

Numerical Modeling of Diffusion for Volatile Organic Compounds through Composite Landfill Liner Systems

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Abstract

Mass flux of solute exiting a landfill liner system is a valuable indicator for evaluating the performance of the Municipal Solid Waste (MSW) landfill liner systems. For the volatile organic compounds, such as toluene, the diffusion occurring inevitably in the intact landfill liner systems can transport toluene through the liner system and finally contaminate the outer groundwater. The diffusion of toluene, through four different municipal solid waste landfill liner systems, i.e., Subtitle D composite liner system, composite liner system with a Geosynthetic Clay Liner (GCL) instead of low permeability compacted soil, Wisconsin NR500 liner system, and a proposed four-component composite liner system that is a combination of the GCL composite liner system and the Subtitle D liner (with a 61 cm (2 feet) or 91.5 cm (3 feet) thick compacted clay liner), were evaluated in terms of mass flux. A one-dimensional block-centered numerical model of the diffusive transport through the four intact liner systems was developed for this purpose. The comparison of mass flux shows that the proposed four-component composite liner system outperforms the other liner systems based on mass flux and can be a preferable alternative for a MSW composite liner system.

Keywords: *solid waste, leachate, composite liner system, geosynthetic clay liner, diffusion, contaminant transport*

1. Introduction

The evaluation of the performance of Municipal Solid Waste (MSW) landfill liner systems is usually based on leakage rate. Several studies suggest that the criterion of only leakage rate might not be sufficient for assessing the performance of composite landfill liner systems (Park and Nibras, 1993; Crooks and Quigley, 1984; Rowe, 1987; Shackelford, 1989; Shackelford and Daniel, 1991; Foose *et al.*, 2002) because advective flow is not the only mechanism of mass transport. Research shows that contaminant diffusion (contaminant migration caused by the difference in concentration between the top and bottom of the liner) is often the dominant mode of transport through engineered barriers including compacted low permeability soil liners (Shackelford, 1990; Toupiol *et al.*, 2002; Willingham *et al.*, 2004; Bezza and Ghomari, 2008), Geosynthetic Clay Liners (GCL) (Malusis and Shackelford, 2002, 2004; Rowe *et al.*, 2005), composite liners (Foose *et al.*, 2002; Kalbe *et al.*, 2002; Edil, 2003).

Three commonly used composite liner systems in MSW landfills are the Subtitle D liner (the liner prescribed in Subtitle D of

the Resource Conservation and Recovery Act, US EPA), the GCL composite liner (a popular alternative liner system to the Subtitle D system), and the Wisconsin NR500 liner (the liner prescribed in the Wisconsin Administrative Code Section NR500). The Subtitle D and the Wisconsin NR500 liners consist of a Geomembrane (GM) underlain by a low permeability compacted soil layer with thicknesses of ≥ 61 and ≥ 122 cm, respectively. The GCL composite liner consists of a Geomembrane (GM) underlain by a Geosynthetic Clay Liner (GCL). Foose *et al.* (2002) analyzed the performance of these three composite liners using estimates of leakage rate and mass flux. Cadmium and toluene were used by Foose *et al.* (2002) as typical inorganic and organic leachate constituents, respectively, in the landfill leachate. The results indicate that the GCL composite liner exhibits the lowest leakage rate and lowest mass flux of the inorganic substances, such as cadmium. However, the mass flux of organic substances, such as toluene, through the GCL composite liner is two to three orders of magnitude greater than that through the intact Subtitle D or Wisconsin NR500 liner systems owing to the small thickness, and thus small attenuation volume, of the GCL.

A four-component liner system comprised of a GM/GCL com-

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posite liner and a GM/ low permeability compacted soil layer (61 or 91.5 cm thick) has been recently used in several new landfill constructions to protect important groundwater resources and facilitate siting of an expansion or new facility. This liner system is believed to satisfy all performance requirements for a landfill liner and provide superior performance to the GCL composite liner, Subtitle D liner, and Wisconsin NR500 liner systems because it combines the benefits of these liner systems, e.g., two GMs, a GCL to reduce advective flow, and a thick layer of low permeability compacted soil to reduce diffusive flow. However, there was no published evidence to demonstrate the performance of this four-component composite liner system and its performance compared to the GCL composite liner, Subtitle D liner, and Wisconsin NR500 liner systems. Another widely used type of composite liner system is the double composite liner system. This is a combination of two single composite liner systems (i.e., the system of a GM and a GCL/compacted soil liner) and a leachate collection system between the two liners which can be analyzed separately. Therefore, the double composite liner system is not considered in this study.

There are many benefits of the proposed four-component composite liner system. In this system the GCL is installed in an unhydrated condition between the two geomembranes and thus should remain in the unhydrated state because it is encapsulated. Hydration may occur at locations of defects in the upper GM if leachate is present. If this occurs, the GCL will hydrate at the defect location and slow or eliminate the leachate flow. In addition, the area that will be hydrated will be small because the GCL will hydrate and seal the defect area with the normal stress applied by the overlying waste. The small area of hydration is beneficial for slope stability purposes because a large amount of the bentonite will remain unhydrated so a large area of reduced shear resistance will not occur within or outside of the GCL. This should result in the critical interface being at the GM/compacted soil liner interface instead of within the GCL or a GCL interface. Estornell and Daniel (1992), Daniel *et al.* (1993), Thiel and Erickson (2001), and Thiel *et al.* (2001) show that the amount of GCL hydration is negligible if the GCL is properly installed and protected/encapsulated by GMs which means a low frequency of small GM holes (i.e., 1 to 2 per acre) occur. In the unhydrated state the GCL also will exhibit better durability and less bentonite migration (Stark *et al.*, 2004).

This study compares the performance of the proposed four-

component composite liner system with other composite liner systems based on the diffusion of Volatile Organic Compounds (VOCs) through the liner systems. A one-dimensional block-centered numerical model was developed to estimate the diffusive mass flux through the four intact composite liner systems (i.e., the GCL composite, Subtitle D, Wisconsin NR500, and four-component liner systems).

2. Review of Existing Solute Transport Analyses

The diffusion of VOCs through intact composite liner systems is described by Mueller *et al.* (1998), and Brown and Thomas (1998). The organic solute transport mechanism is also analyzed by Foose *et al.* (2002) using a numerical approach for intact composite liners. For the organic solute transport analysis, the mass transport through defects is negligible as compared with that through the intact portion of composite liner (Shackelford, 1990; Toupjol *et al.*, 2002; Foose *et al.*, 2002; Kalbe *et al.*, 2002; Malusis and Shackelford, 2002; Edil, 2003; Willingham *et al.*, 2004; Malusis and Shackelford, 2004; Rowe *et al.*, 2005; Bezza and Ghomari, 2008). The diffusion of VOCs through the intact composite liners can be analyzed using a one-dimensional model because the width of the liner is much greater than the thickness. One-dimensional models of organic solute transport in porous media with an infinite thickness are presented by Ogata and Banks (1961) and van Genuchten and Alves (1982). An analytical method for calculating the time to reach steady-state flux for diffusion of organic solute in a composite liner was suggested by Mueller *et al.* (1998). Foose *et al.* (2002) provide a numerical approach for analyzing organic solute transport through intact composite liners. The governing equation for diffusive transport of toluene through a composite liner system was solved herein using a Crank-Nicholson node-centered finite-difference algorithm developed by Foose (1997). Rowe and Booker (2005) developed a computer program POLLUTE v7 to simulate the contaminant transport through the double composite liner systems with the leachate collection system between the primary and secondary composite liner systems. The proposed four-component composite liner system considered herein was analyzed using a block-centered finite-difference model of diffusive transport through intact liners that was developed during this study. This model uses an explicit method to calculate the mass flux of toluene transported through the four liner systems considered.

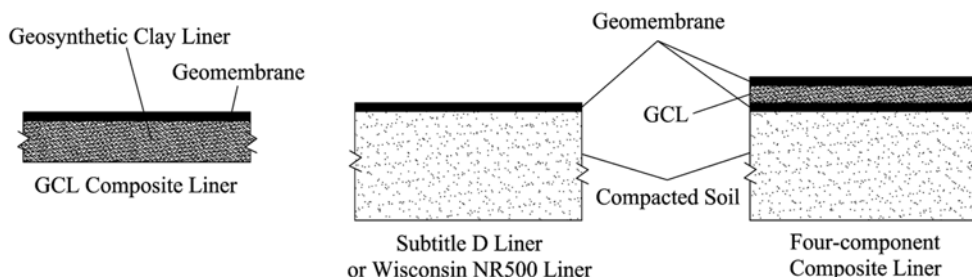


Fig. 1. Profile of GCL Composite, Subtitle D, Wisconsin NR500 Liner, and Proposed Four-component Composite Liner Systems

The four different composite liner systems considered and frequently used in practice are presented below (referring to Fig. 1) and the four liner systems consist of:

1. Subtitle D liner system with a 61 cm (2 feet) or 92 cm (3 feet) thick compacted low permeability soil layer overlain by a GM;
2. Wisconsin NR500 Liner which consists of a GM and underlain by a compacted low permeability soil layer with a thickness of 122 cm (4 feet);
3. GCL composite liner system with a GM underlain by a 6.5 mm thick GCL;
4. Proposed four-component composite liner system consists of four components from top to bottom: GM, 6.5 mm thick GCL, GM, and 61 cm (2 feet) or 92 cm (3 feet) of compacted low permeability soil layer.

3. Diffusive Transport Modeling for Organic Solute

Figure 2 shows the organic solute transport with initial leachate

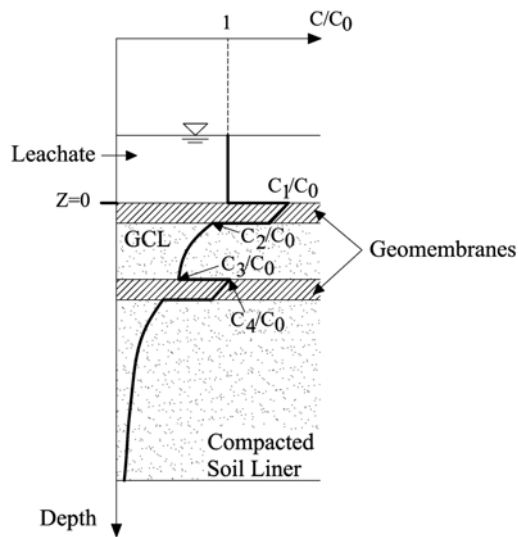


Fig. 2. Concentration Profile of Toluene Transport through the Intact Four-component Composite Liner

concentration C_o through the intact four-component composite liner system. The toluene as an organic contaminant in the leachate initially partitions into the upper GM (the concentration $C_1 = K_{d,gm} C_o$, where $K_{d,gm}$ = partitioning coefficient for the geomembrane and toluene), then diffuses downward through the upper GM and partitions back into the pore water at the base of the upper GM (the concentration C_2). Next, toluene diffuses through the GCL until partitioning again into the lower GM (the concentration $C_4 = K_{d,gm} C_3$). Subsequently, the transport process through the lower GM and the compacted low permeability soil liner is similar to that through the upper GM and GCL.

The mass flux of toluene transported through the intact four-component composite liner system is dominant than through defects because of the small area of defects considered compared to the area of the intact liner. Therefore, only the mass flux of toluene transported through an intact four-component composite liner system is estimated. The block-centered model of organic solute transport through the intact four-component composite liner system was developed to solve the diffusive transport governing equations. The governing equations include the pure diffusion equations (i.e., no advection) for the GM and compacted soil layers and the equations for the continuities of solute flux and concentration at the interfaces between the GM and compacted soil layers. More information on the equations can be found in Foose *et al.* (2002). The concentrations of VOCs in the landfill leachate are usually less than 100 mg/L (Farquhar, 1989; Krug and Ham 1995). In this study, the initial leachate concentration C_o was assumed to be 100 $\mu\text{g/L}$. The other parameters for the modeling of diffusion transport of toluene through the composite liner systems are presented in Table 1.

Foose (1997) and Foose *et al.* (2002) solved the diffusive transport governing equations for the Subtitle D, GCL, and Wisconsin NR500 composite liners using a one-dimensional finite-difference model. The model adopts a node-centered grid system and the implicit scheme. The difficulty with this model is handling a sharp change in concentration at the interface between the GM and the compacted low permeability soil liner. Foose (1997) and Foose *et al.* (2002) overcame this difficulty using a normalized concentration and coordinate in the GM and imaginary concen-

Table 1. Parameters for Diffusive Transport Modeling of Toluene

	GM	GCL	Low permeability compacted soil liner	References
Partitioning coefficient, $K_{d,gm}$, of toluene into GM (phase change)	135			Park and Nibras (1993)
Diffusion coefficient, D_{gm} , of toluene for GM	$3.0 \times 10^{-9} \text{ cm}^2/\text{s}$			Park and Nibras (1993)
Distribution coefficient, K_d		2.6 mL/g	1.0 mL/g	Edil <i>et al.</i> (1995); Benson and Lee (2000)
Total porosity, n		0.70	0.54	Shackelford and Daniel (1991b); Kim <i>et al.</i> (1997); Benson <i>et al.</i> (1999)
Bulk density, ρ_b		0.79 g/cm ³	1.24 g/cm ³	Shackelford and Daniel (1991b); Estornell and Daniel (1992)
Apparent tortuosity, τ_a		0.074	0.24	Shackelford (1989); Rowe <i>et al.</i> (1997)
Free solution diffusion coefficient, D_o , for toluene		$8.47 \times 10^{-6} \text{ cm}^2/\text{s}$	$8.47 \times 10^{-6} \text{ cm}^2/\text{s}$	Yaws (1995)

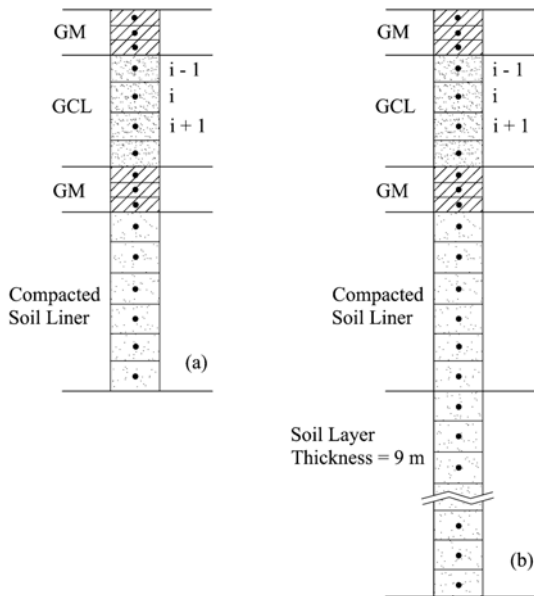


Fig. 3. Block-centered Model for Transport of Toluene through Intact Four-component Composite Liner: (a) Zero Concentration at Base, (b) Semi-infinite Bottom Boundary Condition

trations at the interface, which were removed in the calculation process. However, the three interfaces present between four components (the upper GM, GCL, lower GM, and soil liner) in the four-component composite liner causes a singular matrix in adopting the implicit method. Therefore, a block-centered formulation with an explicit solution scheme was developed. Each layer was divided into a sufficient numbers of blocks, which are represented by the properties denoted at the center of each block. This approach has an advantage that the concentration of solute can be directly calculated without any numerical difficulty (i.e., singular matrix appears in the implicit method). However, the selected time step should be small enough to avoid numerical instability of the solution. The continuities of solute flux and concentration at the interfaces between the GM and compacted low permeability soil layers are adopted as in Foose (1997) and Foose *et al.* (2002). A period of 100 years is used in the simulations performed herein. The two bottom boundary conditions for the block-centered models as shown in Fig. 3, were chosen as follows:

- The bottom boundary is located at the base of the liner. The constant concentration at the bottom boundary is zero. This condition accounts for the situation where the organic solute can be conveyed away by the flow of groundwater present at the base of liner.
- The bottom boundary is located 9 m downward from the base of liner, at which the concentration is set to zero. To apply this condition, the liner system is underlain by a 9 m thick layer of soil which has the same diffusion coefficient as the compacted soil liner and retardation factor, R_d , of unity. The bottom boundary is at the base of the additional soil layer, which represents the semi-infinite bottom boundary.

4. Comparison of Block-centered and Node-centered Models

Figures 4 and 5 present the diffusive transport results for the three commonly used intact composite liner systems (i.e., GCL, Subtitle D, and Wisconsin NR500 liner systems) with the two bottom boundary conditions. The mass fluxes of toluene for the three composite liner systems obtained by using the node-centered model developed by Foose *et al.* (2002) are compared with those that are calculated herein with the block-centered model. The comparison shows an excellent agreement between the block-centered and node-centered models and also verifies the block-centered formulation with an explicit solution scheme developed herein.

5. Diffusive Transport Results

For the case of a constant concentration of zero at the base of the liner systems, Fig. 6 shows that the proposed four-component composite liner is the most effective liner system in terms of mass flux of toluene after 100 years. The intact four-component composite liner allows the smallest amount of toluene diffusion

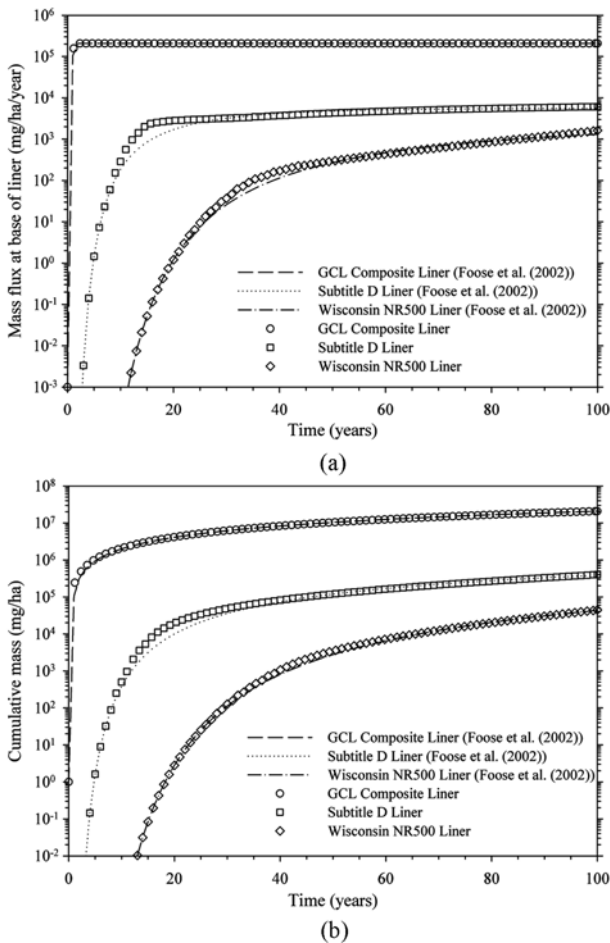
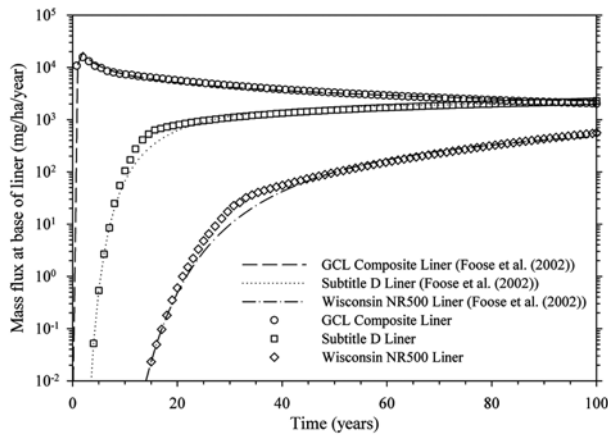
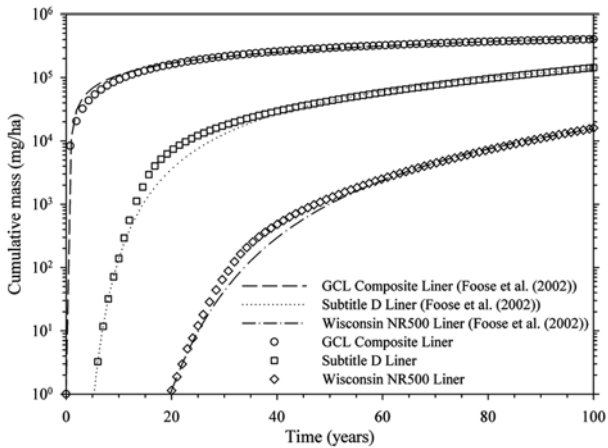


Fig. 4. Verification of Block-centered Model for Transport of Toluene in Three Composite Liners with Zero Concentration at Base: (a) Mass Flux, (b) Cumulative Mass



(a)

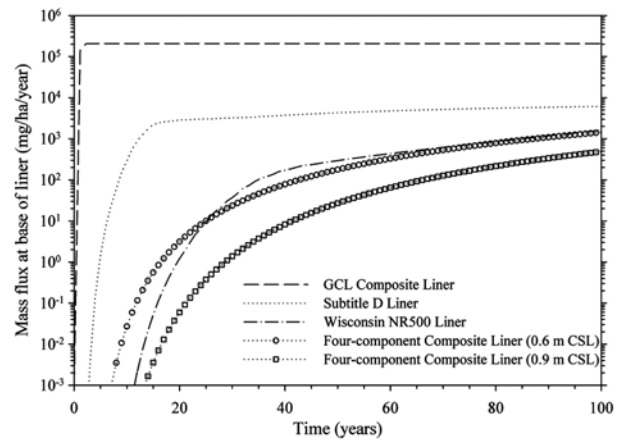


(b)

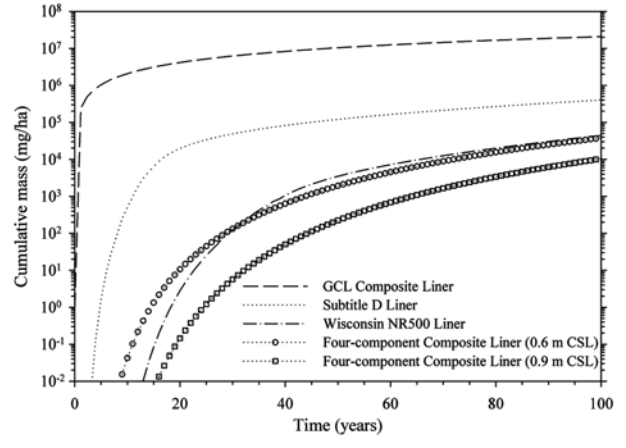
Fig. 5. Verification of Block-centered Model for Transport of Toluene in Three Composite Liners with Semi-infinite Bottom Boundary Condition: (a) Mass Flux, (b) Cumulative Mass

through among the composite liner systems considered. The mass fluxes of toluene through the intact four-component composite liner at the end of the simulation are 1,432 and 489 mg/ha/year for CSL layers having a thickness of 0.6 and 0.9 m, respectively. It was found that the four-component composite liner having a 0.6 m thick compacted soil liner has essentially the same mass flux as the Wisconsin NR500 liner, which has a GM overlain by 1.2 m thick compacted soil liner. Similar trends were obtained for cumulative mass through these four different liner systems (Fig. 6b). The cumulative mass for the four-component composite liner with 0.6 m of compacted soil liner is 37,735 mg/ha at 100 years while the value for 0.9 m of compacted soil liner in the four-component composite liner is 10,366 mg/ha.

For the case of a semi-infinite bottom boundary condition, which is represented by the bottom boundary being at a depth of 9 m below the base of the liner system, Fig. 7 shows that the proposed four-component composite liner is an extremely protective liner system for MSW landfills. The four-component composite liner with 0.6 m of compacted soil liner, the mass flux and cumulative mass after 100 years are even lower than those of the



(a)



(b)

Fig. 6. Transport of Toluene in Five Composite Liners with Zero Concentration at Base: (a) Mass Flux, (b) Cumulative Mass

Wisconsin NR500 liner system. In this case, the mass fluxes at the end of the simulation are 445 and 153 mg/ha/year for the cases of CSLs having thicknesses of 0.6 and 0.9 m, respectively. The cumulative masses are 11,678 and 3,280 mg/ha at 100 years for the cases of 0.6 and 0.9 m of compacted soil liner in the four-component composite liner system, respectively. The addition of 0.3 m of compacted soil liner, for a total thickness of 0.9 m, further improves performance of the four-component composite liner in terms of diffusion of organic solutes. The mass flux and cumulative mass after 100 years for the case of 0.9 m of compacted soil liner are about 3 times lower than those for a 0.6 m of compacted soil liner in the proposed four-component composite liners. However, the four-component composite liner system is still the most protective liner system with only 0.6 m of compacted low permeability soil based on the mass flux of transported toluene.

6. Conclusions

The performance of the proposed four-component composite liner system was analyzed and compared with three other

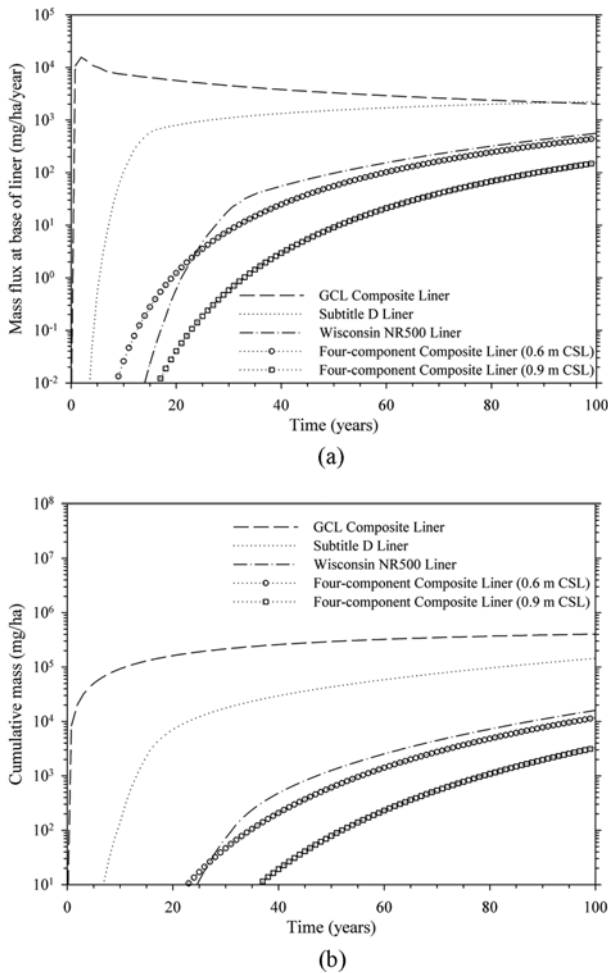


Fig. 7. Transport of Toluene in Five Composite Liners with Semi-infinite Bottom Boundary Condition: (a) Mass Flu, (b) Cumulative Mass

composite liner systems in terms of mass flux of VOC diffusion. The block-centered finite-difference models provide a useful means for evaluating the performance and protectiveness of these composite liner systems because there is limited field mass flux data published to date to evaluate these liner systems.

The mass flux and cumulative mass after 100 years of toluene transported through an intact four-component composite liner system is significantly smaller than the amounts through the other three composite liner systems considered. Under the identical conditions, the cumulative mass of toluene transported through the four-component composite liner system is around one to two orders of magnitude smaller than that through the GCL and Subtitle D composite liner systems. The four-component composite liner system with a 0.6 m thick compacted low permeability soil layer at the bottom even has equivalent mass flux and cumulative mass of transported toluene to that of the Wisconsin NR500 liner, which consists of a GM and 1.2 m of compacted soil liner. The proposed four-component composite liner system with a 0.9 m thick compacted low permeability soil layer at the bottom has smallest mass flux and cumulative mass

of transported toluene and thus exhibits the best performance of the composite liner systems considered. With the above advantages, it can be concluded that the proposed four-component composite liner system represents a more protective design for MSW landfills than the other three composite liner systems considered. Diffusive transport analyses for other VOCs through the four-component composite liner system are being performed to evaluate the effectiveness of this system.

Notations

- D_{gm} : Diffusion coefficient of toluene through geomembrane
- D_o : Free solution diffusion coefficient of toluene
- K_d : Distribution coefficient for soil liner and toluene
- $K_{d, gm}$: Partitioning coefficient for geomembrane and toluene
- n : Porosity
- ρ_b : Bulk density of soil liner layer
- τ_a : Apparent tortuosity

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