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The role of thermodiffusion in organic pollutant transport in landfill composite liner system

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ABSTRACT

This paper presents a new analytical model for forecasting one-dimensional transient transport of organic pollutant in a typical landfill composite liner including a geomembrane (GMB), a geosynthetic clay liner (GCL) and a soil liner (SL). The model can consider the coupling effect of molecular diffusion, thermodiffusion, advection and linear adsorption processes. Zero concentration gradient condition is adopted at the bottom boundary. The separation of variables method and superposition principle are utilized to derive the solution. The solution is first validated against the results of experimental test and a numerical model. Then, the influence of thermodiffusion is investigated on pollutant migration in typical composite liner. The results reveal that thermodiffusion may significantly influence the pollutant migration. If the Soret coefficient exceeds 0.01 K^{-1}, regardless of the combination conditions in terms of molecular diffusion, advection and adsorption, the influence of thermodiffusion generally cannot be ignored. The analytical model also can be utilized to design composite liner systems and to verify other numerical models.

1. Introduction

Composite liners are widely used in modern municipal solid waste landfills as essential barrier structures to isolate the pollutants in landfill leachates from the external natural environment. Even though intact geomembranes (GMBs), the important components of composite liners, are nearly impenetrable by leachates, they could be readily diffused through by some organic pollutants (e.g., volatile organic compounds (VOCs)) (Chao et al., 2007). Moreover, organic pollutants are generally toxic even at low concentration compared to many inorganic pollutants (Edil, 2003; Xie et al., 2015a). Thus, studying the transport of organic pollutants through landfill composite liner is of great significance for rational design and barrier performance assessment.

Molecular diffusion is broadly recognized as one of the dominant transport mechanisms of pollutants in composite liner system. Plenty of pure diffusion analytical models have been proposed to investigate this issue (Chen et al., 2009; Cleall and Li, 2011; Chen et al., 2019; Foose et al., 1999; Foose, 2002; Pu et al., 2020; Qiu et al., 2021; Xie et al., 2015b; Zhan et al., 2014). However, the transport mechanism of pollutant is diverse and complicated. Apart from molecular diffusion, there are advection, adsorption, biodegradation and thermal diffusion etc. that may also be involved (Chen et al., 2015; Feng et al., 2019a, 2019b; Lu et al., 2021; Lu and Feng, 2022; Pu et al., 2018, 2021; Rosanne et al., 2006; Yan et al., 2021).

In practice, there usually exist defects in GMBs, such as holes, wrinkles and seams due to large construction machinery, solar radiation and insufficient overlaps. Landfill leachates could break through GMBs via these defects by advection. Thus, some diffusion–advection models were also developed (Feng et al., 2019b; Foose, 2010; Xie et al., 2015a, 2018). For example, Xie et al. (2010, 2011) proposed a quasi-steady state analytical solution for pollutants diffusion and advection through GMB/CCL (note: CCL is compacted clay liner) and GCL/SL composite liners. Xie et al. (2015a) then extended this solution to GMB/GCL/SL composite liner system. Feng et al. (2019a, 2019b) further presented a transient solution to diffusion–advection transport of pollutant in the three-layer composite liner system. However, all these models assume that the composite liner works in an isothermal environment. Namely, these models can not consider the thermodiffusion impact on solute movement in liner systems.

Actually, there generally exists temperature gap between the

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leachate heaping on the geomembrane and the bottom surface of modern landfill composite liners (Rowe, 2012). Thus, the effect of thermodiffusion is of growing concern for the transport issue in landfills (Engelhardt et al., 1997; Leaist and Lv, 1990; Rahman and Saghir, 2014). Unfortunately, the model able to consider the thermodiffusion impact on solute migration in multi-layer media is rare (Peng et al., 2021; Xie et al., 2015c). For this topic, Xie and his coauthors (Xie et al. 2015c; Yan et al., 2020) presented a transport model suitable for monolayer porous media. Moreover, Peng et al. (2020, 2021) proposed transport models for organic pollutants in GMB/CCL and GMB/GCL/SL composite liners, respectively. However, all these models are unable to consider the effect of advection. So far as is known to the authors, there is currently no analytical model for the migration of organic pollutant in triple-layer landfill composite liners that account for the coupling effect of molecular diffusion, advection, adsorption and thermal diffusion.

The aim of this paper is to present a diffusion–advection-adsorption-thermodiffusion analytical model for organic pollutant transport through a GMB/GCL/SL landfill composite liner system. The developed model is validated against the results of experimental test and a numerical model. The solution is then applied to systematically explore the influence of thermodiffusion, especially when the thermodiffusion must be considered.

2. Mathematical model development

2.1. Geometric model

Fig. 1 is a conceptual model that describes the process of organic pollutant transport in the composite liner system via molecular diffusion, advection, adsorption and thermodiffusion. This system is mainly composed of GMB, GCL and SL, the three individual horizontal layers. Molecular diffusion, advection, and thermodiffusion occur in all layers. Instantaneously equilibrium linear absorption occurs in the system except for GMB layer. Two degrees of freedom are considered at any z position. They are the concentration of pollutant C and the temperature T. T_{b} is the bottom surface temperature of the liner system, and T_{u} is that for the upper surface. L_{gmb}, L_{gcl}, L_{sl} and L mean the thicknesses of the geomembrane layer, GCL, SL and composite liner system, respectively. The substratum layer underneath the liner system is an obstructed groundwater collection and detection layer (GCDL) or an aquitard layer (MCPRC, 2007; MOHURDPRC, 2013) (Note: MCPRC is the abbreviation for the Ministry of Construction of the People's Republic of China, and MOHURDPRC is for the Ministry of Housing and Urban-Rural Development of the People's Republic of China.). In fact, the model is still valid for inorganic pollutant, which the difference is that the magnitude of the parameters for the calculation are different. In addition, the main improvement of the present model in method is including advection in the previous solution proposed in Peng et al. (2021).

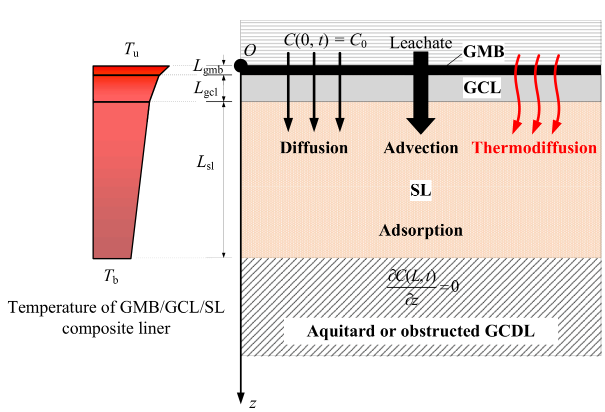

Fig. 1. Mathematical model of organic pollutants diffusion–advection-adsorption-thermodiffusion in a composite liner.

2.2. Basic assumptions

Several assumptions are adopted to simplify the problem: (1) fixed constant concentration of the representative pollutant in leachate, C_0, is assumed (Peng et al., 2021); it is conservative and reasonable for the design of the liner system; (2) the organic pollutant transports along the z axis (Peng et al., 2020); (3) SL and GCL are saturated (Peng et al., 2021); (4) all layers are homogeneous (Xie et al., 2015a); (5) the influence of biodegradation of the pollutants is neglected (Foose, 2002); thus, the forecast concentration would be higher than the reality, which is conservative and safe for the design; (6) a reversible and instantaneously equilibrium linear adsorption process is assumed (Foose, 2002); it is fair for dilute solution whereas the sorption capacity would be overestimated for concentrated solution; (7) the properties of the liner materials remain invariable over time (Peng et al., 2021); (8) steady-state heat conduction is assumed (Thomas et al., 2012).

2.3. Mathematical model

Based on energy conservation law and Fourier's law, for the steady-state heat conduction process in the liner system, the governing equation can be expressed as (Peng et al., 2021)

$$\nabla^2 T_i(z) = 0, \quad i = 1, 2, 3 \tag{1}$$

in which, i is the property for the composite liner of the i^{th} layer, $i = 1$, 2, 3 refers to the properties for GMB, GCL and SL, respectively; $T_i(z)$ is the temperature of the i^{th} layer at z position.

The solution is

$$T_i(z) = G_{T,i} z + I_i, \quad i = 1, 2, 3 \tag{2}$$

in which, I_i is a coefficient.

The temperature gradient of each layer is (Peng et al., 2021)

$$\begin{bmatrix} G_{T,1} \\ G_{T,2} \\ G_{T,3} \end{bmatrix} = \frac{(T_{\mathrm{u}} - T_{\mathrm{b}})\kappa_2}{\kappa_2^2 z_1 z_2 - \kappa_1\kappa_2 z_1 z_2 + \kappa_1\kappa_3 z_1 z_2 - \kappa_2\kappa_3 z_1 z_2 - \kappa_1\kappa_2^2\kappa_3 z_3} \begin{bmatrix} \kappa_3 \\ \kappa_1\kappa_3 \\ \kappa_1\kappa_2\kappa_3 \end{bmatrix} \tag{3}$$

Base on steady-state heat conduction hypothesis as well as the law of mass conservation, the governing equation for a certain organic pollutant transport in the GMB layer with defects via molecular diffusion, advection and thermodiffusion is (Peng et al., 2020)

$$\frac{\partial C_1(z,t)}{\partial t} = D_1 \frac{\partial^2 C_1(z,t)}{\partial z^2} - v_{\mathrm{g}} \frac{\partial C_1^{\mathrm{h}}(z,t)}{\partial z} + G_{T,1} S_T D_1 \frac{\partial C_1(z,t)}{\partial z} \tag{4}$$

where $C_1(z,t)$ and D_1 are the mass concentration and molecular diffusion coefficient of the organic pollutant in the GMB layer, respectively; S_T represents the Soret coefficient of the pollutant in landfill leachate; $C_1^{\mathrm{h}}(z,t)$ represents the organic pollutant concentration of the solution in the holes in geomembrane, which is $1/K_{\mathrm{g}}$ times $C_1(z,t)$ in accordance with Henry's law; K_{g} is the partition coefficient for the pollutant to the GMB; v_{g} equals to the Darcy velocity for the leakage flow in the composite liner, which can be determined by Rowe's equation (Eq. (14) in Feng et al. (2019a)).

The governing equation for thermal diffusion, advection and molecular diffusion of pollutant through GCL and SL is (Xie et al., 2015c)

$$\frac{\partial C_i(z,t)}{\partial t} = \frac{D_i}{R_{\mathrm{d},i}} \frac{\partial^2 C_i(z,t)}{\partial z^2} - \frac{v_i - G_{T,i} S_T D_i}{R_{\mathrm{d},i}} \frac{\partial C_i(z,t)}{\partial z}, \quad i = 2, 3 \tag{5}$$

where $C_i(z,t)$ is the pollutant concentration in the i^{th} layer; $R_{\mathrm{d},i}$ is the retardation factor for the organic pollutant in the i^{th} layer (it is a coefficient regarding to the adsorption capacity of the i^{th} layer to the pollutant. It can be obtained by $R_{\mathrm{d},i} = 1 + \rho_{\mathrm{d},i} K_{\mathrm{d},i}/n_i$ in this model that the adsorption is a linear process, where $K_{\mathrm{d},i}$, n_i and $\rho_{\mathrm{d},i}$, are the distribution coefficient, the porosity and the dry density of the i^{th} layer, respectively); D_i and v_i are the pollutant effective diffusion coefficient and seepage velocity of the flow respectively in the i^{th} layer.

The continuity condition of the organic pollutant concentration at the GMB-GCL interface is

$$\frac{C_1(z=z_1,t)}{K_g^{'}} = C_2(z=z_1,t) \tag{6}$$

As for the continuity condition of the mass flux, it is

$$D_1\frac{\partial C_1(z_1,t)}{\partial z} = n_2 D_2 \frac{\partial C_2(z_1,t)}{\partial z} \tag{7}$$

where $K_g^{'}$ is the partition coefficient, which generally equals to K_g (Rowe et al., 2016).

Similarly, for the GCL-SL interface, the continuity conditions are

$$C_2(z=z_2,t) = C_3(z=z_2,t) \tag{8}$$

$$n_2 D_2 \frac{\partial C_2(z_2,t)}{\partial z} = n_3 D_3 \frac{\partial C_3(z_2,t)}{\partial z} \tag{9}$$

The initial condition is

$$C_i(z,0) = C_{i,\text{ ini}}, \quad i=1,2,3 \tag{10}$$

where $C_{i,\text{ini}}$ represents the initial concentration of organic pollutant in the i^{th} layer.

The concentration of the organic pollutant in landfill leachate C_0 has the relationship with the concentration at the upper surface of GMB (Peng et al., 2021; Sangam and Rowe, 2005):

$$C_1(0,\ t) = C_0 K_g \tag{11}$$

where K_g is the partition coefficient.

Neumann type boundary condition is chosen to simulate the transport process at the bottom surface:

$$\frac{\partial C_3(z=z_3,t)}{\partial z} = 0 \tag{12}$$

this expression is applicable for the scenario that the substratum underneath the liner system is an obstructed GCDL or an aquitard layer (Peng et al., 2021). Moreover, the model can be used as a design tool to depict pollutant transport in liner system (Peng et al., 2021).

A normalized model is preferred for facilitating deriving the solution, which demands the governing equation is written in a unified form, so do the continuity equations for mass flux and the concentration (Feng et al., 2019a; Peng et al., 2021). According to the steps in Peng et al. (2021), the model can be normalized by means of replacing $R_{d,\text{gmb}}$, n_{gmb}, z, $G_{T,\text{gmb}}$ and $C_{\text{gmb}}(z, t)$ by K_g^2, 1, z^*, $G^*_{T,\text{gmb}}$ and $C^*_{\text{gmb}}(z^*, t)$, respectively, where

$$G_{T,i}^* = \begin{cases} K_g G_{T,i}, & i=1 \\ G_{T,i}, & i=2,3 \end{cases} \tag{13}$$

$$C_i^*(z^*,t) = \begin{cases} \dfrac{C_i(z,t)}{K_g}, & 0\leqslant z\leqslant z_1 \\ C_i(z,t), & z>z_1 \end{cases} \tag{14}$$

$$z^* = \begin{cases} \dfrac{z}{K_g}, & 0\leqslant z\leqslant z_1 \\ z+\left(\dfrac{1}{K_g}-1\right)z_1, & z>z_1 \end{cases} \tag{15}$$

The unified governing equation then can be obtained as (the normalization mark '*', for simplify, is omitted hereafter.).

$$\frac{\partial C_i(z,t)}{\partial t} = \frac{D_i}{R_{d,i}}\frac{\partial^2 C_i(z,t)}{\partial z} - \frac{v_i - G_{T,i}S_T D_i}{R_{d,i}}\frac{\partial C_i(z,t)}{\partial z}, \quad i=1,\ 2,\ 3 \tag{16}$$

Submitting Eq. (17) into the unified model, the original problem can be divided into two subproblems (i.e., subproblem 1: $w_i(z,t)$ and subproblem 2: $u_i(z)$) based on superposition principle.

$$C_i(z,t) = w_i(z,t) + u_i(z)C_0, \quad i=1,\ 2,\ 3 \tag{17}$$

For the subproblem 1, the governing equation can be obtained as.

$$\frac{\partial w_i(z,t)}{\partial t} = \frac{D_i}{R_{d,i}}\frac{\partial^2 w_i(z,t)}{\partial z^2} - \frac{v_i - G_{T,i}S_T D_i}{R_{d,i}}\frac{\partial w_i(z,t)}{\partial z}, \quad i=1,\ 2,\ 3 \tag{18}$$

The solution is

$$w_i(z,t) = \sum_{m=1}^{\infty} C_m g_{m,i}(z) e^{a_i z - \beta_m t}, \quad i=1,\ 2,\ 3 \tag{19}$$

the function $g_{m,i}(z)$ is

$$g_{m,i}(z) = \begin{cases} A_{m,i}\sin\left(\mu_i\lambda_{m,i}\dfrac{z}{H}\right) + B_{m,i}\cos\left(\mu_i\lambda_{m,i}\dfrac{z}{H}\right), & \text{when } \beta_m - \dfrac{(v_i - G_{T,i}S_T D_i)^2}{4D_i R_{d,i}} \geqslant 0, \ i=1,\ 2,\ 3 \\ A_{m,i}\sinh\left(\mu_i\lambda_{m,i}\dfrac{z}{H}\right) + B_{m,i}\cosh\left(\mu_i\lambda_{m,i}\dfrac{z}{H}\right), & \text{when } \beta_m - \dfrac{(v_i - G_{T,i}S_T D_i)^2}{4D_i R_{d,i}} < 0, \ i=1,\ 2,\ 3 \end{cases} \tag{20}$$

β_m, a_i and C_m are coefficients.

For the subproblem 2, the governing equation is

$$\frac{D_i}{R_{d,i}}\frac{d^2 u_i(z)}{dz^2} - \frac{v_i - G_{T,i}S_T D_i}{R_{d,i}}\frac{du_i(z)}{dz} = 0, \quad i=1,\ 2,\ 3 \tag{21}$$

The solution is

$$u_i(z) = k_{i,1}e^{r_{i,1}z} + k_{i,2}e^{r_{i,2}z}, \quad i=1,\ 2,\ 3 \tag{22}$$

Thus, the solution is achieved to the normalized model (see Eq. (17)). The solution can be finally obtained by the reverse normalization process defined in Eqs. ((13)-(14)) to the original problem. The organic pollutant concentration C is semi-coupled with the temperature T in the model. In summary, the heat conduction model first provides the magnitude of the temperature gradient G_T, which can be used as the known information to solve C.

3. Model verification

To the best of the authors' knowledge, considering the coupling effect of thermodiffusion, advection and molecular diffusion, there is no experimental data or analytical solution for solute migration in landfill composite liner so far. Only Rosanne et al. (2003) announced an experiment about sodium chloride thermodiffusion-diffusion transport in a compacted clay. Therefore, the model is verified with experimental results to test its performance in monolayer porous media. Then, its competence for solute migration in composite liner is further tested using a numerical method.

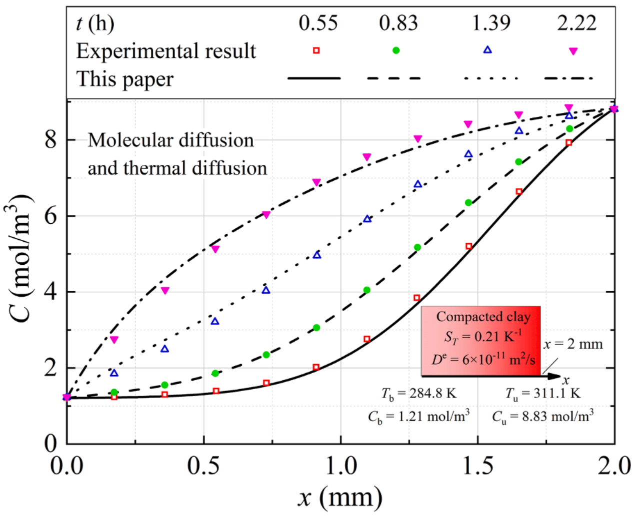

Fig. 2. Comparison of the developed analytical solution with available experimental results (Rosanne et al. [33]) regarding to NaCl concentration profiles.

3.1. Comparisons with available experimental data

As shown in the sketch map in Fig. 2, Rosanne et al. (2003) tested the molecular diffusion and thermodiffusion migration of NaCl in a compacted clay (2-millimeter-thick). A fixed temperature gap of 25°C was applied between the defined upper and lower surfaces of the compacted clay. The gradients of the temperature and the concentration were in the same direction. The required parameters are listed in Fig. 2. Even though the bottom surface was a constant concentration boundary for the test whereas zero concentration gradient boundary is used in the developed model (see Eq. (12)), in fact, the model remains valid for 'constant concentration' situations with minor adjustments. The concentration curves forecasted by the model in this paper for 0.55-, 0.83-, 1.39- and 2.22-hour agree well with experimental data (see Fig. 2), illustrating that the proposed model is well performed in portraying the molecular diffusion and thermodiffusion of pollutant in porous medium.

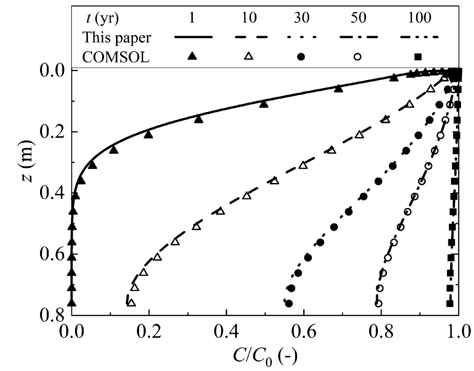

Fig. 3. Comparison of the developed analytical model with COMSOL Multiphysics 5.4 regarding to the concentration profiles in GMB/GCL/SL landfill liner system.

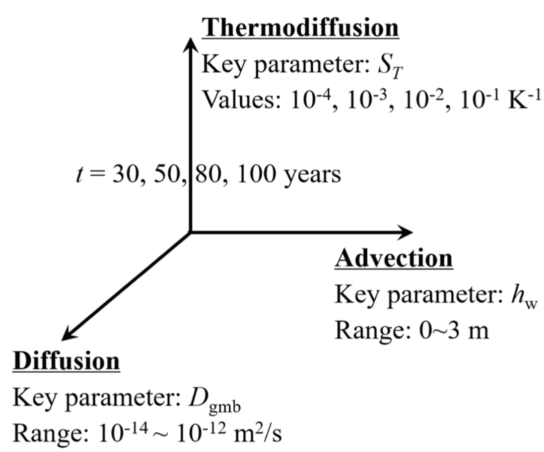

Fig. 4. Key parameters for the analysis.

Table 1
Material characteristics and environmental parameters (the data in branket is the value range of the corresponding parameter).

Key parameters	GMB	GCL	SL	References
Effective diffusion coefficient, D (×10^{-10} m^2/s)	0.003 (0.0001–0.01)	3.0 (1–5)	8.0 (0.02–15)	Feng et al. (2019a)
Partition coefficient, K_g (-)	100 (10–1000)	–	–	Feng et al. (2019a)
Distribution coefficient, K_d (mL/g)	–	8.7 (1–50)	1.60 (0.1–100)	Rowe et al. (2005); Lake and Rowe (2005)
Thickness, L (mm)	0.0015 (0.001–0.003)	0.01 (0.005–0.01)	0.30 (0.3–2)	Feng et al. (2019a)
Porosity, n (-)	–	0.70 (0.6–0.8)	0.40 (0.3–0.6)	Feng et al. (2019a)
Dry density, ρ_d (g/cm^3)	–	0.79	1.62	Xie et al. (2011); Feng et al. (2019a)
Hydraulic conductivity, k (×10^{-9}m/s)	–	0.01 (0.005–0.5)	100 (1–100)	Feng et al. (2019a)
Leachate head, h_w (m)	1 (0.1–3)			Feng et al. (2019a)
Soret coefficient, S_T (/K)	0.03 (0.0001–0.1)			Feng et al. (2019a)
Frequency of holes, m_h (-)	2.5 (2.5–50)			Feng et al. (2019a)
Wrinkle length, L_w (m)	500 (10–1000)			Feng et al. (2019a)
Transmissivity, θ (×10^{-10} m^2/s)	2 (0.02–2)			Feng et al. (2019a)
Threshold concentration, C_a (mg/L)	0.7			Feng et al. (2019a)
Concentration of toluene in leachate, C_0 (mg/L)	5			Feng et al. (2019a)
Background concentration in liners, C_{ini} (mg/L)	0			Feng et al. (2019a)

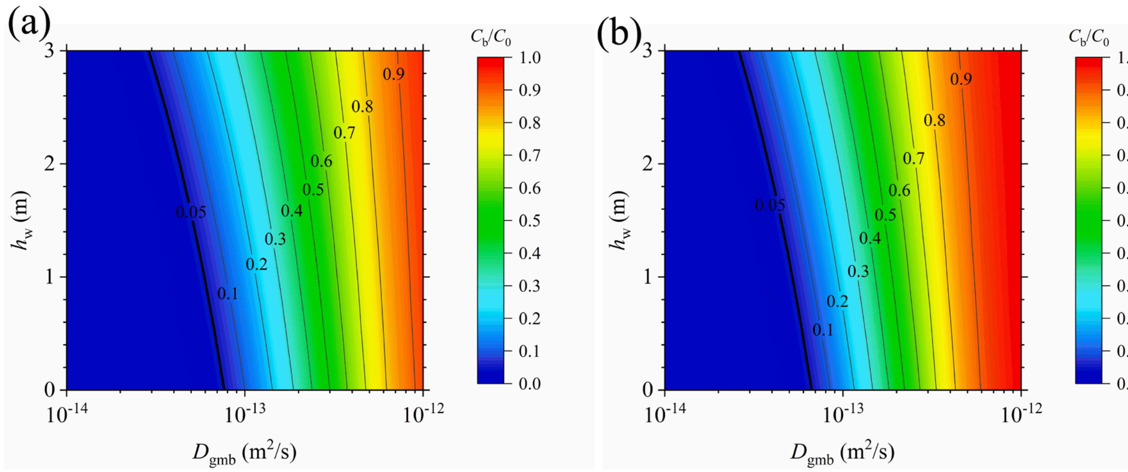

Fig. 5. Relative bottom concentration of the organic contaminant for the GMB/GCL/SL liner system with (a) $S_T = 0$ and (b) $S_T \neq 0$ ($S_T = 0.03$ K^{-1}) when the elapsed time is 30 years.

3.2. Comparison with numerical simulation result

A commercial finite element software, COMSOL Multiphysics v5.4, is chosen to further validate the developed model in this part. Toluene is adopted as the target organic pollutant in the leachate (Kjeldsen et al., 2002; McWatters and Rowe, 2014; Rosanne et al., 2003). The transport of toluene in a MCPRC recommended GMB/GCL/SL composite liner is studied here considering the effect of molecular diffusion, advection, thermodiffusion and linear adsorption. The necessary parameters are summarized in Table 1. Fig. 3 shows the concentration distributions predicted by the present model and the numerical model. The calculated results of the two models are in reasonable agreement, manifesting the fine performance of the developed model in simulating pollutant transport in the composite liner.

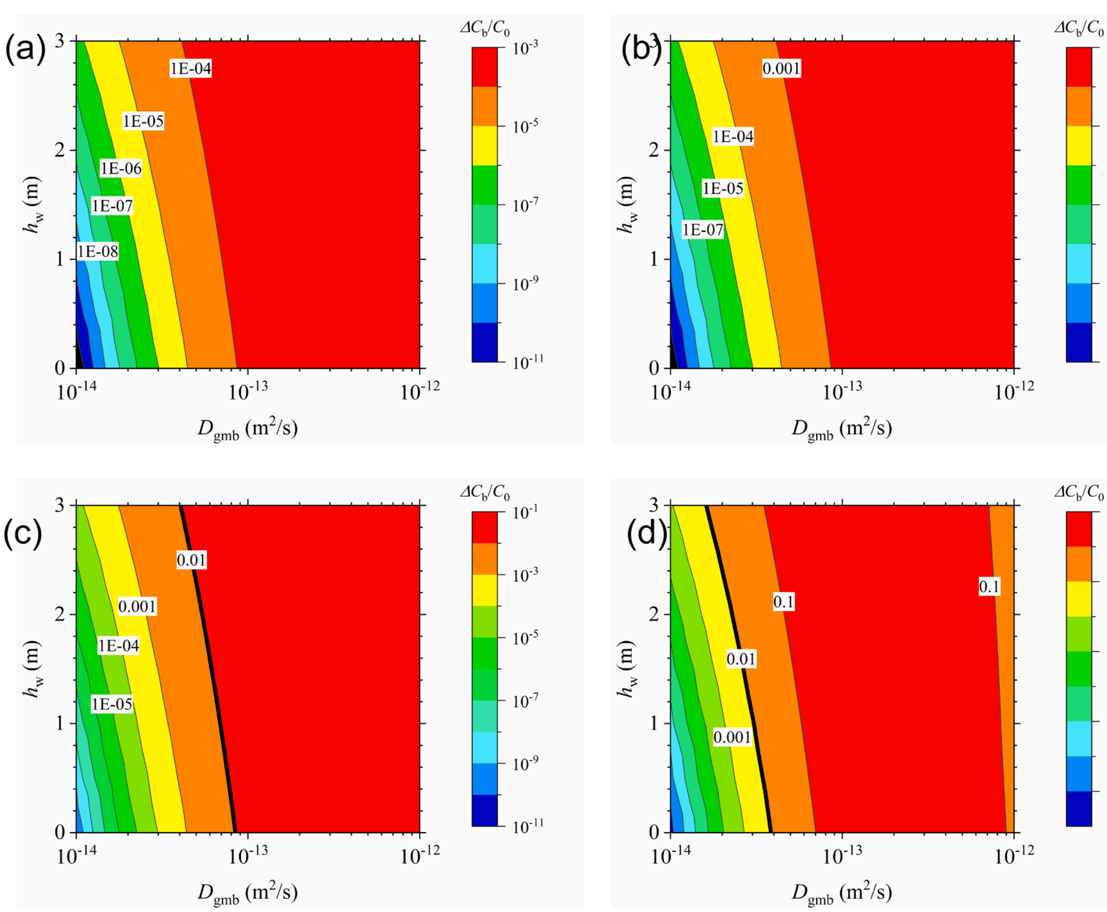

Fig. 6. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 30 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

4. Influence of thermodiffusion on pollutant migration in typical composite liners

The governing transport mechanisms of pollutants in landfill liner system generally include molecular diffusion, advection, adsorption and thermodiffusion. Great attentions have been paid to the first three mechanisms. For example, the effect of advection on pollutants transport in liner system has been synthetically studied in literatures (Feng et al., 2019a; Guan et al., 2014; Xie et al., 2015a). However, the investigation into the last mechanism, i.e. thermodiffusion, is rare, because most analytical studies assumed that the landfill liner works in an isothermal environment. Peng et al. (2020, 2021) studied the influence of thermodiffusion on the barrier performance of landfill liner system. However, these models cannot consider the coupling effect of advection and thermodiffusion. Therefore, using the present model, this paper attempts to systematically quantify the condition under which the thermodiffusion influence on pollutant transport in liner system cannot be ignored.

Toluene is one of the most common organic pollutants in landfill leachate (Feng et al., 2019a; Peng et al., 2020; Xie et al., 2015b). In addition, the transport speed of toluene is relative faster due to its weak polarity and small molecular diameter. Thus, toluene is selected here as the representative pollutant to assess the barrier performance of typical composite liners.

4.1. Parameter selection

The key transport parameters for the molecular diffusion of pollutants in the GMB/GCL/SL liner system are D_{gmb}, D_{gcl} and D_{sl}, the ranges of which are $1 \times 10^{-14} - 1 \times 10^{-12}$ m^2/s, $1 \times 10^{-10} - 5 \times 10^{-10}$ m^2/s and $2 \times 10^{-12} - 1.5 \times 10^{-9}$ m^2/s, respectively (see Table 1). The range of D_{gcl} is relative narrow and the influence of its variation on the barrier performance of the composite liner is mild. Thus, D_{gcl} herein is set to a constant, 3×10^{-10} m^2/s. Fig. 4 shows the key parameters for the analysis. For simplify, D_{sl} is set to 1000 times D_{gmb}.

The key parameter dominating the advection process in the composite liner is Darcy velocity, which is mainly related to the defect frequency, landfill leachate head, porosity and hydraulic conductivity (Feng et al., 2019a, 2019b; Xie et al., 2010, 2011,2018; Zhan et al., 2014). The defect frequency is usually assumed to be 2.5–5, which has proved to be reasonable in calculating the leachate flux for composite liner in good quality assurance (Feng et al., 2019a; Giroud and Bonaparte, 2001; Rowe, 2005). The leakage rate depends on the hydraulic conductivity of GCL layer, k_{gcl}, for GMB/GCL/SL liner system. Usually, the hydraulic conductivity of GCL layer is over a narrow range. Thus, it is assumed 1×10^{-11} m/s here (see Table 1). The other parameters, for example, the porosity of GCL and SL is 0.7 and 0.4, respectively (see Table 1). Apart from k_{gcl}, the leachate head h_w has great effect on the leakage rate of the landfill leachate. Therefore, this part pays special attention to the leachate head, and it is assumed $0 \sim 3$ m (see Fig. 4).

The key parameters governing the thermal diffusion process in multilayer porous media system are Soret coefficient, temperature difference and thermal conductivity (Peng et al., 2020, 2021). For landfill composite liner systems, the thermal conductivity has proved to have little influence on the barrier performance of liner system (Peng et al., 2020, 2021). A fixed temperature difference, i.e. 25 K, is chosen in this section. And the effect of Soret coefficient on the liner breakthrough time is studied considering its range of $1 \times 10^{-4} - 1 \times 10^{-1}$ K^{-1}. Thus,

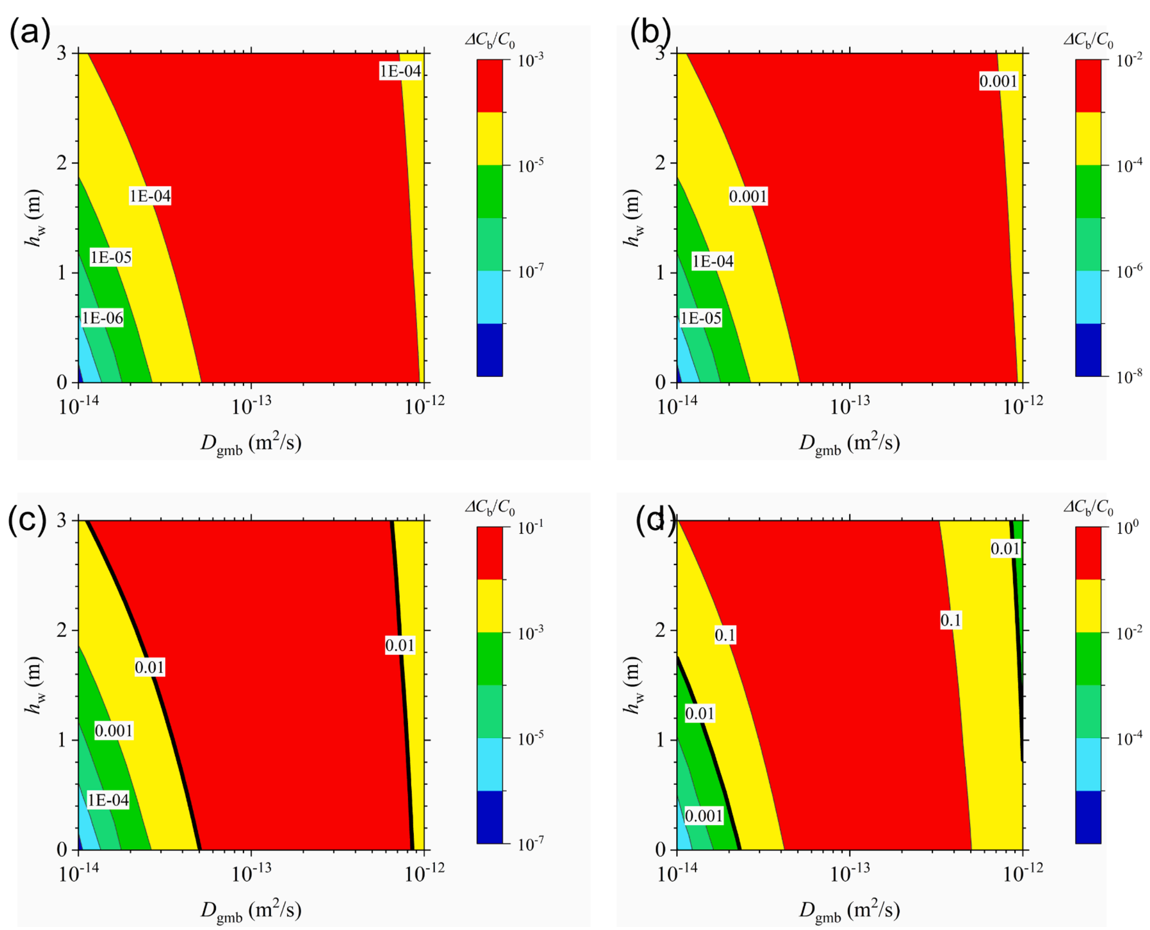

Fig. 7. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 50 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

four Soret coefficients (10^{-4}, 10^{-3}, 10^{-2}, 10^{-1} K^{-1}) are chosen for the analysis (see Fig. 4).

The key parameter dominating the linear adsorption process is distribution coefficient or partition coefficient (Lake and Rowe, 2005; Rowe et al., 2005). Herein, the distribution coefficients of GCL and SL are 8.7 and 1.6 mL/g, respectively (see Table 1). And the partition coefficient of the GMB is 100.

4.2. Influence of considering/ignoring thermodiffusion

The relative concentrations of toluene at the bottom boundary surface of the GMB/GCL/SL liner system at $t = 30$ years with $S_T = 0$ and 0.03 K^{-1} are shown in Fig. 5(a) and 5(b), respectively. The bottom concentration at upper-right area is higher than that at lower-left area (see Fig. 5), illustrating that the liner system with high leachate head and high molecular diffusion coefficient would be broken through within a relatively short time. Given identical level of D_{gmb} and h_{w}, the bottom concentration considering the influence of thermodiffusion (see Fig. 5b) is higher than that ignoring the effect (see Fig. 5a). For example, with $D_{\text{gmb}} = 1 \times 10^{-13}$ m^2/s and $h_{\text{w}} = 1$ m, the bottom relative concentration is 0.159 and 0.224 for $S_T = 0$ and 0.03 K^{-1}, respectively. Omitting the impact of thermodiffusion leads to underestimating the bottom concentration by 29 %. The following part would study this issue in a more general situation.

4.3. Quantitative analysis about the condition under which the influence of thermodiffusion should not be ignored

For the elapsed time $t = 30$, 50, 80 and 100 years, the corresponding underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion is given in Figs. 6-9, respectively. For the elapsed time $t = 30$ years, the distribution of the underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion with $S_T = 10^{-4}$, 10^{-3}, 10^{-2} and 10^{-1} K^{-1} is shown in Fig. 6 (a)-6(d), respectively. All the underestimation ratio for $S_T = 10^{-4}$ and 10^{-3} K^{-1} are lower than 1 % (see Fig. 6a and 6b, defined as "neglectable area"), and the same results can be observed in Figs. 7-9, illustrating that the thermodiffusion impact on organic pollutant migration in the liner system can be ignored. In contrast, when S_T increases to 10^{-2} K^{-1} (see Fig. 6c), a large area where the underestimation ratio $\Delta C_b/C_0$ is higher than 1 % appears at the upper-right conner (defined as "unneglectable area"). In this area, the influence of thermal diffusion should not be ignored. When S_T further increases to 10^{-1} K^{-1} (see Fig. 6d), the underestimation ratio increases and the unneglectable area enlarges toward the lower-left conner. When the elapsed time further increases to 50 years and S_T is not smaller than 10^{-2} K^{-1} (see Fig. 7c and 7d), the unneglectable area moves toward lower-left conner. A neglectable area appears at the upper-right conner because the transport reaches the steady state. Thus, at the early time, thermodiffusion effect still should be considered in this area. Similar phenomena also can be observed for $t = 80$ and 100 years (see Fig. 8c, 8d, 9c and 9d).

In addition, the unneglectable areas with $S_T = 10^{-1}$ K^{-1} is larger than

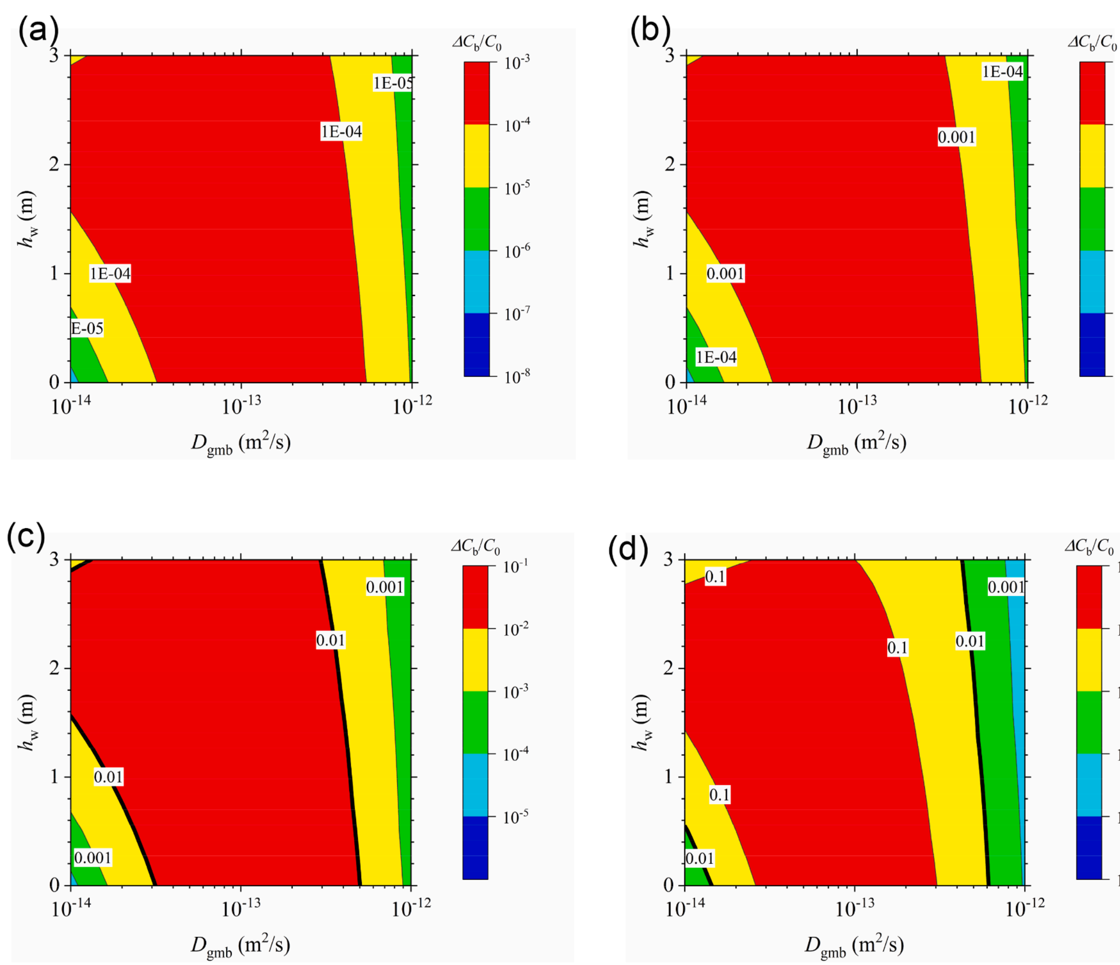

Fig. 8. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 80 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

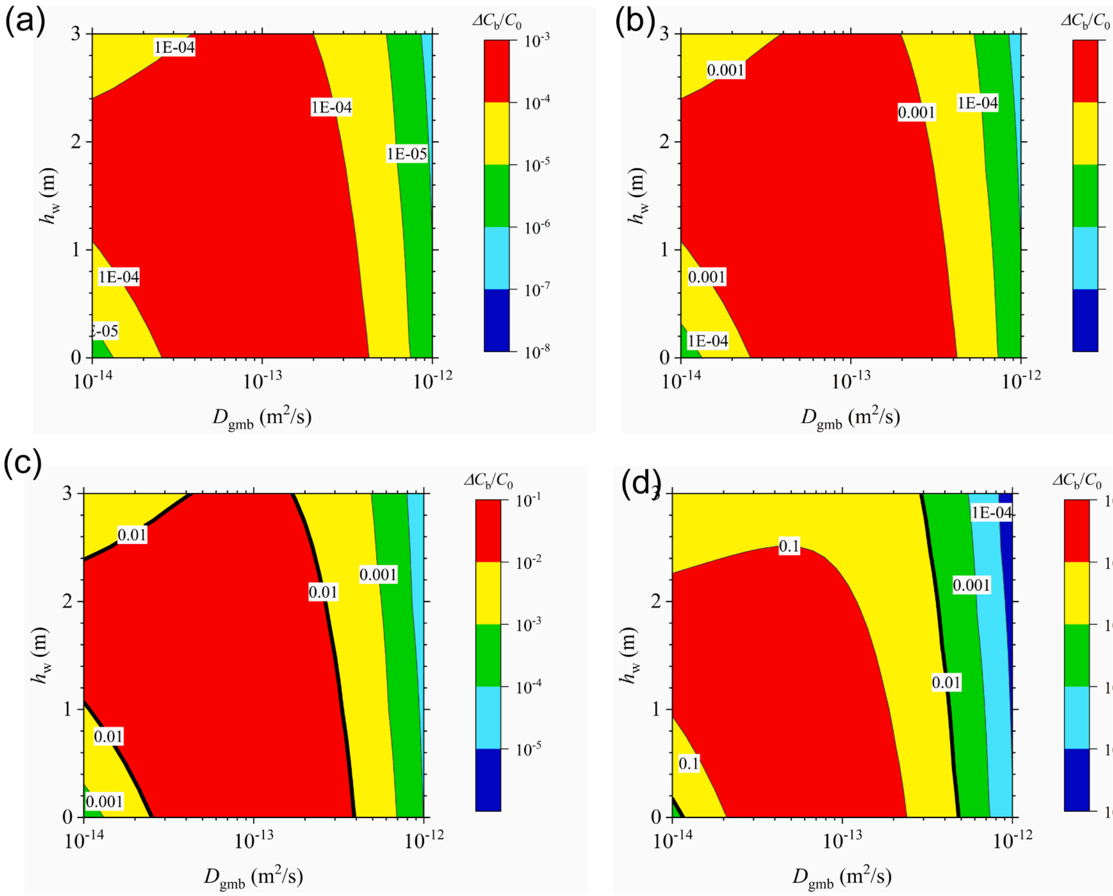

Fig. 9. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 100 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

that with $S_T = 10^{-2}$ K^{-1}. For the scenario with $S_T = 10^{-2}$ K^{-1}, only when h_w and D_{gmb} are very small (see Fig. 9c), the thermodiffusion effect can be ignored. This area will be overwhelmed by unneglectable area when S_T increases to 10^{-1} K^{-1} (see Fig. 9d). Therefore, the thermodiffusion should not be boldly ignored when the S_T exceeds 0.01 K^{-1}. The conclusions hereinbefore are obtained when D_{sl} is 1000 times as large as D_{gmb}. What needs to be emphasized is that these conclusions generally are still applicable when D_{sl} is 500 times as large as D_{gmb} (see the Appendix).

5. Summaries and conclusions

Considering the mechanisms of molecular diffusion, advection, thermodiffusion and linear adsorption process, a new analytical model is proposed for the transient migration of organic pollutant in a GMB/GCL/SL composite liner in this paper. The separation of variables method and superposition principle are adopted to tackle this transport problem. The model can be used to design composite liner systems and verify numerical models.

The model is validated against experimental results and a numerical model first. The influence of thermodiffusion on pollutant migration in the typical composite liner is then investigated. For simplify, two representative ratios of D_{sl}/D_{gmb} (i.e., 1000 and 500) are adopted to conduct the analysis. The results indicate that the thermodiffusion may have great effect on the pollutant transport. When the Soret coefficient exceeds 0.01 K^{-1}, the influence of thermodiffusion generally cannot be ignored on pollutant migration in typical composite liners, regardless of the combination conditions in terms of molecular diffusion, advection and adsorption.

CRediT authorship contribution statement

Ming-Qing Peng: Conceptualization, Methodology, Software, Writing – original draft. **Shi-Jin Feng:** Funding acquisition, Supervision, Conceptualization, Writing – review & editing. **Hong-Xin Chen:** Writing – review & editing. **Zhang-Long Chen:** Methodology, Validation, Software.

Declaration of Competing Interest

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.

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Appendix A. Distribution of underestimation ratio $\Delta C_b/C_0$ when D_{sl} is set to 500 times D_{gmb}

See Figs. A1-A4.

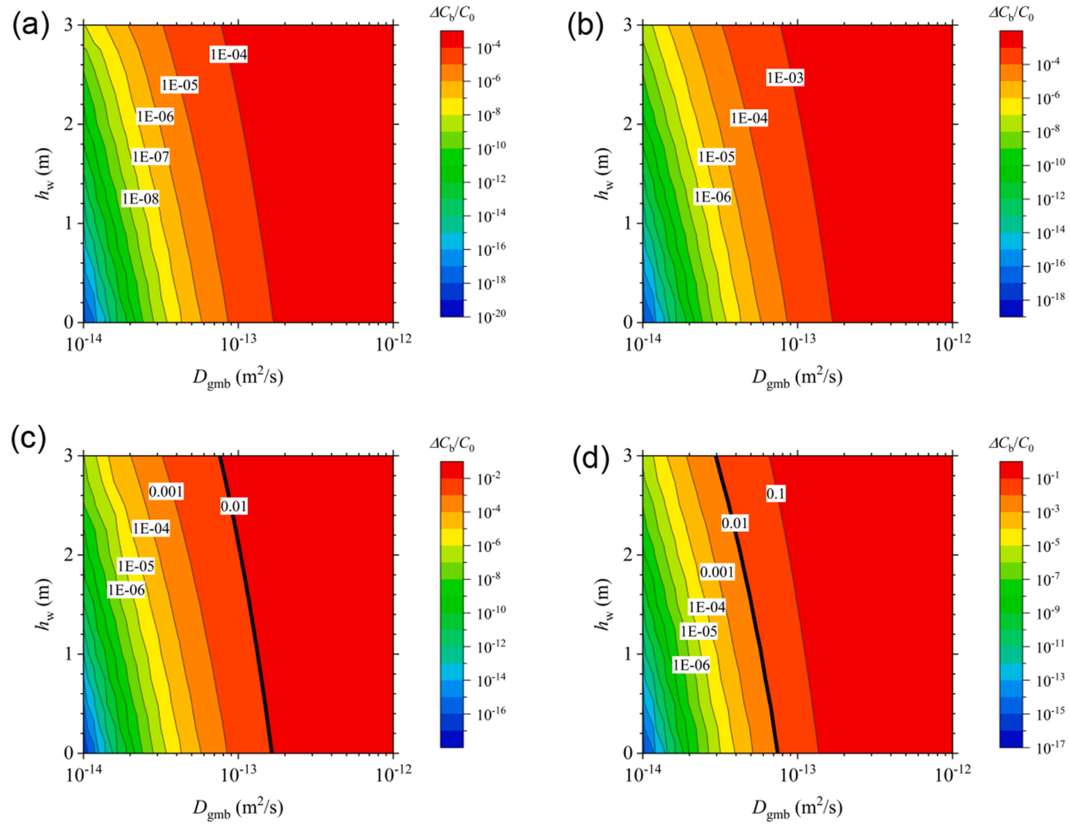

Fig. A1. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 30 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

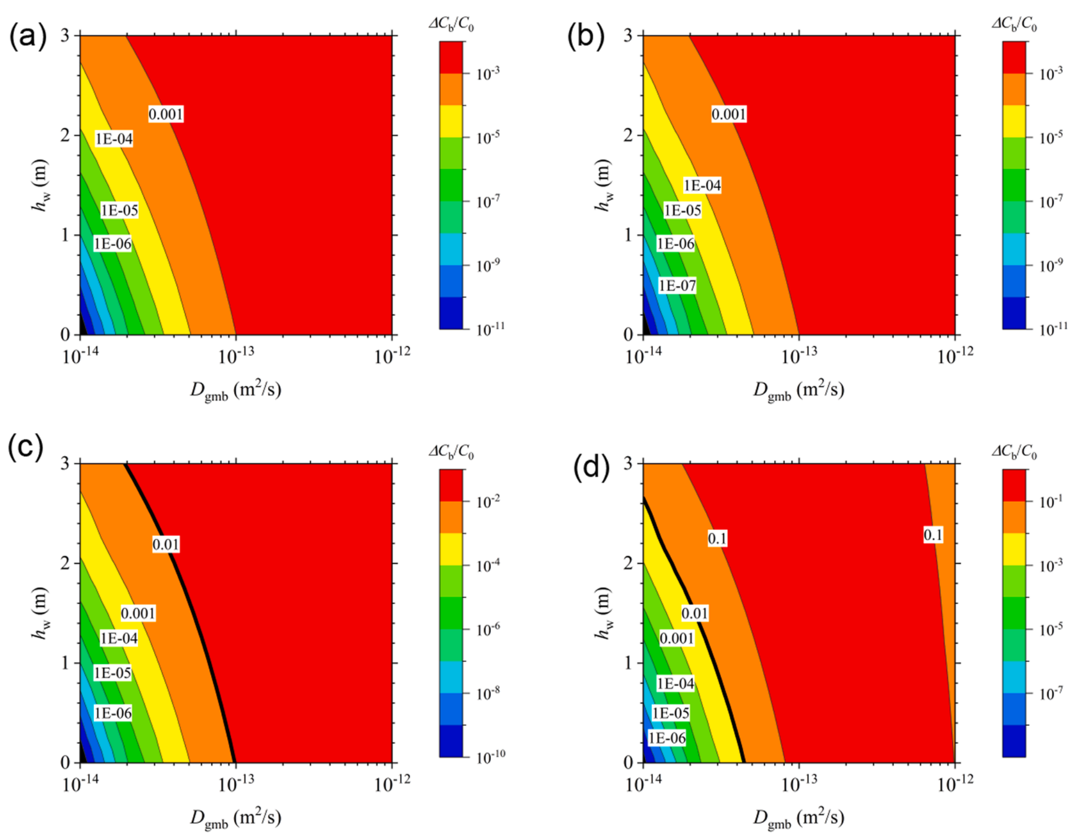

Fig. A2. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 50 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

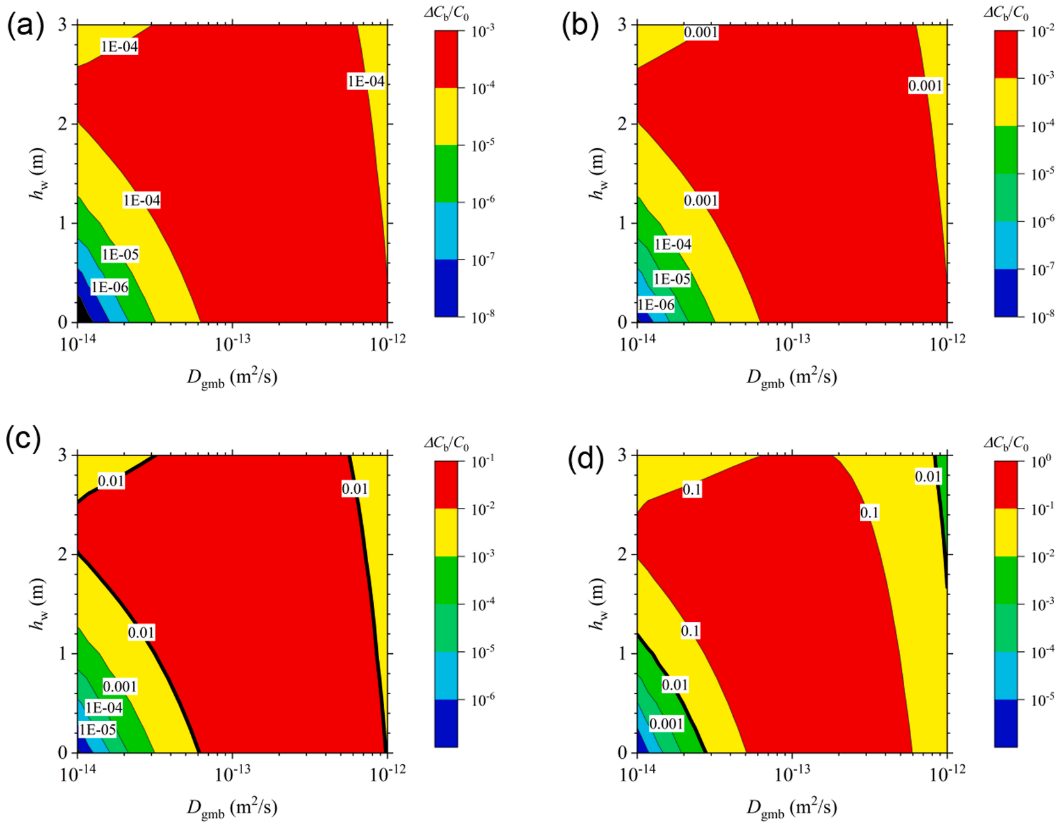

Fig. A3. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 80 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

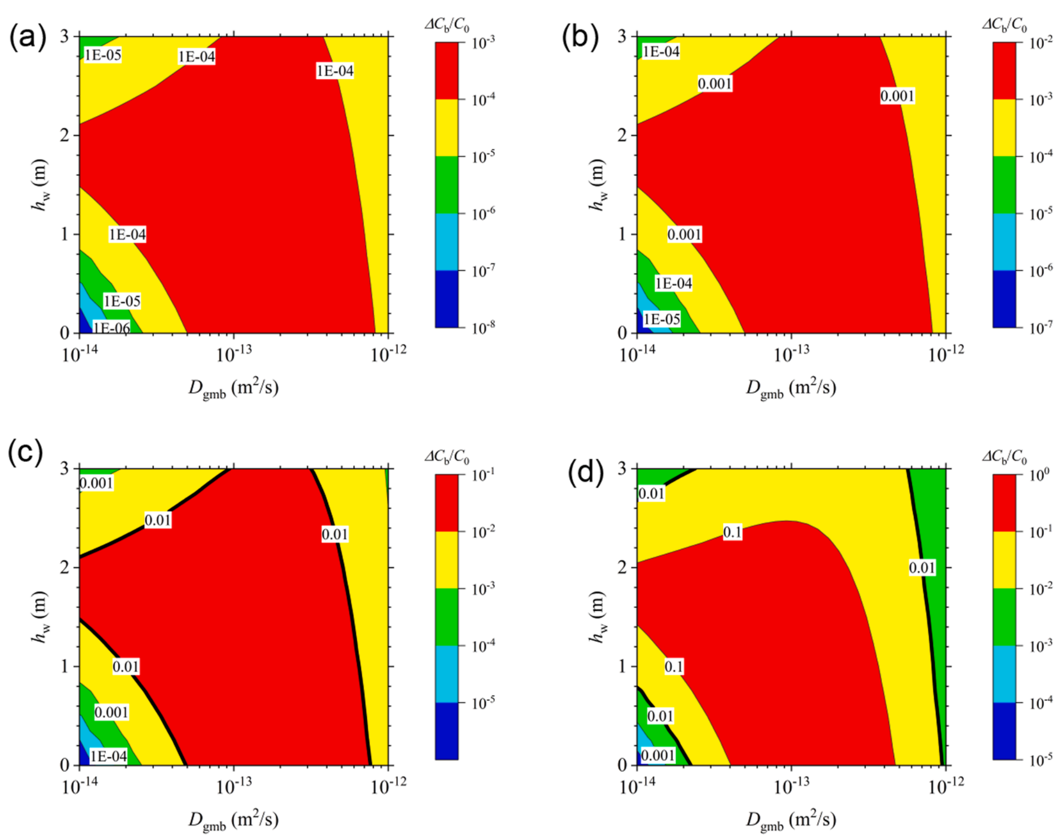

Fig. A4. Underestimation ratio $\Delta C_b/C_0$ due to neglecting thermodiffusion when the elapsed time is 100 years for (a) $S_T = 10^{-4}$ K^{-1}; (b) $S_T = 10^{-3}$ K^{-1}; (c) $S_T = 10^{-2}$ K^{-1}; (d) $S_T = 10^{-1}$ K^{-1}.

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