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ABSTRACT

Numerical and analytical models were used in this study to simulate contaminant transport of toluene in composite liners. Methods for demonstrating equivalency based on contaminant transport are also presented. Toluene was selected for this study because the transport properties of toluene are representative of those for volatile organic compounds (VOCs) commonly found in landfill leachate. Results from the model indicate that the mass flux of toluene through a GCL composite liner is as much as two orders of magnitude greater than that through a composite liner with a 61 cm thick soil liner (e.g. a Subtitle D liner) because a GCL composite liner is thinner. Performance of a GCL composite liner can be improved by adding an additional soil layer beneath the GCL, which reduces the concentration gradient across the base of the liner system. For the materials considered in this study, an additional 90 cm is sufficient to reduce the mass flux and increase the contaminant breakthrough time through the GCL composite liner to be comparable to that for a Subtitle D liner. A thinner layer can be used depending on the material properties of the soil and in particular, the porosity of the layer.

INTRODUCTION

Composite liners having a GCL are a popular alternative to the Subtitle D composite liner required in municipal solid waste landfills. A GCL composite liner typically consists of 5 to 6.5 mm of bentonite having low hydraulic conductivity overlain with a geomembrane liner. The Subtitle D liner consists of 61 cm of compacted clay having a hydraulic conductivity $< 1 \times 10^{-7}$ cm/s overlain with a geomembrane. Designers and owners are inclined to propose using GCL composite liners because suitable clayey soil may not be available, the GCL composite liner uses less air-space in the landfill, and building a composite liner with a GCL may be more economically viable.

Subtitle D of the Resource Conservation Recovery Act includes a provision that permits an alternative liner, provided that that the liner is equivalent to the Subtitle D liner. Traditionally, comparisons of liner systems have been made based on leakage rate through defects, with equivalency being defined as a liner system having a leakage rate less than or equal to that for the Subtitle D liner. However, leakage rates are not always indicative of performance of composite liner (Foose 1997). In fact, diffusion can be a significant mechanism of contaminant transport in soil liners and geomembranes (e.g. Shackelford 1990 and Park and Nibras 1993).

As shown in Fig. 1, VOCs can migrate through composite liners via advection and diffusion through defects in the geomembrane and subsequent advection and diffusion through the soil liner. Additionally, VOCs can diffuse through intact geomembranes (Park and Nibras 1993) and then diffuse through the soil liner (Fig. 1). In contrast to contaminant transport through defects, diffusion of VOCs can occur over the entire footprint of the liner system. Thus, mass flux of VOCs in the intact composite liner has the potential to be several orders of magnitude greater than that through defects, depending on the constituents of the leachate, the hydraulic conductivity of the soil liner, and the frequency, type, and size of defects.

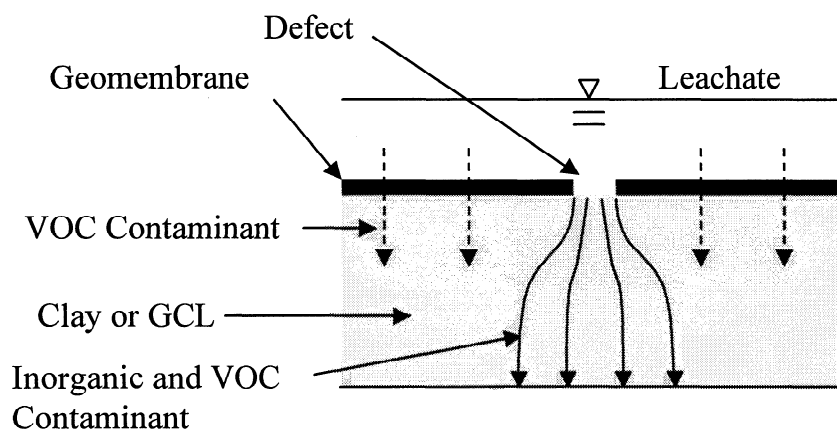


Figure 1. Contaminant Transport Processes in Composite Liners.

An analysis was conducted by Foose et al. (1996) using numerical models for predicting mass flux of a conservative inorganic contaminant in an EPA Subtitle D liner and composite liner having a GCL. Foose et al. (1996) found that the mass flux from defects in the composite liner having a GCL was greater than that in the EPA Subtitle D liner for the conditions analyzed.

A comparison of mass flux of cadmium and toluene through an EPA Subtitle D liner having few defects was performed by Foose (1997) using numerical models. The composite liner was assumed to be constructed with good quality control as defined by Giroud et al. (1992). The concentration of both cadmium and toluene were assumed to be the same. Results from analysis showed that the mass flux of toluene is nearly 5 orders of magnitude greater than that of cadmium. The reason that mass flux of toluene through the intact liner is much greater

than cadmium ion through defects is that the surface area over which transport of toluene occurs is approximately 5 orders of magnitude greater than the area of defects. Thus, provided that the flow rate through defects is small, the dominant pathway for transport of toluene is through the intact liner. Therefore, the remainder of this paper will focus on diffusive transport of VOCs through intact composite liners and transport through defects will not be considered further.

ASSESSING EQUIVALENCY

To compare alternative liner systems based on contaminant transport, the location at which the performance of the liner system will be evaluated must be selected. This location can be at some off-site monitoring well where the impact of the landfill is judged based on exceeding a particular maximum contaminant level (MCL) or immediately underneath the liner system. In many cases, however, the base of the liner system is used because it is usually at the same elevation regardless of the design of the liner. Ultimately, an analysis of groundwater contaminant transport should be conducted to assess how alternative liners impact groundwater quality.

Because a GCL composite liner is thinner than a Subtitle D liner, the contaminant breakthrough time through a GCL composite liner is less than that for a Subtitle D liner. Additionally, the mass flux from a GCL composite liner is greater than that from a Subtitle D liner because the location at which the flux is calculated is closer to the contaminant source for the GCL composite liner. Analyses demonstrating these conditions will be presented in a subsequent section.

When a GCL composite liner is being considered because sufficient clay is not available, the mass flux can be reduced and the breakthrough time increased by adding an additional layer of soil beneath the GCL. The additional layer of soil, which effectively is part of the liner, increases the distance between the source of the contaminant and the base of the liner. As a result, the concentration gradient at the base of the liner decreases and distance of travel increases. An appropriate thickness of the additional soil layer depends on the type of boundary condition existing at the base of the liner, characteristics of the additional soil layer, and the VOC being contained.

COMPOSITE LINER MODELS

Transient Models

Transport of VOCs through composite liners is different than that through porous media because the geomembrane component is not porous. VOCs diffuse through the interstitial spaces between polymer molecules in the geomembrane in a three-step process (Fig. 2): (1) partitioning into the geomembrane, (2) diffusing through the geomembrane, and (3) partitioning out of the geomembrane (Park and Nibras 1993). After the VOC is transported

through the geomembrane, the contaminant diffuses through the soil liner in the same manner that diffusion occurs in porous media.

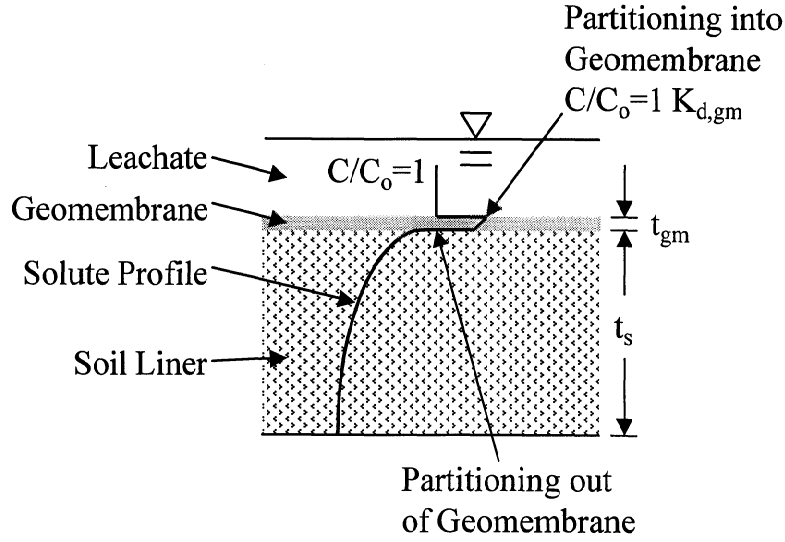


Figure 2. Transport Process for VOCs in Composite Liners.

Because of this multi-step transport process and combination of solid and porous media, application of analytical solutions is limited. Therefore, a numerical model was developed for analyzing transport of VOCs in composite liners. The numerical model includes the three-step process for diffusion of VOCs through geomembranes and maintains continuity of flux at the interface of the geomembrane and soil liner. Linear and reversible sorption and desorption are assumed for the geomembrane based on the findings of Park and Nibras (1993) who show that adsorption and desorption of VOCs onto geomembranes is approximately linear for aqueous concentrations < 100 mg/L.

If transport of VOCs is assumed to be spatially invariant, the governing equations for diffusive transport of VOCs through composite liners are:

$$\frac{\partial C}{\partial t} = D_{gm} \frac{\partial^2 C}{\partial z^2} \quad 0 \leq z \leq t_{gm} \quad (1)$$

$$R_d \frac{\partial C}{\partial t} = D^* \frac{\partial^2 C}{\partial z^2} \quad t_{gm} \leq z \leq t_{gm} + t_s \quad (2)$$

where C is the concentration of solute, t is time, D_{gm} is the diffusion coefficient of the solute in the geomembrane, z is the direction through the profile of the liner, R_d is the retardation factor, D^* is the effective diffusion coefficient of the solute in soil, t_{gm} is the thickness of the

geomembrane, and t_s is the thickness of the soil layer. Linear and reversible sorption and desorption were assumed to apply to the sorption of the solute onto the soil.

Continuity of mass flux at the interface between the geomembrane and soil liner is described by:

$$D_{gm} \frac{\partial C}{\partial z} = D^* n \frac{\partial C}{\partial z} \quad z = t_{gm}, t \geq 0 \quad (3)$$

where n is the porosity of the soil. In this study, it was assumed that the total porosity was equal to the effective porosity. Additional soil layers can be included by applying Eq. 2 for each soil layer and maintaining continuity of flux through the soil layers using equations similar to Eq. 3.

Initially, the solute concentration throughout the entire domain is assumed to be zero:

$$C = 0 \quad z \geq 0, t = 0 \quad (4)$$

and the concentration of the source was assumed to be a constant:

$$C = C_0 \quad z = 0, t \geq 0 \quad (5)$$

where C_0 is the concentration of the leachate. A constant source concentration was used because insufficient field data exist to support a more complicated boundary condition (Rowe 1987, Shackelford 1990).

Comparison of mass flux at the base of the liner requires proper characterization of the bottom boundary condition. Two bottom boundary conditions can be used to bracket the range of field conditions: (1) constant solute concentration at the base of the liner system and (2) a zero solute concentration and zero flux condition infinitely far from the base. The latter case corresponds to a liner system founded on a very thick saturated subgrade.

The bottom boundary was considered to be a constant concentration of zero in this study. The location of the bottom boundary was varied from immediately at the base of the composite liner system to 9 m from the base of the liner system. The latter case approximated a semi-infinite boundary for simulations of 100 years. For example, the concentration gradient at 100 years at a depth of 9 m is 3×10^{-15} mg/L/cm, for a Subtitle D liner, which results in a contaminant flux of essentially zero.

The governing equations were solved using a Crank-Nicholson node-centered finite-difference algorithm. Details regarding development of the composite liner model can be found in Foote (1997).

Steady-State Models

The steady-state diffusive mass flux (J_D) through composite liners in which the base of the liner system is maintained at a constant concentration can be calculated using Fick's first law:

$$J_D = D_{eq} \left(\frac{\Delta C}{t_{gm} + t_s} \right) \quad (6)$$

where D_{eq} is the equivalent diffusion coefficient for the composite liner, and ΔC is the difference in concentration between the solute source and the concentration at the base of the liner system. The equivalent diffusion coefficient (D_{eq}) can be obtained using an approach similar to that for determining the hydraulic conductivity of soil layers in series (Foose 1997). The equivalent diffusion coefficient is:

$$D_{eq} = \frac{t_{gm} + t_s}{\frac{t_{gm}}{D_{gm} K_{d,gm}} + \frac{t_s}{D^* n}} \quad (7)$$

where $K_{d,gm}$ is the partition coefficient for the contaminant and the geomembrane. The partition coefficient of the solute and the geomembrane appears in the denominator of the geomembrane term in the denominator because the solute diffuses in the geomembrane at a higher concentration than the solute source (Fig. 2). Additional soil layers can be incorporated by adding the thickness of the additional soil layer to the numerator and including another term for the additional soil layer in the denominator.

EQUIVALENCY OF COMPOSITE LINERS HAVING A GCL

The transport properties of liner systems and the constituents of leachate can vary significantly. In practice, equivalency comparisons between liners should be based on site-specific data whenever possible. In this study, however, representative material properties were selected (see Table 1) from a summary of material properties contained in Foose (1997). Toluene was the VOC analyzed because it is commonly found in landfill leachate and the properties of toluene are representative of other VOCs. Parameters for the compacted soil liner were taken from data compiled by Foose (1997) for compacted clays. The effective diffusion coefficient for toluene in the GCL is based off of measurements of apparent tortuosity by Shackelford (1989) for a smectitic clay and the porosity is representative of properties of GCLs reported by Ruhl and Daniel (1997). The total and effective porosity were assumed to be equal for all soils in this analysis.

The partition coefficient for toluene and the GCL was assumed to be equal to that for the compacted soil liner because there is a lack of data regarding sorption of VOCs onto GCLs.

Material properties for the additional soil layer are representative of a sandy soil (Foose 1997). The diffusion and partition coefficients for the geomembrane were taken from Park and Nibras (1993). The thickness of the geomembrane was assumed to be 1.5 mm. The thickness of the GCL was assumed to be 6.5 mm and the thickness of the compacted soil liner was 61 cm.

Table 1. Properties used in Analysis of Equivalency.

| Material Property | Compacted Soil Liner | Geosynthetic Clay Liner | Additional Soil Layer | Subgrade Soil | Geomembrane |
|--|----------------------|-------------------------|-----------------------|----------------------|----------------------|
| Effective Diffusion Coefficient (cm^2/s) | 2.0×10^{-6} | 2.0×10^{-6} | 2.4×10^{-6} | 2.0×10^{-6} | 3.0×10^{-9} |
| Retardation Factor | 3.3 | 1.7 | 1 | 1 | - |
| Porosity | 0.54 | 0.75 | 0.32 | 0.54 | - |
| Partition Coefficient (mL/g for Soils) | 0.7 | 0.82 | 0 | 0 | 135 |

Equivalency Based on Concentration at the Base of the Liner System

The numerical model was used to evaluate the concentration at the base of a liner as a function of time. Concentration at the base of the three liner systems founded on a thick subgrade versus time is shown in Fig. 3. As shown in Fig. 3, the concentration at the base of the GCL composite liner is greater than that for the Subtitle D liner for all times. If an additional layer of soil 80 cm thick is added beneath the GCL composite liner, the concentration is similar to that for the Subtitle D liner and, at 100 years is less than that for the Subtitle D liner (Fig. 3). For the concentration at the base of the GCL composite liner to always be less than that for the Subtitle D liner, an additional 120 cm of additional soil is required.

Concentration at the base of the liner is primarily a function of the diffusion coefficient of the soil and the retardation factor. These parameters can vary significantly with soil type. Hence, the thickness of soil required to maintain a lower concentration at the base of a GCL composite liner may vary considerably. This is particularly true if sorptive capacity of the Subtitle D liner is much greater than that for the GCL composite liner because the additional sorptive capacity will increase the contaminant breakthrough time.

Equivalency Based on Mass Flux

The steady-state mass flux, calculated using Eqs. 6 and 7 and a concentration of zero at the base of the liner is shown in Fig. 4 for the Subtitle D liner and the GCL composite liner. The mass flux from the GCL composite liner is nearly two orders of magnitude greater than that from the Subtitle D liner. However, adding an additional 50 cm of soil beneath the GCL results in comparable steady-state mass flux for the GCL composite and Subtitle D liners.

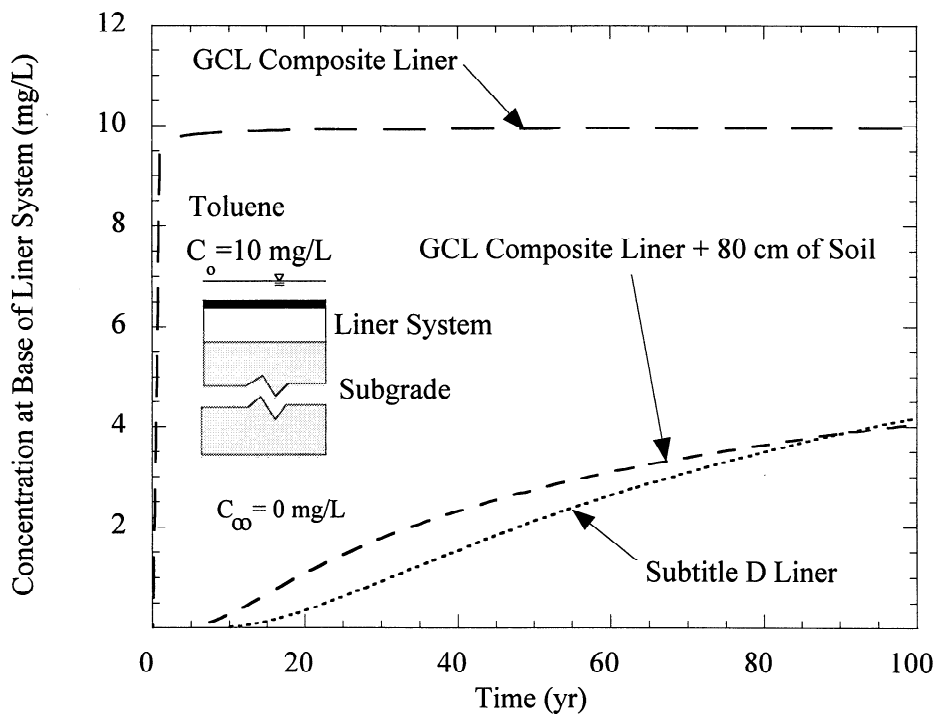


Figure 3. Concentration at Base of Liner Systems.

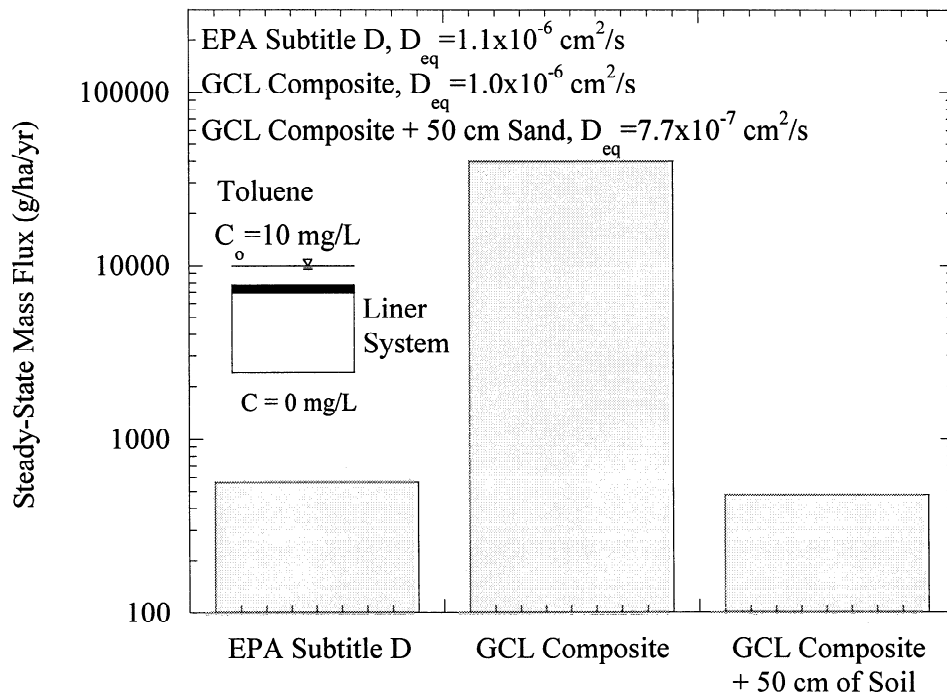


Figure 4. Steady-State Mass Flux through Liner Systems Analyzed.

The effective diffusion coefficient and the porosity of the soil impact the steady-state mass flux through the composite liner (Eq. 6). Decreasing the porosity of the additional soil layer decreases D_{eq} and reduces the mass flux. Thus, a thinner additional soil layer may be possible if the soil is more densely compacted. In addition, D^* of the soil should decrease as the pore space becomes smaller, which will also lower the mass flux.

Graphs of mass flux of toluene versus time for the two liner systems founded on a thick subgrade are shown in Fig. 5. The mass flux for the GCL composite liner is greater than that from the Subtitle D liner at all times. However, if an additional 90 cm of soil is added to the base of the GCL composite liner, the mass flux from the liners is similar for all times and at 100 years, mass flux from the GCL composite liner is less than that for the Subtitle D liner.

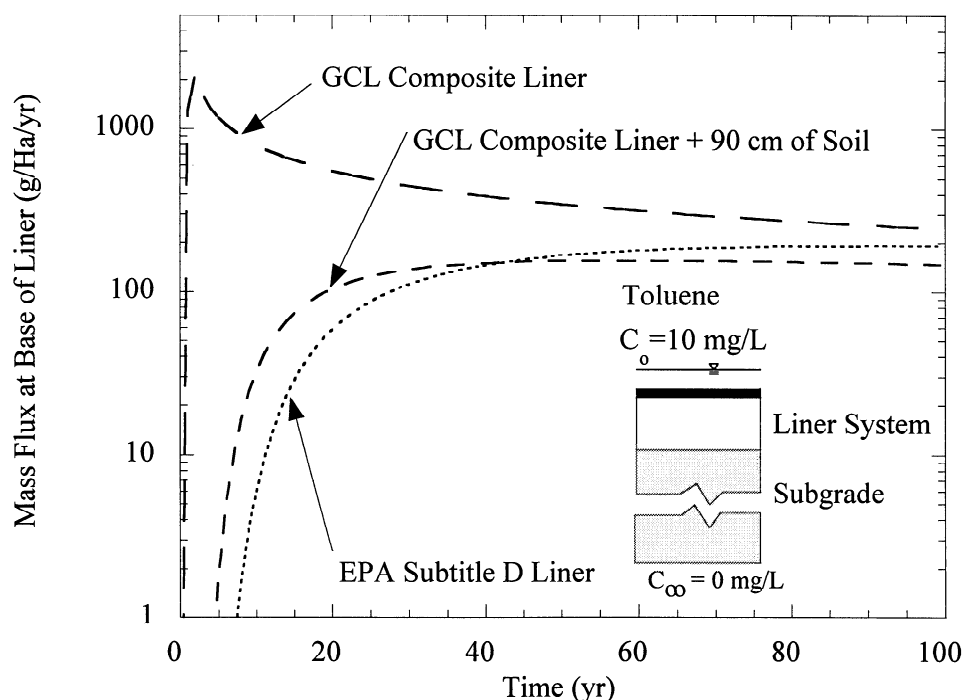


Figure 5. Mass Flux vs. Time through Liner Systems Analyzed.

Because of the peak in mass flux at early times for the GCL composite liner, comparisons based on cumulative mass discharged provide a better comparison. Cumulative mass discharged versus time for toluene for the GCL composite liner, Subtitle D liner, and the GCL composite liner with an additional layer of soil is shown in Fig. 6. The cumulative mass discharged from the GCL composite liner is the greatest because the mass flux during the first 20 years is much greater than that through the GCL composite liner with an additional layer of soil and the Subtitle D liner. Adding 90 cm of soil to the GCL composite liner results in slightly lower cumulative mass discharged at 100 years for the GCL composite liner than that for the Subtitle D liner.

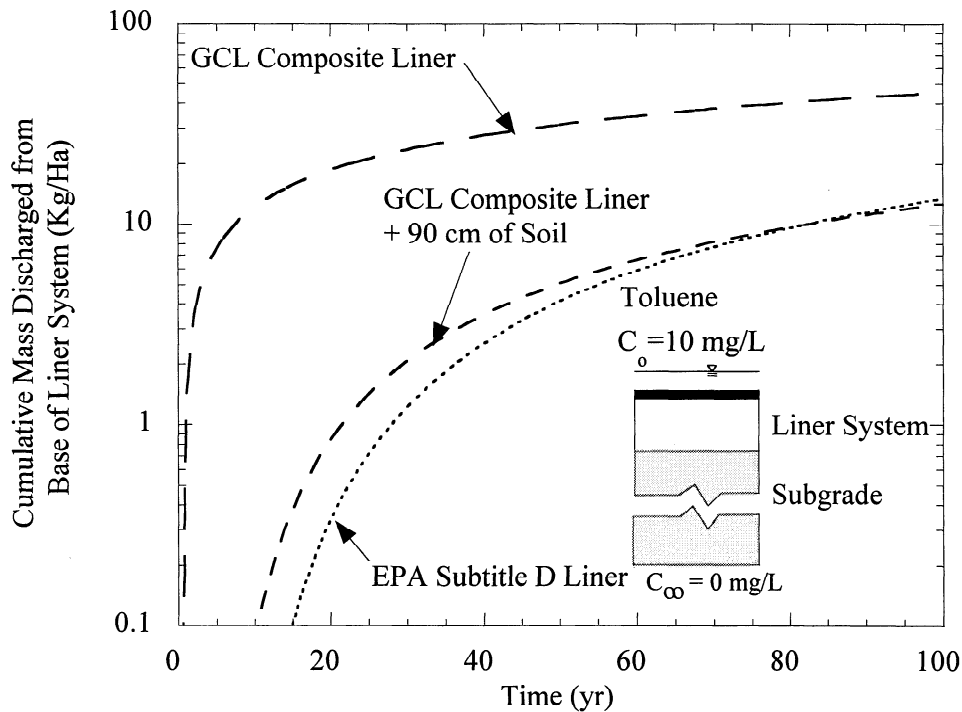


Figure 6. Cumulative Mass Discharged vs. Time through Liner Systems Analyzed.

The results in Figs. 3 through 6 show that an additional soil layer 50 to 90 cm thick is required for a GCL composite liner to be equivalent to a Subtitle D composite liner, with the different thickness corresponding to different equivalency criteria (50 cm based on steady-state mass flux, 80 cm based on contaminant breakthrough, and 90 cm based on mass flux). Thus, adding 90 cm of additional soil beneath the GCL will render the GCL composite liner equivalent or superior to the Subtitle D liner for the conditions that were modeled.

METHOD FOR EVALUATING EQUIVALENCY USING SITE-SPECIFIC MATERIAL PROPERTIES

The results presented in Figs. 3 through 6 are for the assumed material properties listed in Table 1. These properties do not represent the entire range of possible material properties and thus, the 90 cm of additional soil obtained from this analysis may be inadequate. Therefore, analyses should be conducted on a site-specific basis, using site-specific material properties. Unfortunately, there is a lack of tools applicable for analysis of transient transport of VOCs in composite liners. However, analyses of contaminant transport through composite liners using existing one-dimensional models, such as POLLUTE (Rowe and Booker 1983) or analytical solutions, can be conducted provided that the method of analysis is adjusted appropriately.

In the following analysis, an analytical solution to Eq. 2 for calculating mass flux at a particular distance from the contaminant source presented by Shackelford (1990) was used:

$$J_D = D^* n C_o \left[\frac{\exp(-\xi^2)}{\sqrt{\frac{\pi D^* t}{R_d}}} \right] \quad (8)$$

where ξ is:

$$\xi = \frac{t_s}{2 \sqrt{\frac{D^* t}{R_d}}} \quad (9)$$

In a composite liner the geomembrane serves to limit the flow of liquid (i.e. advective transport). In terms of diffusion of VOCs, the geomembrane is relatively thin in comparison to the Subtitle D liner or GCL and thus can be neglected without significant error. Results shown in Fig. 7 for mass flux of toluene in a composite liner having a GCL are from an analytical solution to Eq. 2 and the material properties of the subgrade and a seepage velocity of zero. As shown in Fig. 7, the effect of neglecting the geomembrane and GCL in the analysis of diffusion of VOCs in composite liners having a GCL results in a slightly higher and earlier peak mass flux.

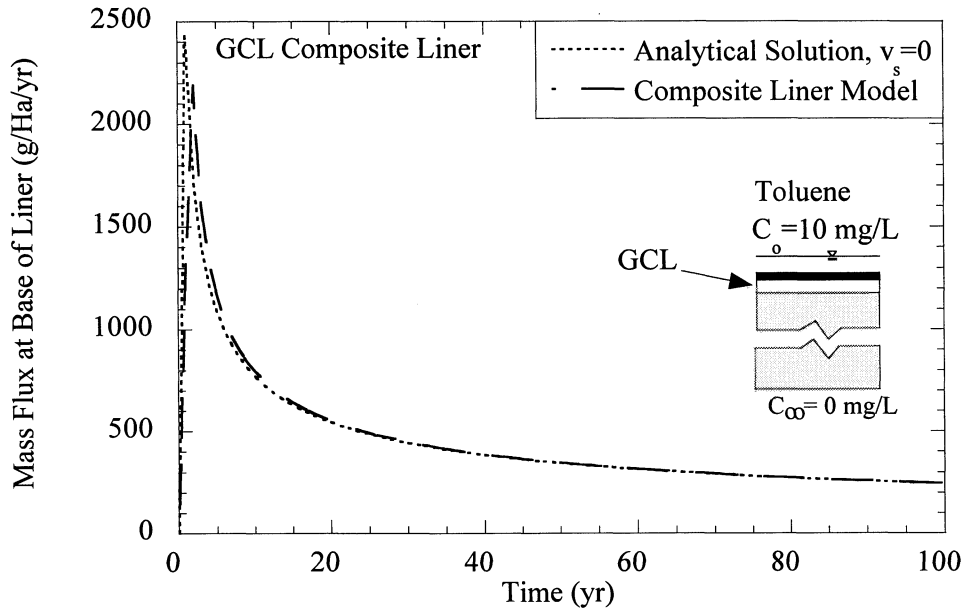


Figure 7. Mass Flux Predicted in a GCL Composite Liner using the Numerical and Analytical Models.

For the Subtitle D liner (Fig. 8), the comparison is not as favorable. The analytic solution for the case of zero seepage velocity and no geomembrane tends to over-predict the mass flux of toluene in the Subtitle D composite liner. The properties of the compacted clay were used in the analytical analysis. Results from Eqs. 8 and 9 tend to over-predict the mass flux in the EPA Subtitle D liner because the chemical gradient is overestimated. The chemical gradient is overestimated because the compacted clay liner is assumed to be semi-infinite and sorption beneath the composite liner (the subgrade) tends to reduce the solute concentration and hence, increase the chemical gradient. Nevertheless, the error from using Eqs. 8 and 9 is conservative in that the mass flux is over-predicted.

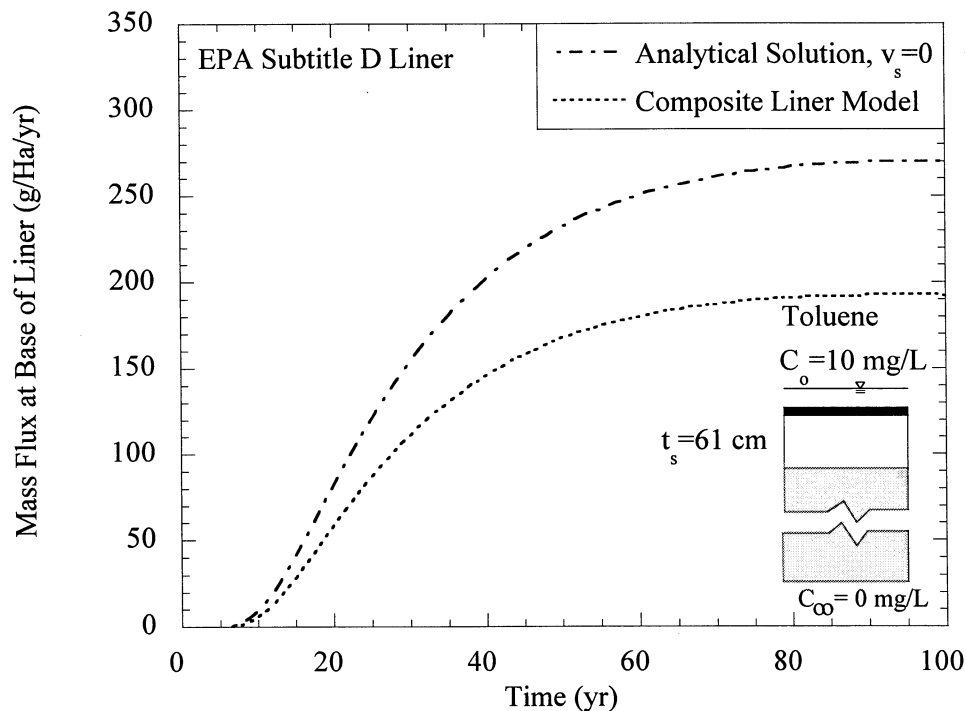


Figure 8. Mass Flux Predicted in an EPA Subtitle D Liner using the Numerical and Analytical Models.

A more detailed analysis can be conducted using POLLUTE (Rowe and Booker 1983) using a seepage velocity of zero. In addition, the geomembrane should not be included in the analysis. Because POLLUTE can be used for multiple layered systems, mass fluxes predicted using POLLUTE are likely to be closer to those from the numerical model because the profile of the liner and underlying soils can be represented in greater detail.

SUMMARY AND CONCLUSION

The current approach for evaluating equivalency of GCL composite and Subtitle D liners based on leakage rate through defects in the geomembrane is inadequate because VOCs can diffuse through intact composite liners. In fact, mass flux of VOCs through the intact liner can

be as much as 5 orders of magnitude greater than that through defects. A more appropriate approach is to employ numerical models and analytical solutions that simulate contaminant transport through intact composite liners.

Simulations conducted using a numerical model specifically developed to simulate VOC transport through composite liners showed that mass flux of toluene through a GCL composite liner can be two orders of magnitude greater than the mass flux through a Subtitle D liner. The mass flux is higher because the GCL composite liner is thinner than the Subtitle D liner and the concentration gradient near the base of the GCL composite liner is greater than that near the base of the Subtitle D liner.

Performance of GCL composite liners can be improved by adding an additional soil layer beneath the GCL. The additional layer reduces the concentration gradient across the base of the liner (which is the base of the additional layer of soil) and results in a decrease in mass flux. The additional soil layer does not have to meet the stringent requirements for compacted soil barriers with regard to hydraulic conductivity and compaction. For soils having diffusion coefficients, retardation factors, and porosity similar to those considered in this study, the additional thickness should be between 50 to 90 cm. In effect, the additional soil layer makes the GCL composite liner as thick as the Subtitle D liner.

The thickness of the additional soil layer needed varies with the diffusion coefficient, retardation factor, and porosity of the soil. The equation for steady-state mass flux (Eq. 6) in a composite liner using an equivalent diffusion coefficient can be used with site-specific material properties to determine a site-specific additional thickness. Additionally, traditional analytical and semi-analytical solutions (Eq. 8) for the advection-dispersion equation can be used to estimate the mass flux provided that the seepage velocity is set to zero. Such an analysis can be readily conducted on a personal computer using a commercially available spreadsheet program.

ACKNOWLEDGEMENT

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