WASTE MIGRATION IN SHALLOW BURIAL SITES UNDER UNSATURATED FLOW CONDITIONS

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ABSTRACT

Unsaturated conditions prevail in many shallow-land burial sites, both in arid and humid regions. Unless a burial site is allowed to flood and possibly overflow, a realistic assessment of any migration scenario must take into account the conditions of unsaturated flow. These are more difficult to observe and to model, but introduce significant changes into projected rates of waste leaching and waste migration.

Column tests have been performed using soils from the Southeastern coastal plain to observe the effects of varying degrees of "unsaturation" on the movement of radioactive tracers. The moisture content in the columns was controlled by maintaining various levels of hydrostatic suction on soil columns whose hydrodynamic characteristics had been determined carefully. Tracer tests, employing Cs-137, I-131 and Ba-133 were used to determine migration profiles and to follow their movement down the column for different suction values. A calculational model has been developed for unsaturated flow and seems to match the observations fairly well. It is evident that a full description of migration processes must take into account the reduced migration rates under unsaturated conditions and the hysteresis effects associated with wetting-drying cycles.

INTRODUCTION

The prediction of the performance of any shallow-land burial site requires a realistic assessment of long-term conditions inside the backfilled trench and the surrounding undisturbed soil. Even with a well-designed trench cap, it is usually assumed that over long periods some water infiltration will take place and give rise to the mobilization of contained radionuclides and their ultimate migration to the biosphere. The magnitude of the predicted levels of released activity, and hence any population dose commitment, depends critically on the flow conditions assumed and the amount of water in contact with the waste (1,2). In a properly designed disposal facility (3), with a well-graded soil forming the backfill material, any water flow existing should be well below saturation conditions. Whereas models assuming saturated flow conditions are fairly easy to handle, unsaturated flow is more difficult to model because the hydraulic conductivity is a power function of the pressure head and the degree of saturation and there is a paucity of experimental data to validate flow models under such conditions (4,5). The equations for soil-water characteristic and hydraulic conductivity obtained by van Genuchten (6) are

$$H = \left[\frac{1}{1 + (\alpha|\Psi|)^n} \right]^m \qquad (1)$$

$$K(H) = K_s H^{1/2} \left[1 - \left[1 - H^{1/m}\right]^m\right]^2 \qquad (2)$$

where α, m, and n are parameters that depend on the shape of $\theta(\Psi)$ curve, and

$$H = \frac{\theta - \theta_r}{\theta_s - \theta_r} \qquad (3)$$

where ψ = pressure head, and θ_s and θ_r refer to saturated and residual values of water content θ. To verify this equation and to obtain numerical data on the magnitude of the dependence of radionuclide migration with flow conditions a series of tests were performed with representative soils from the coastal plain of the Southeastern United States, similar to these described in previous tests (1,7).

As groundwater moves through the site, where radioactive waste is buried, any dissolved radionuclides will tend to interact with the soil matrix surface. Ignoring suspended particulates for the moment, the principal interactions will be by ion exchange on clay minerals and, to a lesser extent, complexation with any organic materials present (8). Depending on the ions involved, the water characteristics and the characteristics of the soil minerals, the dissolved ions will undergo partition between the solid and liquid phases, usually described by the distribution coefficient K_d. The dissolved ions will be subject to a retardation effect as follows:

$$\frac{V_i}{V_w} = \frac{1}{(1 + \rho_b K_d/\theta)} \qquad (4)$$

where V_i = pore velocity of absorbed radionuclides, V_w = pore velocity of groundwater, ρ_b = bulk density of soil, and θ = volumetric water content. This equation is equally applicable to saturated and unsaturated media, but in practice, the K_d factor itself is a function of the wetted area and hence of the moisture content (9). Because K_d depends not only on the fraction of surface wetted, but also significantly on pH, nuclide concentration and species competition, it is important to perform such tests with a soil-equilibrated water under steady-state conditions.

EXPERIMENTAL TESTS

Materials

Since the principal purpose of the tests was to compare saturated and unsaturated flow conditions, most of the work was confined to a single type of soil, designated SP #3, whose principal properties are summarized in Table I. This soil is classified as a sandy clay loam with adequate permeability to serve as a test medium in flow columns.

TABLE I

Soil Properties of Soils Used

		SP #3	SP #1
Bulk Density	:	1.52 ± 0.05 g/cm^3	1.35
Porosity	:	0.43 ± 0.02	0.344
Particle Density	:	2.65 g/cm^3	
Soil Fractions (Sand:Silt:Clay)	:	74:3:23	62:9:29
Hydraulic Conductivity	:	146 cm/d	12.96 cm/d

The hydraulic conductivity was measured in columns 8-12 cm in length by means of both the constant-head and falling-head methods (10). Taking great care to minimize entrapped air, saturated hydraulic conductivities were determined. In both cases these passed through a peak about 2 hours after the start of the tests and reached steady-state conditions of 170 and 146 cm/day, respectively. The residual volumetric water content, for the size fractions employed, was determined by electrical conductivity measurements and found to be about 14%, comparable to values reported for similar soil by de Sousa (1,2).

Surface areas were determined by the modified ethylene glycol - monoethyl ether procedure[11] and found to be 140 ± 9 m^2/g for the SP #3 soil.

Measurement of Distribution Factor

K_d factors were measured by the batch method, in which a suspension of soil particles is contacted with a spiked radioactive solution over a period of time and the absorbed fraction is determined. Care must be taken to have sufficient activity present to reach steady conditions, yet not to overload the receptor soil. For that reason it is useful to run tests in solutions of varying concentrations. For the main tests, triplicate runs were done exposing 4g soil samples in 50 ml centrifuge tubes to the three tracers, Cs-137, I-131 and Ba-133, in 30 ml of soil-equilibrated solution. The results are shown in Table II after 3 days of mild agitation. The pH variation arose from the different tracer solutions employed which were pre-adjusted with NH_4OH. The K_d values observed reflect the expected values for cesium, which is strongly sorbed on clays, and iodine, which will be hardly sorbed at all. The low K_d value for barium had not been expected, as it was supposed to simulate strontium, which has been found to have K_d of several tens (ml/g) in a mild acidic solution of clay soils (11). It is assumed that barium sorption was preempted by higher calcium concentrations in the equilibrated water:

TABLE II

Distribution Factors, K_d, measured for Cs, I and Ba for SP #3 Soil

Radiouclide (Tube Number)	Initial pH of Solution	Final pH of Solution	K_d (ml/g)
Cs-137 (1)	5.60	5.43	404.8
(2)		5.36	416.7
(3)		5.56	401.7
I -131 (1)	5.65	5.21	3.6
(2)		5.39	5.5
(3)		5.24	4.5
Ba-133 (1)	4.78	4.28	0.4
(2)		4.25	0.4
(3)		4.27	0.5

To check the pH dependence of the K_d value for barium, comparative tests were run, at a somewhat higher Ba-133 concentration, with equilibrated water adjusted to a range of pH values. The results are shown in Table III underline the importance of knowing the acidity of the soil and any mobile water in it. At the same time, Ba-133 turned out to be a useful tracer for transport and migration experiments.

TABLE III
Distribution Coefficients of Ba-133
For pH-different Solutions (SP #3 Soil)

Solution Number	Final pH	K_d (ml/g)
1	2.31	1.0
2	4.04	2.0
3	6.95	6.3
4	10.40	644.3

Flow Experiments

To obtain conditions of constant unsaturated flow, it is necessary to provide a means of maintaining constant low pressure or constant suction on the flow system. In the work described here, a constant- head water supply was employed. A 90-cm long plastic tube, I.D. 3.81 cm, was set up, closed at top and bottom by sealed fritted-glass disks with a pore size of 10-15 μm. After filling the column with SP #1 soil a constant head water supply was connected. Moisture profiles could be monitored by means of tensiometers, as shown in the diagram, Fig. 1. Both time-dependent and steady-state conditions could be observed.

One of the questions of concern in the burial sites is the effect of pulsed flow, for instance that following a heavy rainfall on a previously unsaturated soil. The theory associated with the experiment was derived from Darcy's law by Youngs (12). The total volume per unit area of water Q infiltrated at time t is given by

$$Q = \int_0^t [K(\frac{d\Psi}{dz} + 1)]_{z=0}\, dt \quad , \qquad (5)$$

where K is the hydraulic conductivity at a suction Ψ, and z is the distance down from the surface of the column. Eq. (5) explains why, during the first several hours, the infiltration was fast. At first, when water contacted with air-dried soil, the suction gradient $d\Psi/dz$ was very large, but would become smaller and smaller later as time t became large, until the gradient would become zero. When a constant-shaped moisture profile is developed, $d\Psi/dz =$ o and the first term of the integral approaches a constant A. Writing eq.(5) at $t \to \infty$,

$$Q = A + K_{z=0}\, t \qquad (6)$$

Thus, the hydraulic conductivity may be conveniently determined from the asymptotic slope of the plot relating the volume of water infiltrated and time. The moisture content in the region where the constant-shaped profile is developed may be found by plotting the volume infiltrated against the depth of wetting. The asymptotic slope gives the moisture content in the region.

The results obtained are indicated in Fig. 2. The volumetric water content of SP# 1 soil at $\Psi = -30$ cm of water determined in the experiment was 0.344 cm^3/cm^3, and the hydraulic conductivity was determined to be 12.96 cm/day, which was smaller than expected. This small hydraulic conductivity can be explained by the fact that the infiltration of water into a dry soil follows a wetting curve, on the soil-water characteristic, which occurs at lower water contents than a drying curve in the same range of suction.

A series of test were conducted with the apparatus shown in Fig. 1 to observe the moisture profiles on infiltration or drainage. Figure 3 is indicative of the results obtained, showing rapid movement at the beginning and a slowing down deeper as a flat-shaped moisture front developed.

At the end of the infiltration period, with a steady moisture profile in the column, drainage was initiated by disconnecting the water supply. Figure 4 shows what happened. At time t = 0 all six positions were at the same degree of saturation. It was observed that for the first several hours the water content decreased rapidly and then slowed down until it reached equilibrium. The equilibrium water content is that water content the soil can retain at the negative water pressure which is equal to the elevation of the soil, e.g., the soil in the height of 80 cm from the bottom of the column experiences the negative pressure of 80 cm of water. The bottom portion of the column, where no tensiometer probe was embedded, was thought to be almost saturated, since the maximum negative pressure applied in the region was only 30 cm-water (3.05 kPa).

A period of infiltration was applied to the column which had drained to the equilibrium state. The infiltration lasted for six hours and the variation of moisture profile along the column is seen in Fig. 5. As soon as the infiltration started, the column began draining from the bottom, indicating that the column was in a pressure equilibrium state. It is evident that a moisture pulse passed through the column, preceded by a compressed air layer that momentarily reduced moisture, but became more diffused with time. The time constant for the exponential infiltration rate was calculated as 0.044 min^{-1}, whereas the drainage-recovery time constant was 0.007 min^{-1}. This process of rapid wetting and slow drying results in a hysteresis effect which must be taken into account when modeling the process.

Transport Experiments

The purpose of the transport experiment was to analyse migration of radionuclides with different distribution coefficients under various unsaturated flow conditions. The experimental facilities for the transport experiment consisted of 90-cm long soil columns, a water supply system, and a detection system, which comprised a 2" x 2" NaI(Tl) crystal detector, and a multichannel analyser. The detector was placed in a shielded collimator, which was fixed on a small platform, whose height was controllable to scan the soil columns vertically.

The soil columns were prepared the same way as for the flow experiment, except that in this test the tensiometer probes were not connected to the columns. The water supply system employed 250-ml burets with different heights of glass tube in them to give different negative pressures of water for each soil column.

A radioactive source layer was emplaced 5 centimeters below the top of the column. That distance of 5 cm was selected since, by the time the moisture front reached that distance, the flow will reach stability. The thickness of the radioactive source layer was determined to be 0.3 cm, occupying a volume of 3.42 cm^3. 20 ml of radioactive solution was prepared by mixing Ba-133 and Cs-137, which were dissolved in acid in the form of BaCl and CsCl. The pH of the solution was adjusted with NH_4OH to 5.3, which was the pH of the SP #3 equilibrated water. The mixed solution was added to a volume of SP #3 soil such that 3.42 cm^3 of #3 would be loaded with 2 μCi of Ba-133 and Cs-137. The wetted soil was placed in an oven until the soil dried completely. Another volume of SP #3 soil was loaded with 6 μCi of I-131 solution, which had been adjusted to 5.3.

The columns were packed to a bulk density of 1.62 g/cm^3 and arranged in a cluster, so that they could be supplied simultaneously by a set of peristaltic pumps. Negative pressures of 0 cm, -25 cm and -50 cm of water (0, -2.4, -4.8 kPa) were applied to the barium and cesium columns; -25 cm H_2O (-2.4 kPa) to the iodine column. Flow data and moisture front position were recorded periodically, while the columns were scanned to obtain the activity distribution.

By plotting the volume uptake against time, the pore water velocity was obtained for the various suction values that determined the degree of unsaturation. Figure 5 is representative of such plots for volume uptake; the moisture front position varied linearly with pore velocity. Table IV lists the values obtained for hydraulic conductivity and volumetric water content, which is found to agree well with predictions from Eq. (1).

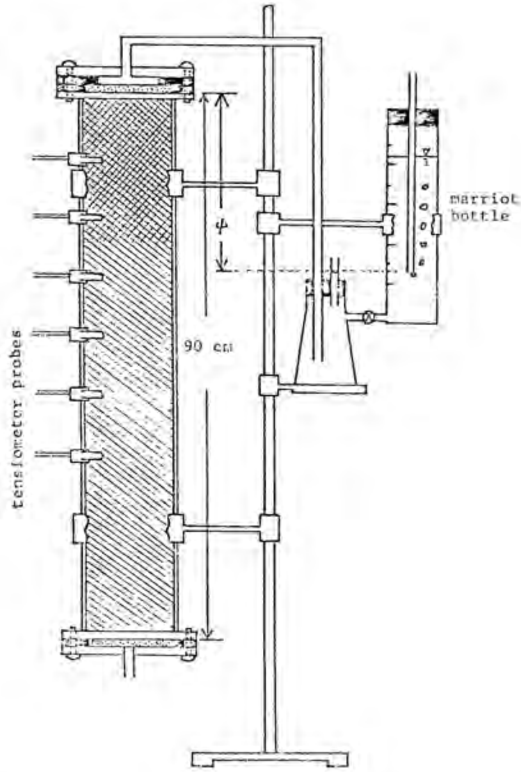

Fig. 1. Diagram of Soil Column With Constant-Head Water Supply System.

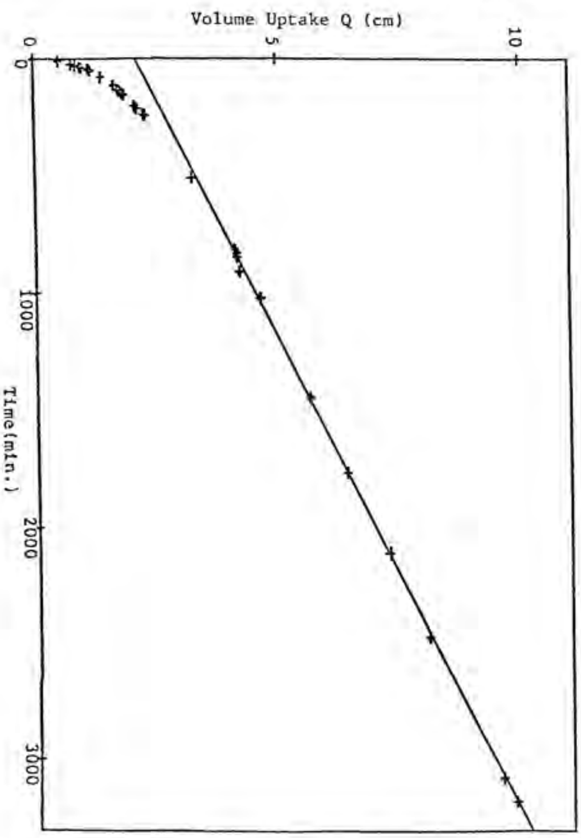

Fig. 2. Hydraulic Conductivity of SP #1: ψ = -30 cm water.

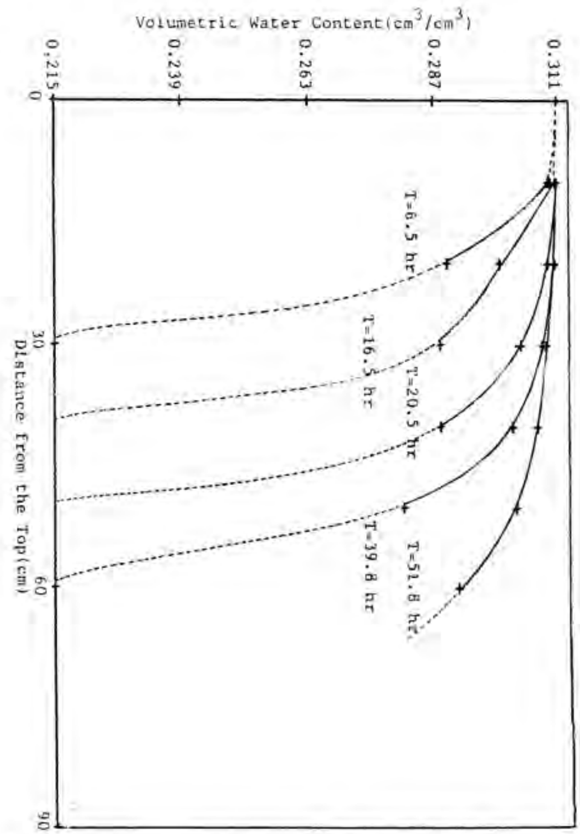

Fig. 3. Movement of Moisture Profiles: ψ = 0 cm water.

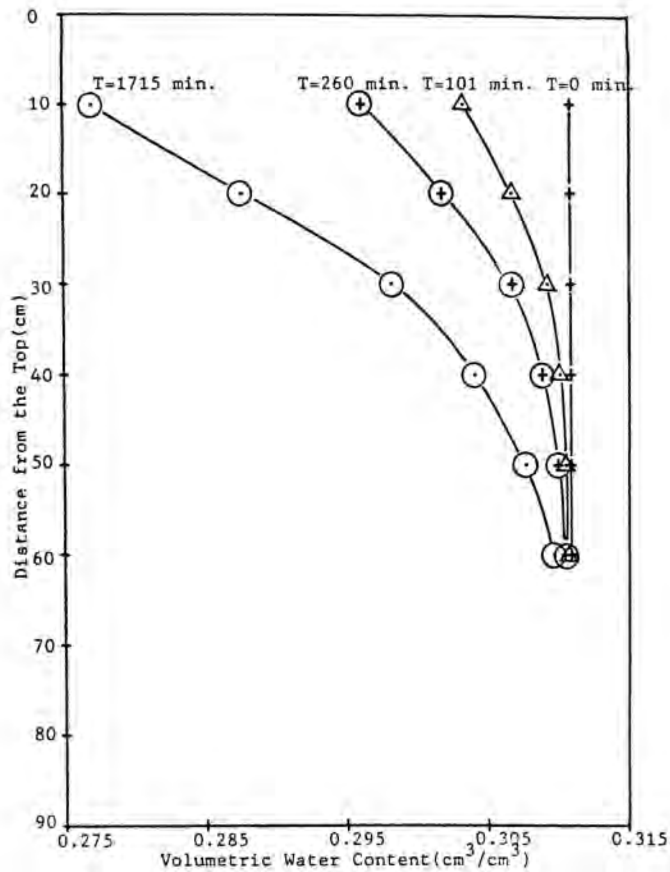

Fig. 4. Moisture Profiles Under Drainage Phase.

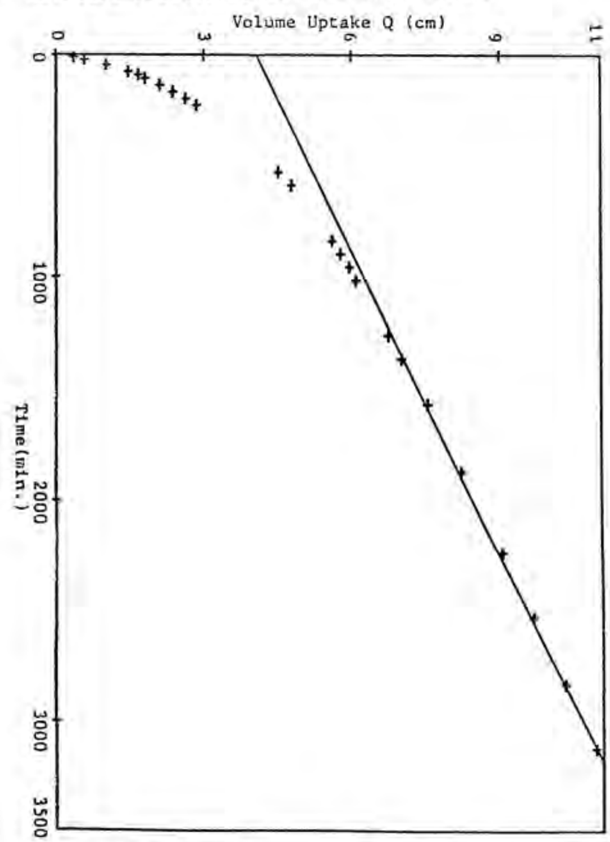

Fig. 5. Volume Uptake vs. Time: = -25 cm water.

TABLE IV

Hydraulic Conductivity and Volumetric Water Content of SRP #3 Soil with Applied Negative Pressure

Negative Pressure (cm-water)	$K(\Psi)$ (cm/day)	$\theta(\Psi)$ (cm^3/cm^3)	$\theta^*(\Psi)$ (cm^3/cm^3)
Ψ = 0	8.490	0.319	0.311
Ψ = -25	3.397	0.293	0.296
Ψ = -50	0.732	0.253	0.253

*values obtained from Eq. (1).

Figures 6 and 7 compare the movement of Ba-133 at 0 and -50 cm of water (0 and -4.8 kPa) of negative pressure. It is evident that movement is considerably slower under unsaturated conditions, as expected; in contrast the iodine movement, even under unsaturated conditions, followed the volumetric flow is shown in Fig. 8. Experimental parameters derived from these experiments, for Ba-133, are as shown in Table V. It is evident that at a suction of -50 cm of water (-4.8 kPa) significant changes in the retardation and K_d factors are obtained, even though that suction results only in a change in moisture content from 100% to 78%. As Fig. 9 shows, fitting the parameters of Table V into the transport model results in a reasonably good fit.

CONCLUSIONS

The work described here has shown the importance of taking into account unsaturated flow conditions in modeling low-level waste burial sites. While the tests necessarily deal with compacted, disturbed soil columns, the conclusions apply equally to backfilled soil and the underlying ground, provided that adequate information can be obtained on the porosity, hydraulic conductivity and sorption parameters. Van Genuchten's equation, equation (1), has been found to be adequate in constructing models that can describe unsaturated flow. Since most backfills and licensable disposal sites are placed in moderately permeable soils, any model based on an assumption of prevailing saturated flow conditions is likely to overestimate grossly the mobility and ultimate environmental impact of any disposed-of radionuclides.

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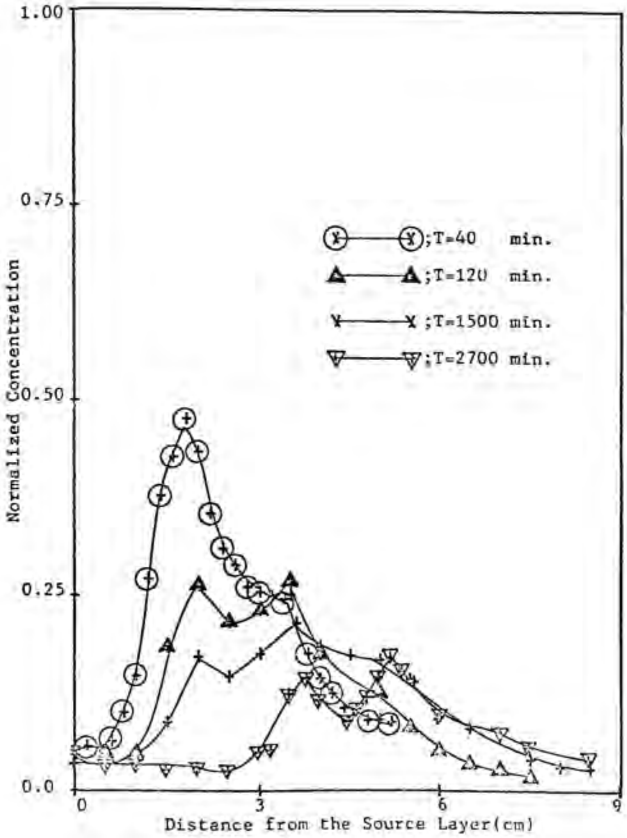

Fig. 6. Ba-133 Concentration Distribution vs. Depth: ψ = 0 cm water.

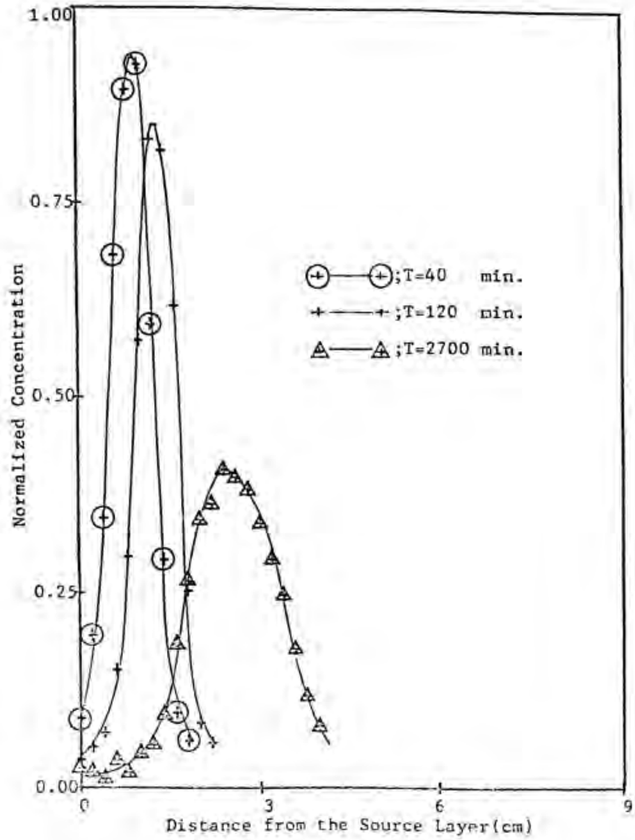

Fig. 7. Ba-133 Concentration Distribution vs. Depth: ψ = -50 cm water.

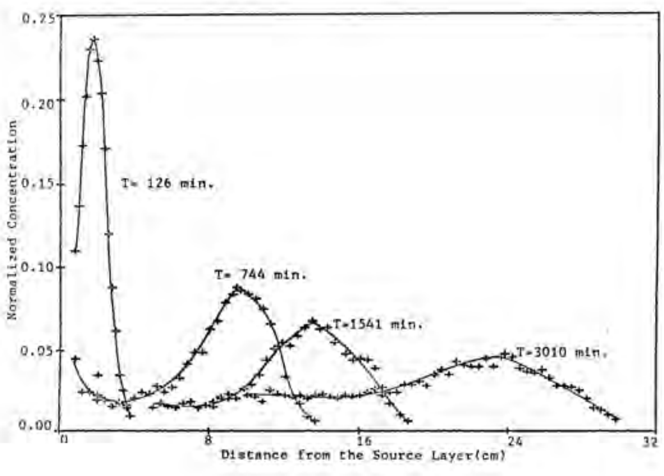

Fig. 8. I-131 Concentration Distribution vs. Depth: ψ = -25 cm water.

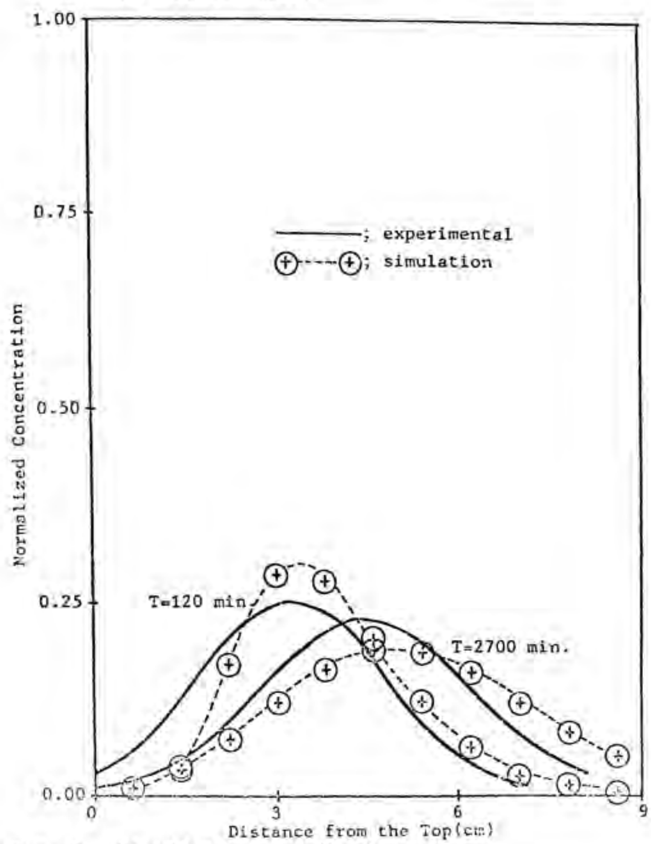

Fig. 9. Ba-133 Transport Simulation.

TABLE V

Va-133 Transport Parameters at t = 40 min, t = 120 min, and t = 2700 min.

t = 40 min

volumetric water content	negative pressure (cm water)	water flux (cm/day)	pore water velocity (cm/day)	retardation factor	distribution coefficient (ml/g)
0.319	ψ = 0	25.52	79.98	1.01	0.002
0.293	ψ = -25	9.48	32.37	1.06	0.01
0.253	ψ = -50	3.98	15.74	0.89	

t = 120 min

volumetric water content	negative pressure (cm water)	water flux (cm/day)	pore water velocity (cm/day)	retardation factor	distribution coefficient (ml/g)
0.319	ψ = 0	18.30	57.36	4.0	0.60
0.293	ψ = -25	6.31	21.55	1.51	0.10
0.253	ψ = -50	2.56	10.13	1.08	0.01

t = 2700 min

volumetric water content	negative pressure (cm water)	water flux (cm/day)	pore water velocity (cm/day)	retardation factor	distribution coefficient (ml/g)
0.319	ψ = 0	8.48	26.57	37.14	7.18
0.293	ψ = -25	3.39	11.56	22.65	3.97
0.253	ψ = -50	0.78	3.07	4.35	0.53