

Numerical Modelling of Tracer Transport in Unsaturated Porous Media

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ABSTRACT

Many types of chemical substances have been used as tracers to estimate the migration of contaminant in a porous media. Inorganic ionic compounds have been applied extensively as hydrogeologic tracers. Sodium chloride is generally used as a tracer since this common salt does not degrade or get removed from the system. Movement of tracer could be described as migration of a non-reactive constituent. A tracer transport numerical model was developed according to the advective-dispersive contaminant transport equation in unsaturated porous media. The governing equation was solved numerically and coded in MATLAB program. The objectives of this study were to develop a model for estimating the non-reactive constituent transport in the unsaturated porous media and to determine the impact of ionic strength of tracer and the effect of the thickness of porous media. The experiments were conducted with two different sodium chloride tracer concentrations (low strength-200 mg/L and high strength-500 mg/L) and for two different soil depths (5 and 20 cm). The observation and simulation data indicate that the interference from soil background concentration is significant, provided that the high strength tracer is applied. As expected, the tracer transport in the thick layer took longer elapse time than in the thin layer. The simulation results using the developed model corresponded very well with the observed data.

Keywords: Advective-Dispersive Transport, Numerical Model, Tracer Test, Unsaturated Soil.

NOMENCLATURE

C	tracer concentration; g cm^{-3}	p	hydraulic properties coefficient; cm^{-1}
D_M	molecular diffusion coefficient; $\text{cm}^2 \text{h}^{-1}$	q_z	Darcy's velocity; cm h^{-1}
D_z	dispersion coefficient; $\text{cm}^2 \text{h}^{-1}$	t	previous time step; unitless
k_{rw}	relative conductivity; ($0 \leq k_{rw} \leq 1$); unitless	$t+\Delta t$	current time steps; unitless
K_{zz}	saturated hydraulic conductivity; cm h^{-1}	z	vertical direction (positive upwards); cm
$K_{zz}k_{rw}$	relative hydraulic conductivity; cm h^{-1}	θ	volumetric water content; $\text{cm}^3 \text{cm}^{-3}$
m	hydraulic properties coefficient; unitless	θ_r	residual soil moisture contents; $\text{cm}^3 \text{cm}^{-3}$
M	specific moisture capacity; cm^{-1}	θ_s	saturated moisture contents; $\text{cm}^3 \text{cm}^{-3}$
n	hydraulic properties coefficient; unitless	λ	dispersivity; cm
N_i	stiffness functions; unitless	v	pore water velocity ; ($= q_z / \theta$); cm h^{-1}
N_j	shape functions; unitless	ψ	pressure head; cm

1. INTRODUCTION

The tracers are useful for characterizing unsaturated water movement in the porous media. Many substances could be applied as tracers into unsaturated porous media. The tracers used could be classified into six categories including ions, dyes, gases, isotopes, water temperature and particulates. The ionic tracers are less toxic than others, and thus they are applied extensively. The ionic tracers include common salts, such as Cl^- , NO_3^- , Br^- , Ca^{2+} , K^+ etc. Most of these ionic tracers are anions, which are less affected by the porous media (Gelhar *et al.* 1992; Boulding and Ginn 2004). The retardation of anion due to absorption and precipitation increases, if the pH of the system decreases. The cation tracer could react to the clay minerals, especially Na^+ and Ca^{2+} , through the ion exchange process (Mattson 1929). The ionic tracers are non-compost materials, and tracer loss is negligible. However, soil sometimes contains large quantities of ions, especially Cl^- and NO_3^- , hence high tracer concentration is required for these high background concentration sites. If the large amounts of tracers are applied, they could potentially affect adversely on water quality similar to the pollutants (Grisak and Picken 1980a, b; Everts *et al.* 1989). The applications of NaCl tracer have been used by many researchers (Mather *et al.* 1969; Warrick *et al.* 1971; Grisak and Picken 1980a, b; James and Rubin 1986; Nützmann *et al.* 2002; Sato *et al.* 2003) to determine the characteristics of the water flow and the solute transport in unsaturated soils.

A number of models have been developed in the literatures for the prediction of the conservative tracer transport. These models are based on the advective-dispersive solute transport equation. This governing equation can be solved by two different techniques namely the analytical solution and the numerical approach. Most of the referenced models (Warrick *et al.* 1971; James and Rubin 1986; Wallach and Steenhuis 1998; Nützmann *et al.* 2002; Sato *et al.* 2003; Toride *et al.* 2003) relied on the analytical solution, which contained many boundary limitations. Some numerical tracer transport models have also been proposed (Grisak and Picken 1980a, b; Picken and Grisak 1981); however, they cannot apply to the unsaturated soil condition. The practical applications of the developed model in this paper are to estimate the tracer travel time and to evaluate the appropriate conditions for tracer applications.

2. GOVERNING EQUATIONS FOR TRACER TRANSPORT MODEL

One-dimensional tracer transport in unsaturated soil in vertical z direction is given using the advection dispersion equation as (Fetter 1992; Schnoor 1996):

$$\frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - q_z \frac{\partial C}{\partial z} \quad (1)$$

The dispersion coefficient couples both mechanical and molecular diffusion, and the equation was given as (Fetter 1992; van Genuchten and Wierenga 1986):

$$D_z = \lambda v + D_M \quad (2)$$

Richards' equation is applied to determine Darcy's velocity, which was presented in Eq. (1). Richards' equation was given as (Ségol 1993):

$$M \frac{\partial \psi}{\partial t} = \frac{\partial}{\partial z} \left(K_{zz} k_{rw} \left(\frac{\partial \psi}{\partial z} + 1 \right) \right) \quad (3)$$

Richards' equation contained non-linear hydraulic parameters: M and $K_{zz} k_{rw}$, which could be evaluated using van Genuchten (1980) model. The relevant equations are given as follows.

$$\theta = \theta_r + \frac{\theta_s - \theta_r}{\left(1 + (p|\psi|)^n \right)^m} \quad (4a)$$

$$K_{zz} k_{rw} = K_{zz} \frac{\left[1 - (p|\psi|)^{n-1} \left[1 + (p|\psi|)^n \right]^{-m} \right]^2}{\left[1 + (p|\psi|)^n \right]^{m/2}} \quad (4b)$$

3. NUMERICAL SOLUTIONS FOR TRACER TRANSPORT MODEL

The advective-dispersive solute transport (Eq. (1)) and Richards' (Eq. (3)) equations are solved by the Galerkin technique. The numerical solution is arranged in systematically algebraic matrices using the finite element method (FEM) (Segelind 1984; Ségol 1993). The numerical solution and the algebraic matrix system are given as follows.

Tracer transport model:

$$\{P_{ij}\} + \{Q_{ij}\} C_j + \{Q_{ij}\} \frac{\partial C_j}{\partial t} = \{S_i\} \quad (5)$$

where

$$[P_{ij}] = \sum_e \int_{z=0}^{z=L} \left(D_z \frac{\partial N_i}{\partial z} \frac{\partial N_j}{\partial z} + N_i \frac{\partial N_j}{\partial z} q_z \right) dz$$

$$[Q_{ij}] = \sum_e \int_{z=0}^{z=L} N_i N_j \theta dz$$

$$\{S_i\} = N_i D_z \left(\frac{\partial C}{\partial z} \right)_{z=0}^{z=L}$$

Richards' equation:

$$[A_{ij}] \psi_j + [B_{ij}] \frac{\partial \psi_j}{\partial t} = \{E_i\} \quad (6)$$

where

$$[A_{ij}] = \sum_e \int_{z=0}^{z=L} K_{zz} k_{rw} \left(\frac{\partial N_i}{\partial z} \frac{\partial N_j}{\partial z} \right) dz$$

$$[B_{ij}] = \sum_e \int_{z=0}^{z=L} N_i N_j M dz$$

$$\{E_i\} = N_i K_{zz} k_{rw} \left(\frac{\partial \psi}{\partial z} + 1 \right) \Big|_{z=0}^{z=L} - \sum_e \int_{z=0}^{z=L} K_{zz} k_{rw} \frac{\partial N_i}{\partial z} dz$$

The algebraic matrix system is formed with linear Lagrange function. The iteration process is investigated by the single Picard iteration as follows (Ségol 1993).

Tracer transport model:

$$\left\{ \left(\frac{1}{2} [P_{ij}] + [Q_{ij}] \right) + \frac{1}{\Delta t} [Q_{ij}] \right\} \{C\}^{t+\Delta t} - \{C\}^t$$

$$= \{S_i\} - ([P_{ij}] + [Q_{ij}]) \{C\}^t \quad (7)$$

Richards' equation:

$$\left(\frac{1}{2} [A_{ij}] + \frac{1}{\Delta t} [B_{ij}] \right) \{\psi\}^{t+\Delta t} - \{\psi\}^t = \{E_i\} - [A_{ij}] \{\psi\}^t \quad (8)$$

4. MATERIALS AND METHODS

The laboratory scale soil column set up is illustrated in Figure 1. The infiltration column was fabricated from a plexi-glass tube of 6.59 cm inside diameter, and the total length was 30 cm. All soil column tests were packed with the disturbed topsoil sample and the bulk density was controlled at 1.25 g/cm³.

The soil hydraulic properties test was undertaken using the dynamic method (Klute 1986). The pore pressure head at particular elevations were measured using tensiometers. The water content was analysed using a gravimetric method (Rayment and Higginson 1992). Three series of tracer tests were conducted that included: 1) low strength tracer with thin soil layer, 2) high strength tracer with thin soil layer and 3) high strength tracer with thick soil layer. The sodium chloride concentration was determined using the Electrical Conductivity (EC) 1:5 soil water extraction method (Rayment and Higginson 1992). The details of soil column tests are provided in Table 1.

5. RESULTS AND DISCUSSIONS

The soil characteristics are evaluated and the summary is given in Table 2. Soil contains some minerals that may interfere with the EC measurement. This background EC concentration was found to be equivalent to 117 mg/L sodium chloride solution. Hence higher concentration of 200 and 500 mg/L NaCl tracer solution were chosen and their equivalent EC values are 0.130 and 0.503 mohs/cm, respectively.

5.1 Soil hydraulic properties

The water retention curves are illustrated in Figure 2. θ_s and θ_r are found to be 0.42 and 0.04, respectively. The coefficients: m , n and p were evaluated as 0.4026, 1.6740 and 0.0249 cm⁻¹, respectively.

5.2 Unsaturated water movement

The input parameters for Richards' equation are presented in Table 3.

The pressure head profiles for the thin soil layer are shown in Figure 3. The simulation results indicate that about half of soil column reached the full saturation condition within 8 hours. This implies that the whole column may reach the full saturation condition within 16 hours based on simulation. This statement was remarkably close with the observation. During the experiments, the first drop of filtrated water was percolated out after 18 hours of continuous feeding. These pressure head boundary conditions provide excellent support to the time-varying infiltration flow rate.

The simulated pressure head profiles for the thick soil layer are shown in Figure 4. By estimation, the travel time of feeding water in the thick soil layer should be 64 hours. The first drop of filtrated sample was observed after applying the tap water for 66 hours. Water moved through the thin and thick soil layer within 16-18 and 64-66 hours, respectively. The movement of water through the thin soil layer was four times faster than in the thick soil layer. Coincidentally, the thickness of thin layer was four times less than the thick layer. Therefore, there was no retardation effect of migration of water through both soil thicknesses.

5.3 Tracer transport in unsaturated soil

The input parameters used for the tracer transport model are summarised in Table 4. The tracer concentration profiles for T-1 are presented in Figure 5. The estimation of tracer retention time in the thin soil layer was 16 hours. Regarding the observed data, the maximum tracer concentration was 176 mg/L, which was less than the feed concentration of 200 mg/L. This loss might be a result of the ion exchange process onto soil minerals (Mattson 1929).

The tracer transport under the advection-dispersion mechanisms, will consume longer retention time than the water. The averaged D_z values for each time intervals are given in Table 5. The calculated coefficient of 0.01-0.2 cm²/h, lines within the acceptable range. The estimated effective diffusion coefficient of NaCl tracer is 0.037 cm²/h (Picken and Grisak 1981). The D_z is affected by time varying parameters, which depends on the hydrodynamics dispersion. Darcy's and pore velocities are the key parameters, which vary with time and the hydrodynamics dispersion is non-linear (Boulding and Ginn 2004).

The concentration profiles for T-2 are shown in Figure 6. These results indicate that tracer might travel through 70% of the height of the column depth within 11 hours. However, the estimated travel time of the tracer is 16 hours which is closer to the retention time of water. Moreover, the maximum tracer concentrations were observed in the range of 494-497 mg/L.

The migration of tracer through these thin unsaturated soil layers could be clearly observed, when the high strength tracer was applied. The estimated D_z values of these different tracer concentrations are not significantly different. Thus, the advection was the main mechanism, which controls the tracer transport. Obviously, the marked difference between simulation and observation data in Figures 5 and 6 may be primarily due to the use of a single tensiometer to obtain the pressure head measurement. The estimated Darcy's velocities of the tests T-1 and T-2 contain some truncation error due to the measurement of pressure gradient. Therefore, the advection-dispersion tracer transport parameters may be underestimated. Better match between simulation and experimental observations is obtained when 5 tensiometers are used for T-3 as shown in Figure 7.

The travel time of tracer in the thick soil layer is approximately 80 hours. The estimated tracer travel time was 14 hours longer than the water travel time. This indicates that the thickness of soil layer might potentially affect the tracer transport. Besides, the maximum concentration of tracer was found to be in the range of 488-497 mg/L. These maximum tracer concentrations were not different what was found in T-2. This indicates that the losses of tracer concentration could be negligible. The dispersion coefficient for this experiment is given in Table 6.

The D_z values for the thick soil layer are higher than the thin soil layer. This fact suggests that the large scale spreading of tracer may occur and this may be the reason for the difference in elapsed travel time. By statistical comparison, the developed model could precisely predict the pressure head and tracer concentration profiles, especially for the case T-3. This claim relies on the number of tensiometers used as increase a number of tensiometers could precisely identify the boundary conditions and hence accurately measure Darcy's and pore velocities. Thus, the advection-dispersion parameters involved the model could be estimated precisely.

The ionic tracer might be applied to the soil sample contains large quantities of ion; however, the applied tracer concentration must be significantly higher than the background concentration. The advection-dispersion parameters could be estimated accurately, if the pressure head boundaries are measured precisely. A long elapsed travel time in the thick soil layer is observed because of a large scale spreading of tracer.

6. CONCLUSION

It was shown that a tracer test could be used to evaluate the retention and movement of water in unsaturated soil

conditions. The advective-dispersive solute transport was the main governing equation for the developed model. The model was solved numerically using the Galerkin finite element technique. Unsaturated infiltration flow velocity was estimated using Richards' equation. From the results, it is clear that the developed tracer transport model could generate accurate simulation results for all tracer transport conditions and also effectively estimates the diffusion-dispersion coefficients. The high strength tracer concentration and the thin soil layer could be the best solution for the estimation of unsaturated water movement in this topsoil sample. The high strength tracer concentration could overcome the interference of background concentration contained in the soil sample. In the thick soil layer, the tracer travel time could be extended by the diffusion mechanism and the high dispersion could potentially cause large spreading of the tracer.

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Table 1- Details of soil column tests

Tests	Packing depth; cm	NaCl dosage; mg/L	Feeding rate; cm ³ /h	Location of tensiometers; cm	Location of EC(1:5) measurements; cm
HP	5	0	7.18	2.5	None
T-1	5	200	7.18	2.5	0.5, 1.5, 2.5, 3.5, 4.5
T-2	5	500	7.18	2.5	0.5, 1.5, 2.5, 3.5, 4.5
T-3	20	500	7.18	2.5, 6.5, 14.0, 17.5	2.0, 6.0, 10, 14, 18

Note: HP is a hydraulic properties test. T-1, T-2 and T-3 are the tracer tests no.1, 2 and 3, respectively.

Table 2- Characteristics of topsoil sample

Parameters	Values	Parameters	Values
Particle size analysis		Bulk density (g/cm ³)	1.28
Sand content (%)	37.51	Void ratio	0.74
Silt content (%)	43.79	Moisture content (%)	5.00
Clay content (%)	18.70	Soil EC (1:5) (mohs/cm)	0.129
Texture	Loam	Sodium ion (meq/100g soil)	
K _{zz} (cm/h)	0.662		0.783
Specific gravity	2.55		

Table 3- Input parameters for Richards' equation

Parameters	Values
Domains	5 cm (thin) and 20 cm (thick)
Boundary conditions	Upper and lower ψ are -0.005 and -624 cmH ₂ O, respectively. Initial ψ is -624 cmH ₂ O.
Time domain	14 hours (thin) and 72 hours (thick)
Number of time steps; nt	800 steps per hour (thin) and 1272 steps per hour (thick)
Nodal spacing; dz	0.125 cm (thin) and 0.25 cm (thick)

Table 4- Input parameters for tracer transport model

Parameters	Values
Domains	5 cm (T-1 and T-2) and 20 cm (T-3)
NaCl conc.	200 mg/L (T-1) and 500 mg/L (T-2 and T-3)
Nodal Darcy's velocity	Direct load from the Richards' equation
D _M	0.04788 cm ² /h
Fraction for D _M	0.4
Boundary conditions	Concentrations at the upper boundary are 200 (T-1) or 500 (T-2 and T-3) mg/L and at the lower boundary is 0 mg/L. The initial concentrations are 200 (T-1) or 500 (T-2 and T-3) mg/L.
Time domain	14 hours (thin layer) and 72 hours (thick layer)
Number of time steps; nt	800 steps per hour (thin) and 1272 steps per hour (thick)
Nodal spacing; dz	0.125 cm (thin) and 0.25 cm (thick)

Table 5- Average diffusion-dispersion coefficients for T-1

Time (hour)	D_z (cm ² /h)	Time (hour)	D_z (cm ² /h)
3	0.0095	9	0.0356
6	0.0133	11	0.1950

Table 6- Average diffusion-dispersion coefficients for T-3

Time (hour)	D_z (cm ² /h)	Time (hour)	D_z (cm ² /h)
8	0.335	48	2.997
16	0.746	56	3.028
24	1.703	72	3.489

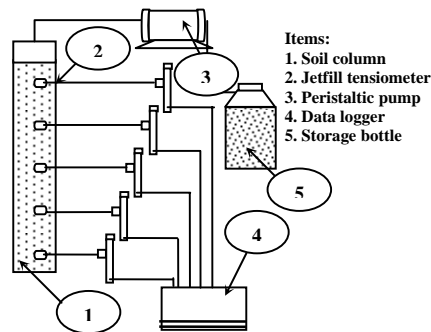


Fig. 1-Column set up.

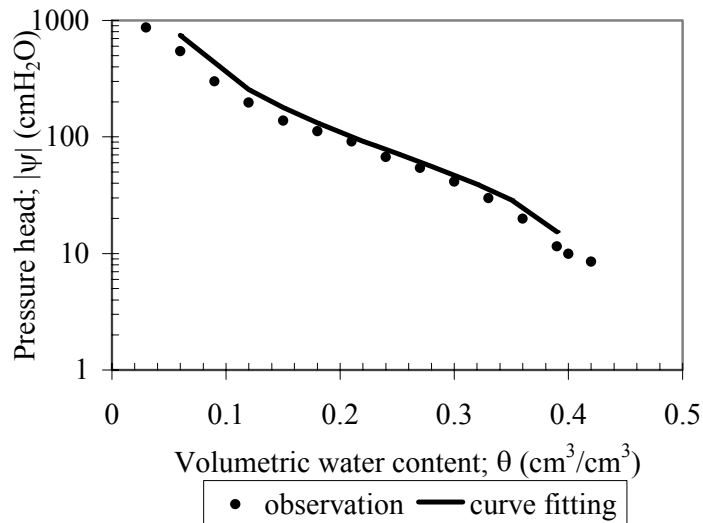


Fig. 2- Soil water retention curve fitting.

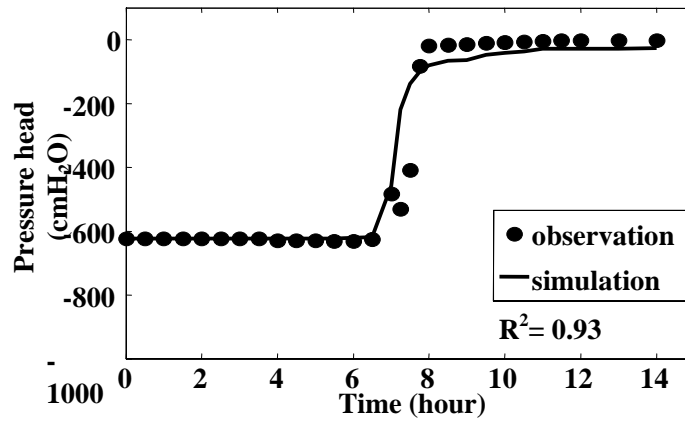
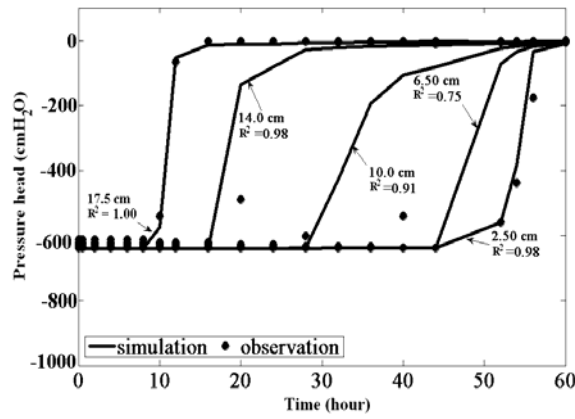
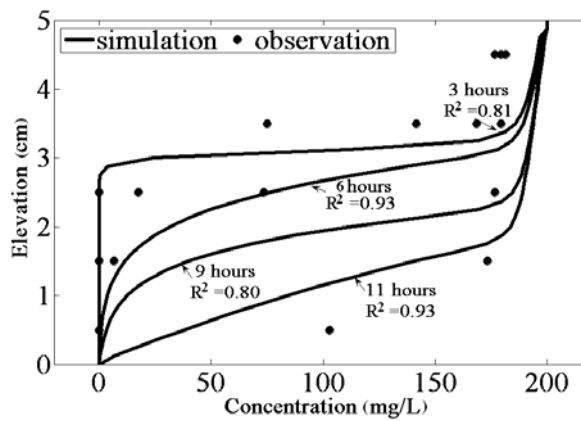


Fig. 3- Pressure head versus time for a thin layer unsaturated soil column.



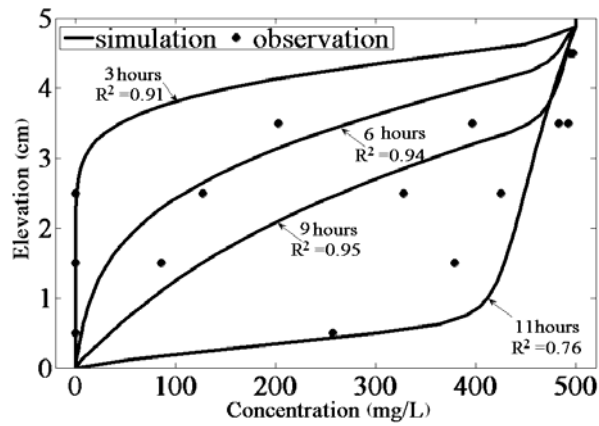
(Datum is at the column base and R^2 is the residual square.)

Fig. 4- Elevated pressure head versus time for a thick soil layer unsaturated soil column.



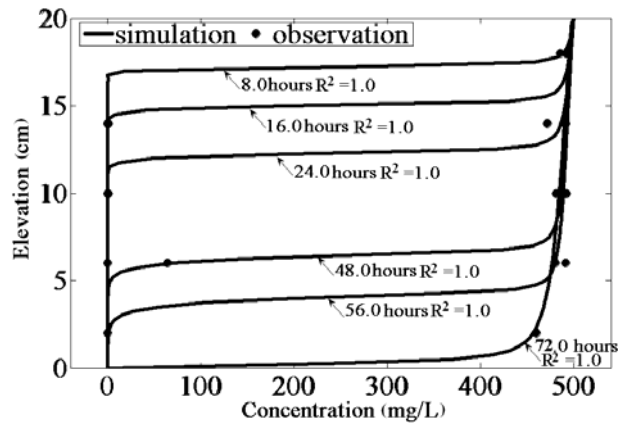
(Datum is at the column base and R^2 is the residual square.)

Fig. 5- Concentration profiles for T-1.



(Datum is at the column base and R^2 is the residual square.)

Fig. 6- Concentration profiles for T-2.



(Datum is at the column base and R^2 is the residual square.)

Fig. 7- Concentration profiles for T-3.