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Analytical Model for Contaminant Transport in Composite Liners Considering the Longevity of Barrier Components

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Abstract The pollution from landfills due to the leakage of leachate is an important source of groundwater contamination. The service life of geomembrane and high-permeability leachate collection system, which play a crucial role in the long-term effectiveness of landfill liner systems, can be greatly shortened due to the complex environmental conditions of landfills. The one-dimensional analytical model of contaminant transport in geomembrane and compacted clay composite liners considering the service life of geomembrane and leachate collection system is presented. Influences of the service life of geomembrane on contaminants transport are described by changing boundary conditions. Effects of the service life of leachate collection system are achieved by the fluctuated leachate head. An investigation into the influence of geomembrane and leachate collection system service life on the breakthrough of the composite liner is presented. An empirical formula for the prediction of breakthrough time considering the service life of geomembrane and leachate collection system is proposed, which may provide the reference for the preliminary design of composite liners. The proposed solution can capture the field observed data well. Both the total flux and concentration are underestimated by about three orders of magnitude and 47.5%, respectively, compared to the model that doesn't consider the service life of geomembrane.

Keywords Service Life · Breakthrough Time · Analytical Model · Composite Liner · Contaminant Transport · Leachate Collection System

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1 Introduction

Recent years saw growing attention paid to the pollution from landfills due to the leakage of leachate with the high chemical oxygen demand (COD), the high ammonium nitrogen content, and lasting toxicological characteristics, which are important sources of groundwater contamination (Farzaneh et al., 2021; Han et al., 2016; Ma et al., 2022). A barrier system, including a low-permeability liner system and a high-permeability leachate collection system (LCS), is required to isolate pollutants from the landfill leachate. Field investigation and the experimental study showed

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that the service life of GMB can be greatly reduced by several factors, including high temperature, complex compound of leachate, and long-term stress (Abdelaal et al., 2014; Ewais et al., 2018; Fan & Rowe, 2023). Meanwhile, the LCS can be clogged resulting in a high leachate head (Bian et al., 2014; Yu et al., 2021; Xie et al., 2022). Failure to consider the service life of these components may lead to a significant overestimation of their performance. In order to design a barrier system ensuring facilities reach the contaminating lifespan (e.g., 100 years for a municipal waste landfill) (Yang et al., 2015), the effectiveness of all components and how they affect the barrier system's overall performance should be carefully considered.

The service life of GMB and LCS highly depends on the material properties and the surrounding environmental conditions. Extensive studies have reported that with exposure to elevated temperature and leachate, GMB has a lower service life, oxidation induction time (OIT), and stress crack resistance (SCR) (Abdelaal et al., 2014; Hsuan & Li, 2005; Lake & Rowe, 2005; Wang et al., 2021a). For example, the OIT of GMB in leachate is only 2.6 years at 60 ℃, while it is 7.3 years in water at 60℃ and 12 years in the leachate at 30 ℃. The SCR of GMB in the leachate at 60 ℃, after 13 years of oxidative degradation, was significantly reduced, which was only 50% of the initial value (Ewais et al., 2018). Moreover, the LCS with high overlying stress may contribute to the brittle rupture of GMB (Abdelaal et al., 2014). Field evidence also showed that the leachate collection layer can be clogged due to the growth of biomass, minerals precipitation and deposition of suspended inorganic particles (Levine et al., 2005; Bian et al., 2014). The clogging of the leachate collection system decreased the porosity and the hydraulic conductivity to the point where the leachate head on the liner can be continually elevated (Rowe & Brachman, 2004). In some landfills, the leachate head even exceeded 10 m (Shu et al., 2019a, 2019b; Zhan et al., 2018), which greatly reduced the service life of barrier systems (Chen et al., 2015; Shu et al., 2019a, 2019b).

Lots of efforts have been devoted to investigating contaminant transport in composite liners by analytical solutions in recent years (Cai et al., 2023; Chen et al., 2015; Feng et al., 2019b; Shu et al., 2019a, 2019b; Xie et al., 2022; Yu et al., 2019). Analytical solutions, unburdened by the need for spatial and temporal discretization, present a simpler alternative to numerical solutions (Feng et al., 2020; Xie et al., 2023). Therefore, analytical solutions can be used as a preliminary modeling method and screening tools when data are absent. Furthermore, they have been widely used for the validation of complex numerical models (Neville et al., 2000; Zhang et al., 2018). However, relatively little work has been paid to the influence of the effectiveness of each component on the service life of barrier systems (i.e., the time when the barrier system can no longer control the release of contaminants to a negligible level). For example, the service life of the geomembrane (GMB) and the high-permeability leachate collection system is required to meet the contaminating lifespan of waste disposal facilities (i.e., the period during which the facility can limit contaminants to a desired level).

The paper aims to develop an analytical model considering the service life of GMB and LCS. The parametric analysis is carried out to reveal the internal mechanization of each parameter. In addition, the effects of GMB and LCS on the service life of the composite liner are investigated. A good design is that when one component of the system fails, other components can control the release of contaminants at an acceptable level. Therefore, an equivalence assessment considering the service life of GMB and LCS is carried out to design the thickness of CCL, which may provide the reference for the design of composite liners. The developed analytical model is applied to a field application for the illustration of the applicability of the analytical solution to the risk assessment of landfill sites.

2 Mathematical model

Figure 1 illustrates the transport mechanism of contaminants in composite liners. The one-dimensional model of contaminant transport through the GMB/CCL composite liner was developed based on the following assumptions: (i) the adsorption is an isothermal linear process; (ii) the fluctuating leachate concentration is approximated by piecewise constant segments; (iii) LCS is clogged and ineffective when the operation time exceeds the service life of LCS t_{sl}^{lcs}, increasing leachate level (see Fig. 1); (iv) GMB is oxidized over time, and ceases to be effective when the operation time exceeds the service life of GMB t_{sl}^{gmb} (see Fig. 1), allowing leachate to contact the CCL directly. Assumption (iv) is a conservative

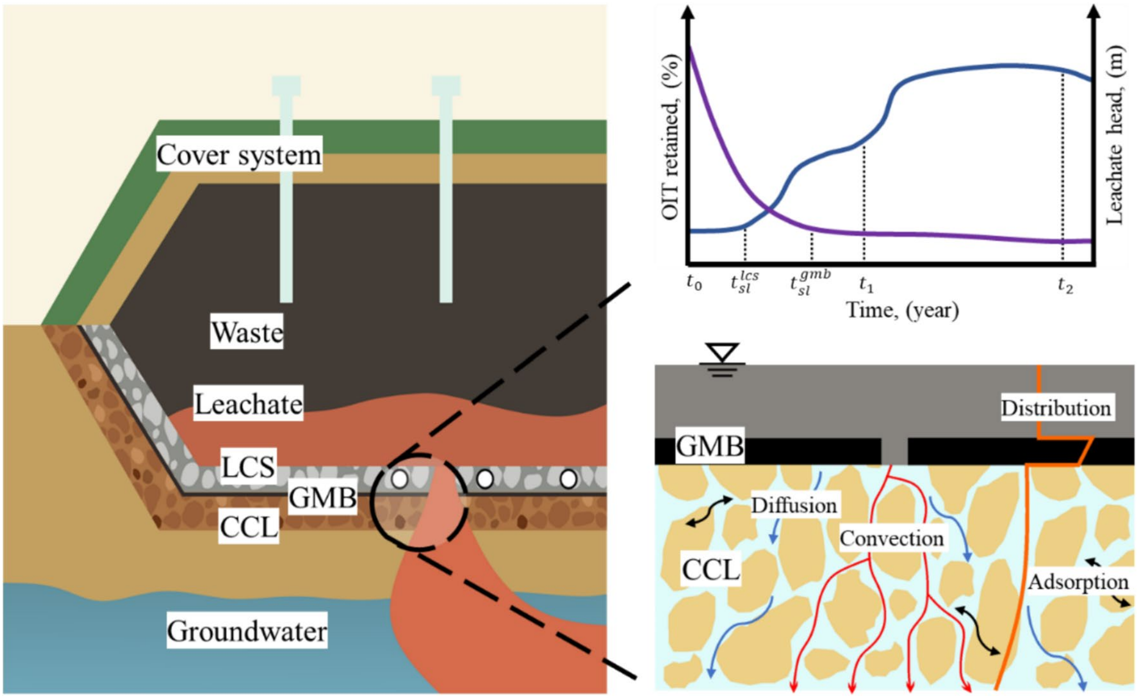


Fig. 1 Mathematical model for contaminants transport through the composite liner

assumption. In reality, GMB tends to be oxidized and damaged gradually over time. However, here we assume that the leachate directly loads on the CCL when the GMB is ineffective (i.e., $t \geq t_{sl}^{gmb}$).

3 Governing equations

The governing equation of contaminants transport through the GMB is expressed as (Feng et al., 2019b; Xie et al., 2023):

$$D_g \frac{\mathrm{d}^2 C_{g,i}}{\mathrm{d}z^2} - \frac{v_{a,i}}{K_g} \frac{\mathrm{d}C_{g,i}}{\mathrm{d}z} = 0 \tag{1}$$

where D_g is the diffusion coefficient in GMB; K_g is the partition coefficient between the concentration of contaminants in the GMB and the leachate; $C_{g,i}$ is the concentration of the contaminant in the GMB layer; the subscript i means the i th period of time; $v_{a,i}$ is the Darcy velocity of the composite liner. The governing equation of contaminants transport through the CCL is expressed as (Booker et al., 2004; Feng et al., 2019a):

$$R_s \frac{\partial C_{s,i}}{\partial t} = D_{s,i} \frac{\partial^2 C_{s,i}}{\partial z^2} - \frac{v_{a,i}}{n_s} \frac{\partial C_{s,i}}{\partial z} \tag{2}$$

where R_s is the retardation factor in the CCL; $C_{s,i}$ is the concentration of the contaminant in the CCL; n_s is the porosity of the CCL; the hydrodynamic dispersion coefficient of the CCL $D_{s,i}$ is calculated by the following function:

$$D_{s,i} = \tau_s D + \alpha_s \frac{v_{a,i}}{n_s} \tag{3}$$

where τ_s is the tortuosity factors of CCL; D is the molecular diffusion coefficient of contaminants in water; α_s is the dynamic dispersity. The Darcy velocity of the composite liner $v_{a,i}$ is calculated by the following function:

$$v_{a,i} = \frac{Q_{L,i}}{A} \tag{4}$$

where A is the area of the landfill site; $Q_{L,i}$ is the leakage rate of the composite liner. When the GMB is effective (i.e., $t < t_{sl}^{gmb}$), $Q_{L,i}$ can be calculated as follows (Touze-Foltz and Giroud., 2003):

$$Q_{L,i} = 0.096mAh_{w,i}^{0.9}a^{0.1}k_s^{0.74}\left[1 + 0.1\left(h_{w,i}/L_s\right)^{0.95}\right], t < t_{sl}^{gmb} \tag{5}$$

where m is the frequency of circular holes, a is the area of the defective hole; $h_{w,i}$ is the leachate head; k_s is the hydraulic conductivity of the CCL; L_s is the thickness of the CCL.

In the complex environment of a landfill, the GMB may cease to be effective with the oxidation and failure of GMB. The leakage rate $Q_{L,i}$ is given by Eq. (6) (Chen et al., 2015; Feng et al., 2019b)

$$Q_{L,i} = \frac{k_s\left(h_{w,i} + L_s\right)A}{n_s L_s}, t \geq t_{sl}^{gmb} \tag{6}$$

3.1 Boundary and initial condition

The contaminant concentration in the leachate is fixed at $C_{l,i}$. The interface condition between the GMB and leachate satisfies (Foose, 2002):

$$C_{g,i}\left(-L_g, t\right) = K_g C_{l,i} \tag{7}$$

where L_g is the length of GMB; and $C_{l,i}$ is the concentration of contaminants in the leachate.

The interface between GMB and CCL needs to meet the continuity of contaminant concentration and flux (Kalbe et al., 2002):

$$C_{g,i}(0,t) = K_g C_{s,i}(0,t) \tag{8}$$

$$v_{a,i}\frac{C_{g,i}(0,t)}{K_g} - D_g\frac{\partial C_{g,i}(0,t)}{\partial z} = v_{a,i}C_{s,i}(0,t) - n_s D_{s,i}\frac{\partial C_{s,i}(0,t)}{\partial z} \tag{9}$$

The general solution of Eq. (1) is:

$$C_{g,i} = C_1 e^{\frac{v_{a,i}}{K_g D_g}z} + C_2 \tag{10}$$

where C_1 and C_2 satisfying boundary conditions (7) and (8) are:

$$C_1 = \frac{K_g\left(C_{l,i} - C_{s,i}(0,t)\right)}{e^{-\frac{v_{a,i}L_g}{K_g D_g}} - 1} \tag{11}$$

$$C_2 = K_g\frac{e^{-\frac{v_{a,i}L_g}{K_g D_g}}C_{s,i}(0,t) - C_{l,i}}{e^{-\frac{v_{a,i}L_g}{K_g D_g}} - 1} \tag{12}$$

Substituting Eqns. (10), (11) and (12) into (7) yields the top boundary condition of CCL when GMB is effective:

$$C_{s,i}(0,t) + \eta_i\frac{\partial C_{s,i}(0,t)}{\partial z} = C_{l,i} \tag{13}$$

and

$$\eta_i = \frac{n_s D_{s,i}\left(e^{-\frac{v_{a,i}L_g}{K_g D_g}} - 1\right)}{v_{a,i}K_g} \tag{14}$$

When GMB is ineffective, the top boundary condition is

$$C_{s,i}(0,t) = C_{l,i} \tag{15}$$

In this way, the top boundary condition can be provided as follows:

$$\begin{cases} C_{s,i}(0,t) + \eta_i\frac{\partial C_{s,i}(0,t)}{\partial z} = C_{l,i} & ,t < t_{sl}^{gmb} \\ C_{s,i}(0,t) = C_{l,i} & ,t \geq t_{sl}^{gmb} \end{cases} \tag{16}$$

The semi-infinite bottom boundary condition was adopted in this study:

$$\frac{\partial C_{s,i}(\infty,t)}{\partial z} = 0 \tag{17}$$

The initial condition is

$$C_{s,i}(z,0) = \begin{cases} 0 & ,i = 1 \\ C_{s,i-1}\left(z, t_{i-1}\right) & ,i > 1 \end{cases} \tag{18}$$

4 Analytical solutions

The following dimensionless variables were defined to facilitate mathematical treatment:

$$Z = \frac{z}{L_s}, T_i = \frac{D_{s,i}t}{R_s L_s^2}, Pe_i = \frac{v_{a,i}L_s}{n_s D_{s,i}}, \Upsilon_i = \frac{\eta_i}{L_s} \tag{19}$$

The inhomogeneous boundary condition was changed into homogeneous conditions by adopting the approach proposed by (Yan et al., 2020):

$$\theta_i = C_{s,i} - C_{l,i} \tag{20}$$

Substituting Eqns. (19) and (20) into Eqns. (2), (16), (17) and (18) yields:

$$\left(\frac{\partial}{\partial T} - \frac{\partial^2}{\partial Z^2} + Pe_i\frac{\partial}{\partial Z}\right)\theta_i(Z, T_i) = 0 \tag{21}$$

$$\begin{cases} \theta_i(0, T_i) + \Upsilon_i\frac{\partial\theta_i(0,T_i)}{\partial Z} = 0 & \text{, GMB is effective} \\ \theta_i(0, T_i) = 0 & \text{, GMB is ineffective} \end{cases} \tag{22}$$

$$\frac{\partial\theta_i(\infty, T_i)}{\partial Z} = 0 \tag{23}$$

$$\theta_i(Z, 0) = I(Z) = \begin{cases} -C_{l,1} & , i = 1 \\ C_{s,i-1}(Z, T_{i-1}) - C_{l,i} & , i > 1 \end{cases} \tag{24}$$

As the Green's Function Method (GFM) has been widely used in the contaminant transport and the transient flow in aquifers (Liu et al., 2021; Sanskrityayn et al., 2017; Wang et al., 2021), we use the GFM to solve this problem. The specific process of solution is in Appendix A. When $\eta_i \neq 0$ (i.e., the GMB is within the service life), the Green's function is

$$G_i(Z, T_i; Z', T_i') = \frac{e^{\frac{Pe_i}{2}(Z-Z')-S_0(T_i-T_i')}}{\sqrt{4\pi(T_i - T_i')}}\left(e^{-\frac{(Z-Z')^2}{4(T_i-T_i')}} + e^{-\frac{(Z+Z')^2}{4(T_i-T_i')}}\right) - \xi e^{\frac{Pe_i}{2}(Z-Z')-\frac{Pe_i^2}{4}(T_i-T_i')+\xi(Z+Z')+\xi^2(T_i-T_i')}\mathrm{erfc}\left(\xi\sqrt{(T_i - T_i')} + \frac{(Z + Z')}{2\sqrt{(T_i - T_i')}}\right) \tag{25}$$

and

$$\xi = -\frac{1}{\Upsilon_i} - \frac{Pe_i}{2} \tag{26}$$

When $\eta_i = 0$ (i.e., the GMB exceeds the service life), the Green's function is

$$G_i(Z, T_i; Z', T_i') = \frac{e^{\frac{Pe_i}{2}(Z-Z')-S_0(T_i-T_i')}}{\sqrt{4\pi(T_i - T_i')}}\left(e^{-\frac{(Z-Z')^2}{4(T_i-T_i')}} - e^{-\frac{(Z+Z')^2}{4(T_i-T_i')}}\right) \tag{27}$$

The analytical solution of Eq. (2) can be written as

$$C_{s,i} = \int_0^\infty G_i(Z, T_i; Z', 0)I(Z')\mathrm{d}Z' + C_{l,i} \tag{28}$$

It should be noted that the initial condition $I(Z')$ is the solution at the end of the previous interval. The Eq. (28) is calculated by the numerical integration due to the complex formula of $I(Z')$ (Liu et al., 2021).

The flux is given by:

$$F_{s,i} = v_{a,i}C_{s,i} - n_sD_{s,i}\frac{\partial C_{s,i}}{\partial z} \tag{29}$$

where $F_{s,i}$ is the flux of the contaminants. The cumulative flux is calculated by integrating the flux over time.

$$F_c = \int_0^{t'}\left(v_{a,i}C_{s,i} - n_sD_{s,i}\frac{\partial C_{s,i}}{\partial z}\right)\mathrm{d}t \tag{30}$$

where F_c is the cumulative flux of the contaminants; t' is the calculation time as the upper limit of the integral.

5 Results and discussions

The effects of the service life of GMB (t_{sl}^{gmb}) and the leachate collection system (t_{sl}^{lcs}) on the performance of composite liners are studied in this section. The composite liner consists of a 1.5 mm GMB + 1 m CCL. Dichloromethane (DCM) and Cl^{-1} are selected to represent typical organic contaminants and inorganic contaminants in the leachate, respectively. The porosity, tortuosity, dispersity and hydraulic conductivity of the CCL are 0.4 (Rowe & Brachman, 2004), 0.24 (Foose et al., 2002), 0.1 m (Gelhar et al., 1992) and 1×10^{-9} m/s (Booker et al., 2004), respectively. The frequency of circular holes and radius of the defective hole of the GMB are 2.5 ha^{-1} and 0.00564 m respectively (Rowe & Brachman, 2004). The input parameters for the verification are provided in Table 1. The breakthrough time (t_b) (Shu et al., 2019a, 2019b) and mass flux of contaminants are introduced to assess the lifespan of waste

Table 1 Transport parameters of Cl^{-1} and DCM

	Cl^-	Dichloromethane (DCM)
Molecular diffusion coefficient (m^2/s)	$2.03\times10^{-9(c)}$	$1.26\times10^{-9(a)}$
Partition coefficient in GMB (-)	$0.0008^{(c)}$	$6^{(a)}$
Diffusion coefficient in GMB (m^2/year)	$1.3\times10^{-6(c)}$	$2\times10^{-5(a)}$
Retardation factor in CCL	$1^{(b)}$	$3.5^{(a)}$
Leachate concentration (mg/L)	$2500^{(c)}$	$3.3^{(c)}$

(a) Booker et al., 2004 (b) Chen et al., 2015 (c) Rowe & Brachman, 2004

disposal facilities. The breakthrough concentration is assumed to be the relative concentration of contaminant reaching 0.1 (i.e., $C_s/C_l=0.1$). The input parameters are provided in Table 1.

Figure 2 shows the spatial concentration profile of DCM at $t=100$ years with t_{sl}^{gmb} = 70 years and ∞. The results of the analytical model proposed (solid lines) agree with the results obtained by COMSOL based on the finite element method (scatters). It is indicated that the present model performs well in predicting contaminant transport in the composite liner considering the service life of GMB t_{sl}^{gmb}.

5.1 *Effects of the service life of GMB (t_{sl}^{gmb}) and LCS (t_{sl}^{lcs})*

Figure 3a and b show the breakthrough curves of Cl^- and DCM. t_{sl}^{gmb} is assumed to be 30 years, 50 years, 70 years and infinite. The leachate head is fixed at 0.3 m as the LCS is assumed to be effective in this case (see Fig. 3a and b). The case of "No GMB" is added to highlight the effects of GMB on contaminants transport. "No GMB" can also represent the case with the service life of GMB being zero. Generally, increasing the service life of GMB can significantly extend the breakthrough time of Cl^{-1} and DCM. The breakthrough time of "No GMB" is 7 years. The performance of the composite liner for isolating Cl^- is largely enhanced when the service life of GMB (t_{sl}^{gmb}) is increased. For example, the breakthrough time of Cl^- for the case with t_{sl}^{gmb} = 70 years and t_{sl}^{gmb} = 30 years can be 11 and 5.3 times larger than that of the case without GMB, respectively. There is no breakthrough observed for the case with t_{sl}^{gmb} = ∞. The plausible explanation for the phenomena is that the GMB can serve as a good diffusion barrier for Cl^-. The breakthrough time of DCM for t_{sl}^{gmb} = 0, 30, and 50 years are 32, 46, and 57 years, respectively. It can be seen that the breakthrough time

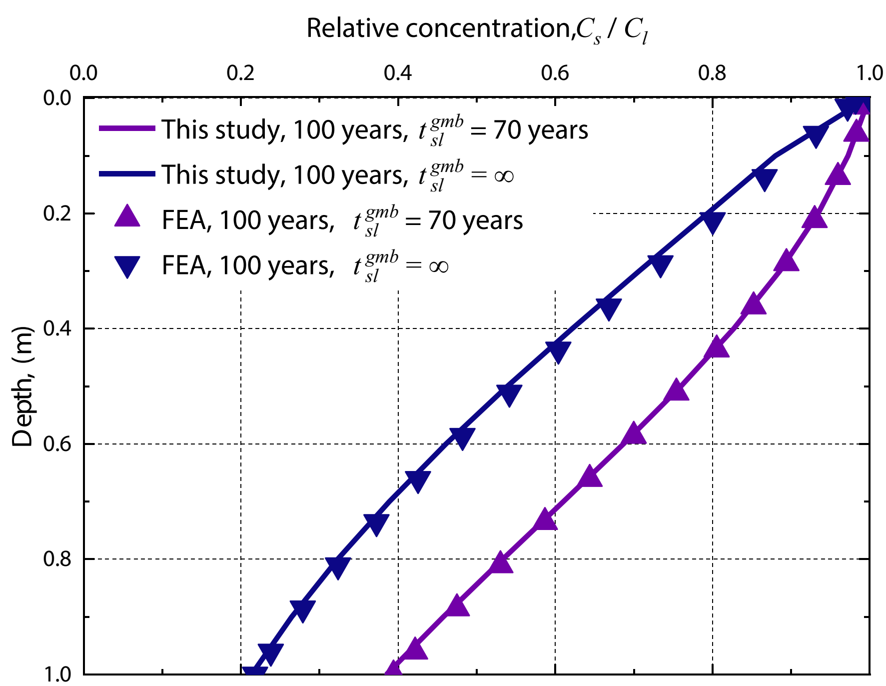
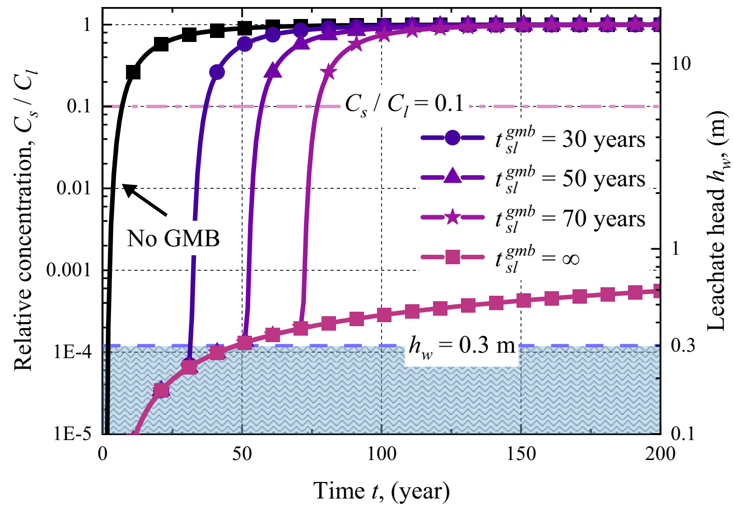
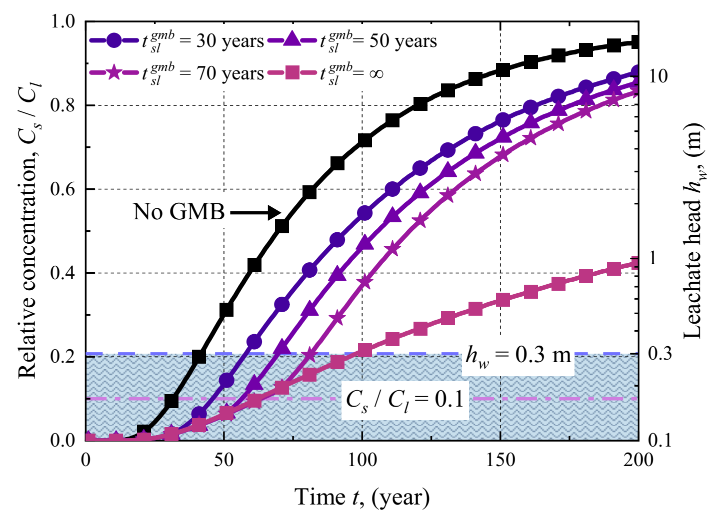


Fig. 2 Concentration profile of DCM at $t=100$ years with t_{sl}^{gmb} = 70 years and ∞

Fig. 3 The breakthrough curves of (**a**) Chloride and (**b**) DCM with different GMB's service life when the LCS is effective



(a)

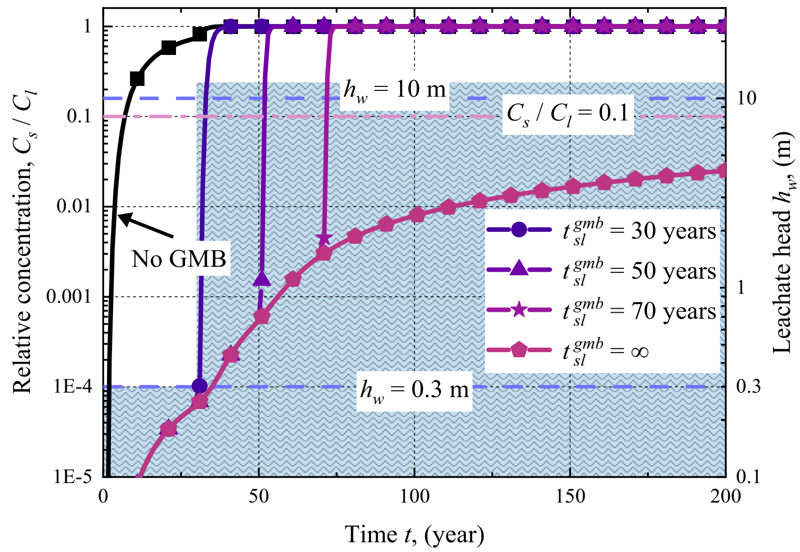


(b)

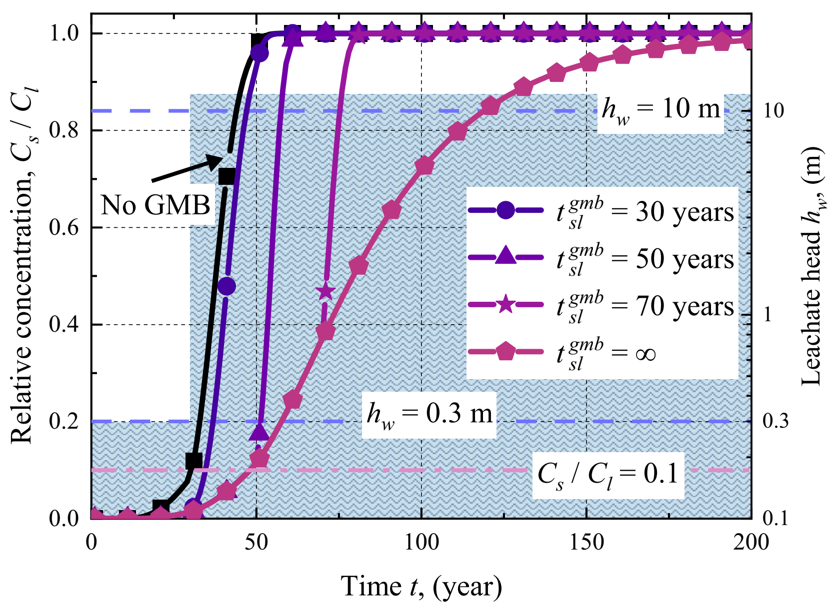
of DCM is less sensitive to variations of t_{sl}^{gmb} compared to Cl^-. On the one hand, adsorption plays a key role in influencing the transport of DCM while there is no adsorption for Cl^{-1}. On the other hand, the partition process of GMB can effectively decrease the flux of Cl^- through the GMB (i.e., K_g = 0.0008 for Cl^-). It is noted that the effects of t_{sl}^{gmb} on the breakthrough time of DCM is negligible when $t_{sl}^{gmb} \geq 70$ years while its impact on the breakthrough curve is significant. The service life of GMB is longer than that of the breakthrough time of DCM (i.e., t_b = 63 years). As a result, t_{sl}^{gmb} has little impact on the breakthrough time of DCM under such conditions. The above results indicated that the service life of GMB plays a key role in assessing the performance of the composite liner.

Figure 4a and b show the effects of the service life of LCS (t_{sl}^{lcs}) on breakthrough curves of Cl^- and DCM, respectively. The leachate head is controlled at 0.3 m within the service life of LCS. However, the leachate head increases when the LCS is continually clogged due to the growth of biomass, minerals precipitation and deposition of suspended inorganic particles (Levine et al., 2005; Bian et al., 2014). The elevation of water level due to the clogging of LCS

Fig. 4 The breakthrough curves of (**a**) Chloride and (**b**) DCM with different GMB's service life when considering the service life of LCS



(a)



(b)

is simplified into a two-step process (see Fig. 4a and b). The leachate head is 0.3 m when t is less than t_{sl}^{lcs}, and 10 m after LCS failure (i.e., $t \geq t_{sl}^{lcs}$). The t_{sl}^{lcs} is assumed to be 30 years in Fig. 4a and b. Although the leachate head may fluctuate with time at this stage, a simplified average leachate head may sufficiently provide a reasonable result to describe the migration processes of contaminants. As can be seen in Fig. 4a, the service life of GMB can significantly promote the breakthrough time of Cl^- and DCM as mentioned above, which isn't reiterated here. This part mainly describes the effects of the service life of LCS t_{sl}^{lcs}. The effect of t_{sl}^{lcs} on the breakthrough time of Cl^- is not significant. For example, the breakthrough time of Cl^- for t_{sl}^{gmb}= 70 years is 77 years while it is 72 years for the case without considering the failure of the LCS (see Figs. 3a and 4a). The phenomena can be explained by that the service life of GMB plays a more important role in the transport of Cl^- than the service life of the LCS. As a result, increasing the service life of the GMB can largely enhance the breakthrough time while maintaining the effectiveness of the LCS only adds about 5 years to the breakthrough time. As can be seen in Fig. 5a, the breakthrough time of Cl^- is almost linearly related to the service life of GMB. A simple empirical equation can be obtained to express the relationships between the breakthrough time of Cl^- and the service life of GMB as follows:

$$t_b = 0.97t_{sl}^{gmb} + 0.02t_{sl}^{lcs} + 3.29(\text{years}) \quad (31)$$

The coefficient of determination (R^2) for Eq. (31) is 0.96.

Effects of t_{sl}^{lcs} on the transport of DCM cannot be ignored. The breakthrough time of DCM for t_{sl}^{gmb}= 70 years is 64 years while it is 49 years for the case without considering the failure of the LCS (see Figs. 3b and 4b). Moreover, considering the failure of LCS leads to the reduction of the time required to reach the steady state. For example, DCM reached its peak concentration before 75 years with $t_{sl}^{gmb} \leq 70$ years, while DCM did not reach the peak concentration over 200 years of the same case without considering the failure of the LCS (see Figs. 3b and 4b). It can be seen in Fig. 5b that the breakthrough time of DCM can be effectively increased by raising both the service life of GMB and the service life of LCS. However, an early failure of one of the components (i.e., GMB and LCS) makes it difficult for the other component to lengthen the breakthrough time. For example, when LCS fails at 10 years, increasing t_{sl}^{gmb} from 0 to 100 years leads to an increase of the breakthrough time by 5.5 years (see Fig. 5b). Similarly, when GMB fails at 10 years, increasing t_{sl}^{lcs} from 0 to 100 years results in an increase of the breakthrough time by 12 years. However, when the service life of LCS is increased to 80 years, increasing the service life of GMB from 10 to 70 years can result in a rise of the breakthrough time by 50.6 years (see Fig. 5b). The above results indicated that the service life of GMB plays a dominant role in the transport of Cl^-. As for the transport of DCM, the effects of the service life of GMB and LCS cannot be neglected.

5.2 The designed thickness of CCL considering the service life of GMB and LCS

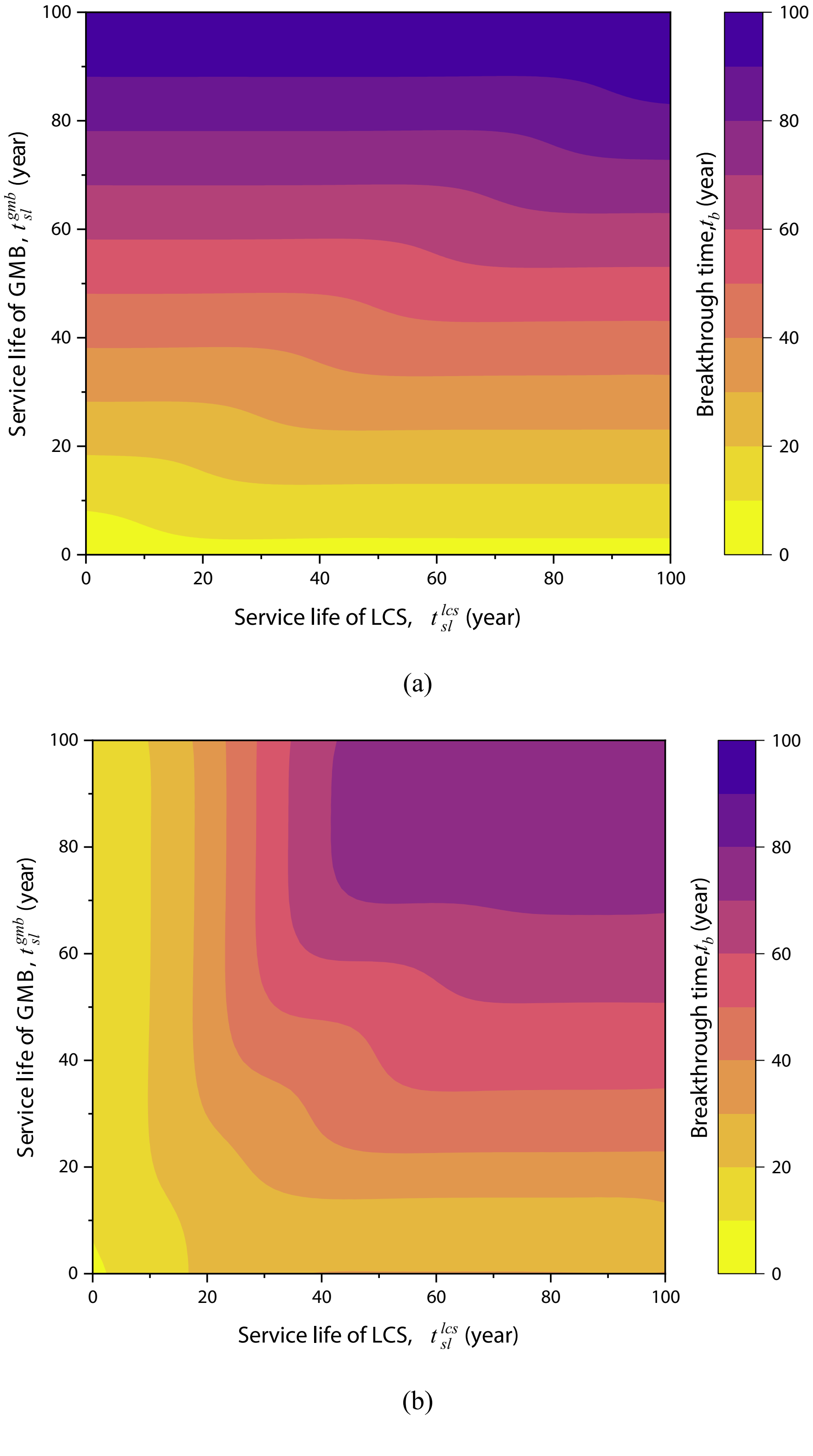
The well-performed liner systems generally recognized that the release of contaminants can be reduced to an acceptable level within the contaminant lifespan of waste disposal facilities. The results of Figs. 3 and 4 indicated the potential failure of the 1.5 mm GMB + 1 m CCL composite liner in the contaminant lifespan (e.g., 100 years). Under such circumstances, a thicker CCL would be required to reduce the release of contaminant from the leachate when the service life of GMB and LCS are considered. The minimum thickness of CCL required to meet the contaminant lifespan under different service life of GMB and LCS is presented in Fig. 6. The required thickness of CCL decreases with the increase in the service life of GMB and LCS. The design thickness of CCL is more sensitive to the variations of the service life of GMB for Cl^- than that for DCM. For example, the required thickness of CCL for Cl^- with $t_{sl}^{gmb} = 0$ years and $t_{sl}^{lcs} = 0$ years can be 2 times larger than that with $t_{sl}^{gmb} = 50$ years and $t_{sl}^{lcs} = 0$ years (see Fig. 6a), while only 28% larger for DCM with the same condition (see Fig. 6b). The designed thickness of CCL for Cl^- can be decreased more than 1 m when the service life of GMB is increased by 20 years. Furthermore, the effect of t_{sl}^{lcs} on the required thickness of CCL cannot be ignored. The required thickness of CCL for DCM with $t_{sl}^{gmb} = 0$ years and $t_{sl}^{lcs} = 50$ years can be 1.3 times larger than that with $t_{sl}^{gmb} = 0$ years and $t_{sl}^{lcs} = 50$ years. However, the influence of the service life of the LCS on the design thickness of CCL can be neglected when the t_{sl}^{gmb} is longer than the t_{sl}^{lcs} (as can be seen in Fig. 6, the curves almost coincide when $t_{sl}^{gmb} \geq t_{sl}^{lcs}$). This is due to the fact that GMB can serve as an excellent advection barrier (Saidi et al., 2008). Consequently, promoting the service life of the GMB can effectively reduce the effects of LCS failure on the performance of the composite liner.

6 Field application

6.1 Site investigation

The site discussed in this study is a lagoon located at Ontario, Canada (Rowe et al., 1998). The lagoon had been used as the landfill for 14 years to store

Fig. 5 The breakthrough time of (**a**) Chloride and (**b**) DCM considering the service life of GMB and LCS



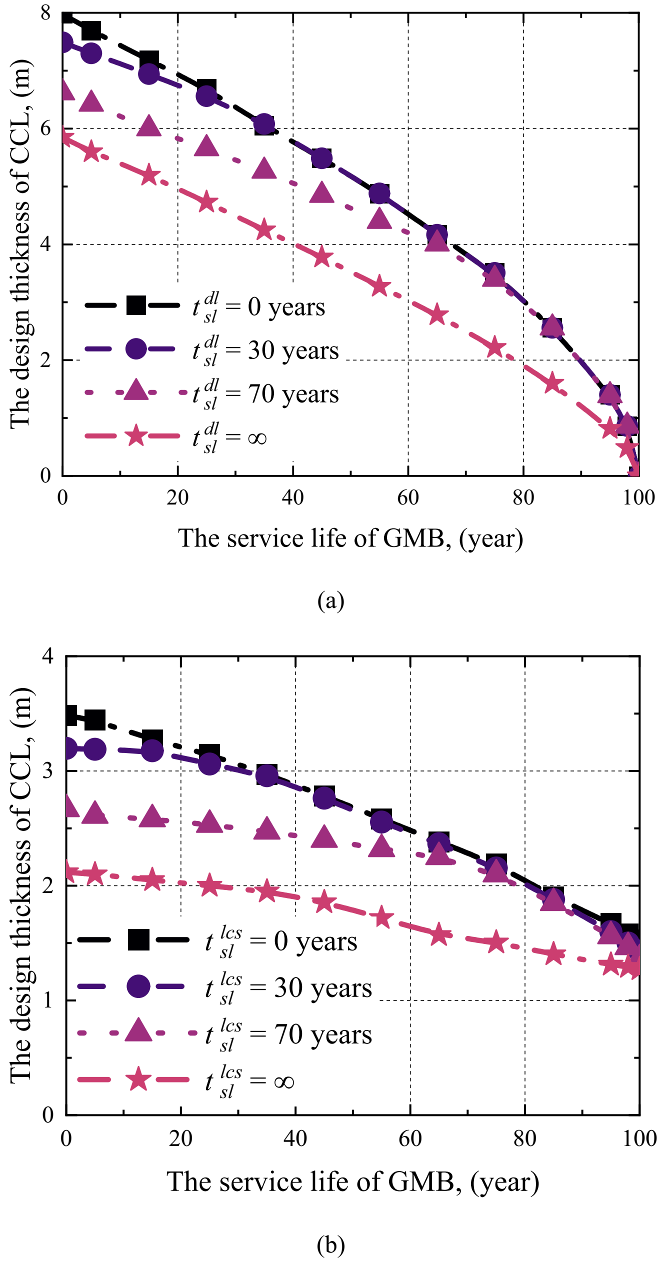


Fig. 6 The designed thickness of CCL considering the service life of GMB and LCS (**a**) Chloride and (**b**) DCM

inorganic constituents. The composite liner contains a smooth 1.5 mm GMB and a 3 m CCL. There were about 180 defects per hectare on the GMB. Long-term field tests were performed at this site to investigate the effectiveness of the liner system (Lake & Rowe, 2005). The measured Cl^- concentration profile is provided in Fig. 8. The concentration of Cl^- was reported to fluctuate during the service of the liner. Cl^-, Na^+, and K^+ concentrations in the leachate were reported by Lake and Rowe (2005). The geometric means of the measured Cl^- concentration was represented as the leachate concentration in time periods of 0 to 4.5 years, 4.5 to 9.5 years, 9.5 to 14 years, and above 14 years (see Fig. 7). The effective diffusion coefficient of Cl^- in CCL is 7×10^{-10} m^2/s. The values of the partition coefficient and diffusion coefficient in GMB are 0.0008 and 1.3×10^{-6} m^2/year, respectively.

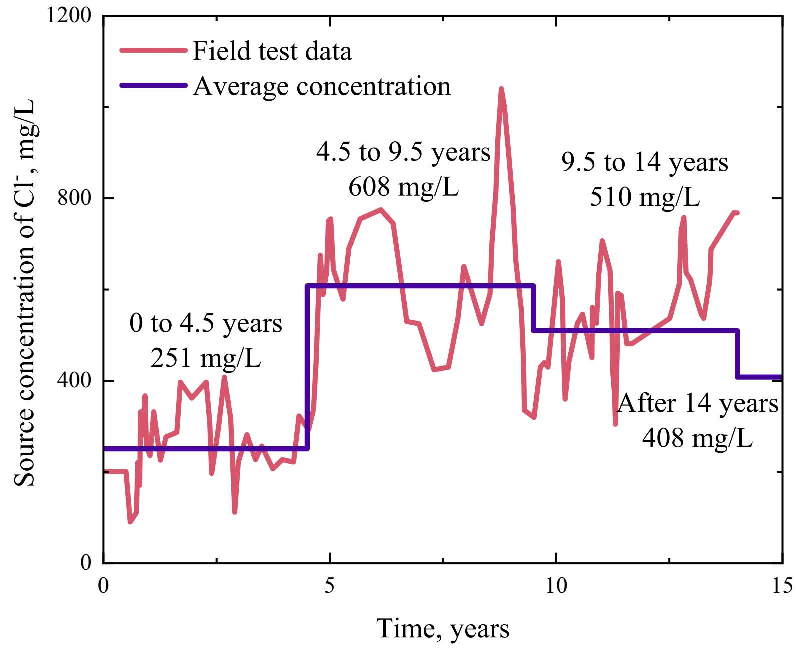


Fig. 7 Time-varying source concentration of Cl^{-}

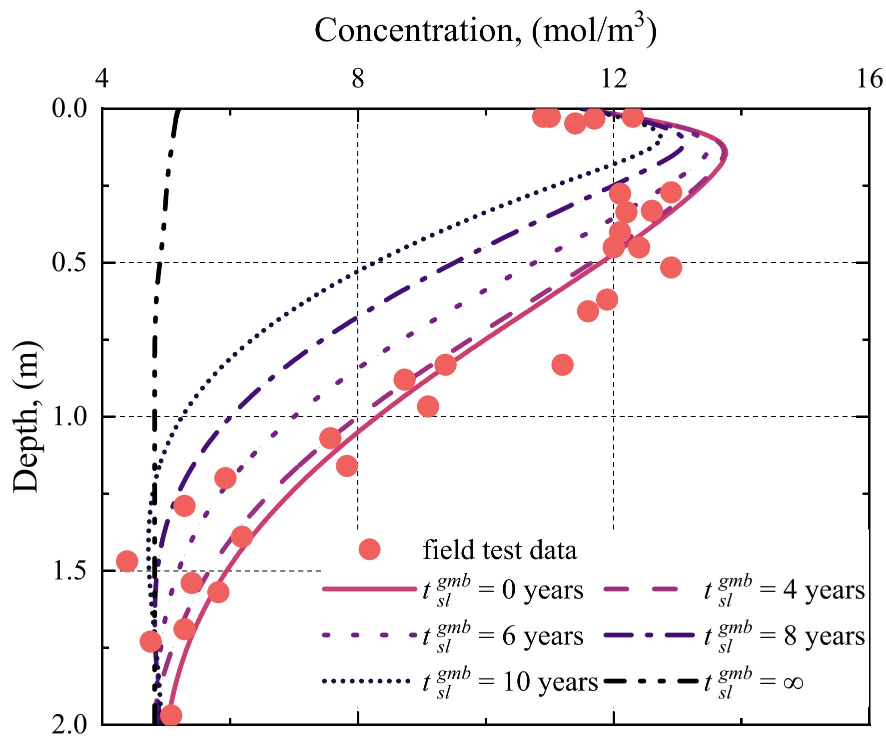


Fig. 8 Prediction of Chloride pore fluid concentrations in CCL for the different assumed service life of the GMB

The Darcy velocity is 3.5×10^{-4} m/year and 0.013 m/year for the cases where GMB was effective and ineffective, respectively, according to the laboratory tests conducted by Rowe et al. (1998).

7 Results and discussion

The simulation results showed that the proposed analytical model can well fit with the field observation

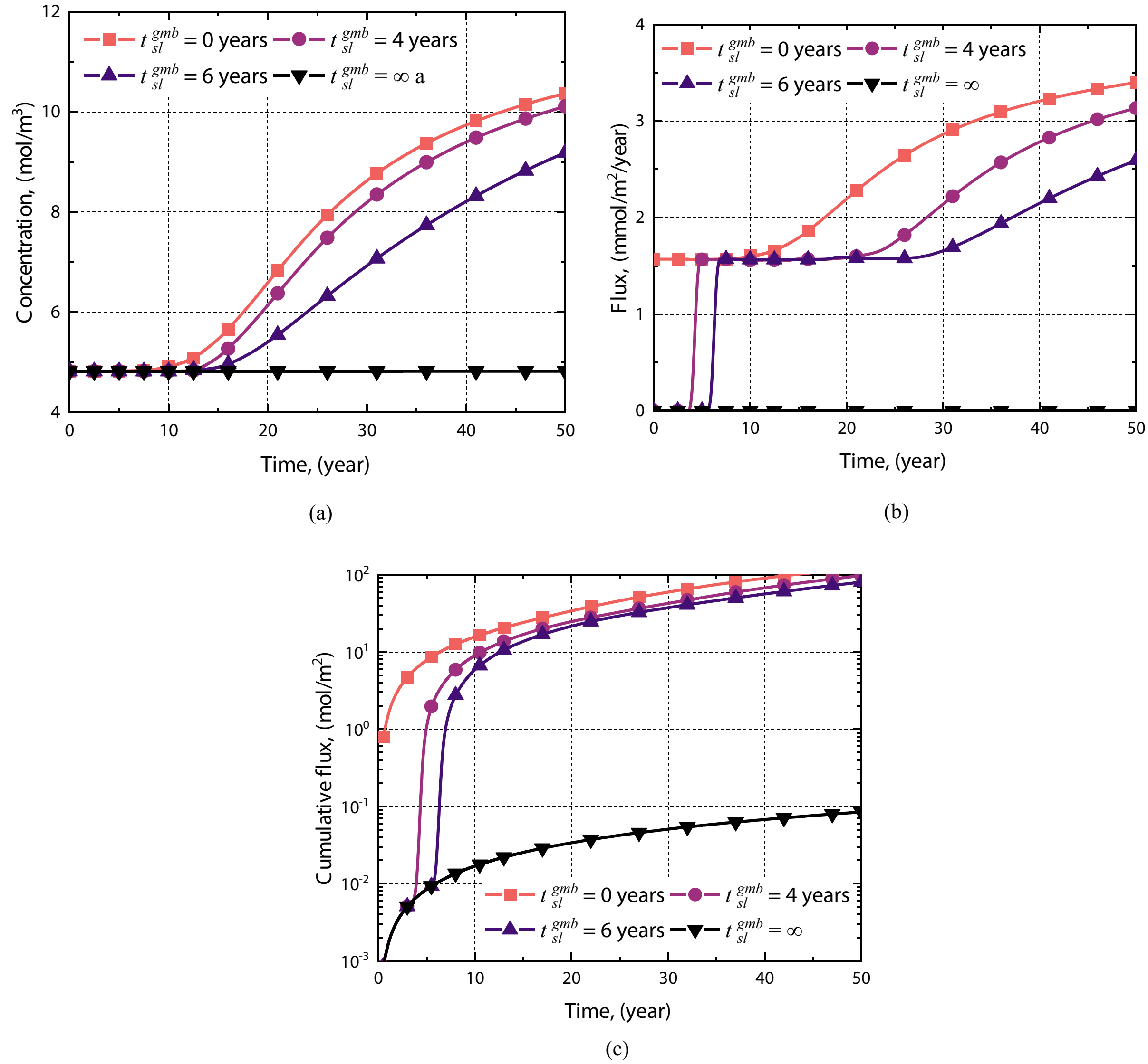


Fig. 9 (**a**) the breakthrough curves; (**b**) the flux curves and (**c**) the cumulative flux of Chloride with different service life of the GMB

data. The back-diffusion phenomenon of Cl^- observed by Lake and Rowe (2005) was accurately captured by the proposed analytical solution. The back-diffusion phenomenon can be attributed to the concentration of Cl^- decreasing after 14 years, which was lower than that in the soil pore water. It can be seen in Fig. 8 that the t_{sl}^{gmb} has significant effects on the concentration profile of Cl^-. For example, the peak concentration with t_{sl}^{gmb} = 4 years is 2.8 times larger than that with $t_{sl}^{gmb} = \infty$. The root mean square error (RMSE) is introduced to analyze the service life of GMB. When the service life of GMB is set as 0, 4, 6, 8, or 10 years, the RMSE is 0.78, 0.76, 1.17, 1.90, or 2.61, respectively. The fitting results indicated that the effective time of GMB is only about 0–4 years. Ignoring the service life of GMB can tremendously overestimate the performance of liner systems. To assess the effects of t_{sl}^{gmb}, A quantitative investigation into the concentration, flux and cumulative flux of Cl^- is conducted.

As shown in Fig. 9a, the concentration of Cl^- at the depth of 2 m is almost the same as the background concentration since the effective GMB is an excellent barrier to advective migration as well as an excellent diffusive barrier against inorganic and polar contaminants (Touze-Foltz et al., 2021). However, according to the above simulation results, the service life of GMB is only about 0–4 years. Without considering the service life of GMB, the concentration of Cl^- can be underestimated by more than 47.5% at 50 years compared with the case with $t_{sl}^{gmb} = 6$ years. When the GMB becomes ineffective, a discontinuity in the flux is observed, resulting in three orders magnitude increase in the flux. It is shown in Fig. 9b that the flux for the case without considering the effect of the service life of GMB (i.e., $t_{sl}^{gmb} = \infty$) is only about 0.0017 mol/m^2/year, while the flux of $t_{sl}^{gmb} = 0$, 4 and 6 years are 3.4, 3.1 and 2.6 mol/m^2/year at 50 years respectively. Figure 9c is the cumulative flux of Cl^-. The cumulative flux is underestimated by three orders of magnitude without considering the service life of GMB. The above results indicated that the service life of GMB is a key factor to evaluate the performance of the liner system.

8 Conclusions

We propose an analytical model of the one-dimensional contaminants transport in the composite liner considering the service life of GMB and LCS. The solution is obtained by the Green's Function method. The parameter analysis is carried out to reveal the impact of the service life of GMB and LCS on contaminant transport in composite liners. In addition, the proposed analytical solution is applied to a field test of a lagoon located at Ontario, Canada, to testify the applicability of the proposed analytical solution. The specific conclusions from this work are as follows:

1. The service life of GMB plays an important role in investigating the performance of composite liners. Without taking into account the impact of the service life of GMB, the breakthrough time of DCM may be overestimated by a factor of two. Additionally, chloride is more sensitive to the service life of GMB compared to DCM. The composite liner can be broken through in 7 years after the cessation of the effectiveness of GMB, while it cannot be broken through without considering the service life of GMB.
2. The 1.5 mm GMB + 1m CCL composite liner is an effective barrier for chloride when the service life of GMB exceeds the lifespan of the landfill. Given the failure of the LCS, the breakthrough of CCL can occur within two years, which represents a 5–6 times faster rate compared to the case without considering LCS failure. A simple linearly empirical formula can be used to describe the relationship between the breakthrough time of Cl^- and the service life of GMB and LCS. This empirical formula is useful for the preliminary design of a composite liner considering the service life of GMB and LCS.
3. CCL plays a key role after the failure of GMB. The breakthrough time of the 1.5 mm GMB + 1m CCL composite liner cannot satisfy the contaminating lifespan of the landfill when the service life of GMB and LCS are considered. For the Cl^-, the designed thickness of CCL can be decreased by more than 1 m when the service life of GMB is increased by 20 years.
4. The proposed analytical model was applied to the site investigation data at a lagoon located at Ontario. The proposed analytical model can effectively capture chloride pore fluid concentration profiles and the back-diffusion phenomenon of Cl^-. The total flux and concentration are underestimated by about three orders of magnitude and 47.5%, respectively, compared to the model that does not take into account the service life of GMB.

Notations

Dimensions of parameters are shown in parentheses (L means length, M means mass, T means time, N means amount of substance). a : Area of the defective hole (L^2); A : Area of the landfill site (L^2); C_g : Concentration of the contaminant in the GMB (NL^{-3} or ML^{-3}); C_l : Concentration of contaminants in the leachate (NL^{-3} or ML^{-3}); C_s : Concentration of the contaminant in the CCL (NL^{-3} or ML^{-3}); D : Molecular diffusion coefficient of contaminants in water (L^{2}T^{-1}); D_g : Diffusion coefficient in GMB (L^{2}T^{-1}); D_s : Hydrodynamic dispersion coefficient of the CCL (L^{2}T^{-1}); F_c : Cumulative flux of the contaminants (NL^{-2} or ML^{-2}); F_s : Flux of the contaminants (NL^{-2} T^{-1} or ML^{-2} T^{-1}); h_w : Leachate head (L); i : Subscript i means the i th period of time (-);

k_s : Hydraulic conductivity of the CCL (LT^{-1}); K_g : Partition coefficient between the GMB and the leachate (-); L_g : Thickness of GMB (L); L_s : Thickness of the CCL (L); m : Frequency of circular holes (L^{-2}); n_s : Porosity of the CCL (-); Q_L : Leakage rate of the composite liner (L^3T^{-1}); R_s : Retardation factor in the CCL (-); t' : Calculation time as the upper limit of the integral (T); t_b : Breakthrough time of contaminants (T); t_{sl}^{gmb} : Service life of GMB (T); t_{sl}^{lcs} : Service life of LCS (T); v_a : Darcy velocity of the composite liner (LT^{-1}); α_s : Dynamic dispersity (L); τ_s : Tortuosity factors of CCL (-)

Abbreviations

CCL: Compacted clay liner; COD: Chemical oxygen demand; DCM: Dichloromethane; FEA: Finite element analysis; GFM: Green's Function Method; GMB: Geomembrane; LCS: Leachate collection system; OIT: Oxidation induction time; SCR: Stress crack resistance

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Data availability Data are available from the authors upon reasonable request.

Declarations

Conflict of interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix

The Green Function and boundary condition of Eqs. (21)-(23) in Laplace domain are as follows:

$$\left(-\frac{\partial^2}{\partial Z^2}+Pe_i\frac{\partial}{\partial Z}+S\right)\overline{G_i(Z,S;Z',0)}=\delta\left(Z-Z'\right) \tag{32}$$

$$\begin{cases}\overline{G_i(Z,S;Z',0)}\Big|_{Z=0}+\Upsilon_i\frac{\partial\overline{G_i(Z,S;Z',0)}}{\partial Z}\Big|_{Z=0}=0 & ,\ \text{GMB is effective}\\ \overline{G_i(Z,S;Z',0)}\Big|_{Z=0}=0 & ,\ \text{GMB is ineffective}\end{cases} \tag{33}$$

$$\frac{\partial\overline{G_i(Z,S;Z',0)}}{\partial Z}\Bigg|_{Z=\infty}=0 \tag{34}$$

The solution of this Green's function can be expressed in the following form:

$$G_i\left(Z,T_i;Z',0\right)=K_i\left(Z,T_i;Z',0\right)+V_i(Z,0) \tag{35}$$

where K_i is the fundamental solution of the Green Function (Eq. 32), and can be easily solved by the Laplace transform method:

$$K_i\left(Z,T_i;Z',0\right)=\frac{e^{-\frac{\left[(Z-Z')-Pe_iT_i\right]^2}{4T_i}}}{\sqrt{4\pi T_i}} \tag{36}$$

The boundary term V_i is the solution of the following homogeneous equation:

$$\left(-\frac{\partial^2}{\partial Z^2}+Pe_i\frac{\partial}{\partial Z}+S\right)\overline{V_i(Z,S)}=0 \tag{37}$$

And the general solution is

$$\overline{V_i(Z,S)}=\varphi_1e^{\frac{Pe_i-\sqrt{Pe_i^2+4S}}{2}Z}+\varphi_2e^{\frac{Pe_i+\sqrt{Pe_i^2+4S}}{2}Z} \tag{38}$$

Substituting (Eq. 36) and (Eq. 38) into (35), we can get the form of the solution in the Laplace domain:

$$\overline{G_i(Z,S;Z',0)}=\overline{K_i(Z,S;Z',0)}+\overline{V_i(Z,S)}$$

$$=e^{\frac{Pe_i}{2}(Z-Z')}\frac{1}{2\sqrt{S+S_0}}e^{-\sqrt{S+S_0}\left|Z-Z'\right|}+\varphi_1e^{\left(\frac{Pe_i}{2}-\sqrt{S+S_0}\right)Z}+\varphi_2e^{\left(\frac{Pe_i}{2}+\sqrt{S+S_0}\right)Z} \tag{39}$$

where

$$S_0 = \frac{Pe_i^{\,2}}{4} \tag{40}$$

Substituting the solution (Eq. 39) into boundary conditions (Eq. 33) and (Eq. 34) can get the value of unknown numbers:

$$\overline{G_i(Z,S;Z',0)} = e^{\frac{Pe_i}{2}(Z-Z')}\frac{1}{2\sqrt{S_0+S}}e^{-\sqrt{S_0+S}|Z-Z'|}$$

$$+e^{\frac{Pe_i}{2}(Z-Z')}\left(\frac{1}{2\sqrt{S_0+S}} - \frac{\xi}{\sqrt{S_0+S}\left(\xi+\sqrt{S_0+S}\right)}\right)e^{-\sqrt{S_0+S}(Z+Z')} \tag{41}$$

where

$$\varphi_1 = e^{-\frac{Pe_i}{2}Z'}\left(\frac{1}{2\sqrt{S_0+S}} - \frac{\xi}{\sqrt{S_0+S}\left(\xi+\sqrt{S_0+S}\right)}\right)e^{-\sqrt{S_0+S}Z'} \tag{42}$$

$$\varphi_2 = 0 \tag{43}$$

$$\xi = -\frac{1}{\Upsilon_i} - \frac{Pe_i}{2} \tag{44}$$

The inverse Laplace transform is performed on $\overline{G_i(Z,S;Z',0)}$ to obtain $G_i(Z,T_i;Z',0)$, and the solution when $T_i' \neq 0$ can be obtained by replacing T_i with $T_i - T_i'$:

$$G_i(Z,T_i;Z',T_i') = \frac{e^{\frac{Pe_i}{2}(Z-Z')-S_0(T_i-T_i')}}{\sqrt{4\pi(T_i-T_i')}}\left(e^{-\frac{(Z-Z')^2}{4(T_i-T_i')}} + e^{-\frac{(Z+Z')^2}{4(T_i-T_i')}}\right) - \tag{45}$$

$$\xi e^{\frac{Pe_i}{2}(Z-Z')-\frac{Pe_i^2}{4}(T_i-T_i')+\xi(Z+Z')+\xi^2(T_i-T_i')}\mathrm{erfc}\left(\xi\sqrt{(T_i-T_i')} + \frac{(Z+Z')}{2\sqrt{(T_i-T_i')}}\right) \tag{46}$$

However, the above solution does not hold for the case of $\Upsilon_i = 0$. Therefore, the Green Function solution in the case of $\Upsilon_i = 0$ needs to be obtained by the above method:

$$G_i(Z,T_i;Z',T_i') = \frac{e^{\frac{Pe_i}{2}(Z-Z')-S_0(T_i-T_i')}}{\sqrt{4\pi(T_i-T_i')}}\left(e^{-\frac{(Z-Z')^2}{4(T_i-T_i')}} - e^{-\frac{(Z+Z')^2}{4(T_i-T_i')}}\right) \tag{47}$$

The process is similar and is omitted here.

On the basis of Green's function, the general solution of Eq. (21) can be written as

$$\theta_i(Z,T_i) = \int_0^\infty G_i(Z,T_i;Z',0)I(Z')\mathrm{d}Z' \tag{48}$$

In this study, the initial condition $I(Z')$ is the solution of concentration at the end of the previous interval $[T_{i-1}, T_i]$. Eq. (48) is evaluated using the numerical integration by MATLAB 2023a.

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