A General Approach for Modeling Solute Transport In Structured Soils

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A GENERAL APPROACH FOR MODELING SOLUTE TRANSPORT IN STRUCTURED SOILS

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Abstract

Classical convective-dispersive type transport models are often found to be of limited use for predicting solute transport in structured soils or fractured aquifer systems. Recently a number of deterministic "two-region" type models have appeared in the literature that consider transport in structured soils from a microscopic (macropore-scale) point of view. In these models, the chemical is assumed to be transported through a single pore or crack of known geometry, or through the voids between well-defined, uniformly-sized aggregates. In addition, diffusion-type equations are used to describe solute transfer from the larger pores into the soil matrix. Analytical solutions are now available for transport between spherical, cylindrical and rectangular aggregates. This paper describes a method that extends the two-region modeling approach to more general conditions involving aggregates of arbitrary geometry. The method is based on the use of a geometry-dependent shape factor (f) that transforms an aggregate of given shape and size (platy, columnar, prismatic) into an equivalent sphere with similar diffusion characteristics as the original aggregate. Values for f were derived empirically by matching average concentrations of individual soil aggregates with those of spherical aggregates. Using conversions between known analytical solutions as test cases, the transformation was found to be very accurate for most aggregate geometries commonly encountered in the field. A similar transformation was also used to quantify the unknown mass transfer coefficient in a previously employed first-order rate expression for solute exchange between "mobile" (interaggregate) and "immobile" (intra-aggregate) regions. An advantage of this last approach is that it can be included easily and effectively in one- or multi-dimensional numerical transport models.

INTRODUCTION

Large macropores in a field soil can significantly alter the rate of water and solute movement [e.g., see reviews by Thomas and Phillips (1979), Bouma (1981) and by Beven and German (1982)]. Macropores may appear in the form of drying cracks, as earthworm or gopher holes, decayed root channels, or more generally as interpedal voids in aggregated soils. It is now being recognized that classical convective-dispersive type transport equations are of limited use when predicting solute movement through soils containing such macropores. Recently, a number of "two-region" models have appeared in the literature that consider transport through structured soils from a microscopic (macropore-scale) point of view. In these models, the chemical is assumed to be transported through a single and well-defined pore or crack of known geometry, or through the voids between well-defined, uniformly-sized aggregates. In addition, diffusion-type equations are used to describe the transfer of solute from the larger pores into the soil matrix (and vice-versa). Assuming long-term steady-state liquid flow, analytical solutions for these two-region type models are now available for transport through rectangular voids (Sudicky and Frind, 1982), hollow cylindrical macropores (van Genuchten et al., 1984), and for transport through the voids between spherical (Rasmuson and Neretnieks, 1980) or solid cylindrical aggregates (Pellett, 1966). In addition, relatively simple solutions are available that neglect solute dispersion in the macropores (Skopp and Warrick, 1974), or that assume matrix diffusion into aggregates of infinite dimensions (Tang et al., 1981; Grisak and Pickens, 1981).

Except for studies by Neretnieks (1972) and Rao et al. (1982), few attempts have been made to extend the two-region modeling approach to more general conditions involving aggregates of arbitrary shape and size. For example, some structured soils contain relatively uniformly-sized columnar, prismatic or blocky aggregates, while others contain a mixture of different sizes. Still other soils contain aggregate geometries that vary with depth or even in time [for reviews on soil structure and soil aggregation, see Baver et al. (1972) and Hillel (1980)]. Thus, the variability of aggregate geometries in the field makes it difficult to formulate one theory that can be applied to all soils irrespective of their specific structure. The purpose of this paper is to present a procedure that can be used to transform soils with different aggregate shapes and sizes (platy, columnar, prismatic) into a reference soil made up of uniformly-sized aggregates of known geometry (e.g., spherical aggregates). A similar transformation is also used to quantify the unknown mass transfer coefficient in a previously employed first-order rate expression to account for the diffusional exchange between "mobile" (interaggregate) and "immobile" (intra-aggregate) regions (van Genuchten and Wierenga, 1976). An important advantage of the method is that the "mobile-immobile" region approach, previously thought to be mostly empirical, can now be formulated in terms of measurable soil-physical parameters.

SUMMARY OF ANALYTICAL SOLUTIONS

First, let us briefly summarize available analytical solutions of the two-region transport model for a number of well-defined aggregates. The general equation describing transport in the macropore system is taken as (van Genuchten et al., 1984)

$$\theta_m R_m \frac{\partial c_m}{\partial t} + \theta_{im} R_{im} \frac{\partial c_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 c_m}{\partial z^2} - \theta_m v_m \frac{\partial c_m}{\partial z} \qquad [1]$$

where θ_m and θ_{im} are the volumetric water contents of the interaggregate (macropore) and intra-aggregate (micropore) liquid phases such that $\theta_m + \theta_{im} = \theta$ is the total water content of the entire soil system, R_m and R_{im} are the retardation factors of the two regions, c_m and c_{im} are the average solution concentrations of the inter- and intra-aggregate liquid phases, respectively, D_m is the dispersion coefficient for transport through the macropore region, v_m is the average pore-water velocity of the macropore liquid phase, z is soil depth and t is time. Transverse diffusion and dispersion processes in the macropore liquid phase are assumed to be so dominant that no cross-sectional concentration gradients are present in this phase. Also, convective transport in the soil matrix is assumed to be negligible.

Transport Around Uniformly-Sized Spherical Aggregates

Equation [1] is formulated independently of the aggregate geometry. For a soil made up of uniformly-sized spherical aggregates, c_{im} represents the average concentration of a sphere:

$$c_{im}(z,t) = \frac{3}{a^3} \int_0^a r^2 c_a(z,r,t)\, dr \qquad [2]$$

where c_a is the local concentration in the spherical aggregate, r is the radial coordinate and a is the radius of the sphere. Solute transfer in the aggregate is governed by the spherical diffusion equation:

$$R_{im} \frac{\partial c_a}{\partial t} = \frac{D_a}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_a}{\partial r}\right) \qquad [3]$$

where D_a is the effective diffusion coefficient of the soil matrix. The transport equations above are augmented with auxiliary conditions requiring that concentrations are continuous at the macropore walls (Eq. 4a), and that the concentration gradient inside the aggregate at r=0 vanishes (Eq. 4b):

$$c_m(z,t) = c_a(z,a,t) \qquad \frac{\partial c_a}{\partial r}(z,0,t) = 0. \qquad [4a,b]$$

For notational convenience, the following dimensionless variables are introduced:

$$T = qt/\theta L \qquad Z = z/L \qquad [5a,b]$$

$$P = v_m L/D_m \qquad \xi = r/a \qquad [6a,b]$$

$$\gamma = \frac{D_a \theta L}{a^2 q R_{im}} \qquad \beta = \frac{\theta_m R_m}{\theta_m R_m + \theta_{im} R_{im}} = \frac{\theta_m R_m}{\theta R} \qquad [7a,b]$$

where T is the number of pore volumes leached through a soil profile (or soil column) of depth L, q is the macroscopic fluid flow density ($q = \theta_m v_m$), P is a Peclet number, β is a dimensionless partitioning coefficient and R the total retardation factor of the soil matrix/macropore system (van Genuchten et al., 1984). All concentrations (c) are assumed to be in dimensionless form: $c = (C - C_i)/(C_o - C_i)$, where C is the actual concentration, C_o is the input concentration at z=0 and C_i is the initial concentration at t=0. The transport model is solved for an initial concentration of zero (Eq. 8a), a semi-infinite profile (Eq. 8b), and a flux-type inlet boundary condition at Z=0 (Eq. 9):

$$c_m(Z,0) = c_a(Z,\xi,0) = 0 \qquad \frac{\partial c_m}{\partial Z}(\infty,T) = 0 \qquad [8a,b]$$

$$\left(c_m - \frac{1}{P}\frac{\partial c_m}{\partial Z}\right)(0,T) = 1. \qquad [9]$$

The analytical solution for the volume-averaged resident concentration of the macropore liquid phase is

$$c_m(Z,T) = \frac{1}{2} + \frac{2P}{\pi}\int_0^\infty \frac{\exp(\frac{PZ}{2} - z_p Z)}{[(\frac{P}{2} + z_p)^2 + z_m^2]}\,[(\frac{P}{2} + z_p)\sin(2\gamma\lambda^2 T - z_m Z) - z_m \cos(2\gamma\lambda^2 T - z_m Z)]\,\frac{d\lambda}{\lambda} \qquad [10]$$

where

$$z_p = [\tfrac{1}{2}(r_p + \Omega_1)]^{1/2} \qquad z_m = [\tfrac{1}{2}(r_p - \Omega_1)]^{1/2} \qquad [11a,b]$$

$$r_p = (\Omega_1^2 + \Omega_2^2)^{1/2} \qquad [12]$$

$$\Omega_1 = \frac{P^2}{4} + \gamma P(1-\beta)R\Psi_1 \qquad \Omega_2 = 2\gamma P\beta R\lambda^2 + \gamma P(1-\beta)R\Psi_2 \qquad [13a,b]$$

$$\Psi_1 = \frac{3\lambda(\sinh 2\lambda + \sin 2\lambda)}{\cosh 2\lambda - \cos 2\lambda} - 3 \qquad \Psi_2 = \frac{3\lambda(\sinh 2\lambda - \sin 2\lambda)}{\cosh 2\lambda - \cos 2\lambda}. \qquad [14a,b]$$

Equation [10] holds for in-situ concentration measurements inside semi-infinite profiles, but gives also a good approximation for macropore concentration distributions inside finite columns (Parker and van Genuchten, 1984). To predict effluent curves from finite systems (Z = 1), the following solution for the flux-averaged concentration (c_e) should be used (see also Rasmuson and Neretnieks, 1980)

$$c_e(T) = \frac{1}{2} + \frac{2}{\pi}\int_0^\infty \exp(\frac{P}{2} - z_p)\sin(2\gamma_s\lambda^2 T - z_m)\,\frac{d\lambda}{\lambda}. \qquad [15]$$

Transport Through Rectangular Voids

For a rectangular void/aggregate system with line-sheet type aggregates, the average micropore liquid concentration c_{im} of Eq. [1] is given by

$$c_{im} = \frac{1}{a} \int_0^a c_a(z,x,t)\, dx \qquad [16]$$

where a now refers to half the width of the line-sheet aggregate and x is the coordinate perpendicular to the aggregate. Solute transfer inside the aggregates is governed by the linear diffusion equation

$$R_{im} \frac{\partial c_a}{\partial t} = D_a \frac{\partial^2 c_a}{\partial x^2} \qquad (0 < x < a). \qquad [17]$$

Auxiliary conditions for the aggregate are now

$$c_m(z,t) = c_a(z,a,t) \qquad \frac{\partial c_a}{\partial x}(z,0,t) = 0. \qquad [18a,b]$$

The analytical solution for this problem is exactly the same as for the spherical transport problem given earlier, except for the following changes (see Eqs. 14a,b)

$$\Psi_1 = \frac{\lambda(\sinh 2\lambda - \sin 2\lambda)}{\cosh 2\lambda + \cos 2\lambda} \qquad \Psi_2 = \frac{\lambda(\sinh 2\lambda + \sin 2\lambda)}{\cosh 2\lambda + \cos 2\lambda}. \qquad [19a,b]$$

An alternative solution for the same problem but with provisions for linear first-order decay was derived earlier by Sudicky and Frind (1982).

Transport Through Cylindrical Macropores

The mathematical problem of convective-dispersive transport through cylindrical macropores with simultaneous matrix diffusion into a finite cylindrical soil mantle surrounding the macropore was recently solved by van Genuchten et al. (1984). The average micropore concentration in this case is

$$c_{im}(z,t) = \frac{2}{b^2 - a^2} \int_a^b r\, c_a(z,r,t)\, dr \qquad [20]$$

where a is the radius of the macropore, b represents the radius of the finite soil cylinder surrounding the macropore, and r is the radial coordinate. Solute transfer in the soil matrix is now described by the cylindrical diffusion equation:

$$R_{im} \frac{\partial c_a}{\partial t} = \frac{D_a}{r} \frac{\partial}{\partial r}\left(r \frac{\partial c_a}{\partial r}\right) \qquad (a < r < b) \qquad [21]$$

The analytical solution can again be expressed in the same format as before, provided the variables Ψ_1 and Ψ_2 of Eq. [14] are redefined as follows

$$\Psi_1 = \frac{2\lambda[N_1(M_1 - M_2) + N_2(M_1 + M_2)]}{(\xi_o^2 - 1)(N_1^2 + N_2^2)} \qquad \psi_2 = \frac{2\lambda[N_1(M_1 + M_2) - N_2(M_1 - M_2)]}{(\xi_o^2 - 1)(N_1^2 + N_2^2)} \qquad [22a,b]$$

$$M_1 = \mathrm{Ber}_1(\xi_o\lambda)\mathrm{Ker}_1(\lambda) - \mathrm{Bei}_1(\xi_o\lambda)\mathrm{Kei}_1(\lambda) - \mathrm{Ker}_1(\xi_o\lambda)\mathrm{Ber}_1(\lambda) + \mathrm{Kei}_1(\xi_o\lambda)\mathrm{Bei}_1(\lambda) \qquad [23a]$$

$$M_2 = \mathrm{Ber}_1(\xi_o\lambda)\mathrm{Kei}_1(\lambda) + \mathrm{Bei}_1(\xi_o\lambda)\mathrm{Ker}_1(\lambda) - \mathrm{Ker}_1(\xi_o\lambda)\mathrm{Bei}_1(\lambda) - \mathrm{Kei}_1(\xi_o\lambda)\mathrm{Ber}_1(\lambda) \qquad [23b]$$

$$N_1 = \mathrm{Bei}_1(\xi_o\lambda)\mathrm{Ker}(\lambda) + \mathrm{Ber}_1(\xi_o\lambda)\mathrm{Kei}(\lambda) - \mathrm{Kei}_1(\xi_o\lambda)\mathrm{Ber}(\lambda) - \mathrm{Ker}_1(\xi_o\lambda)\mathrm{Bei}(\lambda) \qquad [23c]$$

$$N_2 = \mathrm{Bei}_1(\xi_o\lambda)\mathrm{Kei}(\lambda) - \mathrm{Ber}_1(\xi_o\lambda)\mathrm{Ker}(\lambda) - \mathrm{Kei}_1(\xi_o\lambda)\mathrm{Bei}(\lambda) + \mathrm{Ker}_1(\xi_o\lambda)\mathrm{Ber}(\lambda) \qquad [23d]$$

$$\xi_o = b/a. \qquad [23e]$$

Transport Around Solid Cylindrical Aggregates

The last problem for which an analytical solution was derived deals with solute transport through a macropore system that surrounds solid cylindrical aggregates. The average micropore liquid concentration c_{im} in Eq. [1] is now given by

$$c_{im} = \frac{2}{a^2} \int_0^a r\, c_a(z,r,t)\, dr \qquad [24]$$

where a refers to the radius of the solid cylinder, and r is the radial coordinate. The local concentration c_a in the soil matrix is determined by the cylindrical diffusion equation:

$$R_{im} \frac{\partial c_a}{\partial t} = \frac{D_a}{r} \frac{\partial}{\partial r} \left(r \frac{\partial c_a}{\partial r}\right) \qquad (0 < r < a). \qquad [25]$$

An analytical solution for this problem was derived that again has the same format as the solution given earlier for the spherical transport model, provided Ψ_1 and Ψ_2 are redefined as

$$\Psi_1 = \frac{\lambda\sqrt{8}\,[\mathrm{Ber}(\lambda)\mathrm{Ber}'(\lambda) + \mathrm{Bei}(\lambda)\mathrm{Bei}'(\lambda)]}{\mathrm{Ber}^2(\lambda) + \mathrm{Bei}^2(\lambda)} \qquad \Psi_2 = \frac{\lambda\sqrt{8}\,[\mathrm{Ber}(\lambda)\mathrm{Bei}'(\lambda) - \mathrm{Bei}(\lambda)\mathrm{Ber}'(\lambda)]}{\mathrm{Ber}^2(\lambda) + \mathrm{Bei}^2(\lambda)}. \qquad [26a,b]$$

Note that this result is much simpler than an earlier solution derived by Pellett (1966).

AN APPROACH USING SHAPE FACTORS

As shown above, analytical solutions for convective-dispersive transport around aggregates having four different geometries are available. Still, geometries are limited to only four shapes. For example, no solutions are available for aggregates that have a finite prismatic or columnar structure (i.e., aggregates that are finite in the z-direction). To make the analytical solutions useful for transport through soils with aggregates of widely different geometries, an approach using shape factors is now formulated. The method is based on a comparison of the diffusion properties of single aggregates in a non-flowing system.

Analogy With The Diffusion Equation

Consider first the diffusion equation for a single spherical aggregate (Eq. 3). Imposing an initial concentration of zero (Eq. 27a), a surface concentration of one (Eq. 27b),

$$c_a(r,0) = 0 \qquad c_a(a,t) = 1, \qquad [27a,b]$$

and solving for the average concentration $\langle c_a \rangle$ as a function of dimensionless time T yields

$$\langle c_a \rangle = \begin{cases} 6\sqrt{T/\pi} - 3T + 12\sqrt{T} \sum_{n=1}^{\infty} \mathrm{ierfc}\left(\frac{n}{\sqrt{T}}\right) & (T < 0.1) \\ 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{\exp(-n^2\pi^2 T)}{n^2} & (T > 0.1) \end{cases} \qquad [28]$$

where as before the characteristic length a represents the radius of the sphere, and where

$$T = \frac{D_a t}{a^2 R_{im}}. \qquad [29]$$

Note that $\langle c_a \rangle$ has the same meaning as c_{im} in a flowing system (Eq. 2). Similar equations for the average concentration $\langle c_a \rangle$ as a function of dimensionless time T for a wide range of aggregate geometries are available (Carslaw and Jaeger, 1959). Those solutions are not repeated here. Figure 1 gives a semilogarithmic plot of $\langle c_a \rangle$ as a function of T for four aggregate geometries. As before, the parameter a represents the characteristic length of the aggregate: radius of the sphere or solid cylinder, half the width of the plane sheet or cube. Note that the curves are displaced with respect to each other, but that they all have roughly the same sigmoidal shape. A shape factor is now used to convert an aggregate of given shape and size into a differently-sized or shaped aggregate with approximately the same diffusion characteristics as the original aggregate. Using the conversion from a plane-sheet type aggregate into an equivalent sphere as an example, define the shape factor $f_{\ell,s}$ as follows

$$f_{\ell,s} = (T_\ell / T_s)^{1/2} \Big|_{\langle c_a \rangle} \qquad [30]$$

where the subscripts ℓ and s refer to the plane sheet and sphere, respectively. Hence, T_ℓ and T_s represent the dimensionless times for diffusion into a one-dimenisional slab of width $2a_\ell$ (Eq. 31a) and into a sphere of radius a_s (Eq. 31b), respectively:

$$T_\ell = \frac{D_a t}{a_\ell^2 R_{im}} \qquad T_s = \frac{D_a t}{a_s^2 R_{im}}. \qquad [31a,b]$$

A formal but impractical way of evaluating Eq. [30] would be to invert Eq. [28] to obtain T_s as a function of $\langle c_a \rangle$, doing the same for the plane sheet, and then to substitute those expressions into [30] to obtain $f_{\ell,s}$ as a function of $\langle c_a \rangle$. Instead, an easier way is to simply read from plots like those in Fig. 1 for various values of $\langle c_a \rangle$ the dimensionless times T_ℓ and T_s for the line sheet and the sphere, and substituting those values into [30]. This will show that $f_{\ell,s}$ is a slowly decreasing function of $\langle c_a \rangle$. Let us for now ignore this concentration dependency and evaluate Eq. [30] only at $\langle c_a \rangle = 0.5$. Since $T_\ell(0.5) = 0.197$ and $T_s(0.5) = 0.0305$, Eq. [30] leads to $f_{\ell,s} = 2.54$. If we also substitute Eqs. [31a,b] into [30], one obtains

$$a_s = f_{\ell,s}\, a_\ell \quad (= 2.54 a_\ell). \qquad [32]$$

This shows that a plane-sheet type aggregate of half-width a can be replaced with an equivalent sphere of radius 2.54a that has roughly the same diffusion characteristics as the plane sheet. Use of [32] has the effect of shifting the horizontal axis for the sphere in Fig. 1 such that the curves for the sphere and the plane sheet coincide at $\langle c_a \rangle = 0.5$. This is further shown in Fig. 2 for the plane sheet - sphere conversion, as well as for conversion of a rectangular prism with dimensions (2a,2a,2a/3), a cube (2a,2a,2a) and a solid cylinder (diameter 2a) into equivalent spheres. Note that the spherical approximation is excellent for the rectangular prism, while the plane-sheet approximation is considerably less accurate, especially at higher concentrations. The relatively poor approximation of the plane-sheet aggregate is due to the fact that $f_{\ell,s}$ depends quite strongly on the concentration.

This is shown in Fig. 3 which gives a plot of the relative shape factor, f/f_{50}, as a function of the average concentration, $\langle c_a \rangle$, for various aggregate geometries. The relative shape factor is defined as the shape factor at any concentration relative to its value at $\langle c_a \rangle = 0.5$. Clearly, shape factors for the solid cylinder ($f_{c,s}$) and the rectangular prism ($f_{r,s}$) are far less concentration-dependent than $f_{\ell,s}$ for the plane sheet-sphere conversion. The less concentration-dependent a given shape factor, the better the approximation using equivalent spheres will be.

The shape factors above were defined for the transformation of plane-sheet or other aggregate geometries into equivalent spheres. Table 1 lists values of various shape factors for conversion into equivalent spheres, equivalent line sheets, and for use with a first-order type exchange model to be discussed later. The shape factors in Table 1 can also be used for conversion into aggregates other than equivalent spheres or line sheets. This is accomplished with the following rules

$$f_{1,2}\, f_{2,3} = f_{1,3} \qquad\qquad f_{1,1} = 1, \qquad [33a,b]$$

where the subscripts 1, 2, 3 can refer to any of the subscripts (and associated aggregate geometries) listed in Table 1. In particular, note that the subscript assumption 3=1 in Eq. [33a] implies that $f_{1,2} = f^{-1}_{2,1}$; in other words, the shape factor that transforms a sphere into an equivalent plane sheet is the inverse of the shape factor that transforms a plane sheet into an equivalent sphere. Conversions involving the hollow cylinder also require an estimate for ξ_o; this will be shown later.

Application To Two-Region Transport Models

The shape factor approach outlined above can be applied immediately to all two-region type transport models. The transformation of a plane sheet into an equivalent sphere will again be used as an example. Inspection of the analytical solutions (Eqs. 10, 15) and their dimensionless variables shows that the diffusion properties of an aggregate (notably the characteristic length, a, and the diffusion coefficient, D_a, only appear in the dimensionless coefficient γ (Eq. 7a). Restating the dimensionless coefficients γ for the spherical aggregate and rectangular void transport models, we have

$$\gamma_\ell = \frac{D_a \theta L}{a_\ell^2 q R_{im}} \qquad\qquad \gamma_s = \frac{D_a \theta L}{a_s^2 q R_{im}}, \qquad [34a,b]$$

where as before the subscripts s and ℓ refer to the sphere and plane sheet, respectively. Taking the ratio of γ_s and γ_ℓ, solving for γ_s and using [32] yields

$$\gamma_s = \frac{a_\ell^2}{a_s^2}\gamma_\ell = \frac{\gamma_\ell}{f^2_{\ell,s}} = f^2_{s,\ell}\gamma_\ell \qquad (= \gamma_\ell/6.4) \qquad [35]$$

or with [34a]

$$\gamma_s = \frac{D_a \theta L}{f^2_{\ell,s} a_\ell^2 q R_{im}} . \qquad [36]$$

Thus, when Eq. [35] or [36] is used for γ in the spherical transport model, that model should predict in an approximate way also transport through a soil containing parallel rectangular voids. Because analytical solutions are available for both transport models, the accuracy of the approximation can be assessed immediately. Calculated breakthrough curves based on Eq. [15] for one set of parameter values and three values for γ_s are shown in Fig. 4a. Results indeed compare very well. As was noted earlier,

Table 1. Shape factors for the conversion into equivalent spheres, equivalent line sheets, and for use in an equivalent first-order rate model.

Original aggregate	Dimensions (x,y,z) or (2r,z)	Spherical Equivalent	Plane Sheet Equivalent	First-order Equivalent	Comments
Sphere	2a	$f_{s,s}$ = 1.000	$f_{s,\ell}$ = .394	$f_{s,1}$ = .210	2a=diameter
Plane Sheet	2a,∞,∞	$f_{\ell,s}$ = 2.54	$f_{\ell,\ell}$ =1.000	$f_{\ell,1}$ = .533	2a=width
Rectangular Prism	2a,2a,∞	$f_{r,s}$ = 1.49	$f_{r,\ell}$ = .585	$f_{r,1}$ = .312	Rectangle
	2a,2a,16a	1.42	.560	.298	
	2a,2a,8a	1.36	.535	.285	
	2a,2a,6a	1.32	.520	.277	
	2a,2a,4a	1.24	.490	.261	
	2a,2a,3a	1.172	.462	.246	
	2a,2a,2a	1.046	.412	.220	Cube
	2a,2a,4a/3	.892	.351	.187	
	2a,2a,a	.772	.304	.162	
	2a,2a,2a/3	.602	.237	.126	
	2a,2a,2a/4	.491	.193	.103	
	2a,2a,2a/6	.356	.140	.0748	
	2a,2a,2a/8	.279	.110	.0586	
Solid Cylinder	2a,∞	$f_{c,s}$ = 1.44	$f_{c,\ell}$ = .566	$f_{c,1}$ = .302	2a=diameter
	2a,16a	1.38	.543	.289	
	2a,8a	1.32	.521	.277	
	2a,6a	1.29	.506	.270	
	2a,4a	1.21	.479	.255	
	2a,3a	1.149	.453	.241	
	2a,2a	1.030	.406	.216	
	2a,4a/3	.882	.348	.185	
	2a,a	.766	.302	.161	
	2a,2a/3	.599	.236	.126	
	2a,2a/4	.489	.193	.103	
	2a,2a/6	.356	.140	.0747	
	2a,2a/8	.279	.110	.0585	
Hollow Cylinder	2a,2b,∞				ξ_o = b/a
	ξ_o = 2	$f_{p,s}$ = 3.13	$f_{p,\ell}$ =1.23	$f_{p,1}$ = .657	2a=pore diameter
	ξ_o = 5	3.99	1.57	.838	2b=outer diameter
	ξ_o = 10	4.65	1.83	.976	of soil matrix
	ξ_o = 20	5.29	2.08	1.110	
	ξ_o = 50	6.09	2.40	1.28	
	ξ_o = 100	6.66	2.63	1.40	
	ξ_o = 200	7.21	2.84	1.51	
First-Order Rate Model		$f_{1,s}$ = 4.76	$f_{1,\ell}$ =1.88	$f_{1,1}$ = 1.000	

the shape factor $f_{\ell,s}$ for conversion of the plane sheet into an equivalent sphere was found to be considerably more concentration-dependent than the shape factors for conversion of the solid cylinder ($f_{c,s}$) and several other aggregate forms (Fig. 3) into equivalent spheres. Hence, as compared to the rectangular void model, one would expect an even better approximation for transport around solid cylindrical structures using the spherical transport model. This is indeed demonstrated by the near perfect fit in Fig. 4b. Judging from Fig. 3, similar accuracies are expected when the spherical model is used to predict transport through soils containing all types of finite or infinite (in the z-direction) cylindrical or rectangular aggregates (i.e., aggregates of columnar or prismatic structure).

Extension To First-Order Type Rate Models.

Early attemps to deal with transport through aggregated soils frequently used first-order type rate equations to account for diffusional exchange between "mobile" and "immobile" regions (Coats and Smith, 1964; van Genuchten and Wierenga, 1976). For that purpose, Eq. (1) was augmented with the first-order rate expression

$$\theta_{im}R_{im}\frac{\partial c_{im}}{\partial t} = \alpha(c_m - c_{im}) \qquad [37]$$

where α is an empirical rate coefficient that depends in some way on aggregate size and the diffusion coefficient. For the same initial and boundary conditions as before, the analytical solution for this transport model is

$$c_m(Z,T) = \int_0^T g(Z,\tau)\ [1 - e^{-w}\int_0^u e^{-\sigma}I_o(2\sqrt{w\sigma})\ d\sigma]\ d\tau \qquad [38]$$

where

$$g(Z,\tau) = (\frac{P}{\pi\beta R\tau})^{1/2}\exp[-\frac{P(\beta RZ-\tau)^2}{4\beta R\tau}] - \frac{P}{2\beta R}\exp(PZ)\ \mathrm{erfc}[(\frac{P}{4\beta R\tau})^{1/2}[\beta RZ+\tau)] \qquad [39]$$

and

$$u = \frac{\omega T}{\beta R} \qquad\qquad w = \frac{\omega(T-\tau)}{(1-\beta)R}. \qquad [40a,b]$$

The solution above uses the same dimensionless variables as before, except for γ (Eq. 7a) which must be replaced by

$$\omega = \alpha L/q \qquad [41]$$

For the effluent curve from a finite column (or soil profile), $g(Z,\tau)$ in [38] is given by

$$g(Z,\tau) \equiv g(1,\tau) = (\frac{P\beta R}{4\pi\tau^3})^{1/2}\exp[-\frac{P(\beta R-\tau)^2}{4\beta R\tau}]. \qquad [42]$$

The same procedure using shape factors can also be applied here if we associate [37] with a given aggregate of known size and shape. Integrating [37] for an initial concentration of zero and keeping c_m at unity yields

$$\langle c_a\rangle \equiv c_{im}(T_1) = 1 - \exp(-T_1) \qquad\qquad T_1 = \frac{\alpha t}{\theta_{im}R_{im}} \qquad [43a,b]$$

where the subscript 1 is used to identify T here with the linear rate model. Figure 5 shows a semilogarithmic plot of Eq. [43a], as well as of plots of average concentrations for several aggregate geometries (see also Fig. 1). Included are two curves for diffusion from a hollow cylindrical macropore into a finite soil mantle surrounding the macropore; the ratio $\xi_o = b/a$ for those curves refers to the ratio of the radii of the outer and inner cylinders (Eq. 23e). Note that the dimensionless time for diffusion into the finite soil mantle surrounding the cylindrical macropore is given by

$$T_p = \frac{D_a t}{(b-a)^2 R_{im}}. \qquad [44]$$

Similarly as before, we derived shape factors for conversion of various aggregate geometries into "equivalent aggregates" that accumulate solute according to [43a]. For the conversion from spherical aggregates, the shape factor $f_{s,1}$ is now given by

$$f_{s,1} = \left(\frac{T_s}{T_1}\right)^{1/2}. \qquad [45]$$

Substitution of [29] for T_s and [43b] for T_1 into [45] leads to the following definition for α in the mobile-immobile model (Eqs. [38]-[42]) when that model is applied to spherical aggregates

$$\alpha = \frac{D_a \theta_{im}}{f_{s,1}^2 a_s^2}. \qquad [46]$$

Making use the Eqs. [34b] and [41] leads to the following relation between the dimenionless coefficients ω and γ_s:

$$\omega = \frac{(1-\beta)R\gamma_s}{f_{s,1}^2}. \qquad [47]$$

Similar equations apply to conversions from other aggregate geometries. In particular, the conversion from a hollow cylinder to an equivalent first-order exchange model is given by [45] with the subscript s replaced by p. Using [43b] and [44] yields then

$$\alpha = \frac{D_a \theta_{im}}{(b-a)^2 f_{p,1}^2} \qquad [48]$$

where $f_{p,1}$ depends on the value of $\xi_o = b/a$ as shown in Table 1. The dimensionless coefficients ω and γ_p are in this case related by

$$\omega = \frac{(1-\beta)R\gamma_p}{(\xi_o - 1)^2 f_{p,1}^2}. \qquad [49]$$

Figure 6 shows the concentration dependency of several relative shape factors for conversion into "equivalent first-order rate equations". Note that the shape factors for the hollow cylindrical structures are only weakly dependent upon the concentration, while those for the plane sheet and sphere are strongly concentration-dependent. To verify the accuracy of the much simpler Eq. [37] in predicting transport through structured soils, Fig. 7 compares results based on Eqs. [38] and [42] with the exact solutions for the spherical and rectangular void transport models. Similar comparisons for the cylindrical macropore model are shown in Fig. 8. Notice the excellent approximation for the cylindrical macropore problem, especially when $\xi_o = 100$. The approximations in Fig. 7 are far less accurate. Considering the uncertainty in the many transport parameters that must be quantified in these type of two-region models, it is unclear whether or not the approximations for the sphere and line-sheet in Fig. 7 are acceptable for actual calculations. It may very well be that the match shown in Fig. 7 is as good as reasonably can be expected when predicting transport through structured field soils. If so, then the advantages of using the much simpler Eq. [37] instead of the complete two-region formulation should be clear. For example, Eq. [37] can be included easily in one- or multi-dimensional transport models without greatly affecting the numerical complexity of the code. When doing so, the coefficients (including the shape factors) can be made depth-dependent, an important consideration when simulating transport through layered profiles. In any case, it is evident that the otherwise empirical parameter α can now be expressed in terms of measurable soil-physical parameters.

SUMMARY AND CONCLUSIONS

This paper describes a method to transform (scale) soils containing aggregates of widely different shapes and sizes (platy, columnar, prismatic) into a reference soil made up of uniformly-sized spherical aggregates. The transformation is accomplished through the introduction of a geometry-dependent shape factor that converts an aggregate of given shape and size into an equivalent sphere with similar diffusion characteristics as the original aggregate. Using known solutions for transport around spherical, cylindrical and rectangular aggregates as test cases, the method was found to be very accurate for most aggregate forms commonly encountered in the field. A similar transformation was also used to quantify the unknown mass transfer coefficient in a previously employed first-order rate expression for solute exchange between "mobile" (interaggregate) and "immobile" (intra-aggregate) regions. This method proved to be extremely accurate for predicting transport through hollow cylindrical macropores. The transformation into an "equivalent first-order rate model" was found to be far less accurate for spherical and line-sheet type aggregates. Considering the uncertainty in the many transport parameters needed in two-region type models, the accuracy of the first-order rate approximation is probably good enough for most field simulations. The transformations proposed in this paper should make the two-region modeling approach applicable to more general conditions involving soils with widely different aggregate geometries.

The transformations at present are still limited to soils containing uniformly-sized aggregates: no provisions are given to deal with aggregate mixtures. Utility of the transformation could be greatly enhanced when the method can be extended to experimentally derived aggregate size distributions involving a mixture of aggregates of different shapes and sizes [see Rao et al. (1982) for a potential lead]. Soil physicists and soil morphologists over the years have collected a vast amount of information on soil structure and soil aggregation (Brewer, 1964; Baver et al., 1972; Hillel, 1980; Bouma, 1981). Perhaps some results of that research can be applied successfully to this or similar theoretical work.

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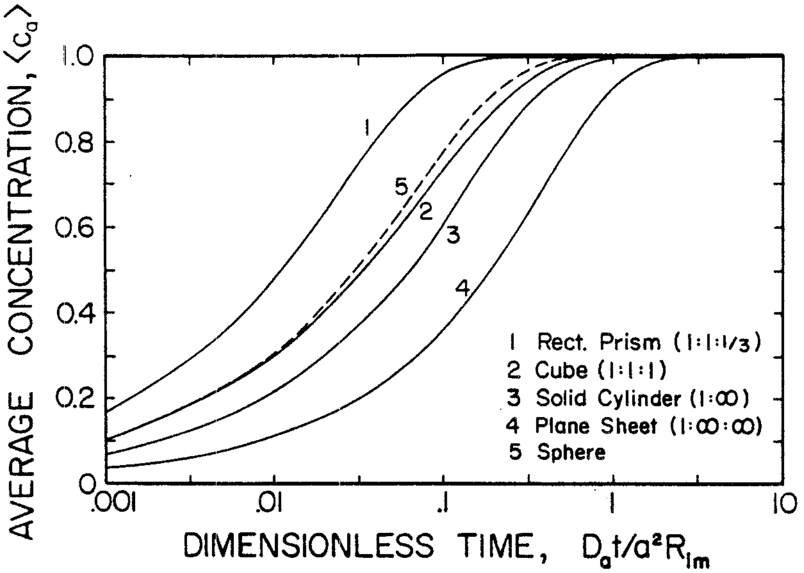

Figure 1. Average concentrations for diffusion in various soil aggregates.

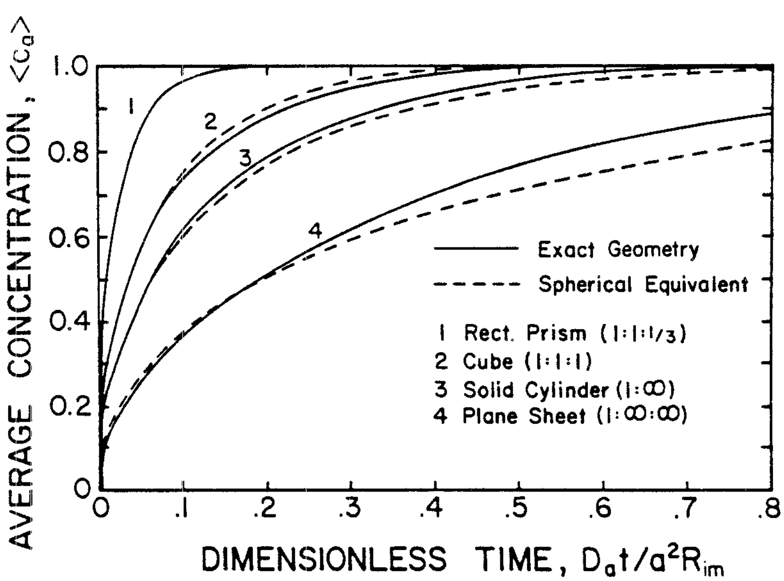

Figure 2. Exact and approximate average concentrations as calculated with equivalent spherical aggregates.

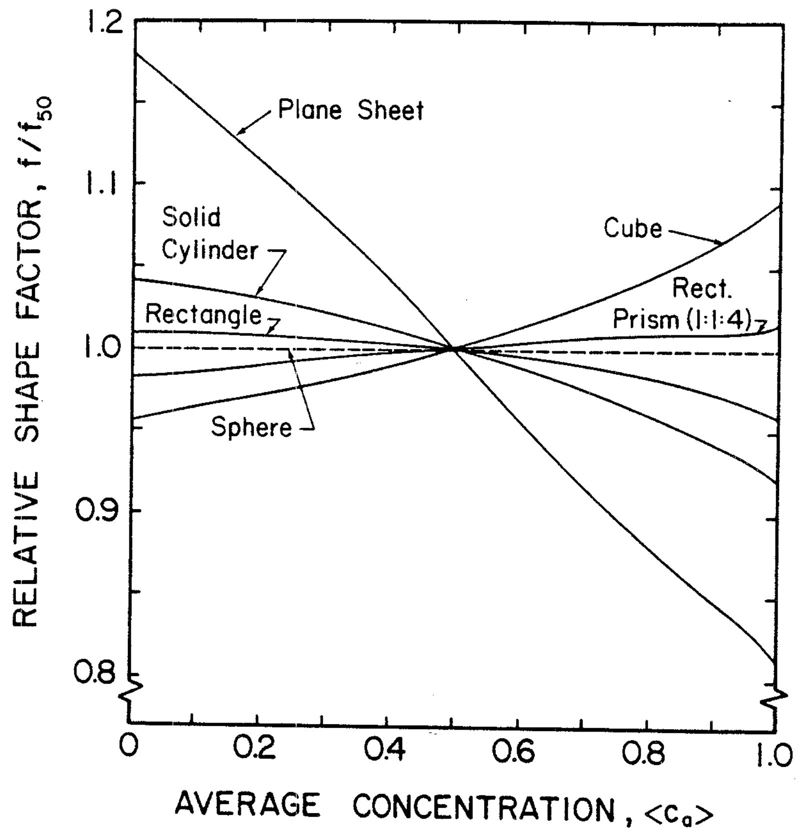

Figure 3. Effect of the average concentration on the relative shape factor for conversion of several aggregate geometries into equivalent spheres.

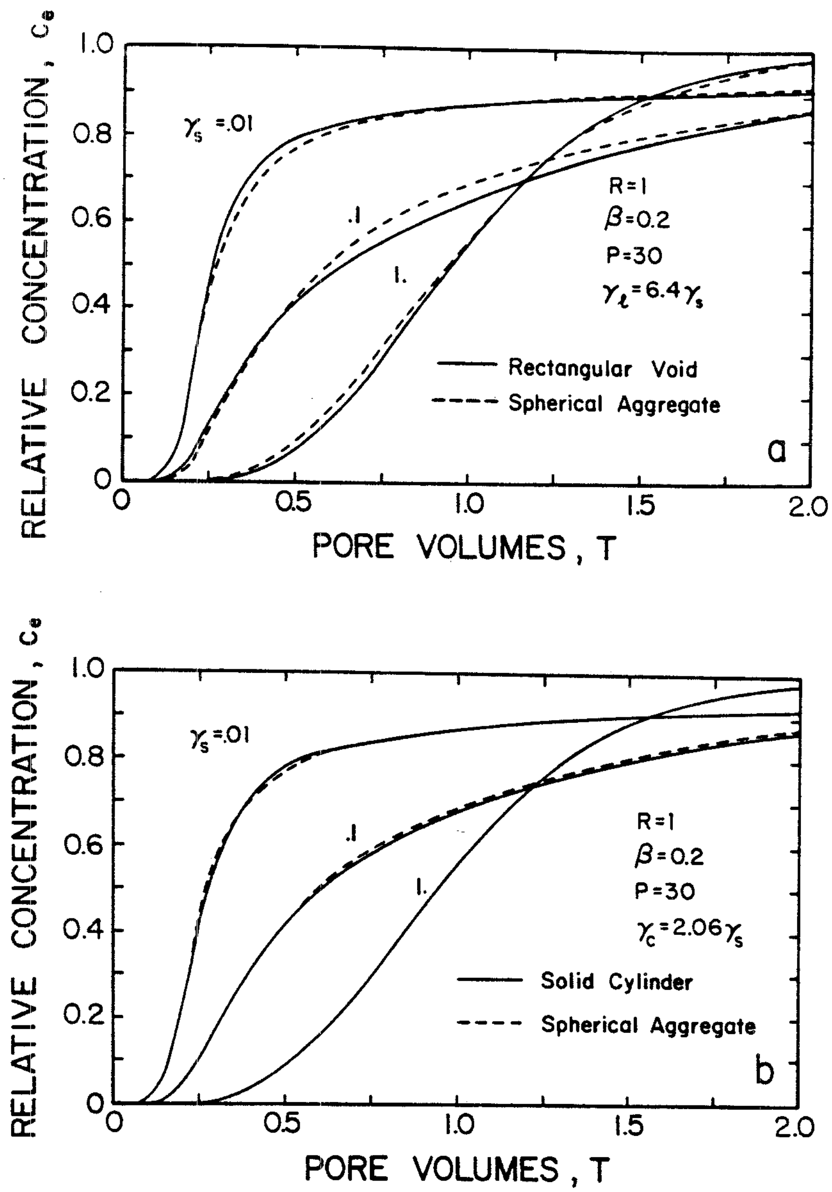

Figure 4. Breakthrough curves obtained with the equivalent spherical transport model and the exact curves for (a) the rectangular void model and (b) the solid cylindrical transport model.

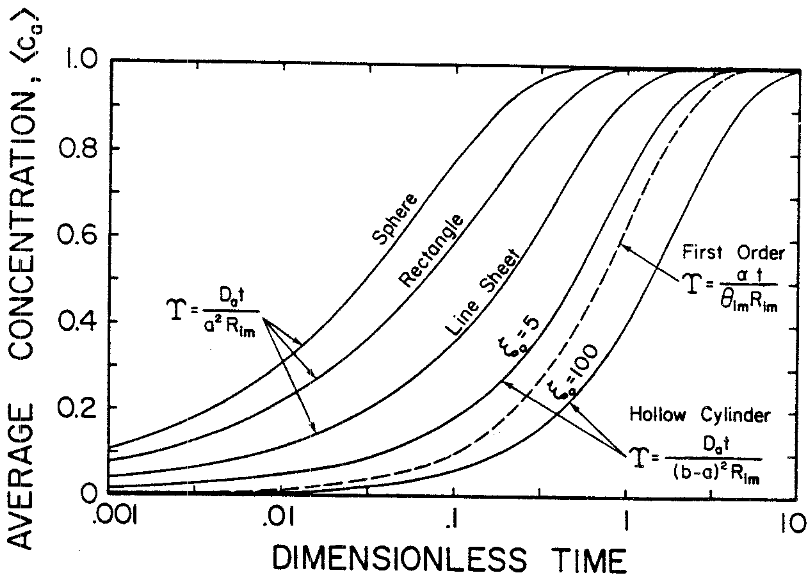

Figure 5. Average concentrations for diffusion into various soil aggregates. The dashed line is based on Eq. [43].

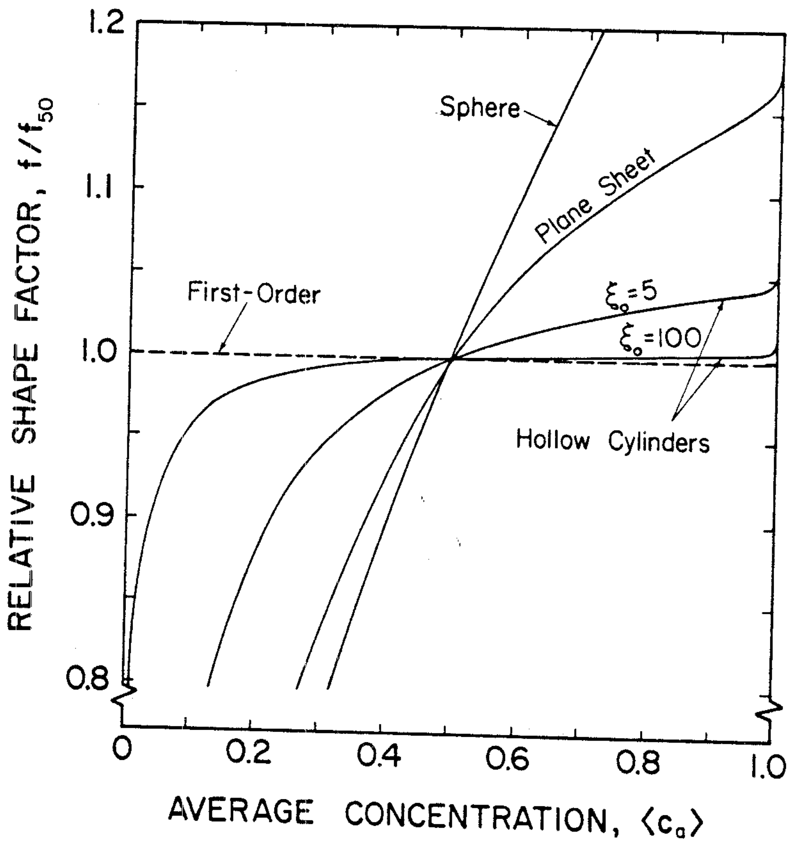

Figure 6. Effect of the average concentration on the relative shape factor for use in an "equivalent" first-order rate model.

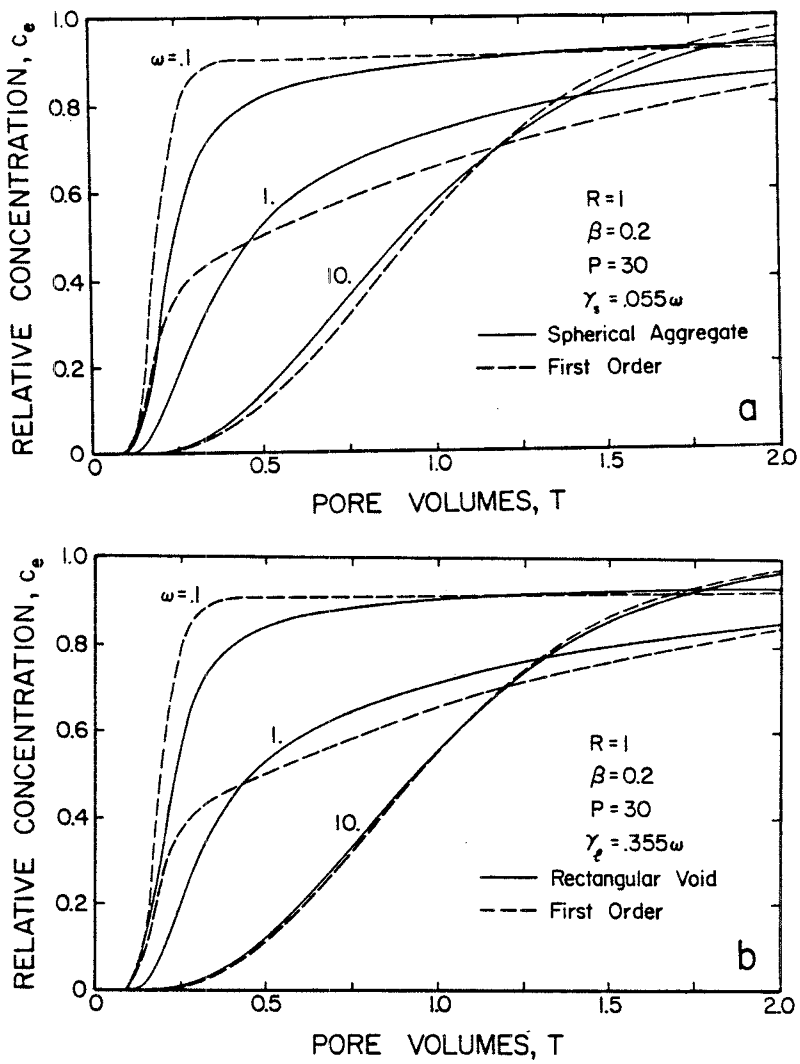

Figure 7. Breakthrough curves obtained with the approximate first-order rate model and exact curves for (a) the spherical transport model and (b) the rectangular void transport model.

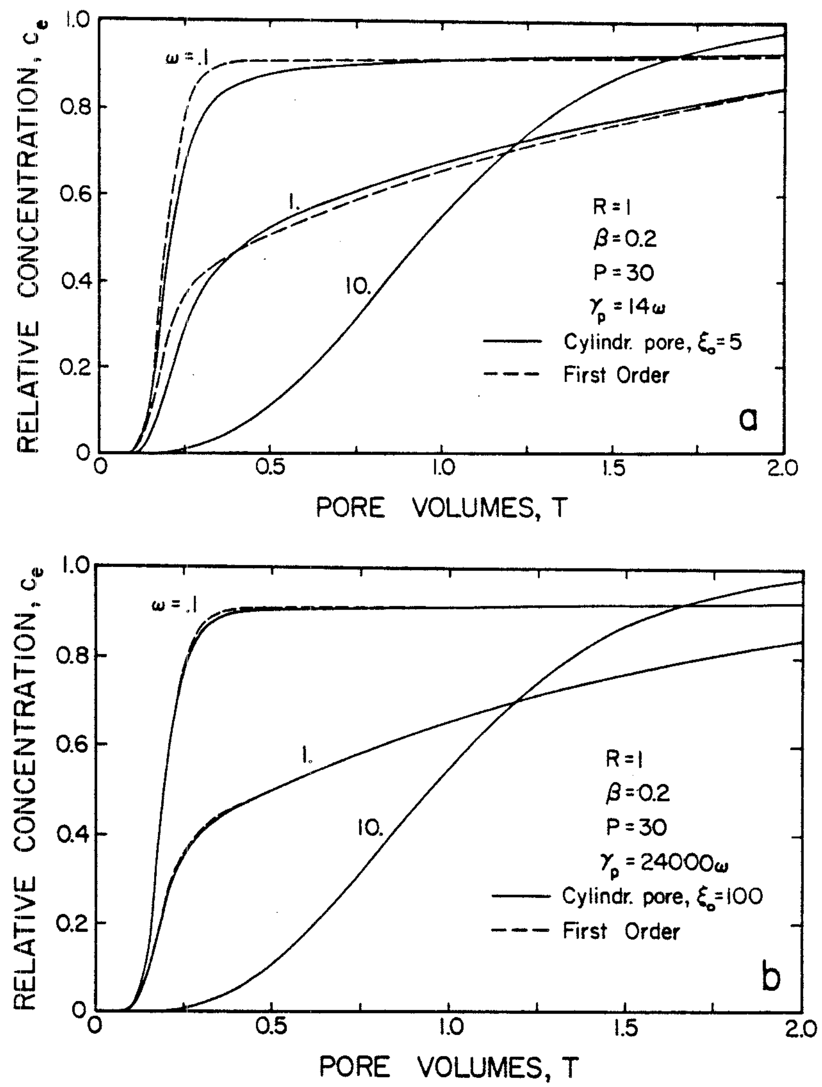

Figure 8. Breakthrough curves obtained with the approximate first-order rate model and exact curves generated with the cylindrical macropore model for two value of ξ_o.