Centrifuge Modelling Criterion for Solute Transport in Partially Saturated Flow

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Abstract Non-sorbing solute transport through partially saturated media results in asymmetric breakthrough curves. A model based on the concept of "mobile" and "immobile" fluid regions (or Two-Region Model, TRM) is often used to model tailing in the solute breakthrough curve. In this paper, we apply the TRM to non-equilibrium, non-sorbing solute transport in a partially saturated porous medium during centrifuge modelling. We discuss the behaviour of the TRM parameters, and how two sources of "immobile" water, dead-end pores and boundary layers, affect the non-equilibrium solute transport. The TRM parameters are: pore water velocity, V (L/T), dispersion coefficient, D (L^2/T), ratio of mobile water to total water content, β, and rate of mass transfer between mobile and immobile regions, α (1/T). Modelling criteria are developed using inspectional analysis. The criteria state that for an N-fold increase in centrifugal/gravitational acceleration level, V and D increase proportionally to N, while β is constant. α is proportional to 1/N if it is affected by local advection, but scales to 1 if it is diffusion-controlled. Two types of experiments are reported. In one set, for which N was constant, α and the moisture content, θ, increased with V, while β decreased. A second set of experiments was conducted at a constant flow rate, Q (L^3/T). For these experiments, α was constant, while β increased and θ decreased with V. For all experiments, the relationship between V and D was linear, irrespective of N. The variation of α and β with V or N is qualitatively linked to the configuration of the pore water. As θ increases, larger pores, with higher hydraulic conductivity, fill up. In relative terms, the smaller pores do not contribute as much to the average flow, reducing the fraction of pore fluid (β) contributing to the flow. Changes in α during the first set of experiments are likely due to velocity variations within the porous medium, while during the constant Q experiments V and θ vary inversely, balancing the effect on α, and reducing β as θ decreased. Both mass transfer and the mobile water fraction are functions of the moisture content and the magnitude of the pore velocity. The modelling criteria developed and the behaviour of the TRM parameters indicate that centrifuge modelling is a useful tool for investigating solute transport in partially saturated porous media.

1. INTRODUCTION

Predicting the movement and fate of dissolved substances is of great importance in subsurface hydrology. Mathematical models, used to describe solute movement and spreading, can be physical, phenomenological or stochastic in origin. Notwithstanding the method of derivation, the Advection-Dispersion Equation (ADE) and its extension, the Two-Region Model (TRM), are widely used predictive models in hydrology and porous media flow.

Understanding and quantifying the behaviour of model parameters can be achieved by laboratory experimentation. Centrifuge modelling of porous media flow and transport processes is a relatively recent physical modelling technique. In essence, the centrifuge is used to replicate a prototype stress field by increasing body forces on a scaled-down version of the prototype, the model, see Figure 1 (Schofield, 1980). The model is subjected to an acceleration equal to $R\omega^2$ = Ng (see Notation List for parameter definitions), where N is the acceleration level, so that the characteristic prototype length, *L*, can be reduced by N. By changing the magnitude of the acceleration level, different models result, each of which is a replica of the prototype. Of interest are the changes of system parameters, such as dispersion, due to these different acceleration levels, as they are indicative of the relationship between model and prototype. The relationship between parameters can be predicted using appropriate scaling criteria. In this case, we develop the scaling criterion using the governing equations.

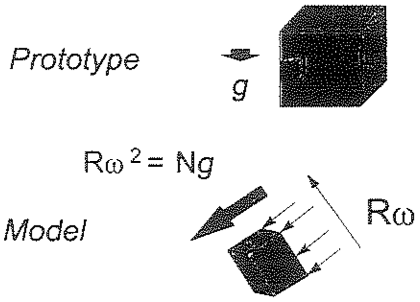

Figure 1. Acceleration effect in prototype and model (after Schofield, 1980).

Centrifuge modelling has been used to simulate a number of environmental processes, for example, column drainage (Cooke and Mitchell, 1991), saturated solute transport in homogeneous media (Hensley and Schofield, 1991) aggregated porous media (Li et al., 1994a), and unstable infiltration (Banno, 1996; Griffioen et al. 1997a). Here, we use centrifuge modelling as a tool to examine solute transport during steady flow in a partially saturated porous medium. We present the governing equations for solute transport and develop scaling conditions under which scaling of centrifuge model data to the prototype is possible.

Further, we present two series of experiments against which the model's performance can be assessed. Finally, we give a physical interpretation of the TRM parameters and how they affect solute transport in a partially saturated porous medium.

2. TWO-REGION MODEL (TRM) SOLUTE TRANSPORT

Solute transport models often assume the process to be an equilibrium one. This is indeed true for non-sorbing, homogeneous porous media. However, many soils, especially field soils, are far from homogeneous, and solutes are frequently sorbed onto, and even into, the soil. Two types of non-equilibrium can be identified: chemical and physical non-equilibrium. The latter is the case considered here. In a generic sense, physical non-equilibrium occurs because the time-dependent processes taking place within the medium are time-averaged by the model. Below, we consider two possible mechanisms for such time-dependent processes.

For non-sorbing solute transport in partially saturated porous media, non-equilibrium transport could result from the pore water configuration. A partially saturated porous medium contains both air and water, but is preferentially wet by water. Thus, water films can be found on most, if not all, the soil particles. Stagnant regions exist near the surfaces of soil particles, due to the boundary-layer effect (Figure 2a). As the solute concentration of the free flowing water increases, diffusion into the boundary layer causes loss of solute mass. If diffusion into the layer is faster than concentration changes at the film boundary and treating the film as a planar layer ($\delta << r$), the time scale for diffusion is $t_d \sim r\delta/D_{mol}$ (Helfferich, 1962). However, the thickness of the boundary layer is a function of the pore water velocity, $\delta \sim v/V$, so that $t_d \sim r/V$, since v and D_{mol} are assumed constant. At the same time, a partially saturated porous medium can also contain pockets of pore water that do not conduct any flow, but are hydraulically connected to the total pore water domain (Figure 2b). These are known as stagnant or dead-end pores. A water-borne solute can only diffuse into these stagnant regions, so that $t_d \sim r^2/D_{mol}$. These two sources of non-equilibrium have different parameters affecting their time scales and so can be expected to scale differently during centrifuge modelling.

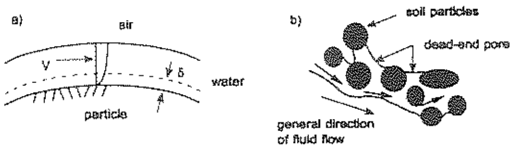

Figure 2. a) Boundary layer and stagnant zone on a soil particle, b) Dead-end pores in a partially saturated porous medium.

The TRM models the complex processes of physical or chemical non-equilibrium in a simple mathematical form. It assumes that the pore water domain can be divided into two regions: in the first, the pore water moves with a velocity, V_1, while in the second region, sometimes called the 'immobile' region, the pore water is assumed stagnant, i.e., $V_2 = 0$. Advection-dispersion accounts for solute spreading in the first region, but is augmented to include the soute exchange of the second region. The exchange is modelled as a first-order kinetic process. Therefore, the TRM is written as (van Genuchten and Wierenga, 1976):

$$\theta_1 \frac{\partial C_1}{\partial t} + \theta_2 \frac{\partial C_2}{\partial t} = \theta_1 D \frac{\partial^2 C}{\partial x^2} - \theta_1 V_1 \frac{\partial C}{\partial x} \qquad (1)$$

$$\theta_2 \frac{\partial C_2}{\partial t} = \alpha (C_1 - C_2) \qquad (2)$$

where it is assumed that the moisture content in each region is constant. The TRM introduces two parameters not present in the ADE. They are the mass transfer rate, α, and the mobile water fraction ($\beta = \theta_1/\theta_1+\theta_2$). It is clear that the TRM reduces to the ADE if mass transfer is instantaneous ($\alpha \rightarrow \infty$), or there is no hydraulic contact between the two phases ($\alpha = 0$), or there is no second region ($\theta_2 = 0$).

3. SCALING OF THE TRM

In order to apply the TRM to centrifuge modelling, we developed a scaling criterion for each parameter. Table 1 lists the scaling of the parameters. A scale factor for a variable, X, is defined as X^* and is equal to the ratio of model to prototype values, X_m/X_p. In the experiments reported here, the acceleration level, $g^* = N$. However, the column length scale, L^*, equals 1, since the column length remained constant as the acceleration level increased. This means that the stress distribution was not similar between experiments. However, the effect of stress on compression/ consolidation of the porous media was found to be negligible.

Table 1. Centrifuge scaling criterion for physical and TRM parameters.

Parameter	Dimension	Scaling Criteria (model/prototype)
particle size, d	L	1
column length, *L*	L	1
hydraulic conductivity, K	L/T	N
pore water velocity, V	L/T	N
moisture content, θ	L^3/L^3	1
mobile water fraction, β	1	1
dispersion coefficient, D	L^2/T	1 if $Pe_g < 1$ N if $Pe_g > 1$
molecular diffusion, D_{mol}	L^2/T	1
mass transfer rate coefficient, α	1/T	N
solute concentration, C	M/L^3	1
fluid density, ρ	M/L^3	1
kinematic viscosity, ν	L/T^2	1
dynamic/absolute viscosity, μ	M/LT	1
time, t	T	1/N
diffusion time scale, t_d	T	1/N for boundary layer 1 for dead-end pore

By using the same soil in model and prototype, the microscopic length scale, d^*, is 1. Since the porous medium is effectively incompressible, it follows that the porosity and moisture content are the same in model and prototype, $n^* = \theta^* = 1$. Using the same solute in model and prototype, means that the solute concentration scale, C^*, is also 1.

The scaling of flow velocity is established using the definition of hydraulic conductivity: $K(\theta) = k(\theta)\rho g/\mu$. Since the intrinsic permeability $k(\theta)$ density (ρ) and viscosity (μ) are not affected by acceleration changes, the hydraulic conductivity scale, $K^* = K_m/K_p$, is N. From Darcy's law, $V = Ki/n$, we see that $V^* = K^*$ if hydraulic gradient, i, and porosity, n, are not affected by the acceleration level.

Next, we develop condition for which centrifuge modelling using the TRM is possible. Using the scaled parameters, t^*, V^* etc., the model parameters can be substituted in the prototype equation,

$$t^* \frac{\partial C_1}{\partial t} + t^* \frac{1-\beta}{\beta} \frac{\partial C_2}{\partial t} = \frac{(L^*)^2}{D^*} D_m \frac{\partial^2 C_1}{\partial x^2} - \frac{L^*}{V_1^*} V_{1m} \frac{\partial C_1}{\partial x} \quad (3)$$

$$t^* \frac{1-\beta}{\beta} \frac{\partial C_2}{\partial t} = \frac{1}{\alpha^*} \alpha_m (C_1 - C_2) \quad (4)$$

Model and prototype are equal if the scale factors are equal. Thus, t^*, $(L^*)^2/D^*$, L^*/V_1^* and $1/\alpha^*$ must be equal. For example, $t^* = L^*/V_1^*$. Since $V_1^* = N$ and $L^* = 1$, t^* must equal 1/N. Also, $(L^*)^2/D^* = 1/N$, if $D^* = N$. For grain Péclet numbers, Pe_g, greater than 1, $D \propto V_1$ (Bear and Verruijt, 1987), so that $D^* = V_1^* = N$. When $Pe_g < 1$, $D \propto D_{mol}$ and direct scaling of the TRM is not possible.

The additional parameters of the TRM, α and β, must also scale correctly for centrifuge modelling to proceed. From (3) and (4), α^* must be equal to $1/t^*$, so that $\alpha^* = N$. Many studies have found $\alpha \propto V_1$ (Rao et al., 1980; de Smedt and Wierenga, 1984; Kookana et al., 1993; Griffioen et al., 1997b), so that the scaling for α is correct when $\alpha^* = V_1^* = N$.

The mobile pore water fraction, β, is often assumed to be a function of the pore water geometry (Li et al., 1994b). For a given moisture content β would, therefore, scale as $\beta^* = 1$. In many instances, however, the moisture content is not constant between experiments. In these cases, assumptions have to be made about the dependence of θ_1 and θ_2 on θ. Below, we use experimental data to develop such a relationship.

The two time scales associated with physical non-equilibrium, viz., boundary-layer diffusion and dead-end pore diffusion, scale differently. For dead-end pore diffusion, $t_d \sim r^2/D_{mol}$, therefore, $t_d^* = 1$. The boundary-layer diffusion time scale, $t_d \sim r/V$, we expect to scale like 1/N during centrifuge modelling.

4. EXPERIMENTS

The experiments were conducted on the 40-gton geotechnical centrifuge facility at the Department of Civil Engineering, University of Western Australia, shown in Figure 3. A schematic diagram of the centrifuge package contained within a strongbox is given in Figure 4. The major features of the setup are: the column, infiltration assembly, resistivity probes and collection tanks. The peristaltic pump used to pump fluid onto the package, was located outside the centrifuge chamber. Details of the apparatus and column preparation are discussed by Griffioen and Barry (1998). The soil used is a silica silt, with an average particle size of 45μm.

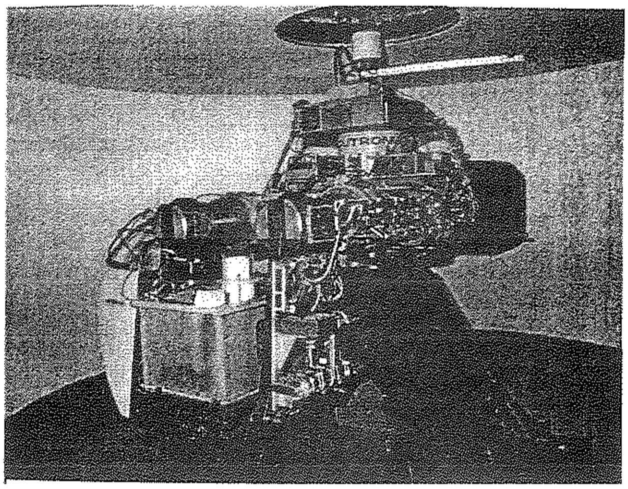

Figure 3. View of centrifuge facility at the Civil Engineering Department, University of Western Australia.

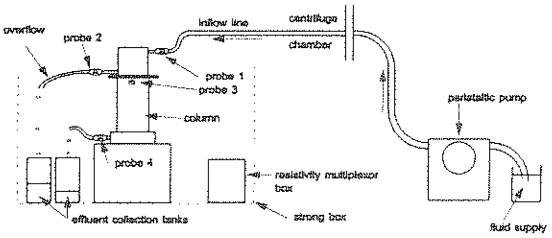

Figure 4. Centrifuge setup as used in the experiments reported here.

Each test proceeded in the following manner: The initially saturated column was drained at N = 100 until very little water drained out. The centrifuge was then stopped in order to weigh the column and estimate the average moisture content. The column was replaced in the strongbox and the centrifuge accelerated to the desired level. At this level, the peristaltic pump was turned on to supply de-ionised water at the desired rate. When a constant effluent flow rate was established, the de-ionised water was replaced with a saline solute of known concentration, NaCl at 3 g/L. The flow rate was calculated using pressure transducer data, while the solute concentration was determined from the resistivity data. A sample breakthrough curve (BTC) is shown in Figure 5.

We report experiments conducted under two conditions: constant acceleration level, N, and constant flow rate, Q. Experiments 1 to 5 were conducted at an acceleration level

Table 2. TRM parameters obtained by curve fitting.

Parameter	Experiment 1	2	3	4	5	6	7	8
Acceleration level, N	100	100	100	100	100	50	50	150
Darcy flux, q (= Q/A, cm/min)	0.1084	0.7451	0.0627	0.1074	0.0408	0.0372	0.0382	0.0502
Moisture content, θ (cm^3/cm^3)	0.2675	0.2484	0.2611	0.2865	0.2358	0.2548	0.2548	0.2167
Grain Péclet number, Pe_g	2.34	1.75	1.55	2.31	1.03	0.72	0.67	1.50
Pore velocity, V (cm/min)	0.430	0.305	0.264	0.388	0.188	0.135	0.128	0.254
Dispersion coefficient, D (× 10^{-2} cm^2/min)	3.64	4.55	4.58	5.36	2.50	0.62	0.56	5.14
Mobile water fraction, β	0.917	0.872	0.854	0.839	0.913	0.930	0.955	0.848
Dimensionless transfer rate, ω	0.765	0.631	0.970	0.797	0.416	0.553	0.147	0.396
Transfer rate, α (× 10^{-3} 1/min)	6.47	3.67	4.75	6.68	1.33	1.61	0.44	1.55

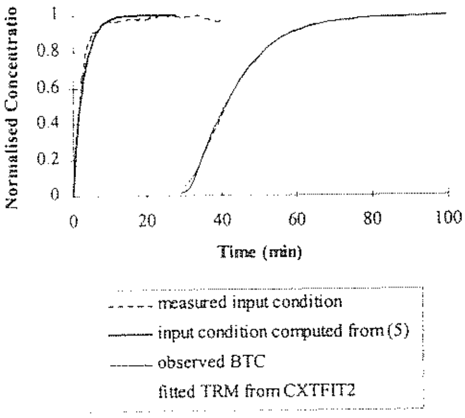

Figure 5. Breakthrough curve and fitted TRM for experiment 2.

of N = 100, while experiments 5 to 8 were subject to a constant Q. Since experiment 5, at N = 100, is applicable to both groups, 8 experiments in all are reported here.

In order to determine the solute transport parameters, a curve-fitting procedure, CXTFIT2 (Toride et al., 1995) was used. This program conducts a search for model parameters by a least-squares fit to effluent data, and can be programmed with a user-defined input condition. In our case, the input condition was measured by a resistivity probe (probe 3 in Figure 4) and matched with the equation,

$$C = \left(1 - \exp\left(\frac{-Q_{tot}t}{Vol}\right)\right) \qquad (5)$$

where t is the time since introducing the solute into the infiltration assembly, and Q_{tot} is the rate at which the fluid is pumped onto the centrifuge. For the constant Q experiments, Q is equal to Q_{tot} less the overflow flow rate. This equation is based on the assumption of a well-mixed reservoir, subject to a step change in input concentration. Figure 5 shows how the measured input condition is matched with (5) for experiment 2.

5. RESULTS AND DISCUSSION

5.1 Dispersion Coefficients (D)

The TRM parameters obtained by curve fitting for the 8 experiments are listed in Table 2. Figure 6 shows the dispersion coefficients, D, for the experiments as a function of the mobile region velocity, V_1. For both types of experiments (constant N and Q) the dispersion coefficient increases approximately linearly with V_1. The general trend, including the experiments for which $Pe_g < 1$, shows that $D \propto V_1$.

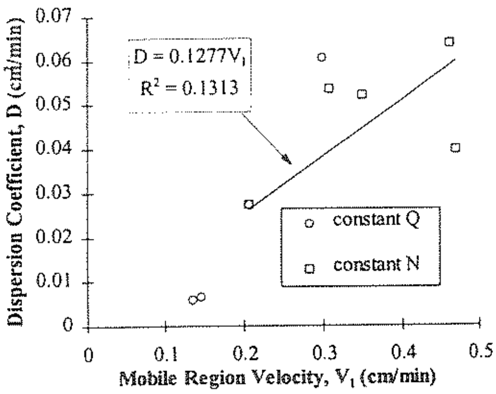

Figure 6. Dispersion coefficient, D, as function of mobile region velocity, V_1.

5.2 Mobile Water Fraction (β)

For the constant acceleration (N) experiments (Figure 7), β decreases as the moisture content increases, so that at saturation β is less than 1. However, for a saturated, homogeneous porous medium, all the pore water is expected to be mobile, i.e., β = 1. On the other hand, the constant Q experiments (Figure 7), show β increasing as the moisture content increases. This increasing trend has been observed during laboratory experiments (N = 1) on partially saturated porous media. Griffioen et al. (1997a) have compared the results of a number of experiments reported in the literature, and observed that β remains constant for different values of

θ when the porous medium is well rounded and uniformly sized (e.g., glass beads), but increases in angular porous media (e.g., sands).

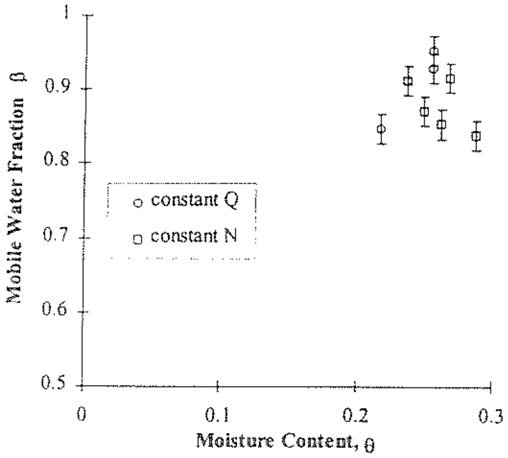

Figure 7. Relationship between β and θ.

A constant θ value must coincide with a constant β value, if the scaling criteria, $\beta^* = 1$ at a constant θ, is correct. From the above observations it is evident that different values for β are possible at a given moisture content. However, when the scaling of β is considered, these differences are negligible. For example, experiments 3 and 7 have very similar moisture contents, $\theta = 0.26$, while $\beta = 0.854$ and 0.955, and $N = 100$ and 50, respectively. If experiment 3 is considered the model, then $N^* = 2$, while $\beta^* = 0.854/0.955 = 0.894$. Thus, β^* is close to 1, but very different from N.

An interpretation for β has been suggested by Bajracharya and Barry (1997), who found β to reflect deviations in the hydraulic conductivity field. This interpretation agrees with our observations, since in partially saturated media the moisture content determines the hydraulic conductivity distribution. A simpler version of this approach is to assume that the mobile and immobile regions are characterised by a certain ratio of their hydraulic conductivities (Zurmühl and Durner, 1996). Then at a given moisture content, the largest pores constitute the mobile region, while the smallest pores are considered immobile. Thus, if we know how K varies with θ, we can predict β qualitatively.

5.3 Mass Transfer Rate Coefficient (α)

The scaling criterion, $\alpha^* = V^*$, is shown in Figure 8 to hold for the constant acceleration experiments, while for the constant Q experiments α is approximately constant.

During the constant Q experiments, $V\theta/A$ remains constant, so that as V increases, θ must decrease. However, α remains constant (≈ 0.0015 1/min). We interpret this to mean that in these experiments the rate of mass transfer per unit volume of pore water increases with increasing V, while the decreasing moisture content nullified this effect to produce a constant mass transfer rate (α). The mass transfer coefficient, α, is therefore a function of the pore water velocity, V, and the moisture content, θ. The same argument can be used for the constant N experiments, where V and θ both increase, amplifying the effect on α. As shown in Figure 9, there is no difference between the two types of experiments when α is considered a function of Q.

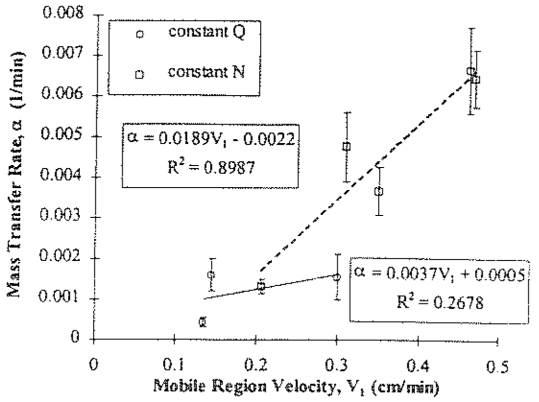

Figure 8. Mass transfer rate, α, variations for the various experiments.

Concerning the mechanisms of mass transfer identified above, we note that the mass transfer rate scale is $1/\alpha^* = 1/N$. The boundary-layer time scale, $t_d^* = 1/N$, matches the observed time scale, although the moisture content is not constant between the experiments. If mass transfer is a diffusion-limited process, experiments with the same moisture content would exhibit the same transfer rate. Thus, although $\alpha^* = 1$ for the constant Q experiments, the non-equilibrium is not due to diffusion as θ was not constant.

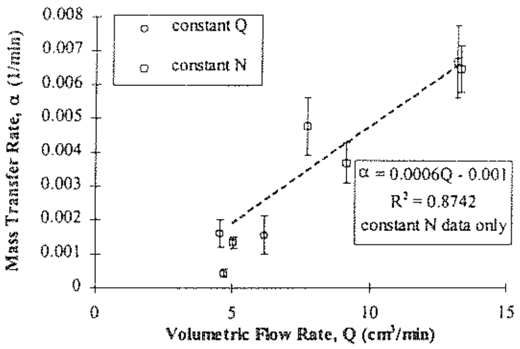

Figure 9. Mass transfer, α, as a function of the volumetric flow rate, Q.

6. CONCLUSION

The centrifuge is a suitable tool for investigating non-equilibrium flow. Each of the experiments reported here was modelled using the TRM. By maintaining N or Q constant, we were able to investigate how the TRM parameters varied. The scaling for β using the experimental data suggests that β^* is approximately equal to 1. The relationship between α and V indicates that the mass transfer rate scales in the expected manner. At the same time, the process of mass transfer is assumed to be controlled by film diffusion since the scaling criterion for α is 1/N.

7. ACKNOWLEDGEMENTS

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9. NOTATION LIST

ADE	Advection-Dispersion Equation
BTC	breakthrough curve
C	solute concentration $[M/L^3]$
d	particle size [L]
D	hydrodynamic dispersion $[L^2/T]$
D_{mol}	molecular diffusion $[L^2/T]$
g	gravitational acceleration $[L/T^2]$
i	hydraulic gradient
k	intrinsic permeability $[L^2]$
K	hydraulic conductivity [L/T]
K_s	saturated hydraulic conductivity [L/T]
L	column length [L]
n	soil porosity
N	acceleration level
Pe_g	grain Péclet number $(V_1 d/D_{mol})$
Q	flow rate through column $[L^3/T]$
Q_{tot}	pump rate of peristaltic pump $[L^3/T]$
r	particle radius (= d/2) [L]
R	centrifuge radius [L]
t	time [T]
t_d	diffusion time [T]
TRM	Two-Region Model
V	pore water velocity [L/T]
Vol	volume of water in reservoir $[L^3]$
x	spatial length [L]
X	general parameter
α	mass transfer rate [1/T]
β	mobile water fraction (= θ_1/θ)
δ	boundary-layer thickness [L]
μ	dynamic viscosity [M/LT]
ν	kinematic viscosity $[L/T^2]$
ρ	fluid density $[M/L^3]$
θ	moisture content $[L^3/L^3]$
ω	angular velocity of centrifuge [1/T], or dimensionless mass transfer rate ($\alpha L/q$)

Common Subscripts

1, 2	mobile and immobile region, respectively
m, p	model and prototype, respectively

Superscript

*	scaled parameter, defined as model/prototype