TRACER TRANSPORT THROUGH A FIELD-SCALE COMPACTED CLAY LINER OVER A 12 YEAR PERIOD

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Abstract: A field-scale earthen liner was constructed over 12 years ago on the southern campus of the University of Illinois at Urbana-Champaign and has been monitored since that time to determine chemical transport rates through the compacted clay liner. The results from this long-term study are presented in this paper.

After construction in 1988, water was ponded on the liner surface to a depth of 30 cm to simulate the maximum allowable head on the compacted clay liner, and was maintained at that depth over the life of the liner. Five tracers: tritium, bromide (Br⁻) and three organic tracers were added to the ponded water to measure chemical transport rates. Tritium was uniformly ponded over the entire liner surface except inside large ring infiltrometers (LRIs); transport for this compound was vertical and one dimensional. Br⁻ and the Benzoic Acid Tracers were ponded only inside LRIs; transport for these compounds were both vertical and horizontal. From 1999 through 2001, soil cores were collected from the liner and analyzed to determine tracer concentration depth profiles (CDP).

CDPs for tritium, Br⁻, and ortho-Trifluoromethyl Benzoic Acid (o-TFMBA) were evaluated to determine transport rates and mechanisms. Simulation results indicate that (1) transport was diffusion controlled and uniform; (2) vertical and transverse diffusion rates were the same; (3) the governing mechanisms of diffusion for Br⁻ and tritium were the same and were represented by the bulk diffusion rate (different for tritium and Br-) times the same tortuosity factor; and (4) transport of the organic tracer o-TFMBA was governed by diffusion and degradation. These results are significant because they evaluate transport through a composite liner from both *distributed* and *localized source zones*. The results from this study indicate that over many continuous years of operation, preferential transport through horizontal lift interfaces did not occur, that transport through the system was diffusion controlled, and that vertical and radial diffusion rates were the same.

Key Words: compacted soil clay liner, preferential transport, bromide, ortho-trifluoromethyl benzoic acid, tritium, core sampling, advection, diffusion.

INTRODUCTION

Most modern landfills employ a composite liner system consisting of a compacted clay liner (CCL) overlain by a geomembrane (GM). In an intact GM, diffusion is considered to be the dominant pathway for chemical transport; however, current engineering practice assumes that "holes" or "punctures" in the GM can form from GM deployment, construction of the leachate collection and recovery system, defects or weaknesses in the welding process, or settlement due to the placement of waste. As a result of these localized source zones, GMs are underlain by CCLs which are capable of retarding the transport of leachate from the composite system. As a result, transport through the composite liner system can be governed by chemical transport through the CCL from a localized source zone. Hence, evaluating and understanding the key parameters for transport from localized source zones in CCLs is critical for being able to accurately evaluate transport through composite liner systems.

Chemical migration through soil barriers is a result of advection, dispersion and diffusion (Manassero and Shackelford 1994, Shackelford 1992). Extensive field and laboratory research has focused on advective flow through soil liners (Daniel 1984, Elsbury et al. 1990, Benson et al. 1999) and on diffusive transport in low conductivity soils (Shackelford 1994 a & b, Crooks and Quigley 1984, Manassero and Shackelford 1994); however, very few studies have focused on multi-dimensional transport in CCLs from localized source zones.

Daniel (1989) reported that horizontal hydraulic conductivities (k_h) could be 5 to 10 times greater than vertical hydraulic conductivities (k_v) in field-scale soil liners based a Boutwell-type test. This suggests that

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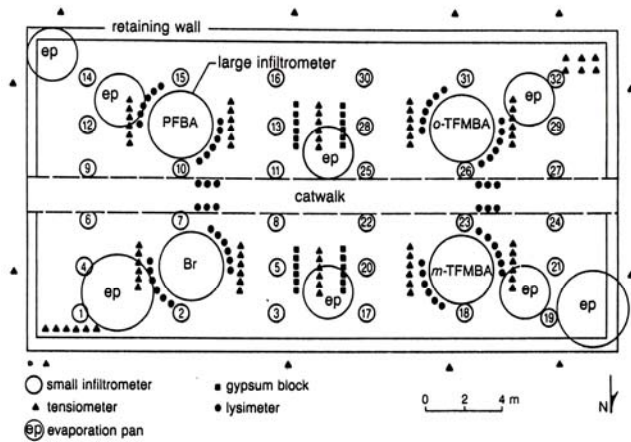
Figure 1. Field Instrumentation for soil liner; plan view showing locations of Large Ring Infiltrometers (LRIs), Small Ring Infiltrometers (SRIs), evaporation pans, tensiometers, and gypsum blocks.

preferential flow between lifts can affect chemical migration.

In the current study, concentration profiles and modeling results for o-TFMBA are presented and compared to modeling results from tritium and Br⁻. Br⁻ and o-TFMBA were only added to localize regions on the liner surface (i.e. inside LRIs) (Figure 1), thus allowing assessment of migration in both the vertical and radial directions. Our objectives are to determine: (1) vertical and horizontal transport rates through the compacted clay liner based on a localized source zone; (2) whether these rates vary spatially from one region to another in the field scale liner; (3) the effect of microbial degradation on chemical transport through a CCL and (4) the variability in chemical migration rates for different chemical tracers through a CCL. This work is unique because chemical transport in CCLs from localized source zones has not been adequately addressed in the literature and no studies have evaluated radial diffusion rates in a field-scale CCL.

Liner Background
In 1986, a prototype liner was built by the Illinois State Geological Survey (ISGS) and USEPA on the southern campus of the University of Illinois in Urbana-Champaign. The prototype liner provided crucial quality control and construction information which was employed during the construction of the current field-scale compacted liner. Construction of the current field-scale compacted liner began in October 1987. Soil for the field-scale compacted liner was a Basetown Till Member obtained from the stockpiled borrow material for the Urbana Landfill in Urbana IL, where it is used as the landfill liner material. The Basetown Till borrow material had a mean water content of 10.4%, a specific gravity of 2.74 g/cm³, Maximum dry density of 2.08 g/cm³, a liquid limit of 21.1%, and plasticity index of 7.0%. The material is classified as a loam to clay-loam using the unified soil classification system and has a particle size distribution of 33.9% sand, 35.4% silt and 30.7% clay (Krapac et al. 1991, Albrecht et al. 1989). In April 1988, water was ponded on the liner surface, and bromide and three benzoic acid tracers were each added to one of four large ring infiltrometers (LRIs). Tritium was added to the pond water (excluding the infiltrometers) in July 1989. These tracers were then allowed to migrate through the liner.

A schematic of the liner facility is shown in Figure 1. The liner facility consists of a compacted soil liner with dimensions of 10 x 17 x 1 m, which includes an instrumented and ponded test area with dimensions of 7.3 x 14.6 x 0.9 m. The entire facility is contained inside an enclosed heated shelter, which minimizes weather effects and prevents infiltration from rainfall. There are four sealed double ring LRIs, 32 open double small ring infiltrometers (SRI), 8 evaporation pans, as well as other monitoring equipment. Br⁻ and o-TFMBA were added to the LRIs 1 and 4, respectively (Krapac et al. 1991).

Recently, soil cores both inside and outside of the LRIs were collected and analyzed for tritium, o-TFMBA and Br⁻. Concentration profiles and modeling results for tritium and Br⁻ have been presented in previous papers (Toupiol et al. 2002, Willingham et al. 2003). These results indicated that the transport of tritium, which was uniformly distributed over the liner surface, was controlled by diffusion, and that the rate of diffusion was spatially uniform throughout the liner. Br⁻ results demonstrated that (1) transport was diffusion controlled and spatially uniform, (2) vertical and radial diffusion rates were the same, and (3) small positioning errors due to compression during soil sampling did not affect the best-fit advection and diffusion coefficients. Results for o-TFMBA are presented here for the first time.

Large Ring Infiltrometers (LRI)
The LRIs are 1.5 m in diameter, and were imbedded 8 cm into a bentonite-cement slurry filled trench, 13 cm wide by 10 cm deep (Krapac et al. 1991). To maintain an equivalent head between the inside of the LRIs and the pond, a small flexible intravenous (IV) bag was attached to the inner ring of the LRI via a tube (Daniel 1989) as shown in Figure 2. Daniel (1989) reports that sealed double ring infiltrometers are able to reliably measure in-situ saturated hydraulic conductivity (k_s) values down to 10^{-8} cm/s. To calculate the advective infiltration flux, the flux of infiltration water was determined by weighing the IV bags every two weeks (Panno et al. 1991); the IV bags were refilled with a solution containing the appropriate tracer to replenish the volume infiltrated as needed during the first
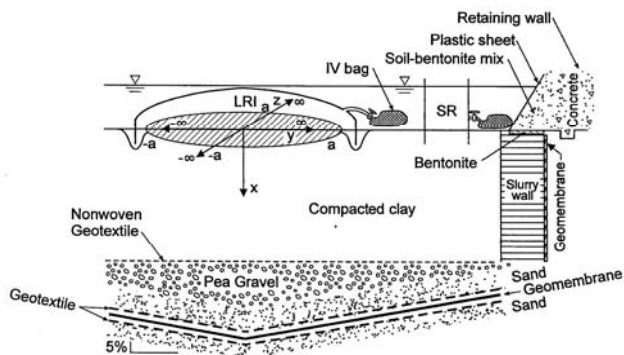
Figure 2. Cross sectional profile showing a LRI, SRI, IV bags, the underdrain collection system and the perimeter retaining wall and slurry wall.

five years (100 mg/l of Br⁻ and 75 mg/l of o-TFMBA).

Figure 2 shows a cross sectional view of one quadrant of the liner system including a LRI and SRI. Liner construction also included an underdrain system, pan lysimeters, drainage collection pits, a cutoff wall, and a concrete retaining wall. The underdrain system is divided into four quadrants (7.3m x 3.6m and one LRI per quadrant). Each quadrant was graded to a 5% slope toward the center of the quadrant where a drainage cup was used to collect water that passed through the liner. From bottom to top, the underdrain system consists of (1) a 3 cm layer of sand covered by a nonwoven geotextile, (2) 30-mil high-density polyethylene GM, (3) a 3 cm layer of sand covered by a nonwoven geotextile, and (4) pea gravel (10 mm in diameter) filled to the level of the foundation of the liner (figure 2).

Ponding
On April 12, 1988, water was ponded on the liner surface to a depth of 29.5 cm; concurrently, tracers were added to each of the LRIs. The water level was raised to a depth of 31 cm on May 25, 1989 and maintained at that depth until January 30, 1998 (Krapac et al. 1991, Cartwright and Krapac 1990, Panno et al. 1991). After January 1998, the pond depth was allowed to decrease due to evaporation and infiltration during core sampling; the water depth never reached the surface of liner.

METHODS

Core Sampling
Prior to core sampling from a LRI (Br⁻ and o-TFMBA sampling), the water level inside the LRI was pumped to approximately the same level as the pond, and the top of the LRI was removed by cutting the LRI at an elevation slightly higher than the pond. The Br⁻ and o-TFMBA LRIs were opened in June 2000 and March 2000, respectively.
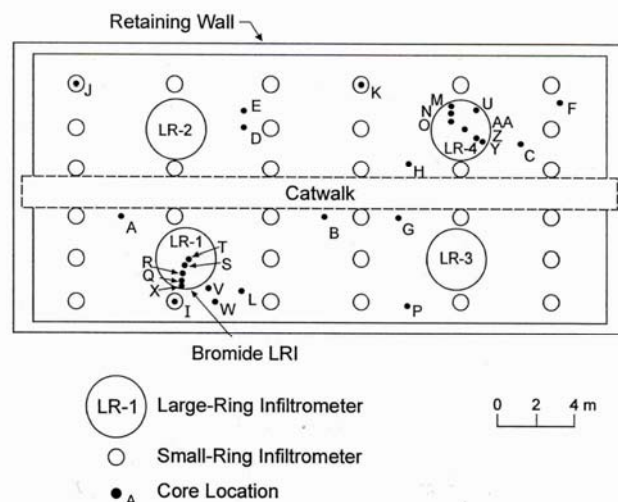
Figure 3. Plan view of liner showing location of core samples for tritium (pond area), Br- (LRI-1), and o-TFMBA (LRI 4).

The locations of soil cores inside of and adjacent to the Br⁻ and o-TFMBA LRIs are shown in Figure 3. To take core samples (for tritium as well as Br⁻ and oTFMBA), a 50 cm diameter steel pipe was driven 5 cm into the liner surface to create a coffer dam; standing water was removed from within the dam. Three soil cores were taken in 30 cm increments from the same hole using a 76.2 mm ID 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 (30cm, 60cm, or 90cm) 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 hours.

Tracer Analysis
Coring tubes were cut open and tracer 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, sections were taken with a spatula by cutting four to six 2-cm thick disks from selected locations along each core length. The sides of each section (disk) were shaved to eliminate smearing effects, and each section was separated into smaller samples. After removing any large rocks (> 6mm), soil samples (approximately 10 g) were put into pre-weighed 50 ml Nalgene centrifuge tubes and weighed. 10 ml of de-ionized water (Barnstead Nanopure De-Ionizer) was added to each centrifuge tube, rapidly mixed for several seconds (vortex mixer), and tumbled end-to-end (NBS rotating tumbler) for 24 hours to equilibrate the original pore water with the de-ionized water (Shackelford 1991, Roy et al. 1991). After

Table 1: Mean and standard deviations of Dry Density and Moisture Content of compacted lifts at construction and excavation of the field-scale liner.

Lift No.	Sample Size	Mean Moisture Content[a] (%)	Stdv	Sample Size	Mean Density[b] (g/cm³)	Stdv
		Construction				
1	8	12.19	1.06	8	1.85	0.07
2	8	12.08	0.52	8	1.87	0.04
3	8	10.76	0.93	8	1.78	0.07
4	8	10.96	0.80	8	1.80	0.04
5	8	11.34	0.86	8	1.87	0.07
6	7	11.33	0.95	7	1.83	0.09
Overall	47	11.44	0.98	47	1.83	0.07
		Excavation				
1	32	10.92	1.83	1	1.99	NA
2	40	10.28	0.55	5	2.00	0.04
3	36	10.04	0.58	1	2.01	NA
4	37	10.41	0.84	2	2.05	0.03
5	39	11.18	0.76	4	2.00	0.02
6	19	11.00	0.76	4	2.02	0.04
Overall	203	10.60	1.05	17	2.01	0.04

Construction density measurements were made using a Seaman Nuclear density meter; excavation density measurements were made using brass rings (5.7 cm diameter by 3 cm high).

a: Mean Moisture Content is defined as Mass Water divided by Mass Dry Solids
b: Mean Density is defined as Mass Dry Solids divided by Total Volume

Table 2: Average Linear Velocities from Large-Ring Infiltrometers (cm/s)

	LRI 1	LRI 2	LRI 3	LRI 4	Average
1st Year	2.5×10^{-8}	8.3×10^{-8}	4.7×10^{-8}	4.1×10^{-8}	4.9×10^{-8}
2nd Year	6.0×10^{-8}	9.1×10^{-8}	1.2×10^{-7}	5.4×10^{-8}	8.0×10^{-8}
3rd Year	5.2×10^{-8}	7.3×10^{-8}	6.2×10^{-8}	5.7×10^{-8}	6.1×10^{-8}
4th Year	3.4×10^{-8}	5.9×10^{-8}	2.9×10^{-8}	3.4×10^{-8}	3.9×10^{-8}
5th Year	1.0×10^{-8}	1.9×10^{-8}	3.3×10^{-8}	2.2×10^{-8}	2.1×10^{-8}
*6th Year	1.2×10^{-8}	3.2×10^{-8}	2.3×10^{-8}	6.0×10^{-8}	3.2×10^{-8}
Average	3.3×10^{-8}	6.1×10^{-8}	5.3×10^{-8}	4.4×10^{-8}	4.8×10^{-8}

*Sampling ceased on the 263rd day of the 6th year.

tumbling, samples were centrifuged (Beckman J2-HC centrifuge) at 16,000 rpm for 20 minutes and approximately 6 to 8 ml of supernatant were removed for analysis. These centrifuge parameters were shown to yield adequate supernatant volumes to perform analysis for all chemical tracers (tritium, bromide, and benzoic acids).

Tracer Analysis

Methodology for the analysis of Tritium and Br⁻ is presented in Toupiol et al. (2002) and Willingham et al. (2003), respectively. For analysis of o-TFMBA, a Hewlett Packard High Pressure Liquid Chromatograph (HP 1090 HPLC) with a reverse phase C18 column and a variable wavelength UV diode array detector was used. To calculate the concentration of the tracers, the following mass balance was used:

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

where C_{meas} is the measured tracer concentration, V_{total} is the total volume of water in a given sample ($V_{pw} + V_{amended}$), C_{back} is the background tracer concentration in the nanopure de-ionized water, $V_{amended}$ is the volume of nanopure water (10 ml), and V_{pw} is the original pore water volume.

The background concentration of o-TFMBA in the nanopure de-ionized water is negligible, hence, equation one reduces to:

Table 3: Tracer Properties

Tracer	V_x (cm/s)	D^* (cm^2/s)	D_{mol} (cm^2/s)	τ []	half life(yr)	μ (yr^{-1})
Tritium	4.2×10^{-8} [a]	6×10^{-6} [a]	24.4×10^{-6} [b]	0.25	12.4[a]	0.0559
Bromide	6.5×10^{-10}	5×10^{-6}	20.4×10^{-6} [c]	0.27	NA	NA
o-TFMBA	6.5×10^{-10}	1.79×10^{-6}	7.3×10^{-6} [d]	0.25	.5	1.39

a: Toupiol et al., (2002)
b: Phillips and Brown (1968)
c: Average of Bulk diffusion coefficient from Table 2
d: Bowman (1984); estimated using the Hayduk and Laudie method

$$C_{pw} = \frac{C_{meas} V_{total}}{V_{pw}} \qquad (2)$$

The volume of pore-water (V_{pw}) was determined from the weight difference between fresh samples (soil samples before addition of de-ionized water) and soil samples baked dry after decanting the supernatant (ASTM 2000b). o-TFMBA concentrations in replicate soil-samples varied by an average of 9.1%.

TRITIUM AND BR$^-$ RESULTS AND DISCUSSION
Analysis of Tritium CDPs revealed that transport of tritium through the liner was controlled by diffusion and spatially uniform (except near LRIs, where the CDPs were affected by radial diffusion due to tritium not having been added to the LRIs). Modeling of Tritium CDPs indicated that advective transport was negligible and well below the regulatory limits of $k_{sat} \leq 10^{-7}$ cm/s. The best-fit diffusion coefficient, D^*, was equal the molecular diffusion coefficient of tritium (D_{mol}) times a tortuosity factor (τ) of 0.25.

$$D^* = D_{mol}\tau \qquad (3)$$

For Br$^-$ analysis, an axially symmetric solution to the cylindrical coordinate system form of the advection dispersion equation was used. Unlike tritium, Br$^-$ was ponded as a localized source (inside of a LRI); hence, transport from the Br$^-$ LRI was governed by vertical and radial transport.

For samples taken at r$\leq$75 cm (e.g. inside of the LRI), Br$^-$ CDPs decreased with depth exponentially from the liner surface. In addition, concentrations at a given depth are lower in cores taken further from the center of the LRI. This is explained by radial diffusion of Br$^-$ away from the LRI, and is reasonable given that the concentration of Br$^-$ in the pond water outside of the LRI was negligible compared to the concentration inside the LRI.

Best-fit values of V_x and D^* for Br$^-$ and Tritium are shown in Table 3. Values of D_{mol} and τ for Br$^-$ and tritium are also listed. While values of D^* for the two tracers are different, the values of τ for the two tracers are nearly coincident (varying by less than 8%). This suggests that the mechanisms controlling diffusion are similar, and that both Br$^-$ and tritium are effective tracers with respect to evaluating transport rates and mechanisms in the field.

A sensitivity analysis for Br$^-$ transport was performed by (i) keeping the average linear velocity at the best fit value and varying the effective diffusion coefficient and (ii) by keeping the effective diffusion coefficient at the best fit value and varying the average linear velocity. Results indicate that transport is sensitive to changes in the effective diffusion coefficient. However, modeled CDPs for average linear velocities less than the best-fit value were coincident, indicating that transport was not sensitive to decreased values of velocity and hence was diffusion controlled. In support of this, a Peclet number (Pe = $V_x *$ L / D^*) of 6.5×10^{-2} was calculated using the best-fit V_x, D^* and the length of the liner, L. Peclet numbers ≤ 1 indicate that diffusion controls mass transport (Shackelford 1994a).

o-TFMBA RESULTS AND DISCUSSION
The o-TFMBA concentration profiles obtained for core samples are shown in Figure 4. Like Br$^-$, o-TFMBA concentrations decrease exponentially with depth. However, unlike Br$^-$, o-TFMBA decreases much quicker, dropping in concentration by an order of magnitude within the first 40 cm, as compared to a factor of two reduction in Br$^-$ over the same distance. Furthermore, the o-TFMBA concentrations decrease by approximately two orders of magnitude from the LRI pond concentration (~30mg/l) to the bottom of the liner (~0.3 mg/l). These results indicate that o-TFMBA transport was substantially slower than that of Br$^-$. This appears reasonable, as the molecular diffusion coefficient of o-TFMBA is approximately 3 times smaller than that of Br$^-$. Also, the concentration profiles in Figure
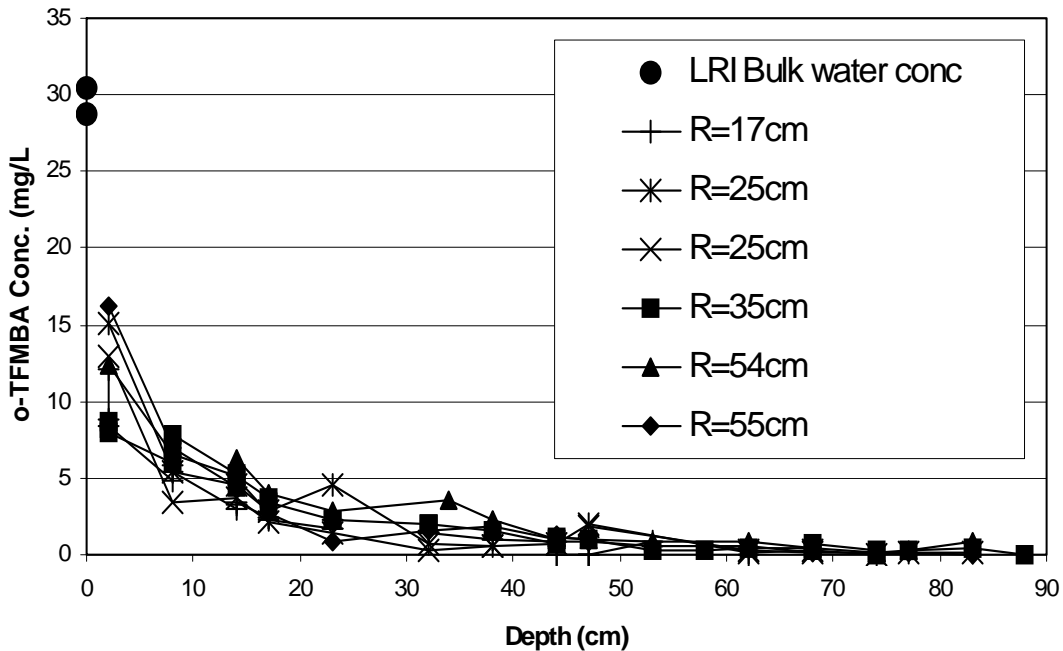
Figure 4. o-TFMBA concentration depth profiles radially away from center of LRI.

4 overlap, indicating that transport of o-TFMBA in the liner was less dependent on radial location than that of Br⁻.

As with Br⁻, an axially symmetrical analytical solution to the cylindrical coordinate system form of the advection dispersion equation from a localized source zone was applied to evaluate transport. However, unlike Br⁻ and tritium, o-TFMBA proved not to be a conservative tracer. In initial modeling simulations, concentration profiles which only considered advection and dispersion dramatically overestimated the field CDPs. As a result, other factors were evaluated to account for the discrepancy. Bowman and Gibbens (1992) found that o-TFMBA can degrade under unsaturated field conditions. Bowman and Rice (1986) observed a 26% mass loss of o-TFMBA over a sampling period of approximately 114 days during intermittent flood irrigation. Hornberger (1990) found that o-TFMBA was not conserved in a Maine hillslope study. Bentley (1983) found that o-TFMBA readily degraded under aerobic conditions. From these studies, it follows that microbial degradation could be responsible for the observed discrepancy in mass. To simulate possible microbial degradation, the degradation rate constant was adjusted to obtain the best fit CDPs for o-TFMBA while V_x was held constant at the value determined from Br⁻, and D^* was evaluated based on $D^*=D_{mol}*\tau$, where τ is 0.25 (Table 3). The best-fit reaction rate constant was found to be 1.39 yr⁻¹ which corresponds to a half-life of ½ yr. Analysis of o-TFMBA revealed that

in cases where degradation of contaminants or tracers occur in low permeability CCLs, very low concentrations (Figure 4) occur at the base of the liner and hence, the flux exiting the liner can be negligibly low.

Conclusions

A field-scale soil liner was constructed over 12 years ago. Bromide and three benzoic acid tracers were ponded inside the LRIs on the liner surface, while tritiated water was ponded on the liner surface outside of the LRIs. Previous work (Toupiol et al. 2002) demonstrated that vertical migration of tritium was uniform, and that diffusion controlled the overall rate of transport (i.e. advective transport was negligible). Willingham et al. (2003) evaluate transport of Br⁻ from the localize source zone inside the LRI. Results from the analysis of Br⁻ indicate that transport was spatially uniform, and governed by diffusion. In addition, the analysis of Br⁻ allowed independent evaluation of the vertical and radial diffusion coefficient and indicated that the effective diffusion coefficient was the same in both the vertical and radial directions. For both tritium and bromide, best-fit profiles were obtained with effective diffusion coefficients that were equal to the molecular diffusion coefficient multiplied by the same tortuosity factor (within 8%), indicating that the governing mechanisms of transport were the same for both tracers. Analysis of the o-TFMBA CDPs revealed that over the life of the experiment, o-TFMBA was not a conservative tracer and hence is not a

good tracer for long term studies. Since o-TFMBA is not a conservative tracer, independent quantitative analysis of transport properties could not be performed. Instead, the effective diffusion coefficient and average liner velocity were fixed based on results from tritium and Br⁻ and the reaction rate constant was varied to obtain adequate modeling fits. From the analysis of o-TFMBA, it is observed that the CDPs of microbially degradable constituents can decrease in concentration very rapidly, and hence, the flux of biodegradable constituents from liners can be significantly reduced if adequate retention time is obtained.

Results from this study indicate that: (1) current engineering practices can be used to construct diffusion controlled barriers that are capable of mitigating chemical transport from localized leaks or source zones; (2) transport of tritium and Br⁻ were governed by diffusion and were spatially uniform (e.g. no preferential transport was detected); (3) the effective diffusion coefficient for Br⁻ transport in the vertical and horizontal directions were the same; (4) the flux of biodegradable constituents from liners can be significantly reduced if adequate retention time in the liner is obtained and (5) o-TFMBA transport through the field-scale earthen liner was controlled by microbial degradation and diffusion.

Evaluation of chemical transport through compacted clay liners is essential to understanding transport through composite liner systems. Breach of the geomembrane protection layer of a composite liner can occur due to membrane and construction deficiencies such as holes in the geomembrane seams from the welding process, creasing of the geomembrane during placement of the leachate collection and recovery system, equipment operator error (insufficient cover to distribute equipment weight/lack of low ground pressure equipment), punctures due to rocks or protrusions from the CCL or from long-term stresses such as settlement due to waste placement. This breach of the geomembrane protection layer can create localized source zones on the surface of the CCL. Hence, results from this study are especially important because they indicate that best engineering practices can be used to construct a composite soil liner that acts as a diffusion barrier to contaminants from a localized source zone. Previous work has not been able to demonstrate this because this is the first time that transport in both the vertical and horizontal directions from a localized source zone has been analyzed in a field-scale liner.

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