 3a) Relative Saturation and (Fig. 3b) Relative Conductivity for Soils (a), (b), and (c).

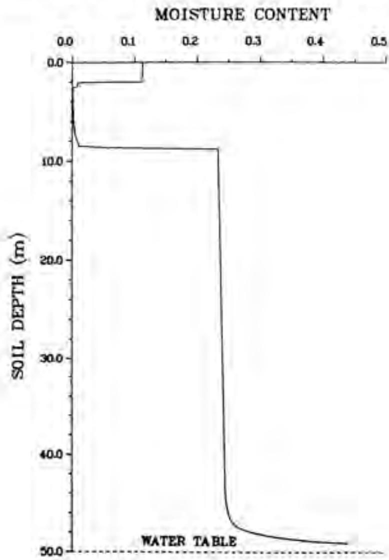

Fig. 4. Steady-State Moisture Content Profile.

The normalized concentration profiles are shown in Fig. 5a at times t = 5.09, 10.1, and 20.1 years for the three species migrating from the base of the trench. The mobility of these radionuclides is influenced to a large extent by their distribution coefficients. These were selected as 40, 400, and 0 cm^3/g for Sr-90, Cs-137, and H-3, respectively. The half-lives used were 28.1, 30.2, and 12.4 y, respectively, and the coefficient of molecular diffusion was chosen as 0.0008 m^2/month for all three nuclides. The dispersivity of the host formation was assumed to correspond to 1 m. Note that H-3 (as water) is almost in contact with the water table at time t = 20.1 years. Moreover, concentrations of the various species at the source seem to be declining after a period of 10 years. This is due largely to our assumption that the waste has reached 86% of its maximum release after 10 years.

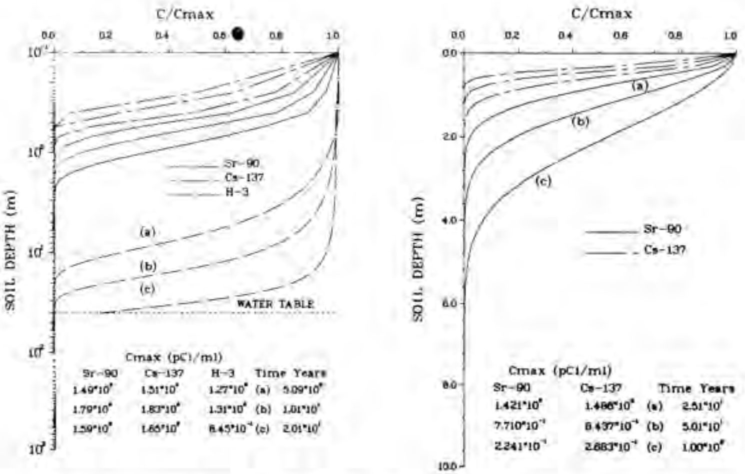

Fig. 5. Concentration Profiles of (Fig. 5a) Sr-90, Cs-137 an H-3 at 5.09, 10.1 and 20.1 years, and of (Fig. 5b) Sr-90 and Cs-137 at 25.1, 50.1 and 100 years.

Because the predictive capability of the one-dimensional model is restricted to the pathway of migration in the vadose zone, H-3 was eliminated from further analysis.

The normalized concentration profiles for Sr-90 and Cs-137 at time t = 25.1, 50.1 and 100 years are shown in Fig. 5b. The trend in the migration rate of both species remains unchanged; however, an appreciable drop in concentration is registered relative to the distance traveled by the solute front. For this hypothetical case, it is expected that the tip of the plume for Sr-90 and Cs-137 would reach the water table within 1875 and 445 years, respectively, from the onset of the burial operations. By then, the maximum concentrations at the source for Sr-90 and Cs-137 would have decreased through decay by 6 and 19 orders of magnitude, respectively.

It appears that the adsorption mechanism plays a dominant role in attenuating the concentration of the radionuclides. As conservative as we have tended to be in our assumptions, we have disregarded the impact of a soil-water solution with a pH lower than its neutral value on the buffering performance of the host media. This particular risk factor should not be overlooked in future studies.

A mathematical model for investigating the suitability of a low-level solid radioactive waste site has been presented. The computer code, written in Fortran IV, was run on an IBM 370/195, and the execution time for the 100-year simulation was approximately 7 minutes.

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