Long-Term Tritium Transport through Field-Scale Compacted Soil Liner

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Abstract: A 13-year study of tritium transport through a field-scale earthen liner was conducted by the Illinois State Geological Survey to determine the long-term performance of compacted soil liners in limiting chemical transport. Two field-sampling procedures (pressure-vacuum lysimeter and core sampling) were used to determine the vertical tritium concentration profiles at different times and locations within the liner. Profiles determined by the two methods were similar and consistent. Analyses of the concentration profiles showed that the tritium concentration was relatively uniformly distributed horizontally at each sampling depth within the liner and thus there was no apparent preferential transport. A simple one-dimensional analytical solution to the advective–dispersive solute transport equation was used to model tritium transport through the liner. Modeling results showed that diffusion was the dominant contaminant transport mechanism. The measured tritium concentration profiles were accurately modeled with an effective diffusion coefficient of 6 $\times 10^{-4}$ mm^2/s, which is in the middle of the range of values reported in the literature.

Introduction

Although compacted soil liners are widely used in landfill liner and cover systems, there is little information available on their long-term performance in limiting chemical transport (U.S. Environmental Protection Agency) (1988). It is believed that molecular diffusion controls transport in low-permeability materials, and diffusion through clays has been extensively studied in the laboratory (Shackelford and Daniel 1991; Rowe et al. 1995; Shackelford and Redmond 1995). However, field scale studies have been more limited because diffusion occurs over long time scales. In the limited field studies that are reported (e.g., Barone et al. 1992; Rowe et al. 1998), vertical transport was monitored in only a few locations and thus there was no assessment of the spatial variability of contaminant concentrations.

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In response to this need for in situ field-scale studies, the Illinois State Geological Survey (ISGS) conducted a unique investigation of the transport of tritium and other tracers through a field-scale compacted soil liner over a 13-year period. During the first phase of this study, a prototype liner was constructed using a regional soil to test construction practices and monitoring methods (Albrecht and Cartwright 1989). During the second phase, the ISGS constructed and instrumented a field-scale soil liner in Champaign, Illinois, and monitored the infiltration of water through the liner, as well as collected long-term data on liner performance.

Water was first ponded on the field-scale liner beginning in April, 1988. Four different tracers [bromide, *o*-trifluoromethylbenzoic acid (*o*-TFMBA), *m*-TFMBA, and pentafluorobenzoic acid (PFBA)] were each introduced into one of four large ring infiltrometers (LRIs) installed at the surface of the liner, and a different tracer (tritiated water) was added to the ponded water outside of the LRIs in May 1989. Initial tests (Panno et al. 1991) showed that the liner was constructed with a uniform and low saturated hydraulic conductivity ($K_{sat} < 1 \times 10^{-6}$ mm/s) that met USEPA requirements.

Since the liner is scheduled for removal in 2002, this paper is a summary and analysis of tritium transport over the 13-year history of the liner. Additional manuscripts that focus upon transport of the bromide and benzoic acid tracers are currently in preparation. Specific objectives of the paper are: (1) evaluate the horizontal spatial variability of transport that could arise from naturally occurring heterogeneities in the soil material or from artifacts caused by the liner design or compaction process; (2) compare and evaluate the tritium profiles measured using two different field-sampling procedures (pressure-vacuum lysimeters and core sampling); (3) evaluate whether long-term tritium transport was controlled by advection or diffusion; and (4) evaluate the validity of analytical one-dimensional models to predict tritium transport using independently estimated parameter values. This

study is unique because tracer data collected over many continuous years of transport in a liner have not been analyzed previously.

Methods and Materials

Soil Liner Construction

A $7.3 \times 14.6 \times 0.9$ m compacted earthen liner was constructed in Champaign, Illinois using a local illitic gray glacial till (Batestown Till Member of the Wedron Formation). The till had a loam texture: 30% clay, 33% silt, and 37% sand. The physical properties of the till (Krapac et al. 1991) and preliminary laboratory and field studies indicated that the till could be compacted to achieve a saturated hydraulic conductivity K_{sat} of less than 1×10^{-6} mm/s (Albrecht and Cartwright 1989).

The liner was built in seven lifts, with each lift constructed by spreading a 230-mm-thick layer of loose soil and compacting it to 150 mm. The top lift protected the liner during the construction of a cutoff wall and was removed later. The liner was compacted at an average moisture content of 11.4% ($M_{water}/M_{dry\ solids}$) (1.1% wet of optimum as determined by the Standard Proctor Test); the mean dry density of the liner was 1.84 g/cm^3 (93% of the maximum Standard Proctor Density) (Panno et al. 1991).

Liner construction also included an underdrain system, pan lysimeters, drainage collection pits, a cutoff wall, retaining walls, and a shelter to enclose the liner. The underdrain system consisted of five layers. From the base of the underdrain to the liner–underdrain interface the layers were: (1) a 30-mil geomembrane of high-density polyethylene, (2) geofabric, (3) a 30-mm-deep layer of sand, (4) a layer of 10-mm-diameter pea gravel, and (5) geofabric. The shelter was constructed to provide maximum control over infiltration and evaporation, and to allow for continuous monitoring. The liner was overlain by a catwalk system to provide access for sampling and monitoring without compromising liner integrity.

Fig. 1(a) shows the plan view and Fig. 1(b) a cross-sectional profile of the field-scale liner. More detailed information regarding the construction of the liner and its physical properties are given in Krapac et al. (1991).

Monitoring System and Tracer Input

A total of 212 instruments (including large and small ring infiltrometers, tensiometers, lysimeters, gypsum blocks, and evaporation pans) were used to monitor the infiltration and movement of water and tracers through the liner. The construction design incorporated instrumentation for evaluating the liner at various scales of measurements. Four 1.5-m-diameter sealed double-ring infiltrometers and 32 0.28 m-diameter open double-ring infiltrometers were installed at the surface of the liner to measure in situ infiltration. Evaporation pans were used to determine evaporative water loss from the pond overlying the study area (see Fig. 1). An underdrain system and pan lysimeters were installed beneath each quadrant to collect water exiting the bottom of the liner. A summary of instrumentation in the field-scale liner is presented in Table 1.

The depth of water in the pond was maintained at 295 mm from April 1988 to May 1989. The depth of water was increased to 310 mm in May 1989 and maintained at that depth until January 1998. After January the pond depth was allowed to decrease due to evaporation and infiltration of water from the pond. The pond depth never dropped below 100 mm.
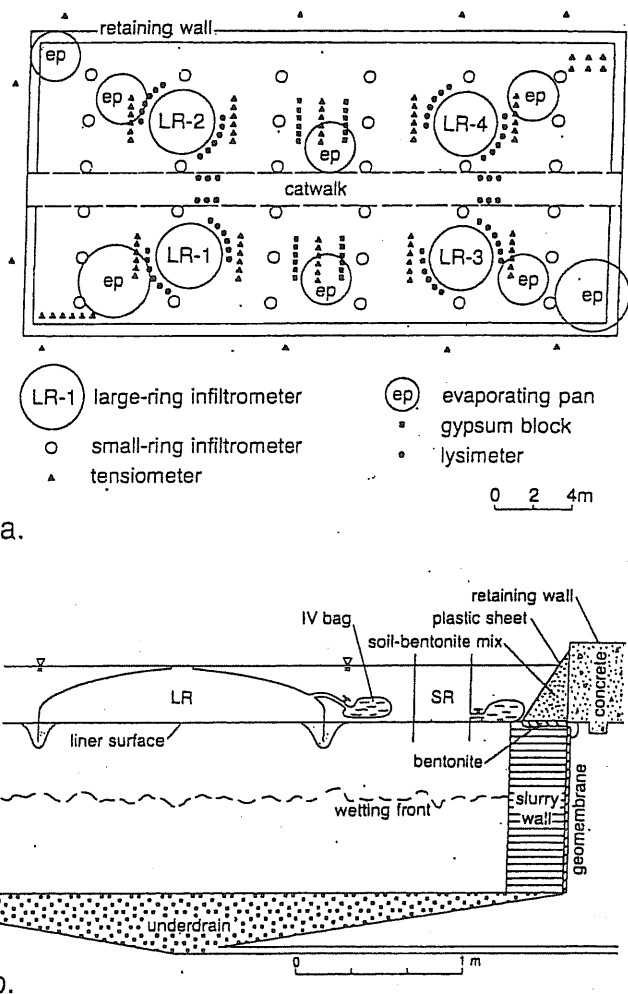

Fig. 1. Field instrumentation for soil liner: (a) plan view showing locations of infiltrometers, evaporation pans, tensiometers, and gypsum blocks; and (b) cross section across retaining wall and infiltrometers (Cartwright et al. 1993)

At the beginning of the experiment, an inorganic tracer (bromide) and three organic tracers (PFBA, m-TFMBA, and o-TFMBA) were each added to one of four large ring infiltrometers (LRI 1, LRI 2, LRI 3, and LRI 4, respectively). Tritium was added to the pond in May 1989 (463 days after water was ponded on the liner). Tritium concentrations in the pond were monitored during the study (Fig. 2). Tritiated water was periodically added to the pond between 1989 and the end of 1990 to maintain an approximately constant concentration, however, the concentration varied between 1150 and 2390 DPM/mL, with an average of 1861 DPM/mL (Fig. 2). After 945 days, tritium was no longer added to the pond, allowing tritium concentrations to decrease exponentially to a current value of 36 disintegrations per minute per milliliter (DPM/mL).

Field Sampling Procedures and Analytical Methods

Two sampling procedures were used to determine tritium concentrations at different depths within the liner. From 1989 to 1998, most of the tritium concentration profiles were determined using pressure-vacuum lysimeters. From 1998 until the present, the tritium concentration profiles were determined from core sampling.

Table 1. Instrumentation in Liner (Krapac et al. 1991)

Instrument	Quantity	Location	Parameter measured
Large-ring infiltrometers	4	Center of each quadrant	Infiltration rates
Small-ring infiltrometers	32	8 quadrant	Infiltration rates
Transducer tensiometers	66	2 nests quadrant 1 nest in NE corner 2 nests along N–S center line	Soil–water tension
Gage tensiometers	18	SW corner and liner perimeter	Soil–water tension
Gypsum blocks	24	4 nests along N–S center line	Soil–water tension
Lysimeters	60	2 nests quadrant 2 nests along E–W center line	Collect soil water for tracer analysis
Evaporation pans	8	Equally spaced in pond and liner perimeter	Evaporation rates

Pressure-Vacuum Lysimeters Sampling Procedure

Prior to 1998, soil pore water samples were collected quarterly from 60 pressure-vacuum lysimeters at ten locations and six depths (0.1, 0.18, 0.33, 0.51, 0.69, and 0.89 m) below the liner surface. Twelve lysimeters (2 nests) were located under the catwalk while the remaining 48 lysimeters were around the periphery of each large ring infiltrometer (2 nests/infiltrometer) so that the nests were directly across from each other [Fig. 1(a)]. Each lysimeter was 0.2 m from the edge of the infiltrometer. To collect samples, each lysimeter was placed under a vacuum of approximately 0.4 m of water for 48 h. The vacuum caused water to flow from the soil pores into a porous cup. The water was then removed from the cup by releasing the vacuum and placing a positive air pressure on the assembly, forcing the water in the cup to travel through a small-diameter tube into sample collection vials (Krapac et al. 1991).

Tritium concentrations were determined by a Packard 2000 CA TriCarb liquid scintillation analyzer. For each sampling time, the tritium concentration at each depth was averaged over all the lysimeters.

Core Sampling Procedure

Core Sampling

Tritium concentrations were measured with depth at 23 locations throughout the line using soil cores (Fig. 3). Prior to core sampling, a 500-mm-diameter steel pipe was driven 50 mm into the liner surface to create a coffer dam; standing water was removed from within the dam. Three soil cores were taken in 0.3 m incre-

ments from the same hole using a 76.2 mm inside diameter thin-walled aluminum coring tube (ASTM 2000a). The coring tube was driven into the liner using a modified post-hole driver until the desired depth (0.3, 0.6, or 0.9 m) was reached. Coring tubes were pulled from the liner using a mechanical jack and sealed on both sides using parafilm and duct tape. After sampling, each hole was filled with bentonite and recompacted. Soil cores were subsampled within 24 h.

Tracer Analysis

Coring tubes were cut open and tritium concentrations were measured in the soil pore water. Coring tubes were opened with a circular saw by making two axial cuts on opposite sides of each soil core. Once the tube was removed, subsamples were taken with a spatula by cutting four to six 20-mm-thick disks from selected locations along each 0.3 m core length. The outer surface of each subsample was shaved to eliminate smearing effects, and each subsample (or disk) was separated into smaller samples. After removing any large rocks (>6 mm), soil samples (approximately 10 g each) were put into preweighed 50 mL Nalgene centrifuge tubes and weighed. Each centrifuge tube was amended with 10 mL of de-ionized water (Barnstead Nanopure De-Ionizer), rapidly mixed for several seconds (vortex mixer), and tumbled end-to-end (NBS rotating tumbler) for 24 h to equilibrate the original pore water with the amended water (Roy et al. 1991). After tumbling, samples were centrifuged (Beckman J2-HC centrifuge) at 16,000 rpm for 20 min and approximately 6–8 mL of supernatant was removed for analysis. These centrifuge parameters yielded reproducible results because an adequate volume of
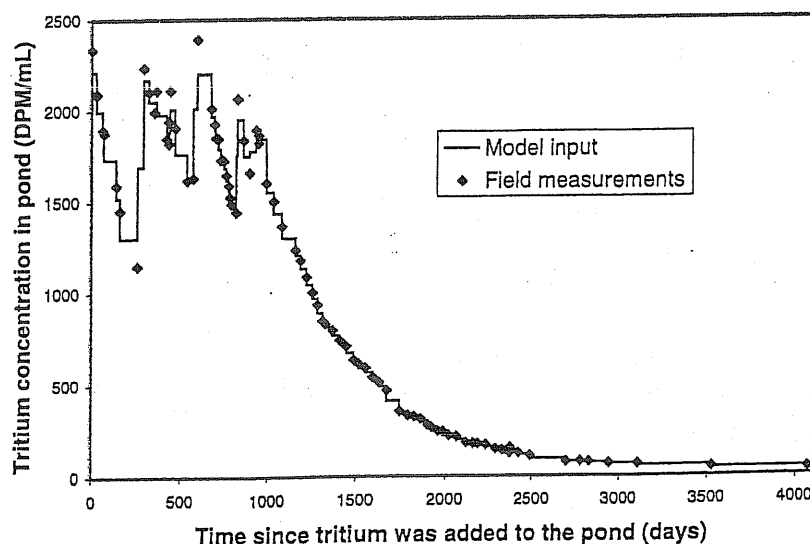
Fig. 2. Tritium concentration measured in pond and input concentration for transport model
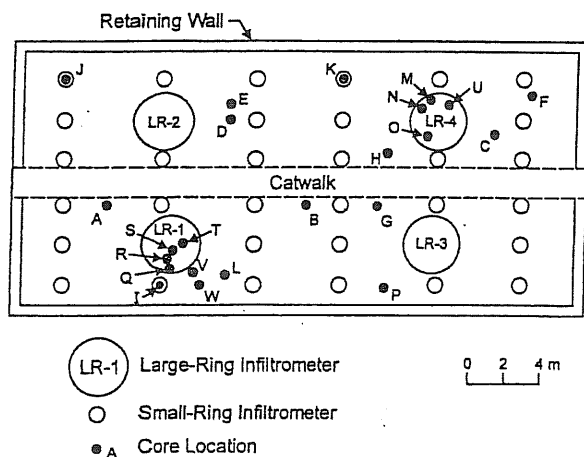
Fig. 3. Plan view of liner showing core locations

water was obtained for analysis of tritium.

A 1 mL aliquot of supernatant was put into scintillation vials containing 10 mL of Ultima Gold liquid scintillation cocktail. A Packard Tricarb 2900TR liquid scintillation analyzer was used to measure the concentration of tritium in each sample. The original pore-water concentration of tritium in each soil sample was then calculated:

$$C_{pw} = \frac{C_{meas}V_{total} - C_{back}V_{amended}}{V_{pw}} \qquad (1)$$

where C_{meas} = measured tritium concentration; V_{total} = total volume of water in a given sample $(V_{pw} + V_{amended})$; C_{back} = background tritium concentration in the amended water; $V_{amended}$ = volume of amended nanopure water (i.e., 10 mL); and V_{pw} = original pore-water volume.

The volume of pore water (V_{pw}) was determined from the following equation:

$$V_{pw} = \frac{1}{\rho_w}\left[M_{wet} - \left(\frac{M_{wet}}{1 + M_c}\right)\right] \qquad (2)$$

where ρ_w = density of water; M_{wet} = mass of soil used in the tritium extraction (with pore water); and M_c = moisture content $(M_{water}/M_{dry\,solids})$ as determined via ASTM D2216-98 (ASTM 2000b). For the majority of cores outside of the LRIs (Method 1), part of the soil subsample was used to determine M_c while another part was used for the tritium extraction (i.e., M_{wet}). For the majority of cores inside of and immediately adjacent to the LRIs (Method 2), M_c was determined from the same soil used in the tritium extraction. Here, centrifuge tubes were baked dry after the supernatant was removed, and M_c was calculated by the difference in the initial weight of the centrifuge tube with the sample (before water amendment) and the weight after baking. This latter method allowed direct measurement of V_{pw} while reducing soil-handling time and error.

Precision and Accuracy

Data precision was ensured by measuring the tritium concentration in multiple soil samples from the same subsample, and by measuring the tritium concentration of each supernatant extraction in triplicate. The tritium measurements in different soil samples (duplicates, triplicates, etc.) from the same subsample varied by an average of 14.1% using Method 1, and by an average of 4.0% using Method 2. Tritium concentrations for the same vial (i.e., triplicate measurements of the same supernatant) varied by an average of 3.4%.

To ensure that the technique used to measure the concentration of tritium in the soil pore water was reliable, five duplicate samples were measured by a vacuum-extraction vapor-trap method. For the vacuum-extraction vapor trap, each soil sample was placed in a weighing bottle and attached to a cold trap (liquid nitrogen) under vacuum, similar to the method described by Malik (1991). After the vacuum was established, soil samples were heated to 100°C and the extraction was run for 3 h. Water vapor captured in the cold trap was then added to a scintillation cocktail and analyzed (Packard 2000 CA/LL). Tritium concentrations from the two methods (vapor trap and Method 2) varied in a nonsystematic manner by an average of 6.4%, suggesting that both techniques provide similar results. Due to the number of samples and expedience of the extraction technique, it was used in this study.

Modeling Approach for Predicting Tritium Transport through Liner

Model Assumptions

Tritium migration within the liner is governed by three main processes: advection, hydrodynamic dispersion, and radioactive decay. It was assumed that tritium transport was one dimensional in the vertical direction, since the data to be reported in the next section did not indicate any significant horizontal variability. Moisture contents measured for the core samples were relatively uniform with depth (Strack et al., unpublished data). Moreover, the measured moisture content was very similar to that determined (11.4%) at the time of liner construction (Panno et al. 1991). Therefore, steady flow at constant moisture content was assumed.

Governing Equations

The simplifying assumptions noted above allow tritium transport through the liner to be modeled using the one-dimensional advective–dispersive solute transport equation (Freeze and Cherry 1979; Van Genuchten and Alves 1982; Rowe et al. 1995)

$$D\frac{\partial^2 C}{\partial z^2} - v_s\frac{\partial C}{\partial z} - \lambda C = \frac{\partial C}{\partial t} \qquad (3)$$

where D = hydrodynamic dispersion coefficient (L^2/T); v_s = seepage velocity (L/T); z = vertical distance from the surface of the liner (L); t = time since tritium has been added to the pond (T); C = tritium concentration (M/L^3); and λ = radioactive decay coefficient for tritium (T^{-1}). The hydrodynamic dispersion coefficient is generally expressed as (Bear 1979; Freeze and Cherry 1979)

$$D = \alpha_L v_s + D_m \qquad (4)$$

where α_L = longitudinal dispersivity (L) and D_m = molecular diffusion coefficient (L^2/T).

Boundary and initial conditions as well as input parameter values are also required to solve Eq. (3). A first-type boundary condition at the surface of the liner is used

$$C(z=0,t) = C_{input}(t) \qquad (5)$$

where $C_{input}(t)$ = input tritium concentration in the pond,

Table 2. Model Input Parameters

Parameter	Input value
Seepage velocity	9×10^{-8}–5×10^{-6} mm/s
Molecular diffusion	4×10^{-4}–8×10^{-4} mm²/s
Radioactive decay coefficient	1.77×10^{-9} s⁻¹

which varied over time as indicated in Fig. 2. The boundary condition at the bottom of the liner was assumed to be a semiinfinite system where

$$\frac{\partial C(z \to \infty, t)}{\partial z} = 0 \qquad (6)$$

Although the liner is finite, studies have shown that the semi-infinite approximation [Eq. (6)] is often adequate (Parker and Van Genuchten 1984). Finally, as the tritium background concentration present in the water used to saturate the liner was low (approximately 25 DPM/mL) compared to the initial tritium concentration added to the pond, the initial tritium concentration was assumed to be zero

$$C(z, t=0) = 0 \qquad (7)$$

Several investigators (e.g., Parker and Van Genuchten 1984; Shackelford 1994) have explained how the concentration in the governing Eq. (3) can be interpreted as either a soil "resident" concentration or a "flux" concentration. In our study we model the resident concentration since we are comparing our simulation results with the soil core and lysimeter data. These same authors argue that continuity of advective–dispersive mass flux at the inlet suggests adopting a third-type boundary condition instead of the first-type condition given in Eq. (5). In our model we have chosen the simpler first-type boundary condition since we found that the third-type condition allows only a negligibly small amount of tritium to enter the liner for diffusion-dominated systems with very low values of the seepage velocity.

General Analytical Solution

The analytical solution for Eq. (3), subject to Eqs. (5)–(7), was developed by Van Genuchten and Alves (1982) for a constant input. To use this solution, the time varying tritium concentration was discretized into multiple steps and the tritium concentration was considered to be constant during each step input. Each step duration was equal to the time between two successive field tri-

Table 3. Infiltration Fluxes Determined from Large-Ring (LR) and Small-Ring (SR) Infiltrometers and From Water Balance

Instrument	Arithmetic mean infiltration flux (mm/s)	Standard deviation
LR (2nd year)	1.8×10^{-8}	6.3×10^{-8}
LR (3rd year)	1.3×10^{-7}	1.9×10^{-8}
LR (4th year)	8.5×10^{-8}	2.9×10^{-8}
LR (5th year)	4.6×10^{-8}	2.1×10^{-8}
SR (2nd year)	7.3×10^{-7}	2.8×10^{-7}
SR (3rd year)	4.2×10^{-7}	2.6×10^{-7}
SR (4th year)	2.9×10^{-7}	2×10^{-7}
SR (5th year)	2.3×10^{-7}	1.5×10^{-7}
Water balance (2nd year)	5.6×10^{-7}	—
Water balance (3rd year)	6.0×10^{-7}	—
Water balance (4th year)	6.7×10^{-7}	—
Water balance (5th year)	5.7×10^{-7}	—
Water balance (6th year)	3.5×10^{-7}	—
Water balance (7th year)	4.3×10^{-7}	—

tium concentration measurements and the concentration associated with it was equal to the average of the two measured tritium concentrations. The superposition principle was then used to combine the analytical solutions as follows:
For $t < t_1$

$$C(z, t) = C_0 A(z, t) \qquad (8)$$
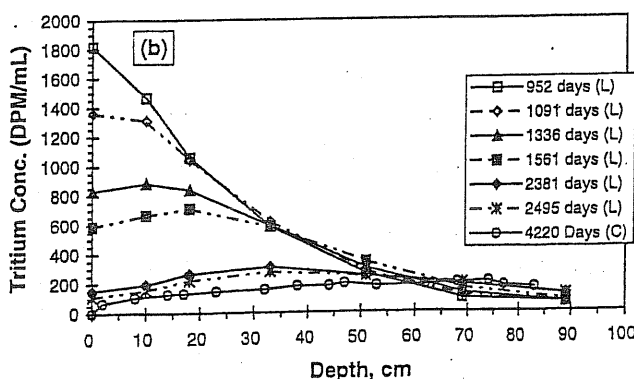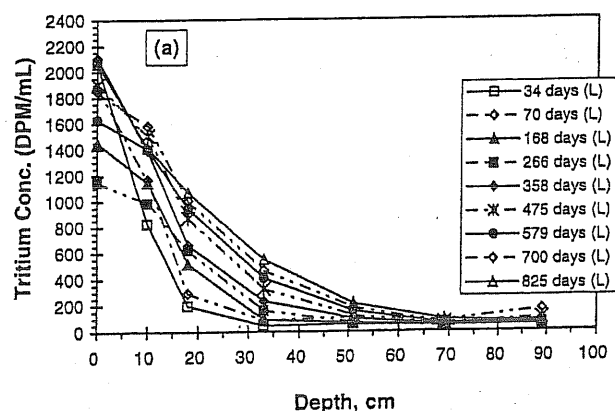
For $t_{n-1} < t \leq t_n$ and $n \geq 2$

Fig. 5. Tritium concentration profiles in liner: (a) during constant source concentration period (less than 945 days); and (b) during decaying source concentration time period (after 945 days)
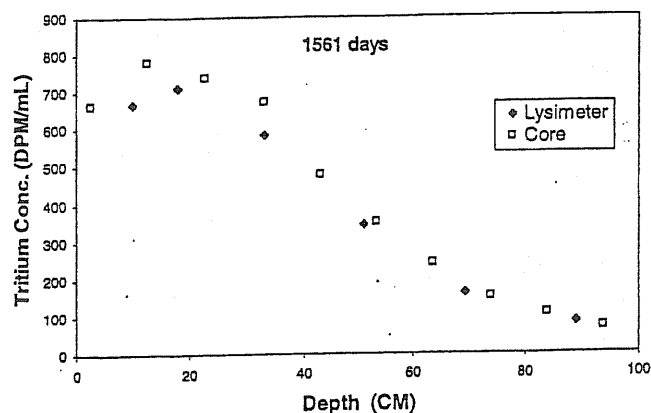

Fig. 4. Comparison of tritium concentration obtained with pressure-vacuum lysimeters and core sampling procedures
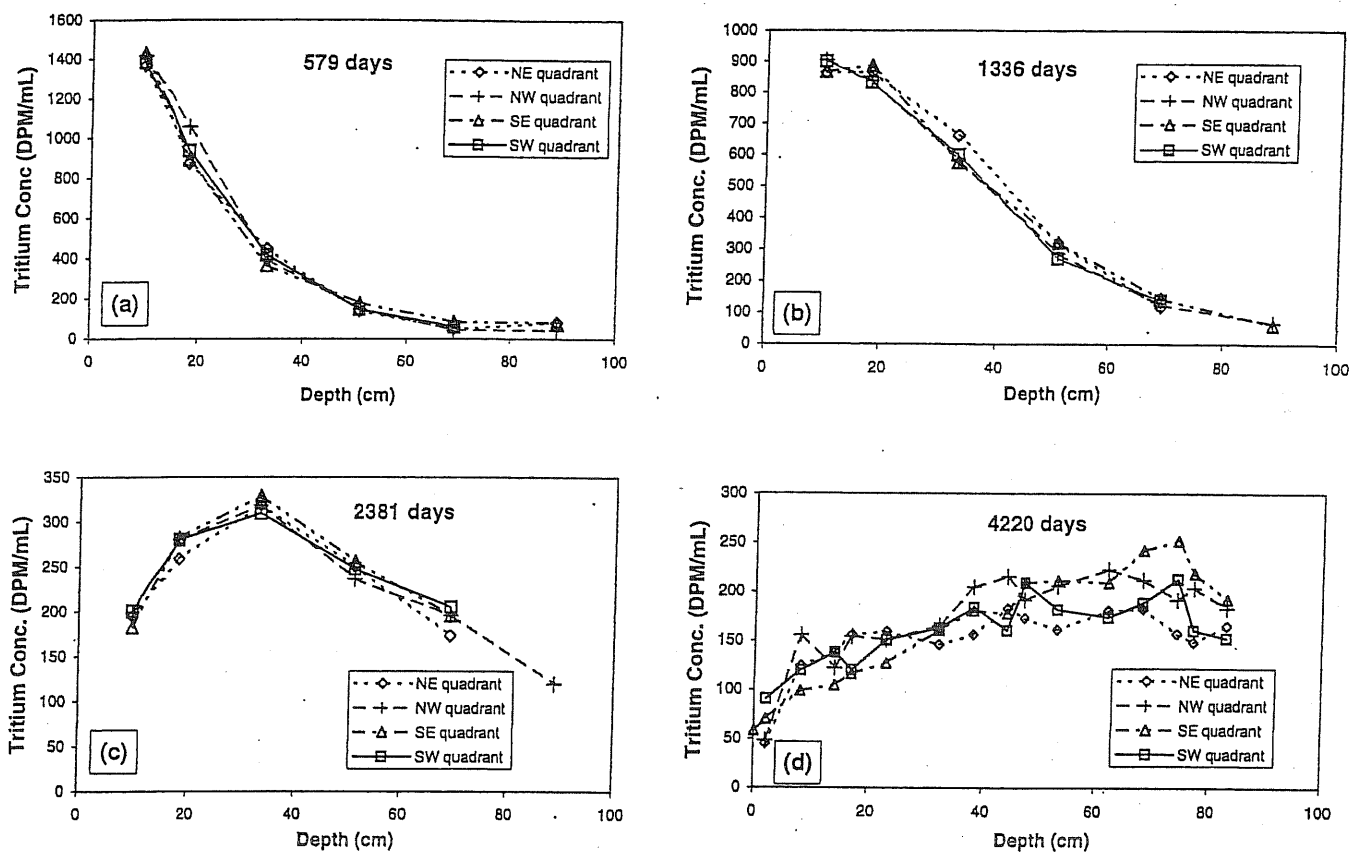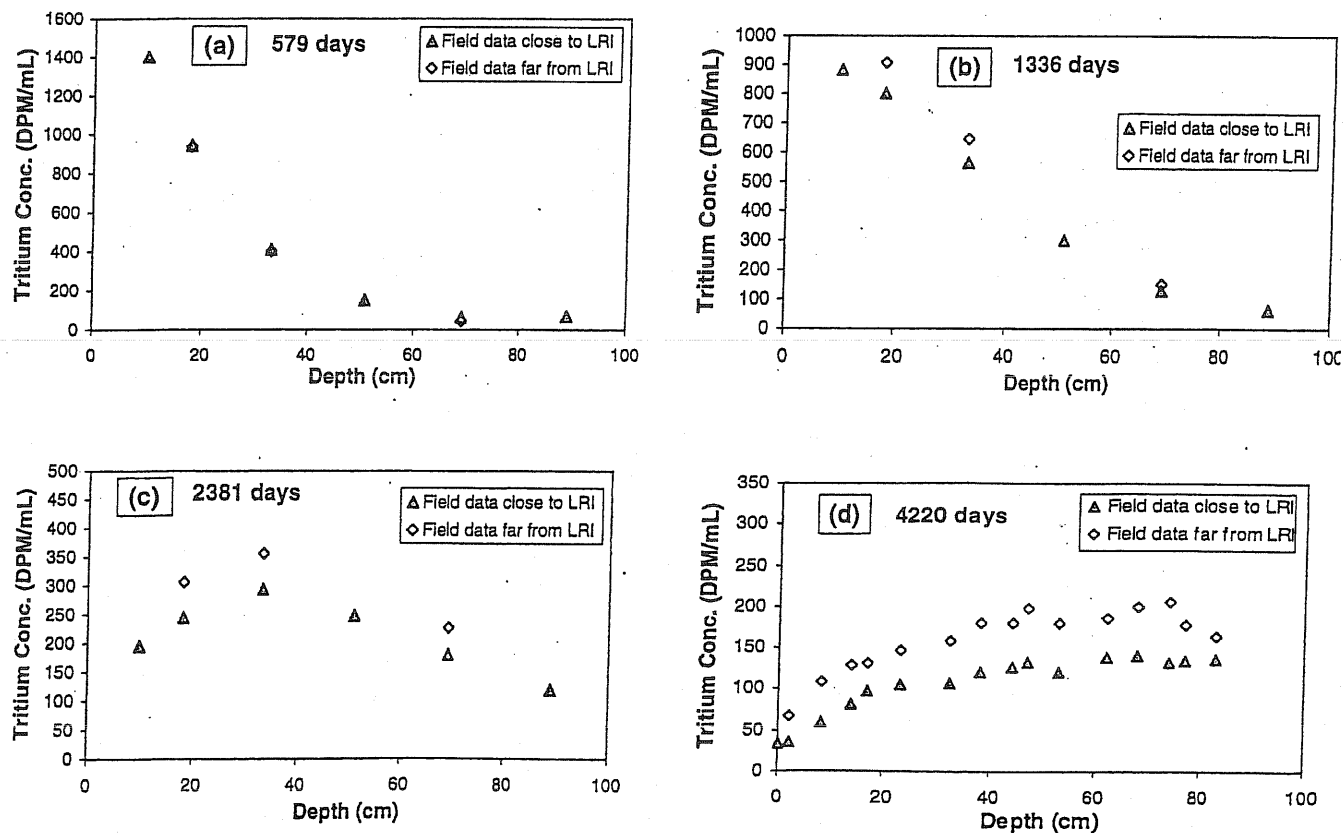
Fig. 6. Comparison of tritium profiles in four quadrants

Fig. 7. Comparison of tritium profiles close to and far from large ring infiltrometers
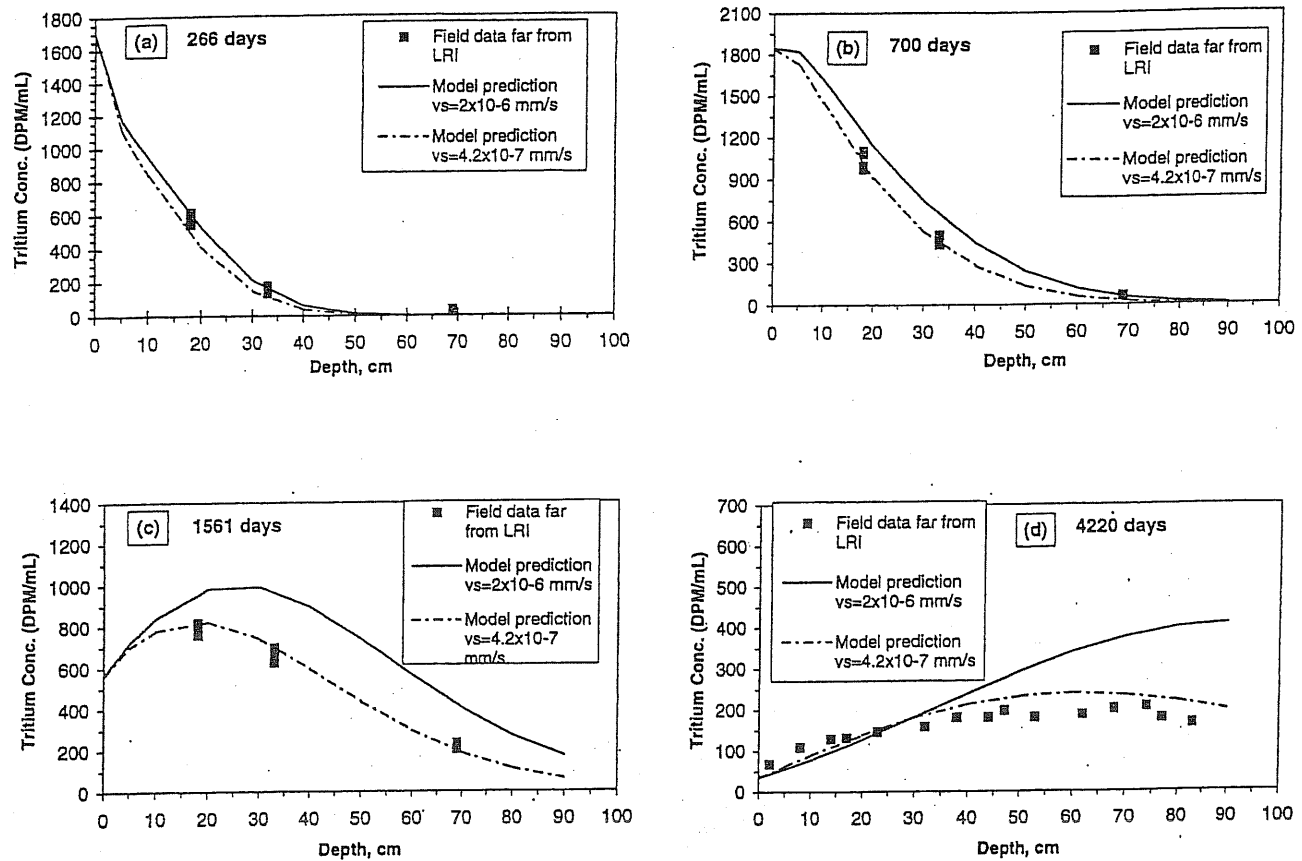
Fig. 8. Comparison of field data and model predictions using diffusion coefficient from literature and seepage velocities based upon measured infiltration fluxes

$$C(z,t) = C_{n-1}A(z, t - t_{n-1})$$

$$+ \sum_{i=2}^{n} [C_{n-i}A(z, t - t_{n-i}) - C_{n-i}A(z, t - t_{n-i+1})]$$

where $A(z,t)$ is defined as

$$A(z,t) = \frac{1}{2} \left\{ \exp\left[\frac{z}{2D}(v_s - u)\right] \text{erfc}\left(\frac{z - ut}{2\sqrt{Dt}}\right) \right.$$

$$\left. + \exp\left[\frac{z}{2D}(v_s + u)\right] \text{erfc}\left(\frac{z + ut}{2\sqrt{Dt}}\right) \right\}$$

with $u = \sqrt{v_s^2 + 4\lambda D}$ where D, v_s, z, t, C, λ were defined previously and C_i = pond tritium concentration between times t_i and t_{i+1} (Fig. 2).

Contaminant Transport Model Input Parameters

Input parameters for Eq. (8) are summarized in Table 2. Some parameter values were relatively straightforward to estimate. For example, the radioactive decay coefficient was equal to 1.77 $\times 10^{-9}$ s^{-1}, which corresponds to a half life of 12.43 years for tritium (Lucas and Unterweger 2000). Other input values required further analyses, as described below.

Seepage Velocity
The seepage velocity v_s was evaluated from the data on the water infiltration fluxes into the liner and the moisture content from the

soil cores. 32 small ring and four large ring infiltrometers were used to measure the infiltration flux. The design and operation of the small and large ring infiltrometers are given in Panno et al. (1991). For each infiltrometer ring, cumulative infiltration volume was plotted against time and the annual average infiltration rate was computed as detailed in Panno et al. (1991). Average infiltration fluxes and standard deviations are summarized in Table 3. The overall infiltration flux of the liner was also calculated using a water balance approach. To estimate the volume of water infiltrating the liner during a 1-year period, the volume of water that evaporated from the pond was subtracted from the amount of water required to maintain a constant pond level (Krapac et al. 1991). A similar approach to that used for infiltration data analysis, in which cumulative infiltration volume was plotted against time, was used to evaluate the average annual infiltration rate for the water balance method.

The arithmetic mean infiltration fluxes for the large-ring and small-ring infiltrometers differ by approximately 1 order of magnitude. Some of the reasons have been detailed by Panno et al. (1991), and include differences in the installation method and accumulation of gas bubbles observed in the LRIs during the initial phase of the study. An alternative explanation could be that the small-ring infiltrometers (SRIs) may not have been completely sealed from the atmosphere and therefore evaporation losses may have occurred (Daniel 1989). Because tritium was added to the liner pond 1 year after the beginning of the study, the infiltration fluxes determined during the first year of ponding are not reported in Table 3. The mean infiltration flux ±1 SD corre-
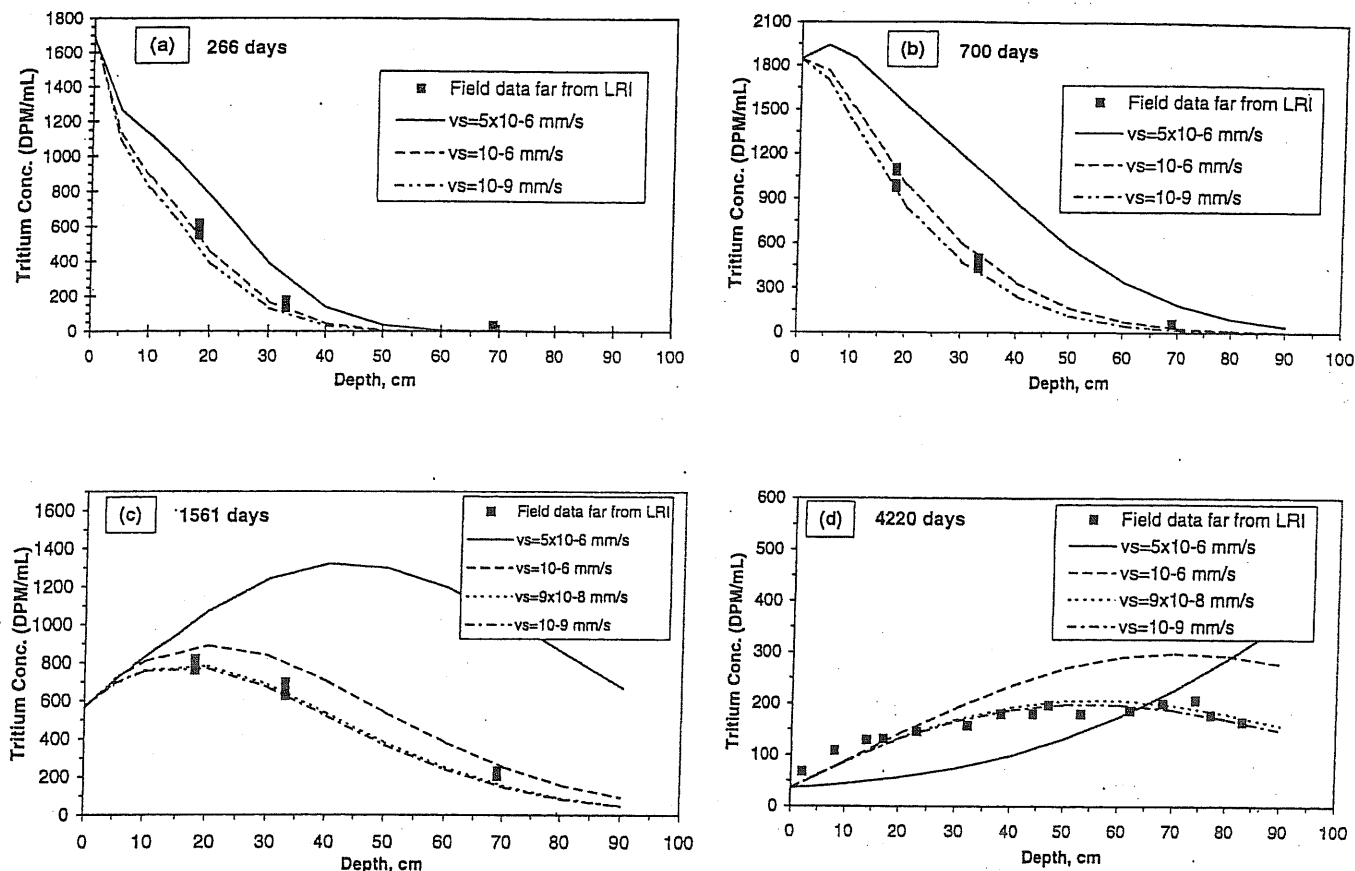
Fig. 9. Effect of seepage velocity on tritium transport at different times (diffusion coefficient $= 6 \times 10^{-4}$ mm^2/s)

sponds to a range from 1.8×10^{-8} to 1×10^{-6} mm/s for all SRI and LRI data.

The volumetric water content, also needed to compute the seepage velocity, was determined from the moisture contents measured on the 30 cores collected during 2000–2001. The moisture content was found to range between 9.9 and 12.5% ($M_{water}/M_{dry\ solids}$) with an average of 10.9%. There was no discernable trend in moisture content with respect to sample depth. Based on these moisture content data and the dry density of the liner (1.84 g/cm^3), the volumetric water content (volume of water/volume of medium) is approximately 0.2. This value is consistent with that determined by Panno et al. (1991) at the beginning of the study. Seepage velocity was estimated by dividing the measured infiltration fluxes by the water saturation value of 0.2. The seepage velocity varied between 9×10^{-8} and 5×10^{-6} mm/s, depending upon the infiltration flux used in the calculation (Table 3).

Hydrodynamic Dispersion and Molecular Diffusion

Hydrodynamic dispersion is defined as the sum of effective molecular diffusion and mechanical dispersion [Eq. (4)]. The effective molecular diffusion coefficient is a function of the diffusion coefficient in free solution and the tortuosity of the medium (Rowe et al. 1995)

$$D_m = D_0 \tau \qquad (9)$$

where $D_0 =$ free solution diffusion coefficient of the solute (L^2/T) and $\tau =$ dimensionless tortuosity factor for an isotropic medium. A molecular diffusion coefficient was not directly determined for the liner, so relevant field and laboratory values published in the

literature (Shackelford 1991) were used to determine a range of possible values. Gillham et al. (1984) reported a value of 2.44 $\times 10^{-3}$ mm^2/s for tritium transport in free solution and an effective diffusion coefficient in the range of $8-17 \times 10^{-4}$ mm^2/s in a sand bentonite soil. Phillips and Brown (1968) reported an effective diffusion coefficient for tritium in the range of $5.3-10.9$ $\times 10^{-4}$ mm^2/s in a kaolinite soil, and in the range of $4.5-9$ $\times 10^{-4}$ mm^2/s in a montmorillonite soil. Young and Ball (1998) found an effective diffusion coefficient for tritium in the range of $3.8-5.9 \times 10^{-4}$ mm^2/s for a soil with 35% clay, 38% silt, and 27% sand, which has a similar composition to the material in the liner. By considering all these data, the effective diffusion coefficient for this study was estimated to range between 4×10^{-4} and 8×10^{-4} mm^2/s.

The mechanical dispersion coefficient is equal to the product of the seepage velocity v_s and the dispersivity α, as shown in Eq. (4). Gillham and Cherry (1982) and Rowe (1987) showed that molecular diffusion controlled the hydrodynamic dispersion at small seepage velocities, as would be expected for low permeability earthen barriers used for waste containment facilities. Hence, mechanical dispersion was assumed to be negligible.

Results and Discussion

Field Data Analysis for Tritium Distribution within Liner

Comparison of Two Field Sampling Procedures

To determine if the removal of pore water from the liner by pressure vacuum lysimeters disrupted solute transport, tritium concen-
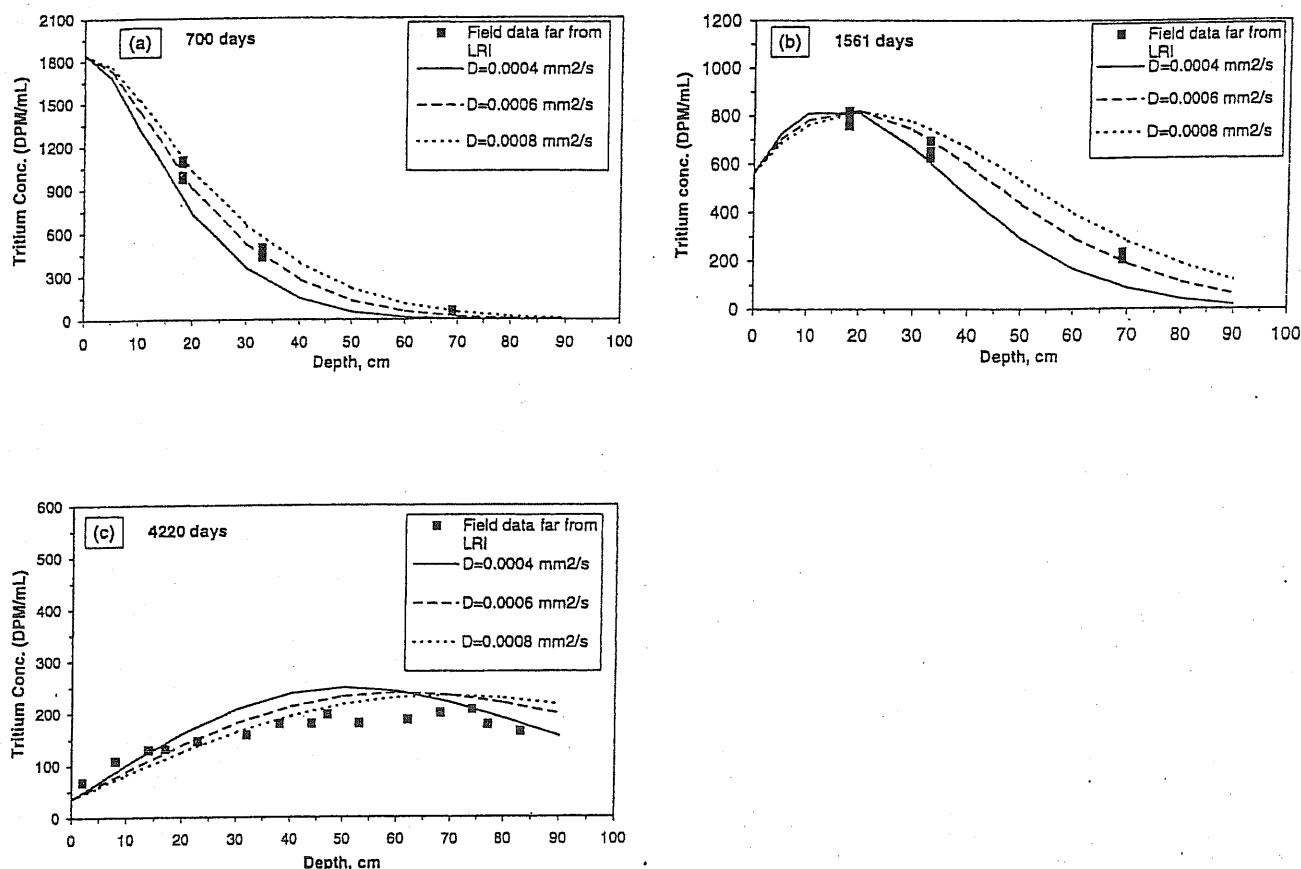
Fig. 10. Effect of diffusion coefficient on tritium transport at different times (seepage velocity $= 4.2 \times 10^{-7}$ mm/s)

trations obtained from a core in 1993 were compared to measurements taken simultaneously from the lysimeters. The lysimeter values at each depth represent an average over the 10 nests (Fig. 1, Table 1). Measured concentrations using both sampling methods were consistent, as shown in Fig. 4. However, at each depth, tritium concentrations obtained by core sampling were slightly greater than those obtained with the pressure-vacuum lysimeters, and this discrepancy was greater toward the liner surface. This could be due to the repeated sampling of the pressure-vacuum lysimeters resulting in tritium concentrations measured close to the surface (at a depth less than 0.1 m) being influenced by the tritium concentration in the pond, as pond water could have been pulled into the lysimeter cups during sampling.

Tritium Profiles in Liner

Tritium concentration versus depth profiles at various times are shown in Fig. 5. In this figure and those to follow, tritium measurements at the last sampling time were obtained from cores (denoted by C in the legend of Fig. 5), whereas measurements at all earlier times were obtained using pressure vacuum lysimeters (denoted by L in the legend of Fig. 5). Profiles during the period when the pond tritium concentration was maintained relatively constant [Fig. 5(a)] decreased from the surface to the bottom of the liner, and the slope of the profiles decreased with time. During the period when the source concentration was decaying [Fig. 5(b)], the tritium profiles changed shape and exhibited a peak concentration that migrated vertically within the liner at later times.

Uniformity of Tritium Transport within Liner

Vertical profiles for the average tritium concentration from all sampling locations in each quadrant are shown in Fig. 6. At each time, tritium concentrations profiles in the four quadrants are similar and are within the variability of a single profile. Earlier work using geostatistical analysis of infiltration fluxes measured in the liner by Krapac et al. (1991) indicated that hydraulic properties of the liner were relatively homogeneous. Our concentration profiles confirm this earlier work and suggest that construction practices used to build the liner were sufficient to minimize preferential flow paths at the scale of our measurements.

Impact of Large Ring Infiltrometers on Lateral Tritium Migration

Tritium concentrations in samples collected near the LRIs (less than 1 m away) were noticeably smaller than those in samples more distant from the LRIs (Fig. 7). Although tritium was not added to the LRIs other than the background concentration (25 DPM/mL) contained in water used to fill the instruments, elevated tritium concentrations were detected in five cores taken within LRI 1 and LRI 4 at 4220 days (data not shown). Since tritium was not added to the LRIs, a horizontal diffusion gradient at each depth likely induced horizontal transport. Thus, the assumption that tritium was essentially transported vertically and that horizontal transport could be neglected was violated near the LRIs.

Tritium Modeling Results and Discussion

Comparison between Observed and Calculated Field Profiles

Measured tritium profiles were used to evaluate the one-dimensional model described previously. Simulations were performed using the parameters listed in Table 2, and the results are

shown in Fig. 8. Only cores located more than 1 m from the LRIs were modeled so that transverse diffusion could be ignored.

Two different seepage velocity values were used, 4.2×10^{-7} and 2×10^{-6} mm/s, which correspond, respectively, to the average determined for the LRIs and SRIs. The molecular diffusion coefficient was adjusted within the range reported in the literature and noted in Table 2 (from 4×10^{-4} to 8×10^{-4} mm²/s) in order to obtain an acceptable fit between the model profiles and the field data. No formal parameter optimization was conducted, but it can be seen in Fig. 8 that an excellent fit was obtained using the lowest value of the seepage velocity (4.2×10^{-7} mm/s) and a diffusion coefficient in the middle of the range of literature values (6×10^{-4} mm²/s).

Sensitivity Analysis

An analysis was performed to determine the sensitivity of the model predictions to the seepage velocity and the diffusion coefficient. The concentration profiles were calculated at different times with a seepage velocity ranging between 9×10^{-8} and 5×10^{-6} mm/s (as computed from the infiltration fluxes reported in Table 3), and a constant molecular diffusion coefficient of 6×10^{-4} mm²/s (Fig. 9). A number of observations can be made from these figures. First, the impact of a change in the seepage velocity is more notable as time increases, since changing the seepage velocity from 10^{-6} to 10^{-9} mm/s had a small effect in the predicted profiles at 700 days, but a relatively larger effect at 4220 days. This impact is reasonable since the location of an advected solute is simply equal to the product of seepage velocity and time. Second the model overpredicted tritium transport for all sampling times when using a seepage velocity of 5×10^{-6} mm/s (largest value calculated from the measured infiltration fluxes), however model predictions agreed very closely with the tritium data when using a seepage velocity of 9×10^{-8} mm/s (smallest value calculated from the measured infiltration fluxes). Finally, the profiles were not sensitive to the seepage velocity when it was below a critical value. Comparing Figs. 8 and 9 shows that seepage velocity values less than 9×10^{-8} mm/s have a minimal impact upon the concentration profiles. This is in the range where the velocity is so small that tritium transport is controlled by diffusion over the spatial and temporal scale of this study. The computed profiles for these small seepage velocities are in fact very close to those reported in Fig. 8 for a seepage velocity of 4×10^{-7} mm/s, which corresponds to the average value determined from the LRI infiltration fluxes. Therefore it appears that molecular diffusion was the dominant transport mechanism in the liner.

The transport model was also used to examine the impact of the diffusion coefficient on the concentration profiles assuming a constant seepage velocity of 4.2×10^{-7} mm/s. A change in the diffusion coefficient from 4×10^{-4} to 6×10^{-4} or from 6×10^{-4} to 8×10^{-4} mm²/s affected the predicted concentration profiles as shown in Fig. 10. This impact is noticeable, but not dramatic, suggesting that transport through the liner can be modeled as diffusion controlled using literature values for the diffusion coefficient.

Conclusions

This paper presents a case study of the long-term performance of a compacted soil liner in limiting chemical transport. Tritiated water was ponded on the liner and allowed to transport into the liner over a 13-year period. Pressure-vacuum lysimeters and core sampling were used to determine tritium concentration profiles in the liner. Both sampling methods provided consistent results. In addition, horizontal spatial analysis of tracer concentration profiles in the liner indicated that tritium concentrations were uniformly distributed, and that there was no apparent preferential flow at the sampling scale. However, the assumption that tritium transport was essentially vertical and that horizontal transport could be neglected was violated in the vicinity of the four large-ring infiltrometers. Two-dimensional transport was occurring close to the large-ring infiltrometers, in which no tritium was added. Thus both vertical and horizontal transport can be important when contaminants are heterogeneously distributed on a liner surface.

A one-dimensional reactive transport model was used to simulate the experimental profiles. Soil core measurements indicated that the liner moisture content was vertically uniform and had changed little since its construction. Therefore, steady flow was assumed in the model and thus a simple analytical solution could be used. The model results were in excellent agreement with the measured core profiles when using a seepage velocity based upon measurements of infiltration fluxes for the liner, and molecular diffusion and decay coefficients based upon literature values. Molecular diffusion was found to be the most important transport mechanism through the liner. An effective diffusion coefficient of 6×10^{-4} mm²/s and a seepage velocity of less than 4.2×10^{-7} mm/s were found to accurately describe the tritium profiles at different times.

This study shows that simplified analytical solutions can be used to assess the uncertainties involved in the design of landfills and predict the transport of contaminants within compacted soil liners. The level of modeling needed to predict the variability in transport is essentially dependent on the design and construction practices as well as on the uniformity of the contaminant input at the surface of the liner.

Acknowledgments

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