JOURNAL OF THE KOREAN SOCIETY OF AGRICULTURAL ENGINEERS DEC. 2006, pp. 55-63

JOURNAL OF THE KOREAN SOCIETY OF AGRICULTURAL ENGINEERS, Vol. 48, No. 7

Evaluation of Bacterial Transport Models for Saturated Column Experiments

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Abstract

Bacterial transport models were evaluated in this study to determine the suitable model at describing bacterial transport in saturated column experiments. Four models used in the evaluation were: advective-dispersive equation (ADE) + equilibrium sorption/retardation (ER) + kinetic reversible sorption (KR) (Model 1), ADE + two-site sorption (Model 2), ADE + ER + kinetic irreversible sorption (KI) (Model 3), ADE + KR + KI (Model 4). Firstly, analyses were performed with the first experimental data, showing that Model 4 is appropriate for describing bacterial transport. Even if Model 1 and 2 fit well to the observed data, they have a defect of not including the irreversible sorption, which is directly related to mass loss of bacteria. Model 3 can not properly describe the tailing observed in the data. However, further analysis with the second data indicates that Model 4 can not describe retardation of bacteria, even if the sorption-related parameters are varied. Therefore, Model 4 is modified by incorporating retardation factor into the model, resulting in the improved fitting to the data. It indicates that the transport model, into which retardation, kinetic reversible sorption, and kinetic irreversible sorption are incorporated, is suitable at describing bacterial transport in saturated column experiments. It is expected that the selected transport model could be applied to properly analyze the bacterial transport in saturated porous media.

Keywords : Bacteria transport, Sorption, Transport modeling, Column experiment

I. Introduction

Remediation of contaminated soils and groundwater using bioremediation technologies attracts considerable attention since they can be applied in situ effectively at relatively low cost (Sturman et al., 1995). Among them, bioaugmentation is practiced by introducing bacteria with specific metabolic capabilities of degrading target contaminants, and thus successful delivery of contaminant-degrading bacteria to the targeted area is a subject of great interest (Gross and Logan, 1995).

Numerous studies have been carried out using various types of bacteria to improve understanding of bacterial transport and attachment in

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porous media (Harvey and Garabedian, 1991; Kinoshita et al., 1993; Mills et al., 1994; Tan et al., 1994; Rijnaarts et al., 1996; Schäfer et al., 1998; Hendry et al., 1999; Li and Logan, 1999; Lahlou et al., 2000). It was found that movement of bacteria is mainly controlled by advective-dispersive transport and attachment to solid matrix (Hornberger et al., 1992). Deposition of bacteria on solid matrix is affected by properties of porous media (e.g., surface charge, grain size), solution chemistry (e.g., ionic strength and pH) and surface characteristics of bacteria (e.g., cell surface charge and hydrophobicity) (Gannon et al., 1991; Fontes et al., 1991).

To examine bacterial transport and sorption in porous media, laboratory column experiments are usually carried out, and observed breakthrough data are analyzed using mathematical models. In the analysis of bacterial data, it is important to apply an appropriate model. Depending on the researchers, however, different transport models, composed of the advective-dispersive equation and various types of sorption terms, are used. It is therefore necessary to determine what type of model is suitable at describing bacterial transport in saturated column experiments. In this study, bacterial transport models were evaluated to find an appropriate model using the reported column experimental data and models from the literature review.

Ⅱ. Materials and Methods

1. Transport Models

Laboratory column experiments are carried out in homogeneous and saturated porous media under steady-flow condition and resting state of bacteria to exclude effects of biological processes such as growth and death on bacterial transport. In saturated porous media, bacterial sorption processes may include both reversible and irreversible sorptions. If the net forces of attractive London-van der Waals forces and repulsive electrostatic forces become attractive, attachment of bacteria to solid surfaces can occur at the secondary minimum, which is situated at a larger distance from solid surfaces. Bacteria may attach to the secondary minimum by the long-range forces and be readily detached from solid surfaces by shear forces (McEldowney et al., 1986). If hydrophobic interactions and polymer bridging affect bacteria attached to the secondary minimum, they may transfer to the primary minimum. In the potential energy curve for bacterial attachment to solid matrix, the primary minimum is located close to solid surfaces and responsible for strong attachment of bacteria. Bacteria held at the primary minimum due to the non-DLVO forces may irreversibly attach to matrix surfaces (McEldowney et al., 1986; Rijnaarts et al., 1995).

According to literature reviews, bacteria transport models were grouped into four models: (a) Model 1: The one-dimensional ADE with linear equilibrium sorption/retardation and first-order kinetic reversible sorption was (McCaulou et al., 1994):

$$\frac{\partial C}{\partial t} = \frac{D}{R}\frac{\partial^2 C}{\partial x^2} - \frac{v_w}{R}\frac{\partial C}{\partial x} - k_a C + k_d \frac{\rho_b}{\theta} S \quad \cdots\cdots (1)$$

$$\frac{\rho_b}{\theta}\frac{\partial S}{\partial t} = k_a C - k_d \frac{\rho_b}{\theta} S \quad \cdots\cdots (2)$$

where C is the concentration of bacteria suspended in the aqueous phase (M/L^3), D is the hydrodynamic dispersion coefficient (L^2/T), v_w is the pore-water velocity (L/T), R is the retardation factor, P_b is the dry bulk density of solid matrix (M/L^3), θ is the water content, k_a is the reversible adsorption rate coefficient (1/T), k_d is the desorption rate coefficient (1/T), and S is the mass of bacteria attached per unit mass of solid matrix (M/M).

(b) Model 2: the one-dimensional ADE with two-site sorption (instantaneous equilibrium sorption and kinetic reversible sorption) (Model 2) was (Lindqvist et al., 1991):

$$\left(1+\frac{f\rho_b K_s}{\theta}\right)\frac{\partial C}{\partial t}=D\frac{\partial^2 C}{\partial x^2}-v_w\frac{\partial C}{\partial x}-\frac{\alpha\rho_b}{\theta}[(1-f)K_sC-S] \quad (3)$$

$$\frac{\partial S}{\partial t}=\alpha[(1-f)K_sC-S] \quad \cdots\cdots(4)$$

where f is the fraction of equilibrium sorption site, α is the mass transfer rate coefficient (1/T) and K_s is the distribution coefficient of bacteria between the aqueous phase and solid matrix (L^3/M).

(c) Model 3: the one-dimensional ADE with linear equilibrium sorption/retardation and kinetic irreversible sorption was (Harvey et al., 1991):

$$\frac{\partial C}{\partial t}=\frac{D}{R}\frac{\partial^2 C}{\partial x^2}-\frac{v_w}{R}\frac{\partial C}{\partial x}-k_{ir}C \quad \cdots\cdots(5)$$

(d) Model 4: the one-dimensional ADE with first-order kinetic reversible and irreversible sorption terms (Model 4) was (Hendry et al., 1997):

$$\frac{\partial C}{\partial t}=D\frac{\partial^2 C}{\partial x^2}-v_w\frac{\partial C}{\partial x}-k_aC+k_d\frac{\rho_b}{\theta}S-k_{ir}C \quad (6)$$

$$\frac{\rho_b}{\theta}\frac{\partial S}{\partial t}=k_aC-k_d\frac{\rho_b}{\theta}S \quad \cdots\cdots(7)$$

2. Numerical Solutions

Numerical solutions of the models were obtained with the Crank-Nicolson Finite Difference Method along with the Thomas algorithm. Initial and boundary conditions used in the analysis were:

$$C(x,0)=S(x,0)=0 \quad \cdots\cdots(8)$$

$$-D\frac{\partial C}{\partial x}(0,t)+v_wC(0,t)=\begin{cases}v_wC_0 & at\ 0<t\le t_0\\ 0 & at\ t>t_0\end{cases} \quad (9)$$

$$\frac{\partial C}{\partial x}(L,t)=0 \quad \cdots\cdots(10)$$

where C_0 is the influent bacterial concentration, L is the column length, and t_0 is the duration of bacterial injection.

Transport parameters (v_w and D) were firstly determined from breakthrough data of conservative tracer, and using these parameters sorption-related parameters were estimated from bacterial data. Mass recovery (M_r) of conservative tracer (e.g. KCl) or bacteria at the effluent could be quantified from breakthrough curves using the following expression:

$$M_r=\left(\frac{\int_0^\infty Cdt}{C_ot_o}\right) \quad \cdots\cdots(11)$$

2. Column Data for Analysis

The first column data used in the analysis were from Hendry et al. (1999), who have performed column experiments in the saturated silica sand column (11.4 cm × 3.3 cm) with various flow rates. The observed data of *Klebsiella oxytoca* used in our analysis had flow rates ranging from 1.9 to 41.3 ml/h with pulse injection (Table 2). The second bacterial data were from Hendry et al. (1997), who have performed the column experiments in the saturated silica sand column (10.0 cm × 4.7 cm) using *K. oxytoca*.

Ⅲ. Results and Discussion

The model fitting to first data from Hendry et al. (1999) using Models 1~4 are given in Fig. 1~4, respectively. The fitted parameters are presented in Table 1. The observed bacterial data had characteristics of no retardation, considerable tailing, and lowered peak concentration

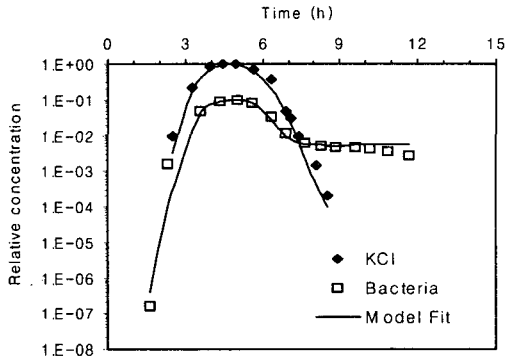


Fig. 2 Model fitting to column data (flow rate = 9.5 ml/h) of *Klebsiella oxytoca* from Hendry et al. (1999) using Model 2.

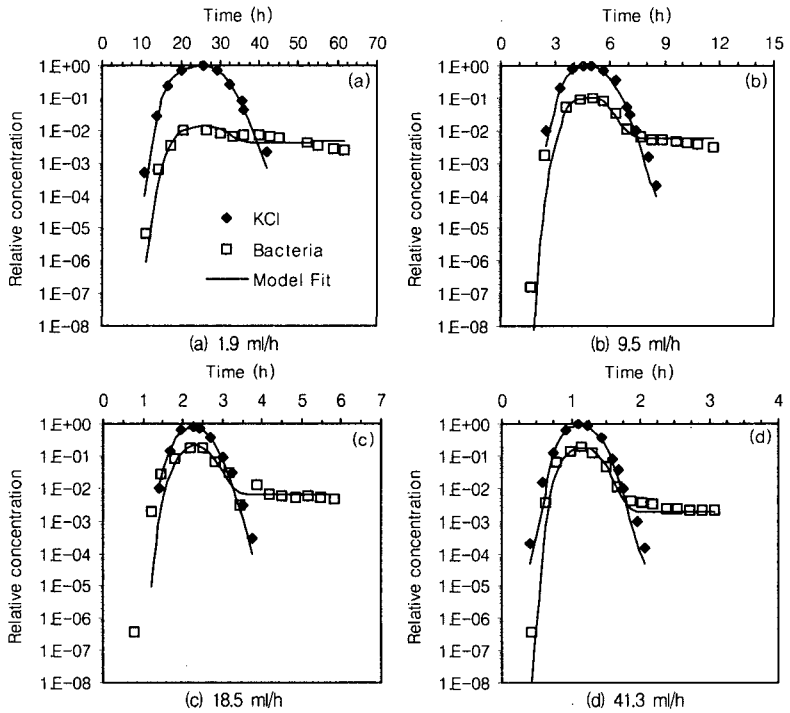


Fig. 1 Model fitting to column data of *Klebsiella oxytoca* from Hendry et al. (1999) using Model 1.

Table 1 Experimental conditions and fitting parameters for column experiments of *Klebsiella oxytoca* from Hendry et al. (1999).

Experimental conditions				
Exp.	Flow rate (ml/h)	Injection time (h)	v_w (cm/h)	D (cm^2/h)
1	1.9	12.3	0.6	0.072
2	9.5	2.2	3.1	0.372
3	18.5	0.7	6.0	0.720
4	41.3	0.5	13.4	1.608
Parameters from Model 1 (Figure 1)				
Exp.	R	k_a (1/h)	k_d (1/h)	SSE
1	1.0	2.25E-1	5.83E-3	5.62E-5
2	1.0	5.94E-1	1.07E-2	1.74E-4
3	1.0	7.06E-1	2.66E-2	1.29E-3
4	1.0	1.10E+0	1.21E-2	2.00E-3
Parameters from Model 2 (Figure 2)				
Exp.	R	a (1/h)	f	SSE
1	36.2	6.55E-4	1.68E-3	3.91E-4
2	59.7	9.36E-4	7.28E-4	1.49E-4
3	23.6	2.93E-3	1.81E-3	1.16E-2
4	67.5	2.54E-3	1.06E-4	7.32E-4
Parameters from Model 3 (Figure 3)				
Exp.	R	k_{ir} (1/h)		SSE
1	1.0	2.25E-1		2.19E-4
2	1.0	5.94E-1		4.13E-4
3	1.0	7.06E-1		1.70E-3
4	1.0	1.80E+0		1.99E-3
Parameters from Model 4 (Figure 4)				
Exp.	k_a (1/h)	k_d (1/h)	k_{ir} (1/h)	SSE
1	1.11E-1	2.28E-2	1.49E-1	3.43E-5
2	2.34E-1	3.52E-2	3.78E-1	7.86E-4
3	3.24E-1	9.65E-3	3.82E-1	4.54E-3
4	7.24E-1	4.50E-2	1.10E-0	1.65E-3

relative to chloride data.

As shown in Fig. 1 and 2, the observed data is simulated well by Model 1 and 2 with two reversible sorption terms, i.e., the equilibrium sorption (retardation) and the kinetic reversible sorption. However, Model 1 and 2 have a defect of not including irreversible sorption, which describes bacterial mass loss quantified in the bacterial data. Mass recoveries of bacteria were determined to be ranging from 2.0 to 19.0 %

(Hendry et al., 1999), indicating that considerable mass loss occurred during column experiments. Unless full mass recovery of bacteria is observed for the column experimental period, inclusion of kinetic reversible sorption only is not enough, requiring irreversible sorption to describe bacterial mass loss. In other words, incorporation of irreversible sorption into the model is the only way to describe bacterial mass loss because potential of mass loss due to bacterial death was excluded in a given column experimental condition. In addition, the two models show the difference regarding retardation factor. The obtained retardation factors from Model 1 ($R = 1.0$) accord well with no retardation observed in the bacterial data, while retardation factors from Model 2 are unreasonable ($23.6 \leq R \leq 67.5$).

The fitting results using the Model 3 are shown in Figure 3. Even though the model includes both reversible (equilibrium sorption) and irreversible sorption terms, it can not properly describe the tailing observed in the bacterial data. This defect comes from the incorporation of equilibrium sorption (retardation) instead of kinetic reversible sorption into the model, indicating that kinetic reversible sorption is essential in simulating the tailing effect (Hornberger et al, 1992; Fontes et al, 1991; Camesano et al, 1999).

The one important thing is that bacterial curve should be presented as a semi-log scale in y-axis in order to precisely examine the behavior of Model 3. As shown in Fig. 3a, the simulated curve declines continuously at the later time in the semi-log scale plot, clearly demonstrating the defect of Model 3. If the plot is presented in a normal scale, however, the defect

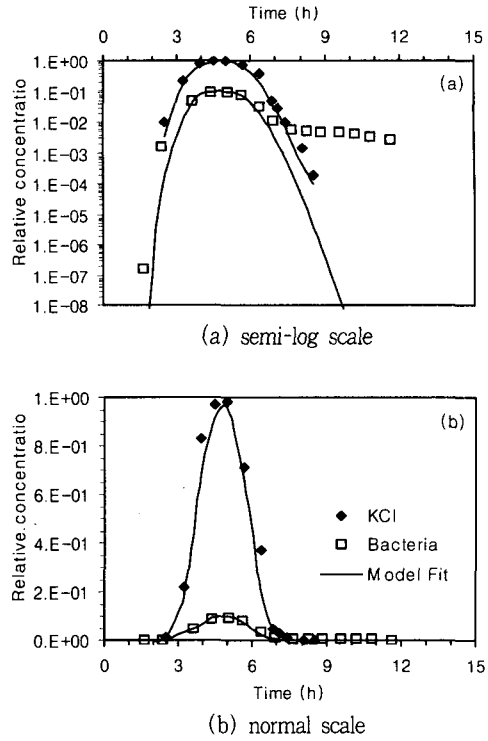


Fig. 3 Model fitting to column data (flow rate = 9.5 ml/h) of *Klebsiella oxytoca* from Hendry et al. (1999) using Model 3.

is masked (Fig. 3b), misleading that Model 3 is appropriate for describing the observed data.

As presented in Fig. 4, Model 4 gives good fitting result to column data, properly describing the tailing and mass loss observed in the bacterial BTC. In Model 4, reversible kinetic sorption instead of the equilibrium sorption is incorporated into the model along with irreversible sorption. Model 4 is considered as the most suitable among the four models at describing the bacterial transport in the saturated column. However, Model 4 has a problem at describing another bacterial data from Hendry et al. (1997), who have performed the column experiments using the same porous medium (silica sand) and bacteria (*K. oxytoca*). The observed data from

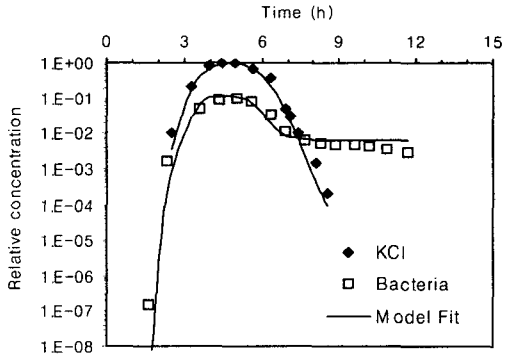


Fig. 4 Model fitting to column data (flow rate = 9.5 ml/h) of *Klebsiella oxytoca* from Hendry et al. (1999) using Model 4.

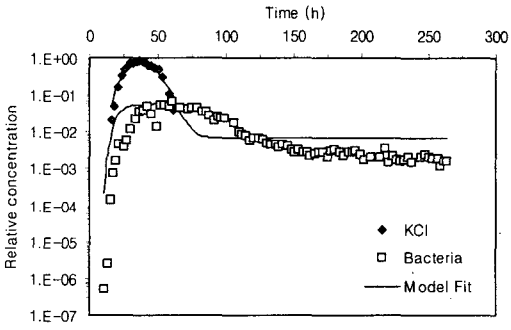
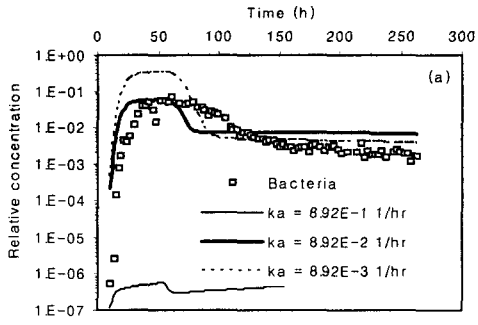


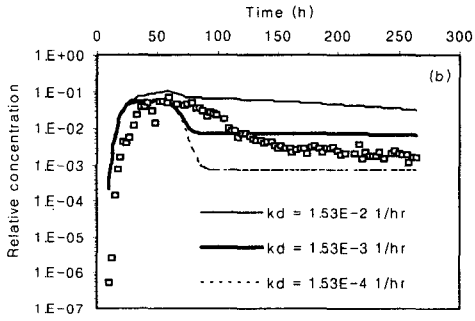
Fig. 5 Model fitting to column data (column length = 10 cm) of *Klebsiella oxytoca* from Hendry et al. (1997) using Model 4.

Hendry et al. (1997) had similar characteristics with those from Hendry et al. (1999), including considerable tailing and lowered peak concentration relative to chloride, but showed retardation compared to those from Hendry et al. (1999).

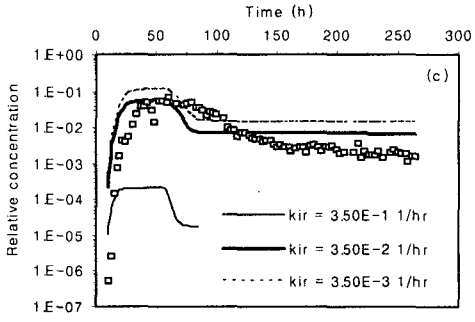
As shown in Fig. 5, Model 4 has a limitation



(a) reversible sorption rate coefficient (k_a)



(b) desorption rate coefficient (k_d)



(c) irreversible sorption rate coefficient (k_{ir})

Fig. 6 Effects of sorption parameters from Model 4 on the bacterial breakthrough curves.

Table 2 Parameters obtained from the model fitting to column data (column length = 10 cm) of *Klebsiella oxytoca* from Hendry et al.(1997)

	v_w (cm/h)	D (cm/h)	R	k_a (1/h)	k_d (1/h)	k_{ir} (1/h)	SSE
Model 4	0.37	0.048		8.92E-2	1.53E-3	3.50E-2	1.76E-2
Modified Model 4	0.37	0.048	1.8	4.25E-2	9.25E-4	2.45E-2	3.79E-3

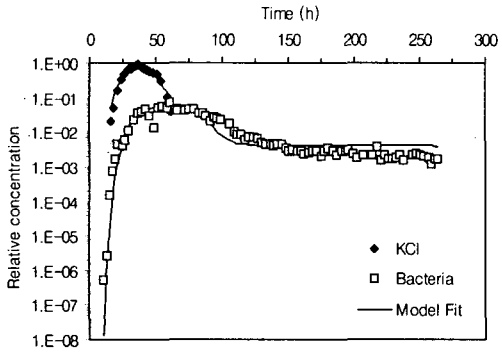


Fig. 7 Model fitting to column data (column length = 10 cm) of *Klebsiella oxytoca* from Hendry et al.(1997) using modified model 4.

of simulating data from Hendry et al. (1997). The obtained sorption parameters are summarized in Table 3. It should be noted that Hendry et al. (1997) have also applied Model 4 to analyze the data with poor fitting result. As shown in Fig. 6, Model 4 can not describe the retardation effect even though the sorption parameters (k_a, k_d and k_{ir}) are varied to fit to the data. The parameter variations only make the curves shift upward or downward. To properly describe the data, simulated curve should shift rightward. The only way to do this is to incorporate the retardation factor into the model (modified model 4):

$$\frac{\partial C}{\partial t} = \frac{D}{R}\frac{\partial^2 C}{\partial x^2} - \frac{v_w}{R}\frac{\partial C}{\partial x} - k_a C + k_d \frac{\rho_b}{\theta} S - k_{ir} C \quad (12)$$

$$\frac{\rho_b}{\theta}\frac{\partial S}{\partial t} = k_a C - k_d \frac{\rho_b}{\theta} S \quad \cdots\cdots (13)$$

Using the modified model 4, the fitting result is considerably improved with R = 1.8 (Fig. 7). The obtained sorption parameters are summarized in Table 2.

V. Conclusions

In this study, bacterial transport models were compared to determine the suitable model at describing bacterial transport in saturated column experiments. It is found that the transport model into which retardation, kinetic reversible sorption, and kinetic irreversible sorption are incorporated is appropriate. It is expected that the selected transport model could be applied to properly analyze the bacterial transport in saturated porous media. From the analysis, the following recommendations are made for the future bacterial column experiments. First, conservative tracer test must be performed during bacteria transport test in order to determine potential retardation of bacteria relative to conservative tracer, and transport parameters should be determined from conservative tracer breakthrough curve, not directly from bacterial curve. Second, mass recovery must be quantified from bacterial data in order to determine potential mass loss of bacteria due to irreversible sorption.

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