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

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
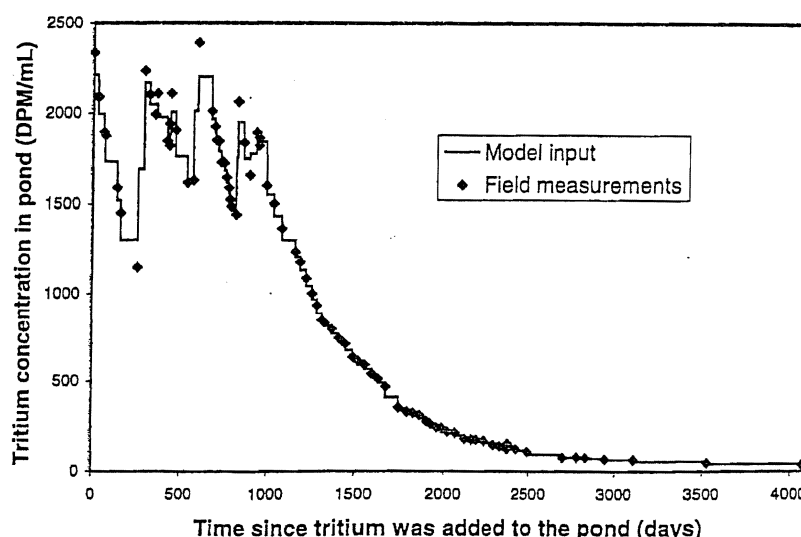

Fig. 2. Tritium concentration measured in pond and input concentration for transport model

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} mm^2/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-
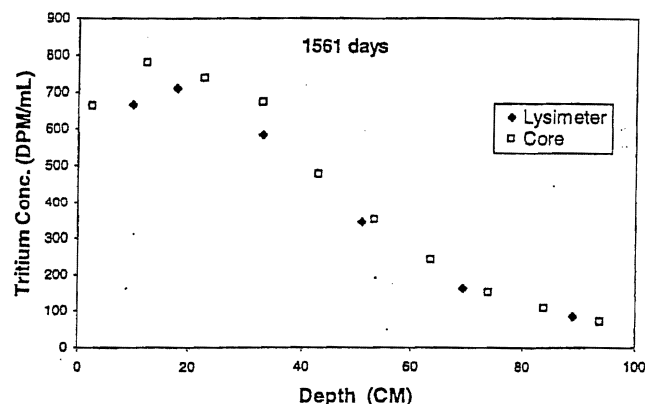
Table 3. Infiltration Fluxes Determined from Large-Ring (LR) and Small-Ring (SR) Infiltrometers and From Water Balance

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 $t < t_1$

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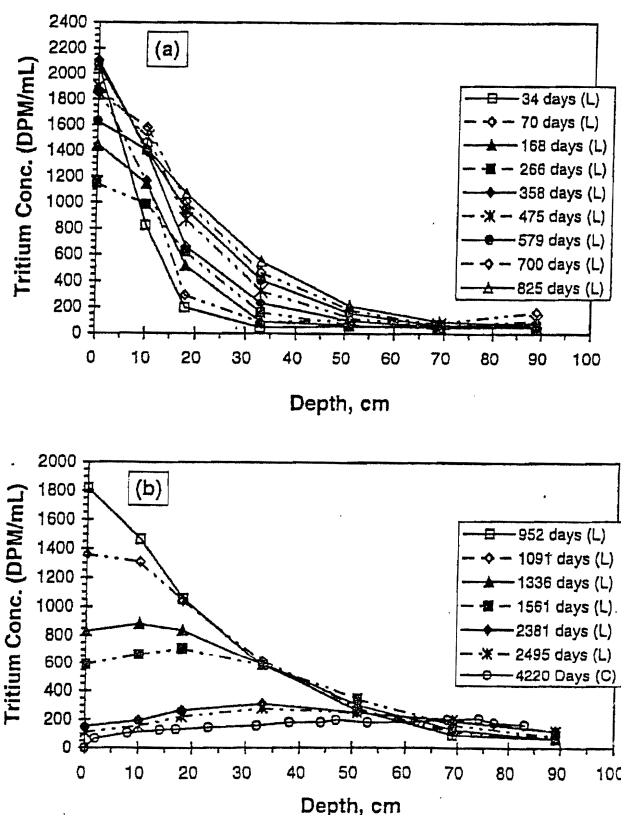
For $t_{n-1} < t \leq t_n$ and $n \geq 2$

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)
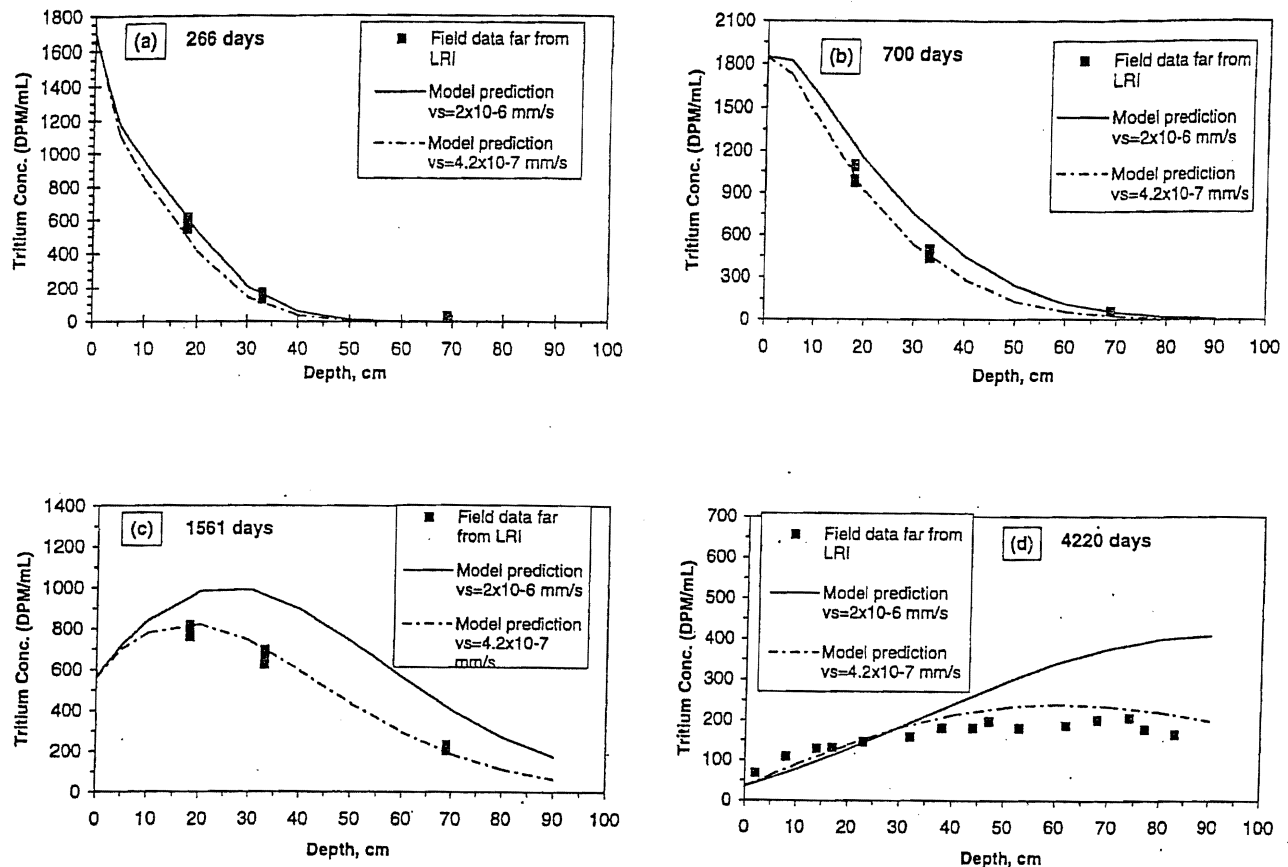
Fig. 4. Comparison of tritium concentration obtained with pressure-vacuum lysimeters and core sampling procedures

Fig. 8. Comparison of field data and model predictions using diffusion coefficient from literature and seepage velocities based upon measured infiltration fluxes

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

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

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

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

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

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-
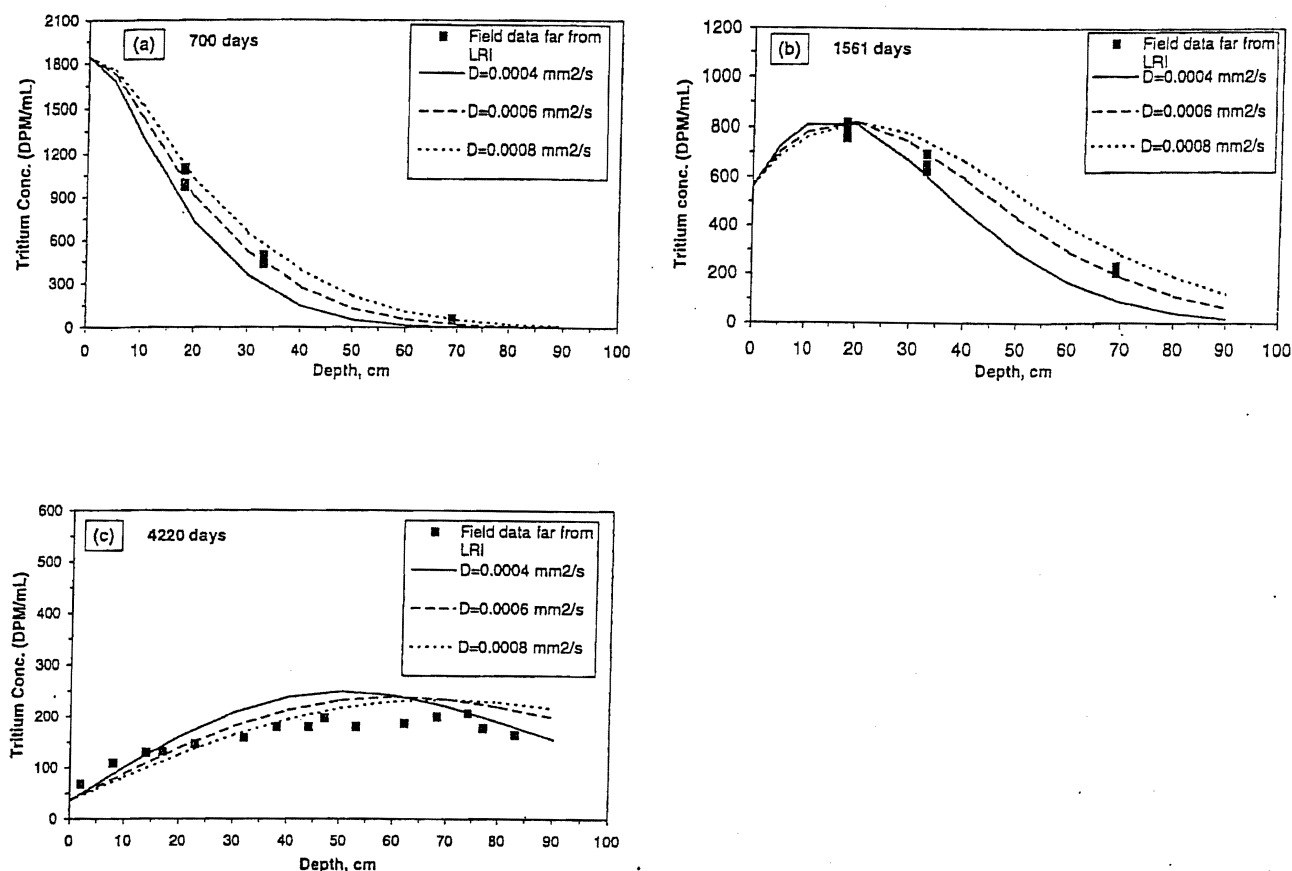
Fig. 10. Effect of diffusion coefficient on tritium transport at different times (seepage velocity=4.2×10⁻⁷ mm/s)

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

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