Simulation of Solute Transport in Aggregated Media

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Organic solute transport in aggregated soil has been modeled by a number of investigators (Passioura, 1971; Passioura and Rose, 1971; Rao et al., 1976; Rao et al., 1979; Rao et al., 1980a; Rao et al., 1980b; Crittenden et al., 1986). Studies using packed columns are useful in investigating solute transport in soil. Rosen (1952), Babcock et al. (1966), Pellett (1966), and Rasmuson and Neretnieks (1980) obtained analytic solutions for models describing diffusion and/or dispersion in packed columns with porous solids. Raghavan and Ruthven (1983) numerically solved a similar set of differential equations, but with different boundary conditions than those considered in this work.

The effect of aggregate size distribution in a column has received very little attention. Moharir et al. (1980) reported that, because of the mathematical complexities involved, the size distribution has never been considered in the theoretical prediction of the breakthrough curves from packed beds. Rasmuson (1985) extended his earlier model (Rasmuson and Neretnieks, 1980) to include the effect of size distribution, and obtained an analytical solution for a system of infinite column length and constant entrance solute concentration. He concluded that size distribution was important for short distances and when film diffusion was assumed.

The model presented in this study describes solute transport in soil columns by considering aggregate size distribution. An iterative numerical scheme is introduced to implement the model.

Theory

Consider an isothermal soil column packed with porous spherical soil aggregates of various sizes, as illustrated in Figure 1. The solute migration in the column is modeled by considering transport between the interaggregate macropore fluid and the intraaggregate micropore fluid. The micropores within aggregates are interconnected and filled with fluid. The solute transport within each aggregate is controlled by radial diffusion in the micropore fluid. The axial mass transport is dominated by the transport (advection and dispersion) in macropores between spherical aggregates. The solute exchange between macropores and micropores occurs through a thin liquid on the exterior aggregate surface.

The governing equations describing transport of nondegrading solute in the column are given below.

The flow pattern in the macropores is modeled by longitudinally dispersed plug flow:

$$\frac{2De}{3R_b}\frac{\partial U}{\partial T} = \frac{1}{Pe}\frac{\partial^2 U}{\partial X^2} - \frac{\partial U}{\partial X} - \frac{De}{Ga}W \tag{1}$$

subject to initial and boundary conditions,

$$\begin{aligned} &I.C. \quad U = 0 && @\, T = 0 \\ &B.C. \quad 1 = U(0_+, T) - \frac{1}{Pe}(\partial U/\partial X)0_+ && @\, X = 0 \\ &B.C. \quad \partial U/\partial X = 0 && @\, X = 1 \end{aligned} \tag{2}$$

The governing differential equation for each single spherical aggregate is:

$$\frac{\partial Q}{\partial T} = 0.5\left[\frac{\partial^2 Q}{\partial^2 R} + \frac{2}{R}\frac{\partial Q}{\partial R}\right] \tag{3}$$

subject to the following initial and boundary conditions:

$$\begin{aligned} &I.C. \quad Q = 0 && @\, T = 0 \\ &B.C. \quad \partial Q/\partial R = 0 && @\, R = 0 \\ &B.C. \quad N_{yk}(\partial Q/\partial R) = U - Q && @\, R = R_k \end{aligned} \tag{4}$$

The sink source in Eq. 1 is defined as:

$$W = \frac{2Ga}{3}\frac{\partial M'}{\partial T} \tag{5}$$

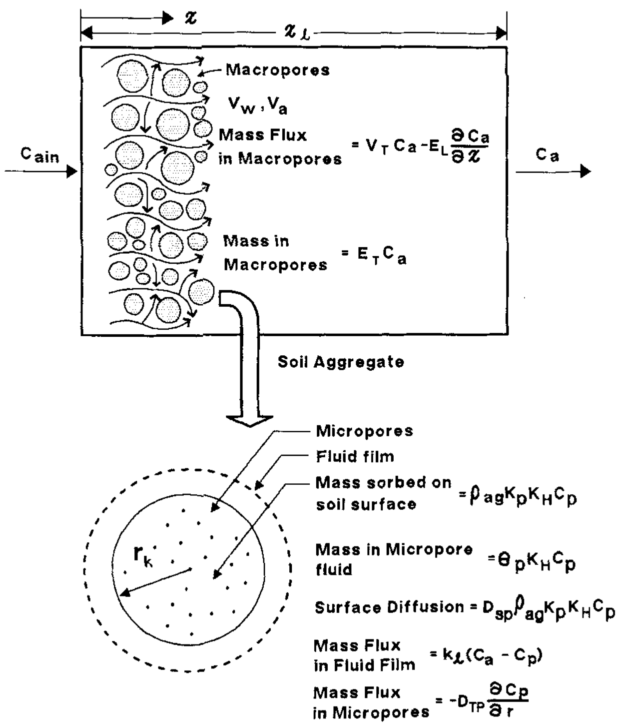

Figure 1. Flow system of a soil column.

where $M(x, t)$ is the solute mass in aggregates,

$$M' = \sum_{k=1}^{N} \left[\frac{3f_k}{R_k^3} \int_0^{R_k} QR^2 \, dR \right]$$

Table 1 lists the definitions of the parameters and dimensionless groups. Those used by Rasmuson and Neretnieks (1980) are also presented for comparison.

Standard finite difference expressions were employed for Eqs.

Table 1. Definitions of Parameters and Dimensionless Groups

Parameters	This Study	Rasmuson and Neretnieks (1980)
Peclet Number (Pe)	$\frac{x_L V_T}{E_L}$	$\frac{zV}{D_L} = Pe$
Bed Length Parameter (D_e)	$\frac{(1 - E_T) x_L G_a}{V_T}$	$\frac{z\gamma}{mV} = \delta$
Distribution Ratio (R_b)	$\frac{E_{TP}(1 - E_T)}{E_T}$	$\frac{K}{m} = R$
Dimensionless Time (T)	$S_e t$	$\sigma_t = \tau$
Film Resistance Parameter (N_{yk})	$G_a R_{fk}$	$\gamma R_f = \nu$
G_a (1/s)	$\frac{3D_{TP}}{a_r^2}$	$3D_s K/b^2 = \gamma$
R_{fk} (s)	$\frac{a_r}{3k_{Lk}}$	$\frac{b}{3k_f} = R_f$
S_e (1/s)	$\frac{2D_{TP}}{E_{TP} a_r^2}$	$\frac{2D_s}{b^2} = \sigma$

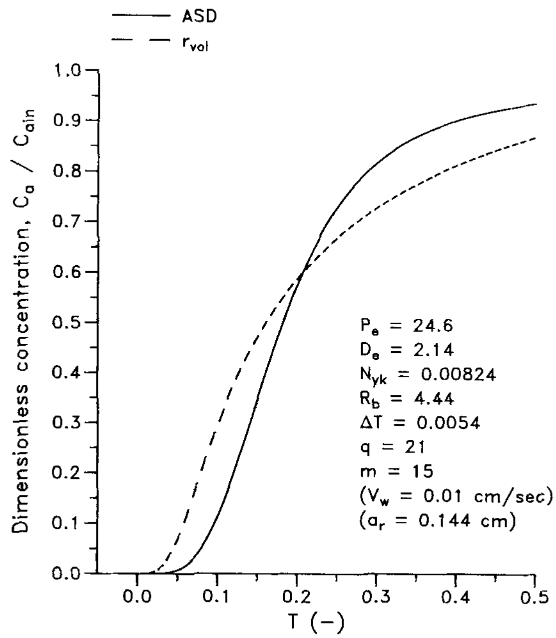

Figure 2. Effect of aggregate size distribution on breakthrough curves at flow velocity of 1*E*-4 m/s.

1 to 4. The time integration was accomplished using the backward Euler or Crank-Nicolson methods.

At each new time level, Q, W, and U were solved iteratively and alternately from the corresponding Eqs., 1, 5, and 3. The details of the computation schemes are available from the authors. The numerical solution was accurate by comparison to the exact analytical solution by Rasmuson and Neretnieks (1980).

Results and Discussions

The effect of aggregate size distribution on breakthrough curves is illustrated via a case study, as shown in Figure 2. The physical data of the solute and soil listed in Table 2 were employed. The soil size distribution and its experimentally measured dispersion coefficient, by Klotz et al. (1980), were chosen for this analysis. The mass transfer coefficient, k_{LK}, was computed by the use of the Wilson and Geankoplis (1966) correlation for low Reynolds numbers (0.0016 to 55). The choice of a correlation for the mass transfer coefficient is not critical because sensitivity analyses determined that varying the values of k_{Lk} by a factor of two, results in minimal change in breakthrough curves. Han et al. (1985) studied dispersion in columns packed with spheres having a wide size distribution, and suggested the use of a volume-averaged mean radius (r_{vol}) for data correlation. (Pertinent data are given in Table 3). The breakthrough curves resulting from this volume-averaged mean ra-

Table 2. Physical Data

X_L = 30 cm	kp = 1.1 cm^3/g
E_T = 0.375	Θ_p = 0.20
Soil Bulk Density = 1.4 g/cm^3	
D_{mol} = 0.623 × 10^{-5} cm^2/s	

Table 3. Aggregate Size Distribution

Source	Curve 4 of Figures 6 and 7 (Klotz et al., 1980)						
$r_{vol}(m) * 100$	0.144						
$r_i(m) * 100$	0.010	0.020	0.0375	0.10	0.20	0.375	0.75
f_i	0.07	0.30	0.11	0.17	0.12	0.18	0.05

dius (r_{vol}) are presented in Figure 2 for comparison. The results clearly show the significant effect of size distribution on solute movement in columns. Rasmuson (1985) reported that size distribution was of primary importance for short distances only. His conclusion, however, does not contradict our result; the differences arise from the difference in aggregate sizes. The Rasmuson study (1985) contained fine particles down to micrometer sizes, which become solute-saturated in a short time period. The effect of size distribution diminishes after a short distance. With the coarser aggregates used in this study, the interparticle processes become more important.

The work presented here clearly demonstrates the importance of including increasingly more detail in modeling solute-soil interactions and suggests that for certain spherical packing material sites, previous analyses of packed beds that ignore aggregate size distribution may be inadequate. Failure to include effects of aggregate size can lead to substantially different breakthrough curves and may incorrectly represent solute movement in contaminated soils or treatment systems.

Notation

a_r = reference length, m
a = aggregate radius, *m*
a_k = aggregate radius of fraction *k*, m
a_o = mean aggregate radius, m
Ca = solute concentration in macropore, mol/m^3
Cp = solute concentration in micropore, mol/m^3
$Cain$ = macropore solute concentration $x = 0$, mol/m^3
D_{mol} = molecular diffusion coefficient, m^2/s
D_{TP} = $Dwp\ \theta_p$ effective diffusion coefficient, m^2/s
Dw = dispersion coefficient of liquid phase in macropore, m^2/s
Dwp = diffusion coefficient of liquid phase in macropore, m^2/s
De = bed length parameter, Table 1
E_L = $E_T Dw$
E_T = water content in macropore
E_{TP} = $\theta p + \rho_{ag} Kp$
Ga = parameter, Table 1, 1/s
f_k = volumetric proportion of fraction *k*
Kp = soil-water partition coefficient, m^3 liquid/kg soil
k = index for size fractions of aggregates
k_{LK} = mass transfer coefficient for aggregate of size, a_k, m/s
M = total mass, mol/m^3
M' = total mass
m = boundary grid on aggregate surface
N = number of size fractions
N_{yk} = film resistance parameter, Table 1
Pe = Peclet number, Table 1
q = boundary grid at x_L
Q = $Cp/Cain$, concentration
r = radius coordinate, m
R = radius, r/a_r
R_{fk} = parameter, Table 1, s
R_b = $(1 - E_T)E_{TP}/E_T$, distribution ratio
R_k = a_k/a_r
r_{vol} = volume-averaged radius, m
Se = parameter, Table 1, 1/s
t = time, s
T = time, $Se \cdot t$
T_p = tortuosity of soil aggregate
U = $Ca/Cain$, concentration
V_T = $E_T V_w$, m/s
V_w = water flow velocity, m/s
W = a source/sink term, Eq. 1, mol/m$^3 \cdot$ s
x = depth coordinate, m
X = column length, x/x_L
x_L = length of soil column, m
$Z(a_k)da_k$ = aggregate fraction of radius a_k

Greek letters

δ = bed length parameter, Table 1
Δ = spacing between grid points
ϵ = tolerance
θp = water content in micropore
ν = film resistance parameter, Table 1
ρ_{ag} = soil aggregate density, kg/m$_3$
σ = parameter, Table 1, 1/s
γ = parameter, Table 1, 1/s

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