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SOLUTE TRANSPORT THROUGH SATURATED SOIL COLUMN WITH TIME-DEPENDENT DISPERSION

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ABSTRACT

Solute transport through porous media is governed by various physical, chemical and biological process that takes place between solute and porous media. Advection, mechanical dispersion, molecular diffusion, decay processes, and solute exchange with the solid phase majorly govern the solute transport through porous media. All these processes play an important role in plume spreading and dilution, therefore quantification of impact of these processes on solute transport is essential to ensure the optimal cleaning operations. Most challenging task is to accurately predict the arrival time and spatial patterns of contaminant plume in the subsurface environment. The difficulty in prediction increases with the heterogeneity and chemical properties of solute and porous media. When solute transport parameters are different at different spatial and temporal scale, then predicting the behavior of solute in porous media become difficult. One of the most challenging aspect of studying solute transport through heterogeneous porous media is ever-growing effect of scale with space/ time on the estimation of dispersion. Influence of distance/ time-dependent dispersion on solute transport is observed at various scales. Therefore, present study focuses on the development of solute transport model which incorporate time-dependent dispersion with physical partitioning of heterogeneous porous media. In this study, numerical solution of Mobile-immobile model (MIM) with time-dependent dispersion is presented to simulate experimental data of conservative solute transport through a heterogeneous saturated soil column. Crank-Nicolson finite difference numerical technique is used to solve the coupled solute transport equations. Simulation capabilities of constant and time-dependent dispersion models are compared using time scale breakthrough curves for various down-gradient distances from input source. It is observed that the perturbation of solute concentration can be better simulated using MIM with time-dependent dispersion in comparison to constant dispersion model. Influence of time-dependent dispersion parameters on breakthrough curve is discussed. Numerical results depicted the early initial breakthrough point with decreasing time-dependent dispersion coefficient. It is concluded that time-dependent dispersion function reduces the inherent uncertainty in estimation of coupling factor (mass transfer coefficient) in mobile-immobile model.

KEYWORDS: *Conservative solute transport, heterogeneous soil column, mobile-immobile model, time-dependent dispersion*

INTRODUCTION

Contaminant transport through soil has been an important research problem in the geo-environmental engineering for decades. Various mathematical models have been developed to understand the transport of chemicals through porous media while considering these processes. Classical advection dispersion equation (ADE) is the commonly used model for describing solute transport through porous media. But, when the system is heterogeneous, early arrival time and long late time tails have been observed from tracer test measurements which attributed to anomalous dispersion behavior (Levy and Berkowitz 2003; Cortis and Berkowitz 2004). It is evident that ADE is less adequate for simulating anomalous or non-equilibrium transport through heterogeneous soils (Levy and Berkowitz 2003; Gao et al. 2009). There are higher modelling approaches (mobile-immobile model, multi-processes non equilibrium model, multi rate mass transfer mode) which consider physical and chemical non-equilibrium of the porous medium (van Genuchten and Wierenga 1976; Brusseau et al. 1989; Haggerty and Gorelick 1995). These models can predict the solute transport even for heterogeneous porous media viz. stratified porous media, fracture media (Kumar et al. 2006, 2008; Swami et al. 2013, 2016; Joshi et al. 2015; Singh et al. 2018; Zhou et al. 2018; Xie et al. 2019). It is observed that the mobile-immobile model can better describe the solute transport in both the homogeneous and heterogeneous porous media as compared to advection dispersion model (Starr et al. 1985; Gao et al. 2009).

Several field and laboratory scale transport studies suggested that the dispersion is not a constant parameter but depends upon mean travel distance/distance along the movement of solute in porous media or temporal scale (Sauty 1980; Pickens and Grisak 1981; Gelhar et al. 1992; Logan 1996). It is observed from stochastic analyses that the dispersion depends directly on travel time until it reaches to an asymptotic value (Dagan 1988; Gelhar et al. 1992). A constant dispersion is unable to capture the broad distribution of time scale breakthrough curves, which is represented by the recently developed time-dependent dispersion models in pre-asymptotic regime (Basha and EL-Habel 1993; Kumar et al. 2006; Sharma and Srivastava 2012; Selim 2014; Yu et al. 2019). Barry and Sposito (1989) solved convection-dispersion equation with time dependent dispersion coefficient using Laplace transformation. Time-dependent dispersivity behavior of non-reactive solute in a system of parallel fractures has been studied by Kumar et al. (2006) using the method of spatial moments. It is observed that the distance-dependent dispersion coefficient resulted in a steeper concentration profile than the time-dependent dispersion coefficient (Zhou and Selim 2002, 2003; Sharma and Srivastava 2012). Basha and El-Habel (1993) presented analytical solutions of the advection-dispersion equation (ADE) with time dependent dispersion coefficient. Analysis carried out by Basha and El-Habel (1993) is restricted to asymptotic dispersion coefficient. It is evident from the literature that the higher mathematical models with time-dependent dispersion functions have not been explored so far to simulate solute transport through heterogeneous media. So, it gives an idea to present numerical approach which can simulate non-Gaussian solute transport behavior. Therefore, in this study an attempt is made to test the applicability of MIM model with time-dependent dispersion coefficient in describing the non-reactive solute transport through saturated porous medium. In this study, one dimensional MIM model governing transport equations with time-dependent dispersion coefficient are solved using finite difference method (FDM) using C++ programming language.

Governing solute transport equations

The mobile-immobile (MIM) model developed by (van Genuchten and Wierenga 1976, 1977) on the basis of two-region or two-site model, which divides the heterogeneous porous media into mobile and immobile regions. Mobile region governs for flow transport processes, i.e. advection, dispersion process and stagnant or immobile region accounts for first-order lumped mass transfer between mobile and immobile regions. Both regions include the first order transformation reaction on specific sites in porous media. Considering linear sorption isotherm for the sake of simplicity, following governing equations can be written as:

$$\left(\theta_m + f\rho_b K_{d_m}\right)\frac{\partial C_m}{\partial t} = \theta_m D_{(t)}\frac{\partial^2 C_m}{\partial x^2} - v_m\theta_m\frac{\partial C_m}{\partial x} - \omega(C_m - C_{im}) - \left(\theta_m\mu_{lm} + f\rho_b K_{d_m}\mu_{sm}\right)C_m \quad (1)$$

$$(\theta_{im} + (1-f)\rho_b K_{d_{im}})\frac{\partial C_{im}}{\partial t} = \omega(C_m - C_{im}) - \left(\theta_{im}\mu_{\text{lim}} + (1-f)\rho_b K_{d_{im}}\mu_{sim}\right)C_{im} \quad (2)$$

Where C_m and C_{im} are the solute concentrations in the mobile and immobile regions (M/L^3) at any time t respectively; x = spatial coordinate (L) taken in the direction of the fluid flow; $D_{(t)}$ represent time-dependent hydrodynamic dispersion coefficient along the flow velocity (L^2/T); θ_m and θ_{im} are volumetric water contents of the mobile and immobile regions respectively, and $\theta = \theta_m + \theta_{im}$; θ is the total volumetric water content of the porous media; v_m = mobile pore water velocity (L/T); $v_m\theta_m$ is equal to q (flow rate (L/T)); ω is the first order mass transfer coefficient (T^{-1}); f and $(1-f)$ represent the fractions of sorption sites that equilibrate instantly with the mobile and immobile regions, respectively; μ_{lm} and $\mu_{\text{lim}}$are the first-order decay coefficients for degradation of solutes in the mobile and immobile solution phases respectively; μ_{sm} and μ_{sim} are the first-order decay coefficients for degradation of solutes in the mobile and immobile region adsorbed solid phases respectively; K_{d_m} = distribution coefficient of linear sorption process (L^3/M) in the mobile region; $K_{d_{im}}$ = distribution coefficient of linear sorption process (L^3/M) in the immobile region; ρ_b = bulk density of the porous medium (M/L^3).

In this study, three cases of dispersion functions are considered to simulate experimental breakthrough curves (BTCs) of chloride transport through soil column. We define the following abbreviations to represent our results: MIMC is the mobile-immobile model with constant dispersion; MIML is the MIM with linear time-dependent dispersion; MIMA is the MIM with asymptotic time-dependent dispersion function.

Constant dispersion function:

$$D_{(t)} = D_0 + D_m \quad (3a)$$

Linear time-dependent dispersion function:

$$D_{(t)} = D_0 \frac{t}{K_L} + D_m \quad (3b)$$

Asymptotic time-dependent dispersion function:

$$D_{(t)} = D_0 \frac{t}{t+K_A} + D_m \quad (3c)$$

Where D_0 = uniform hydrodynamic dispersion coefficient (L^2/T); D_m= effective diffusion coefficient (L^2/T); K_A (T) is the asymptotic time-dependent dispersion coefficient which is equivalent to mean travel time; K_L (T) is the linear time-dependent dispersion coefficient.

Initial and Boundary Conditions

The initial condition assumes that the porous medium is not contaminated and is given as follows:

$$C_m(x,0) = C_{im}(x,0) = 0 \quad (4)$$

Following inlet and outlet boundary conditions have been used:

Dirichlet type boundary condition at inlet – Continuous source concentration

$$C_m(0,t) = C_0 \quad (5)$$

Neumann type boundary condition at the outlet

$$\left(\frac{\partial C_m(x,t)}{\partial x}\right)_{(x=L,t)} = 0 \quad (6)$$

Where C_0 = injected concentration (M/L^3) of solute source at the inlet of the porous medium.

Numerical model and Validation

In the present model, solute transport through saturated porous media is described by coupled partial differential equation in 1-D domain. Crank-Nicolson finite-difference numerical technique has been used to obtain the solution of the mobile-immobile (MIM) transport equation with arbitrary time-dependent dispersion. First-order upwind scheme has been used to discretize advection term in the mobile transport equation because it avoids the artificial oscillations associated with the central weighting scheme (Zheng and Bennett 2002). Dispersive term in the mobile transport equation has been discretized using second-order central difference scheme. The temporal term of both the equations (1) and (2) have been discretized using a first order forward difference scheme. Discretized form of solute transport equations in the mobile-immobile domain is presented below:

$$R_m\left(\frac{C_{m_i}^{t+1}-C_{m_i}^{t}}{\Delta t}\right)=\frac{\theta_m D_{(t+1)}}{2}\left(\frac{C_{m_{i+1}}^{t+1}-2C_{m_i}^{t+1}+C_{m_{i-1}}^{t+1}}{\Delta x^2}\right)+\frac{\theta_m D_{(t)}}{2}\left(\frac{C_{m_{i+1}}^{t}-2C_{m_i}^{t}+C_{m_{i-1}}^{t}}{\Delta x^2}\right)-\frac{\theta_m v_m}{2}\left(\frac{C_{m_i}^{t+1}-C_{m_{i-1}}^{t+1}}{\Delta x}\right)-\frac{\theta_m v_m}{2}\left(\frac{C_{m_i}^{t}-C_{m_{i-1}}^{t}}{\Delta x}\right)-\frac{\omega}{2}\left(C_{m_i}^{t+1}-C_{im_i}^{t+1}\right)-\frac{\omega}{2}\left(C_{m_i}^{t}-C_{im_i}^{t}\right)-\frac{A_1}{2}C_{m_i}^{t+1}-\frac{A_1}{2}C_{m_i}^{t} \quad (7)$$

$$R_{im}\left(\frac{C_{im_i}^{t+1}-C_{im_i}^{t}}{\Delta t}\right)=\frac{\omega}{2}\left(C_{m_i}^{t+1}-C_{im_i}^{t+1}\right)+\frac{\omega}{2}\left(C_{m_i}^{t}-C_{im_i}^{t}\right)-\frac{A_2}{2}C_{im_i}^{t+1}-\frac{A_2}{2}C_{im_i}^{t} \quad (8)$$

Where

$$R_m=(\theta_m+f\rho_b K_{d_m}) \quad (9a)$$

$$R_{im}=(\theta_{im}+(1-f)\rho_b K_{d_{im}}) \quad (9b)$$

$$A_1=(\theta_m\mu_{lm}+f\rho_b K_{d_m}\mu_{sm}) \quad (9c)$$

$$A_2=(\theta_{im}\mu_{\mathrm{lim}+(1-f)\rho_b K_{d_{im}}\mu_{sim}}) \quad (9d)$$

Where, $\Delta t=$ time step; $\Delta x=$ grid size; t denotes the known time and $(t+1)$ denotes the unknown time level.

$$A_7C_{m_i}^{t+1}-\left(A_3D_{(t)}\right)C_{m_{i+1}}^{t+1}-\left(A_3D_{(t)}+A_4\right)C_{m_{i-1}}^{t+1}-A_5C_{im_i}^{t+1}=A_{10}C_{m_i}^{t}+\left(A_3D_{(t)}\right)C_{m_{i+1}}^{t}+\left(A_3D_{(t)}+A_4\right)C_{m_{i-1}}^{t}+A_5C_{im_i}^{t} \quad (10)$$

$$A_8C_{im_i}^{t+1}-A_9C_{m_i}^{t+1}=A_{11}C_{im_i}^{t}+A_9C_{m_i}^{t} \quad (11)$$

Where coefficients are defined below:

$$A_3=\frac{\theta_m}{\Delta x^2}\left(\frac{\Delta t}{2R_m}\right),\ A_4=\frac{v_m\theta_m}{\Delta x}\left(\frac{\Delta t}{2R_m}\right)$$

$$A_5=\frac{\omega\Delta t}{2R_m},\ A_6=\frac{A_1\Delta t}{2R_m}$$

$$A_7=1+\left(2A_3D_{(t)}\right)+A_4+A_5+A_6$$

$$A_8=1+\left(\frac{\omega\Delta t}{2R_{im}}\right)+\left(\frac{A_2\Delta t}{2R_{im}}\right)A_9=\left(\frac{\omega\Delta t}{2R_{im}}\right),$$

$$A_{10}=1-\left(2A_3D_{(t)}\right)-A_4-A_5-A_6,\ A_{11}=1-\left(\frac{\omega\Delta t}{2R_{im}}\right)-\left(\frac{A_2\Delta t}{2R_{im}}\right)$$

$D_{(t)}$ is the time-dependent dispersion coefficient which is updated after each time step in the numerical simulation. The discretized partial differential equations for solute transport through mobile-immobile domain have been solved using Tridiagonal Thomas algorithm. To get the accurate solution from Crank-Nicolson finite difference method, the values of grid Peclet number, $Pe=\left(\frac{v\Delta x}{D_{(t)}}\right)$ are kept between 1-2 and the Courant number, $Cau=\left(\frac{v\Delta t}{\Delta x}\right)$ is kept less than 1. Presented numerical solution is validated with the solutions of Basha and El-Habel (1993) by scaling down mobile-immobile model in to advection-dispersion equation. It is assumed that there is no solute concentration in the porous media initially and continuous solute source is present at the input end. For validation, input model parameters used are as follows: L = 100 m, saturated porosity (θ) = 0.35, pore-water velocity (v_m) = 0.25 m/day, total time = 200 days, retardation factor (R) = 1, Saturated porous media bulk density (ρ_b) = 2.11 gm/cm^3, maximum dispersion coefficient (D_0) of asymptotic time-dependent dispersion function = 1 m^2/day, asymptotic time-dependent dispersion coefficient (K_A) = 50, effective molecular diffusion coefficient (D_m) = 0 m^2/day, and first order decay constant = 0 day^{-1}.

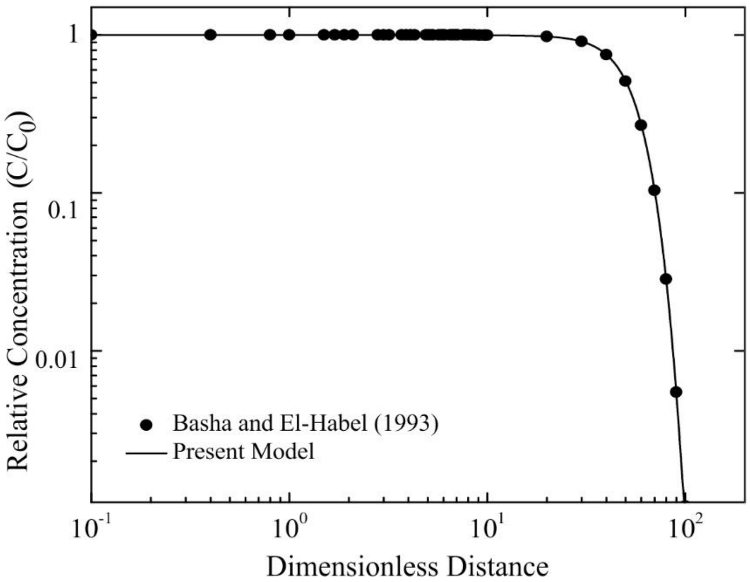

Fig. 1. *Concentration profile for non-reactive solute at time T = 200 for $K_A = 50$*

Figure 1 represents the variation of relative concentration for continuous injection at T = 200 days. Relative concentration decreases gradually with distance which is due to the dispersion processes. The numerical model shows good agreement with results obtained by Basha and El-Habel (1993). Secondly, present numerical solution is validated with the analytical solutions of Barry and Sposito (1989) for advection-dispersion equation with constant dispersion function. It is assumed that continuous solute source is present at the input end. For validation, input model parameters used are as follows: L = 5 cm, pore-water velocity (v_m) = 1 cm/day, total time = 2.5 days, retardation factor (R) = 1, dispersion coefficient (D_0) = 0.05 cm^2/day, effective molecular diffusion coefficient (D_m) = 0 m^2/day, first order decay constant = 0 day^{-1}.

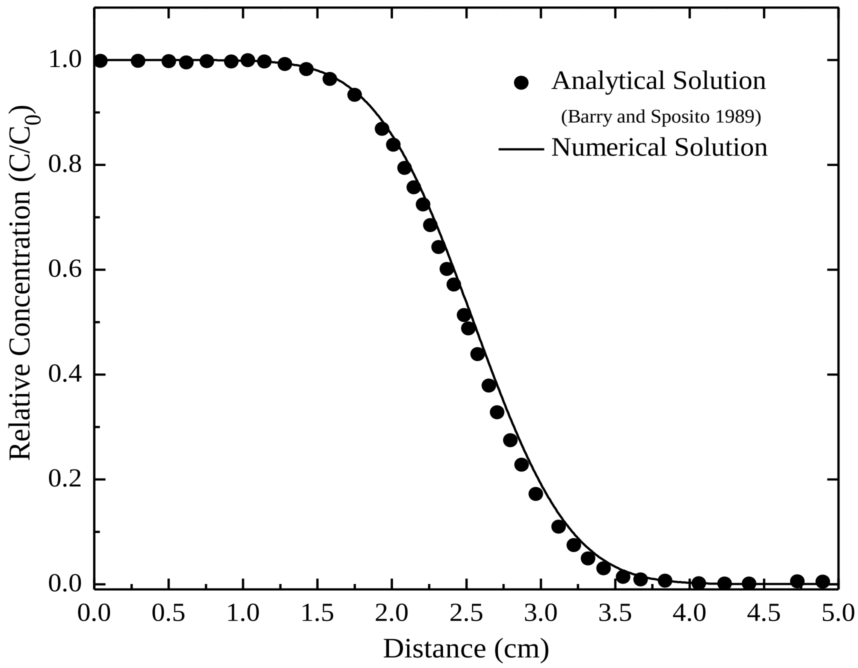

Fig. 2. *Spatial concentration profile for non-reactive solute at time T = 2.5 days*

Figure 2 represents the spatial variation of relative concentration for continuous injection at time T = 2.5 days. The numerical model shows good agreement with the analytical solutions obtained by Barry and Sposito (1989).

RESULTS AND DISCUSSIONS

Concentration profiles with constant and time-dependent dispersion models

In this study, constant and time-dependent dispersion models are used to simulate observed experimental BTCs of non-reactive solute (chloride) transport for analyzing the simulation capabilities of different models. Observed experimental data of chloride transport through heterogeneous soil column is taken from Sharma and Abgaze, (2015). Description of horizontally placed 1500 cm long soil column is presented in the study by Sharma and Abgaze (2015). There are two parameters (ω, D_0) in mobile-immobile model with constant dispersion (MIMC) and three parameters $(\omega, D_0, K_A\ or\ K_L)$ of the linear time-dependent dispersion model (MIML) and asymptotic time-dependent dispersion model (MIMA) which need to be estimated at various down-gradient distances. MIMC, MIML, and MIMA have similar mobile-water fraction and pore-water velocity in mobile region. Constant concentration type boundary condition is considered at inlet. Domain length = 1500 cm; total simulation time (t) = 3000 minute, q = 0.326 cm/min, θ_m = 0.34, θ_{im}= 0.04, v_m = 0.9588 cm/min have been used for simulation. Firstly, the experimental data of chloride at 1500 cm down-gradient distance is fitted using MIMC model. Estimated values of parameters for MIMC are ω = 7.05E-05 min^{-1}, D_m = 0 cm^2/min, and D_0 = 67.98 cm^2/min. Figure 3 shows the simulated BTC at 1500 cm down-gradient distance using MIMC model. It is observed that the BTC simulated using MIMC model over predicts observed data at large transport time. Now, observed breakthrough curve at 1500 cm down-gradient distance is simulated using MIML and MIMA models while keeping measured parameters (q = 0.326 cm/min, θ_m= 0.34, θ_{im}= 0.04, v_m = 0.9588 cm/min) fixed. Value of $K_L = 500$, ω = 5.03E-05 min^{-1}, and D_0 = 40.67 cm^2/min are obtained for MIML model while estimated values of $K_A = 180$, ω = 5.03E-05 min^{-1}, and D_0 = 80.67 cm^2/min are obtained via best fit for MIMA model. Unknown transport parameters (ω, D_0, K_L or K_A) are estimated using inverse optimization method in which Levenberg-Marquardt algorithm is coupled with solute transport model. Inverse optimization procedure is depicted in several studies (Cobb et al. 1982; Joshi et al. 2013; Kool et al. 1987; Ojha et al. 2011; Ratha et al. 2009). Similarly, BTCs at 1200 cm, 600 cm, and 300 cm down-gradient distances are simulated separately for all dispersion models to observe the influence of the travel distance on time-dependent dispersion parameters (D_0 and K_L or K_A). Estimated model parameters at all down-gradient distances for MIMC, MIML, and MIMA models are listed in Table 1. It is observed from figures 5 and 6 that the MIMC tends to overestimate the solute concentration at small transport times. Table 2 shows the goodness of fitting criterion values for all down-gradient distances.

Table 1: Estimated values of Parameters at different down-gradient distances

Distance (cm)	**MIMC**	**MIML**		**MIMA**	
	D_0 (cm^2/min)	D_0 (cm^2/min)	K_L (min)	D_0 (cm^2/min)	K_A (min)
1500	67.98	40.67	500	80.67	180
1200	58.81	38.67	510	70.67	200
600	52.63	35.67	520	65.67	250
300	45.78	32.67	550	63.67	255

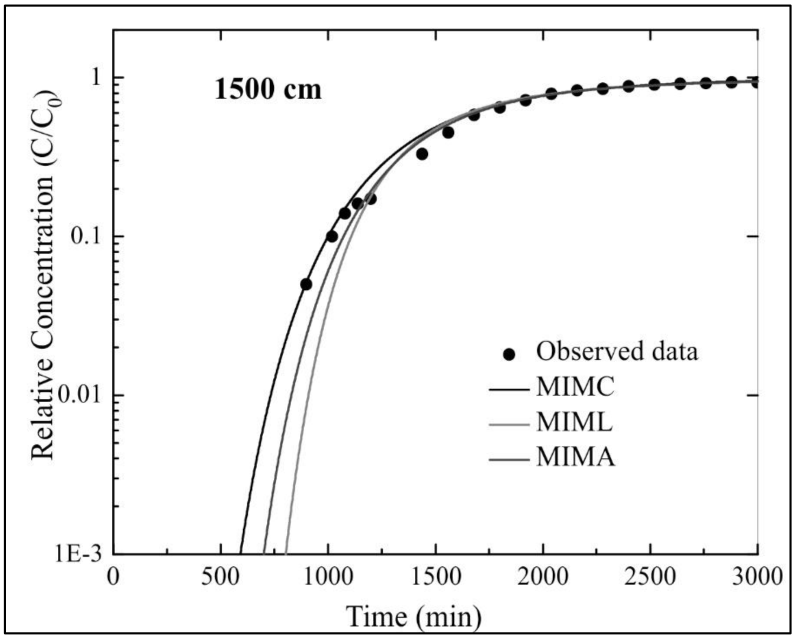

Fig. 3: *Simulation of observed data of chloride transport through soil column at 1500 cm down-gradient distance using constant and time-dependent dispersion models*

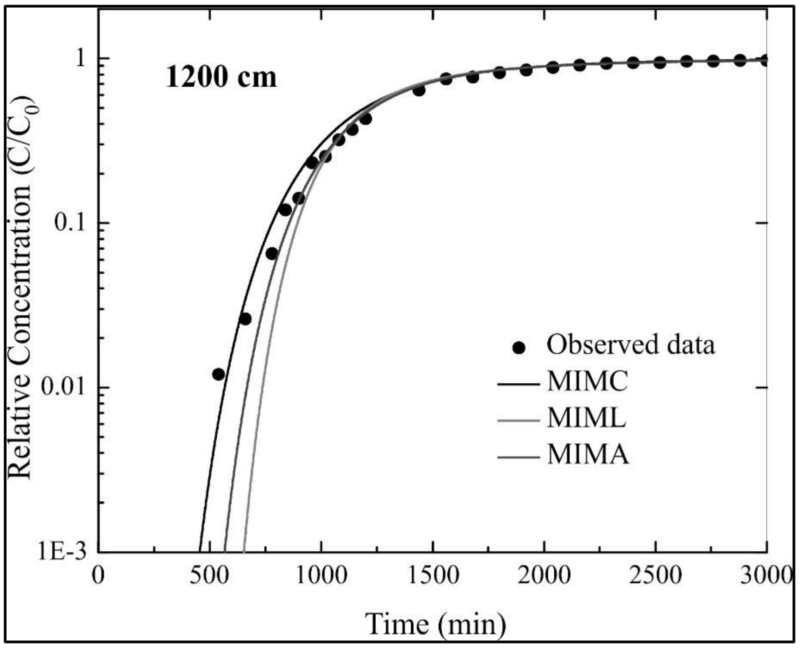

Fig. 4: *Simulation of observed data of chloride transport through soil column at 1200 cm down-gradient distance using constant and time-dependent dispersion models*

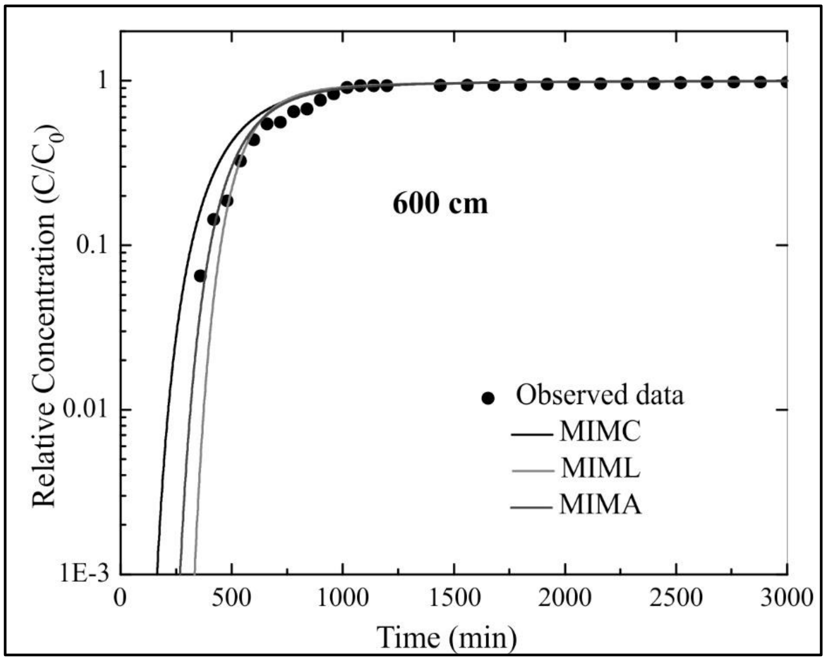

Fig. 5: *Simulation of observed data of chloride transport through soil column at 600 cm down-gradient distance using constant and time-dependent dispersion models*

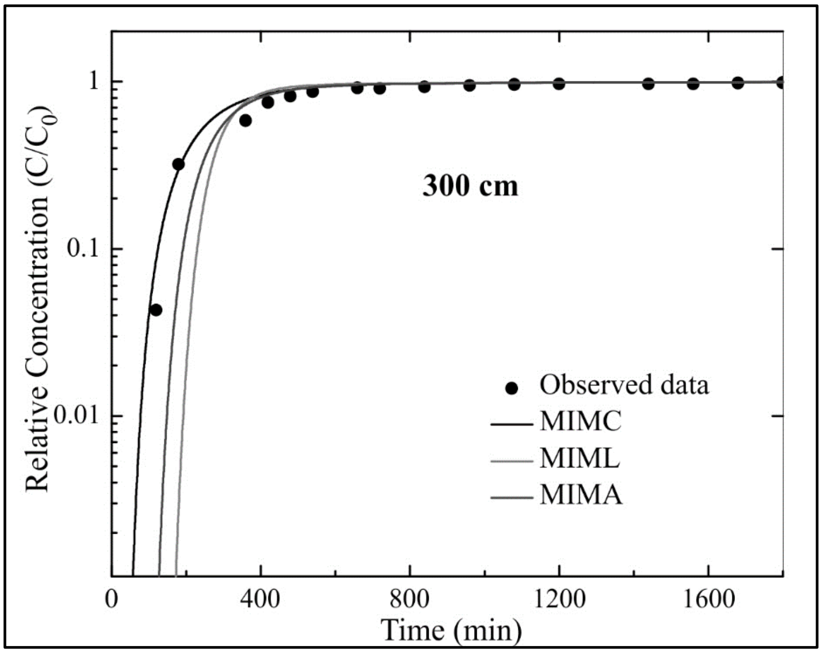

Fig. 6: *Simulation of observed data of chloride transport through soil column at 300 cm down-gradient distance using constant and time-dependent dispersion models*

Table 2: Goodness of fit obtained from simulation of chloride data

Distance (cm)	MIMC			MIML			MIMA		
	r^2	RMSE	NSE	r^2	RMSE	NSE	r^2	RMSE	NSE
1500	0.99	0.03	0.99	0.99	0.03	0.99	0.99	0.02	0.99
1200	0.99	0.03	0.99	0.99	0.03	0.99	0.99	0.02	0.99
600	0.97	0.08	0.95	0.98	0.06	0.97	0.98	0.05	0.98
300	0.98	0.05	0.97	0.95	0.08	0.93	0.97	0.06	0.96

It is observed from Table 1 that the value of dispersion coefficient (D_0) increases with an increase in travel distance while value of K_L or K_A decreases with travel distance. Estimated value of first order mass transfer coefficient (ω) for mobile-immobile model with constant dispersion (MIMC) is 7.05E-05 min^{-1} whereas mass transfer coefficient $(\omega) =$ 5.03E-05 min^{-1} is obtained for MIM with time-dependent dispersion model. It indicates that the time-dependent dispersion model tends to give smaller value of mass-transfer coefficient in comparison to MIM with constant dispersion model because of increase in the number of fitting parameters. It is seen that the values of r^2, NSE are higher for MIMA in comparison to MIMC and MIML models as shown in Table 2. Simulated BTCs and fitting criterion values show that the asymptotic time-dependent dispersion model (MIMA) gives best fit of observed experimental data of chloride transport through heterogeneous long soil column in comparison to MIML and MIMC. It is suggested that the MIM model with time-dependent dispersion function should be used for simulating non-reactive solute transport through saturated porous media.

Effect of time-dependent coefficient on breakthrough curves

The influence of linear (K_L) and asymptotic time-dependent dispersion coefficient (K_A) on BTC of conservative solute transport in heterogeneous soil column is shown in Figure 7 and 8 respectively. In Figure 7, linear time-dependent dispersion function has been considered. Pore-water velocity, $v_m =$ 0.9588 cm/min, $\omega =$ 5.03e-05 min^{-1}, and $D_0 =$ 40.67 cm^2/min, total time= 3000 min, and length = 1500 cm are considered to analyze the influence of K_L on breakthrough curve. Effective dispersion coefficient is higher for K_L=10 than K_L =500, so earlier breakthrough achieved for K_L =10 than K_L =500. It has been observed that the breakthrough point delayed with an increase in K_L value.

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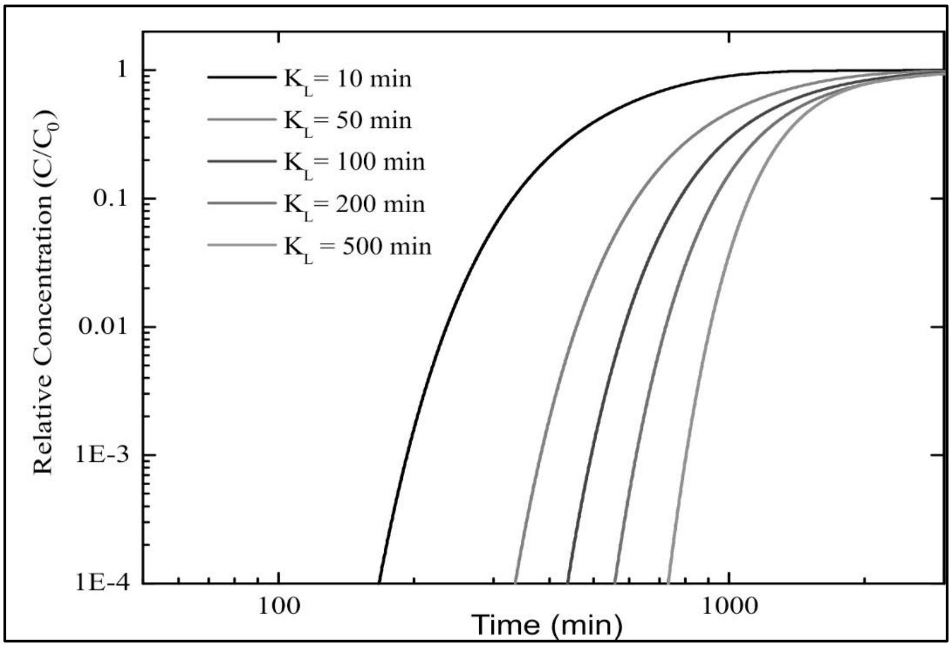

Fig. 7: *Breakthrough curve predicted at 1500 cm down-gradient distance with different values of linear time-dependent dispersion coefficient (K_L)*

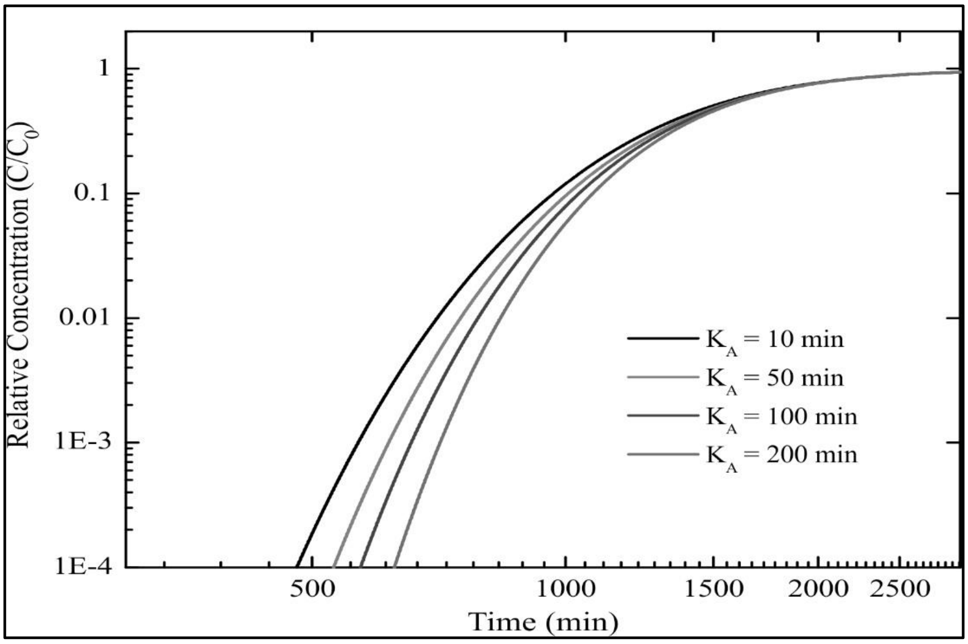

Fig. 8: *Breakthrough curve predicted at 1500 cm down-gradient distance with different values of asymptotic time-dependent dispersion coefficient (K_A)*

Figure 8 shows the effect of asymptotic time-dependent dispersion coefficient (K_A) on BTC. Pore-water velocity, $v_m = 0.9588$ cm/min, $\omega = 5.03\text{e-}05$ min^{-1}, and $D_0 = 80.67$ cm^2/min, total pulse time = 3000 min, and length = 1500 cm are considered for sensitivity analysis. It is observed that the breakthrough point delayed with an increase in K_A value. It is found that the change in the value of K_L for linear time-dependent dispersion model (MIML) significantly affects the BTC in comparison to asymptotic dispersion model (MIMA).

SUMMARY AND CONCLUSIONS

In this study, MIM model with constant and time-dependent dispersion function have been used to simulate experimental BTC for constant concentration type boundary condition. It is observed that the time-dependent dispersion coefficient with mobile-immobile (MIM) model can simulate experimental BTCs of non-reactive solute through saturated heterogeneous soil column very well. MIMA gives the best fit breakthrough curves for conservative solute transport through heterogeneous long soil column in comparison to linear time-dependent and constant dispersion function. In addition, influence of time-dependent dispersion coefficient ($K_L\ or\ K_A$) on breakthrough curve is studied. Large variation in BTC is observed for linear time-dependent dispersion model with K_L value in comparison to asymptotic dispersion model. It is concluded that the MIM model with time-dependent dispersion coefficient should be used for simulating non-reactive solute transport through saturated porous media. In future, semi-analytical solution of the MIM model with time dependent dispersion can be developed, which will be helpful to compare numerical modelling results with analytical results.

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