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A Pollutant Migration Model Considering Solute Decay in Layered Soil

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ABSTRACT: Organic pollutant solute undergoes significant decay during the migration process in clay liner systems and foundation clay. Liner and foundation soil have layered properties. A one-dimensional computational model is established to calculate pollutant migration by considering the decay in layered soil medium. The separation of variable method is used to obtain the analytical solution. To verify the capability of the developed method, a typical example is illustrated by applying this model. The calculated results are compared with the results obtained from the GAEA GAEA Pollute v7. Consistent results demonstrate the reliability and validity of the proposed migration model, which can be a promising tool for landfill liner design when considering the organic pollutant decay.

KEYWORDS: Pollutant migration, Layered soil, Decay, Analytical solution, Numerical validation.

1. INTRODUCTION

Sanitary landfill is one of the most widely used municipal solid waste disposal methods in the world. Recently, China has built nearly 500 formal landfills, over 3000 simple landfills, and more than 10,000 junk yards (Du et al. 2011; Chen et al. 2009). The amount of leachate, which contains large amounts of toxic and hazardous inorganic/organic contaminants, is increasing because of overburden pressure, rainwater infiltration, and garbage compression and degradation. The garbage in China is not strictly classified and sifted through. Therefore, a large number of kitchen waste in landfill leads to high organic acid content in the leachate because of several factors such as pressure, convection, diffusion, and dispersion. Pollutants can easily leak into the surrounding soil of the landfill and cause serious geoenvironmental problems. This phenomenon influences the engineering properties and environmental characteristics of soil and results in serious environmental engineering geological problems. Many landfills causeing significant pollution on the surrounding water or soil were reported. The contaminated landfills in China have constituted serious threats on the surrounding environment (Zha et al. 2012; Du and Hayashi 2005). Thus, it is an urgent need to carry out environmental assessment of contaminated sites with engineering treatment.

Clay is an important part of landfill and is also the main material of foundation soil for many contaminated sites. The migration of contaminants in the clay has physical (convection–diffusion and physical adsorption), chemical (chemical adsorption, ion exchange, adsorption, chemical reactions, etc.), and biological (microbial growth, degradation, and other effects) (Rowe et al. 2004; Zheng 2009) effects on pollutant transportation processes. Organic pollutants are mainly present in clay and may appear in the following states: free, volatile, dissolved, and solid. The vast majority of organic pollutants are volatile organic pollutants. These volatile organic contaminants migrate in the soil, escape into the air or water, or move out of the soil by organism absorption through volatilization, shower, and diffusion of concentration gradient. Analysis of the organic migration shows that organic matter can easily produce the chelating reaction with other heavy metals. Therefore, the decay of organic pollutant solute must be considered to improve the accuracy of pollutant migration estimation. Further research is required to investigate the decay behavior. Nevertheless, the lack of research does not mean that the process is overlooked.

Foose (2002) established a one-dimensional diffusion model of organic contaminants in a composite liner with semi-infinite space boundary solution. Chen et al. (2006) established a one-dimensional diffusion model of pollutants going through any layer composite liner with analytical solution in the boundary of finite thickness. By using the Laplace transform method, Rowe (2004, 2005) solved the free upper boundary conditions and the lower boundary conditions with diving layer in the bottom. They developed the GAEA Pollute Series software for simulating pollutant migration. Li et al. (2011) established a one-dimensional transport model by considering the role of pollutant convection and diffusion. On the basis of double-foundation consolidation theory, they obtained the theoretical solution of contaminant migration. Xie et al. (2009) established the organic pollutant migration diffusion–adsorption model by setting the landfill lining system and analyzed the permeability characteristics of different liner systems. Luan et al. (2005) assumed the biodegradable effects of pollution sources and pollutant adsorption in the soil markings of an aquifer under limited depth on the basis of convection, dispersion, and geochemical reaction mechanism. Then they proposed a one-dimensional model that could consider landfill contaminant transport processes. However, few studies on pollutant migration issues of layered clay have considered pollutant decay. Actual landfill liner systems and polluted foundation soil usually have limited thickness and layered properties.

Under the assumption that molecular diffusion dominates pollutant migration, this paper considered the issue of the proliferation of pollutant decay in layered soil medium and established a one-dimensional diffusion model, and ignored the effects of advection. The paper utilized the variable separation method for the analytical solution.

2. POLLUTANT DISOERSION MODEL

The established one-dimensional diffusion model, which considers the effects of pollutant decay in layered soil, is mainly based on the following assumptions: (1) layered clay is homogeneous and saturated; (2) the retardation factor and the effective diffusion coefficient in the *i*th-layer of soils are constants and don't change with time and space; (3) the diffusion of pollutants is one-dimensional, and the sources of the contaminants are in the upper soil, whereas other external sources of pollution are not considered; (4) the spread of pollutants in the leachate is caused only by the solubility gradient of pollution matter, ignoring the role of hydraulic conductivity while taking the linear adsorption of soil particles into account; (5) the effects of decay on the dispersion process is considered.

With these assumptions, the paper simplified the equations of contaminant migration in the soil and established a one-dimensional diffusion model that considers decay in layered soil (see Figure 1). For the *i*th-layered soil, the pollutant diffusion equation can be written as follow:

$$\frac{\partial C_i}{\partial t} = \frac{D_i}{R_i}\frac{\partial^2 C_i}{\partial z^2} - \lambda_i C_i \tag{1}$$

where $C_i(z,t)$ is the pollutant concentration in the *i*th-layered soil with a unit of mg/L; R_i is the retardation factor of the contaminants in leachate in the *i*th-layered soil; D_i is the diffusion coefficient of pollutant dispersion in the *i*th-layered soil with a unit of m^2/a; λ_i is the decay coefficient of the pollutant in the *i*th-layered soil; $\lambda = \ln 2 / T_{1/2}$ with $T_{1/2}$ as the half-life of the contaminants in soil with a unit of year; n_i is the porosity of the *i*th-layered soil; z is the depth; t is time.

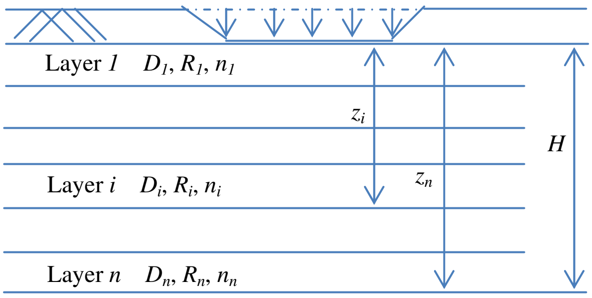

Figure 1 One-dimensional pollutant diffusion in layered soil

The boundary conditions can be expressed as

$$z=0:\quad C_1(0,t)=C_0,\quad t\geq 0 \tag{2}$$

$$z=H:\quad C_n(H,t)=0,\quad t\geq 0 \tag{3}$$

Assuming that the soil has not been contaminated before the existence of the pollution sources, the initial condition can be described as

$$t=0:\quad C_i(z,0)=0\quad (i=1,2,...,n) \tag{4}$$

The continuity conditions of the concentration and flux in the interface of layered soil can be expressed separately as

$$C_i(z_i,t)=C_{i+1}(z_i,t) \tag{5}$$

$$n_i D_i \frac{\partial C_i(z,t)}{\partial z}\Big|_{z=z_i} = n_{i+1} D_{i+1} \frac{\partial C_{i+1}(z,t)}{\partial z}\Big|_{z=z_i+1} \tag{6}$$

3. ANALYTICAL SOLUTION

The definite solution of the nonhomogeneous boundary conditions can be transformed into the definite solution of other unknown function of the homogeneous boundary conditions. By using the principle of superposition, we set

$$C_i(z,t)=u_i(z)C_0+\omega_i(z,t) \tag{7}$$

Substituting Eq. (7) into Eq. (1) yields

$$\frac{\partial \omega_i(z,t)}{\partial t} = \frac{D_i}{R_i}\frac{\partial^2 \omega_i(z,t)}{\partial z^2} - \lambda\omega_i + \frac{D_i}{R_i}C_0 u''_i(z) - \lambda_i C_0 u_i(z) \tag{8}$$

Equation (8) can be reduced to the solution of the following definite problems:

Definite problem 1:

$$\frac{D_i}{R_i}u''_i(z) - \lambda u_i(z)=0 \tag{9}$$

Definite problem 2:

$$\frac{\partial \omega_i(z,t)}{\partial t} = \frac{D_i}{R_i}\frac{\partial^2 \omega_i(z,t)}{\partial z^2} - \lambda_i\omega_i \tag{10}$$

To solve definite problem 1:

The following equation can be obtained on the basis of Eq. (9):

$$u_i(z)=k_{i1}e^{r_i z}+k_{i2}e^{-r_i z} \tag{11}$$

where $r_i^2 = \dfrac{\lambda_i R_i}{D_i}$. The following recursive relationship can be obtained according to the boundary conditions (Eqs. (2) and (3)) and according to the continuous conditions (Eqs. (5) and (6)) of the interface:

$$\begin{bmatrix} k_{i+1,1} \\ k_{i+1,2} \end{bmatrix} = \begin{bmatrix} \frac{1}{2}(1+\alpha_i)e^{(r_i-r_{i+1})z_i} & \frac{1}{2}(1\text{-}\alpha_i)e^{-(r_i+r_{i+1})z_i} \\ \frac{1}{2}(1\text{-}\alpha_i)e^{(r_i+r_{i+1})z_i} & \frac{1}{2}(1+\alpha_i)e^{(r_{i+1}-r_i)z_i} \end{bmatrix} \begin{bmatrix} k_{i1} \\ k_{i2} \end{bmatrix} \tag{12}$$

where $\alpha_i = \dfrac{n_i D_i r_i}{n_{i+1}D_{i+1}r_{i+1}}$.

Set

$$\begin{bmatrix} k_{i+1,1} \\ k_{i+1,2} \end{bmatrix} = N_i \begin{bmatrix} k_{i1} \\ k_{i2} \end{bmatrix} \tag{13}$$

Therefore,

$$\begin{bmatrix} k_{n1} \\ k_{n2} \end{bmatrix} = M_{n-1} \begin{bmatrix} k_{11} \\ k_{12} \end{bmatrix} \tag{14}$$

In Eq. (14), set $M_{n-1} = N_{i-1}N_{i-2}...N_1 = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix}$.

M_{n-1} is obtained from various soil parameters.

Here,

$$\begin{bmatrix} k_{11} \\ k_{12} \end{bmatrix} = \begin{bmatrix} \dfrac{e^{r_n H}M_{12}+e^{-r_n H}M_{22}}{e^{r_n H}(M_{12}-M_{11})+e^{-r_n H}(M_{22}-M_{21})} \\ \dfrac{e^{r_n H}M_{11}+e^{-r_n H}M_{21}}{e^{r_n H}(M_{11}-M_{12})+e^{-r_n H}(M_{21}-M_{22})} \end{bmatrix} \tag{15}$$

On the basis of Eqs. (14) and (15), any soil coefficient k_{i1} and k_{i2} can be obtained. The solution of $u_i(z)$ is determined by substituting the soil coefficients into Eq. (11).

To solve definite problem 2:

According to the solution ideas of Lee et al. (1992), set

$$\omega_i(z,t) = \sum_{m=1}^{\infty} C_m[A_{mi}\sin(\mu_i\lambda_m \frac{z}{H}) + B_{mi}\cos(\mu_i\lambda_m \frac{z}{H})]e^{-\beta_m t} \tag{16}$$

Substituting Eq. (16) into Eq. (10) yields

$$\beta_m = \frac{D_i}{R_i}(\frac{\mu_i\lambda_m}{H})^2 + \lambda_i = \frac{D_1}{R_1H^2}\lambda_m^2 + \lambda_i \tag{17}$$

where $\mu_i = \sqrt{\frac{D_1R_i}{R_1D_i}}$.

On the basis of the continuous conditions of the interface (Eqs. (5) and (6)), the following equation can be obtained:

$$A_{mi}\sin(\mu_i\lambda_m \frac{z_i}{H}) + B_{mi}\cos(\mu_i\lambda_m \frac{z_i}{H})$$

$$= A_{m,i+1}\sin(\mu_{i+1}\lambda_m \frac{z_i}{H}) + B_{m,i+1}\cos(\mu_{i+1}\lambda_m \frac{z_i}{H}) \tag{18}$$

$$n_iD_i\frac{\mu_i\lambda_m}{H}A_{mi}\cos(\mu_i\lambda_m \frac{z_i}{H}) - B_{mi}n_iD_i\frac{\mu_i\lambda_m}{H}\sin(\mu_i\lambda_m \frac{z_i}{H})$$

$$= n_{i+1}D_{i+1}\frac{\mu_{i+1}\lambda_m}{H}A_{m,i+1}\cos(\mu_{i+1}\lambda_m \frac{z_i}{H}) -$$

$$n_{i+1}D_{i+1}\frac{\mu_{i+1}\lambda_m}{H}B_{m,i+1}\sin(\mu_{i+1}\lambda_m \frac{z_i}{H}) \tag{19}$$

Set $\bar{A}_i = \sin(\mu_{i+1}\lambda_m \frac{z_i}{H})$; $\bar{B}_i = \sin(\mu_i\lambda_m \frac{z_i}{H})$; $\bar{C}_i = \cos(\mu_{i+1}\lambda_m \frac{z_i}{H})$; $\bar{D}_i = \cos(\mu_i\lambda_m \frac{z_i}{H})$; $\gamma_i = \frac{n_iD_i\mu_i}{n_{i+1}D_{i+1}\mu_{i+1}}$.

The matrix form of Eqs. (5) and (6) is

$$\begin{bmatrix} \bar{B}_i & \bar{D}_i \\ \gamma_i\bar{D}_i & -\gamma_i\bar{B}_i \end{bmatrix}\begin{bmatrix} A_{mi} \\ B_{mi} \end{bmatrix} = \begin{bmatrix} \bar{A}_i & \bar{C}_i \\ \bar{C}_i & -\bar{A}_i \end{bmatrix}\begin{bmatrix} A_{m,i+1} \\ B_{m,i+1} \end{bmatrix} \tag{20}$$

$$\begin{bmatrix} A_{m,i+1} \\ B_{m,i+1} \end{bmatrix} = \begin{bmatrix} \bar{A}_i\bar{B}_i + \gamma_i\bar{C}_i\bar{D}_i & \bar{A}_i\bar{D}_i - \gamma_i\bar{B}_i\bar{C}_i \\ \bar{B}_i\bar{C}_i - \gamma_i\bar{A}_i\bar{D}_i & \bar{C}_i\bar{D}_i + \gamma_i\bar{A}_i\bar{B}_i \end{bmatrix}\begin{bmatrix} A_{mi} \\ B_{mi} \end{bmatrix} \tag{21}$$

Set $S_i = \begin{bmatrix} \bar{A}_i\bar{B}_i + \gamma_i\bar{C}_i\bar{D}_i & \bar{A}_i\bar{D}_i - \gamma_i\bar{B}_i\bar{C}_i \\ \bar{B}_i\bar{C}_i - \gamma_i\bar{A}_i\bar{D}_i & \bar{C}_i\bar{D}_i + \gamma_i\bar{A}_i\bar{B}_i \end{bmatrix}$.

The following equation can be derived on the basis of the boundary condition in Eq. (2):

$$\begin{bmatrix} A_{m1} \\ B_{m1} \end{bmatrix} = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \tag{22}$$

The following equation can be obtained on the basis of the boundary condition in Eq. (3):

$$[\sin(\mu_n\lambda_m) \quad \cos(\mu_n\lambda_m)].S_{n-1}.S_{n-2}...S_1\begin{bmatrix} 1 \\ 0 \end{bmatrix} = 0 \tag{23}$$

The infinite number of characteristic value λ_m can be determined by using Eq. (23).

The following equation can be derived by using the initial conditions in Eq. (4):

$$\sum_{m=1}^{\infty} C_m[A_{mi}\sin(\mu_i\lambda_m \frac{z}{H}) + B_{mi}\cos(\mu_i\lambda_m \frac{z}{H})] = -u_i(z)C_0 \tag{24}$$

Set

$$g_{mi}(z) = A_{mi}\sin(\mu_i\lambda_m \frac{z}{H}) + B_{mi}\cos(\mu_i\lambda_m \frac{z}{H}) \tag{25}$$

The following equation can be derived on the basis of the orthogonal nature of the quasi-orthogonal function (Schiffman and Stein 1970; Tittle 1965; Bulavin and Kashcheev 1965):

$$\sum_{i=1}^{\infty}\int_{z_{i-1}}^{z_i} n_iR_ig_{mi}g_{ni}\mathrm{d}z = 0 \qquad (\mathrm{m} \neq n) \tag{26}$$

Thus,

$$C_m = \frac{\sum_{i=1}^{\infty}\int_{z_{i-1}}^{z_i} -u_i(z)C_0n_iR_ig_{mi}\mathrm{d}z}{\sum_{i=1}^{\infty}\int_{z_{i-1}}^{z_i} n_iR_ig_{mi}^2\mathrm{d}z} \tag{27}$$

Substituting the above coefficient into Eq. (16), we obtain $\omega_i(z,t)$. The solution of $C_i(z,t)$ is achieved. Furthermore, the solution of pollutant flux is identified:

$$f_i(z,t) = -n_iD_i\frac{\partial C_i}{\partial z} = -n_iD_ir_i(k_{i1}e^{r_iz} - k_{i2}e^{-r_iz}) - \frac{n_iD_i\mu_i}{H}\cdot$$

$$\sum_{m=1}^{\infty} C_m\lambda_m[A_{mi}\cos(\mu_i\lambda_m \frac{z}{H}) - B_{mi}\sin(\mu_i\lambda_m \frac{z}{H})]e^{-\beta_m t} \tag{28}$$

4. VERIFICATIONS AND ANALYSES

The typical example of one-dimensional migration of contaminants in layered soils by Chen et al. (2006) is adopted. Some basic parameters are summarized in Table 1. The boundary conditions include the top concentration (i.e., 1.0) and the bottom concentration (i.e., 0). When $\lambda = 0$, the present model can be reduced to the model of Chen et al. (2006); both calculated results are consistent. The present calculated results are compared with the results of the pollutant analysis software GAEA Pollute V7. Figure 2 shows that for both cases, whether decay is considered or not, the pollutant concentration along the depth at different time in the soil is consistent. This finding demonstrates the reliability of the analytical solution of the proposed model.

Figure 3 provides the order of the distribution of pollutant concentration when soil layers 1 and 2 have changed after 30, 60, and 120 years. Soil distribution affects the distribution of pollutant concentration. When the small diffusion coefficient is in the upper level, the pollutant concentration in the soil will be relatively low. Figure 4 shows the changes in the soil layers after 120 years. Pollutant concentration varies along the depth whether the half-life of pollutants is considered or not. The half-life of pollutants has a significant effect on the distribution of pollutant concentration.

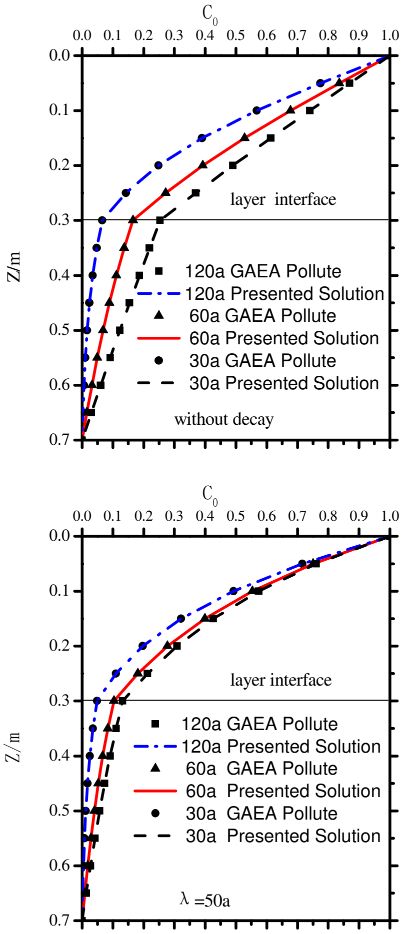

Figure 2 Comparisons between the results of the present model and the numerical results of Chen et al. (2006)

Table 1 Properties of two-layered soil

Property	Layer 1	Layer 2
Diffusion coefficient D(m^2/s)	6.5×10^{-11}	1.3×10^{-10}
Retardation factor R	4.0	2.0
Porosity n	0.3	0.5
Thickness h(m)	0.3	0.4

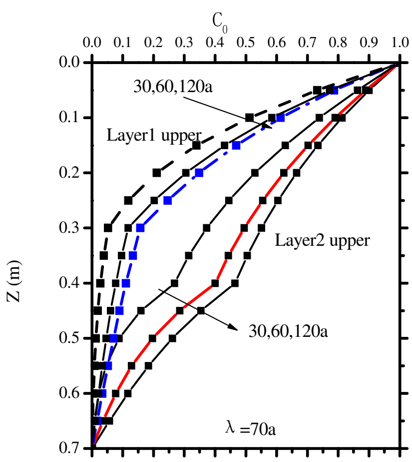

Figure 3 Effect of layered soil on the concentration

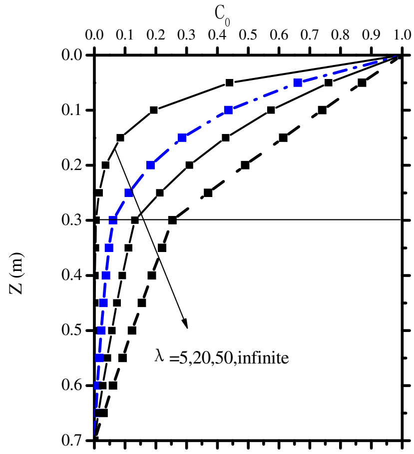

Figure 4 Effect of half-life on the concentration after 120 years

Figure 5 shows the evolution of concentration over time at different depths with a half-life of 50 years. The pollutant concentration decays quickly with the change of depth. After 60 years, the pollutant concentration becomes relatively stable. Figure 6 shows the evolution of concentration over time at the depth of 0.1 m with different half-life periods. With the shortening of the half-life of the pollutants, the concentration significantly decreases and the time for the pollutant concentration to be stable is reduced. However, when the half-life is 5 years or about 10 years, the pollution concentration is relatively stable. Thus, the diffusion caused by upper soil pollutants and the decay of layer are basically balanced.

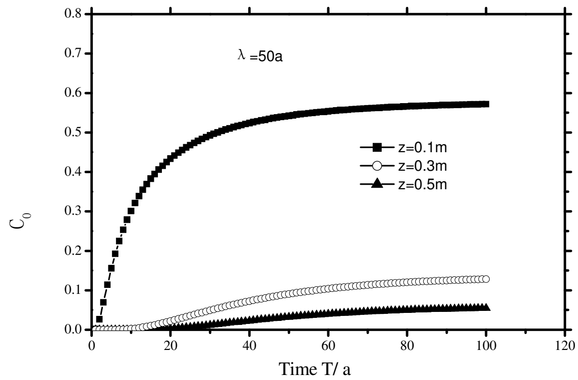

Figure 5 Concentration versus time at different depths

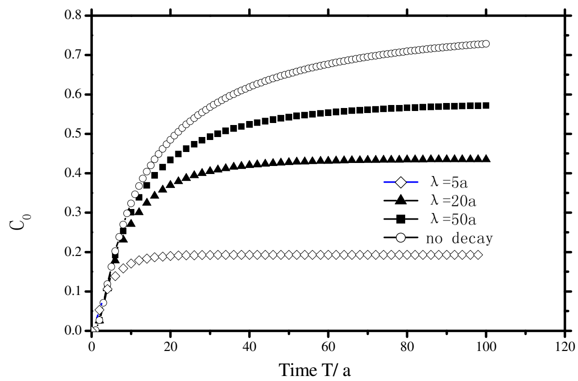

Figure 6 Concentration versus time at depth z = 0.1 m

5. CONCLUSIONS

A one-dimensional diffusion model of pollutants in layered soil that considers pollutant decay was proposed. The analytical solution was obtained by using the method of variable separation and the orthogonal nature of the quasi-orthogonal functions. The model was validated through the calculation of a typical example and by comparing the results with those of the GAEA Pollute software. Analyses show that the half-life of pollutants significantly decreases the concentration of contaminants in the soil layer. In the layered soil, the upper soil has good impermeable performance and can effectively reduce the concentration of contaminants in the soil. The analytical model can be used as a migration model accounting for pollutant decay properties and as preliminary reference for landfill liner design. Given that this calculation model is relatively simple, further considerations such as the nonlinear adsorption effect of convection and clay should be discussed.

6. ACKNOWLEDGEMENTS

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