THE MIGRATION OF LIGHT ORGANIC LIGUIDS IN AN UNSATURATED-SATURATED ZONE OF THE SOIL

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ABSTRACT

A one-dimensional finite difference model for the simultaneous movement of light non-aqueous phase liquid (LNAPL) and water through unsaturated-saturated zone of the soil in a three fluid phase system with air assumed constant at atmospheric pressure is developed. The flow equations described the motion of light non-aqueous phase liquid and water are cast in terms of the wetting and non-wetting fluid pressure heads respectively. The finite difference equations are solved fully implicitly using Newton-Raphson iteration scheme. The present numerical results are compared with results of Kaluarachchi and Parker (1989) and there is a good agreement between them. The present model can be used to simulate various transport problems in a good manner. Results proved that the maximum LNAPL saturation occurred below the source of the contaminant during LNAPL infiltration. During redistribution, the LNAPL saturation had a maximum value at the advancing of the LNAPL infiltration front.

الخـلاصـــة

في هذه الدراسة تم تطوير نموذج عددي ذو بعد واحد يستخدم الفروقات المحددة لوصف حركة الماء والسائل العضوي الأخف منه خلال التربة المشبعة وغير المشبعة مع ثبوت ضغط الهواء عند الضغط الجوي. أن المعادلات التي تصف حركة الماء والسائل العضوي وضعت بدلالة عمود الضغط لتلك السوائل. أن معادلات الفروقات المحددة حلت (fully implicitly) وباستخدام طريقة (Newton-Raphson). تمت عملية اختبار كفاءة النموذج الحالي من خلال مقارنة نتائجه مع نتائج النموذج المقدم من قبل(1989) Kaluarachchi and Parker. أن النموذج الحالي ممكن آن يستخدم لوصف مختلف المسائل بشكل جيد. أن اكبر تشبع بالملوث يحدث اسفل مصدر التلوث خلال التسرب في حين تكون هذه القيمة في مقدمة جبهة الملوث خلال اعادة التوزيع.

KEYWORDS: Multiphase, Unsaturated, Saturated, Modeling And Contaminant

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INTRODUCTION

Groundwater contamination due to surface spills or subsurface leakage of Light Non-Aqueous Phase Liquids (LNAPLs) such as hydrocarbon fuels, organic solvents, and other immiscible organic liquids is a widespread problem which poses a serious threat to groundwater resources. These compounds have some acute and long-term toxic effects. As these compounds are migrated through unsaturated-saturated zone, they will pollute great extents of soil and groundwater. This represents a major environmental problem. According to the Environmental Protection Agency (EPA) records, there are 1.8 million underground storage tanks which are in use in the United States. According to EPA estimates 280000 tanks are leaking, from which more than 20% are discharging their contents

directly to the groundwater (El-Kadi, 1992). Light Non-Aqueous Phase Liquids (NAPLs) are organic fluids that are only slightly miscible with water where the "L" stands for "lighter" than water, i.e., less denser than water.

As the LNAPL (or oil) migrates, the quantity of mobile oil decreases due to the residual oil left behind. If the amount of oil spilled is small, all of the mobile oil will eventually become exhausted and the oil will percolate no further. The column of oil is immobile and never reaches the capillary fringe unless it is displaced by water from a surface source. However, if the quantity of oil spilled per unit surface area is large, mobile oil will reach the water table. Depending on the nature of the spill, a mound of the oil will develop and spread laterally. Fig.1 is a pictorial conceptualization of a subsurface area to which LNAPL is introduced from an oil source, resulting in contamination of the unsaturated and saturated zones. The LNAPL plume (Fig.1)through the unsaturated zone and it forms a free-product mound floating on the water table. This mound will spread laterally and move in the direction of decreasing hydraulic gradient until it reaches residual and can travel no further (Kim and Corapcioglu, 2003).

Floating LNAPL can partly be removed by using a pumping well. The LNAPL will flow towards the well facilitated by the water table gradient and can be pumped into a recovery tank. The movement of oil through unsaturated and saturated zones will accompanied with leaving the residual droplets (or ganglia) in the pore spaces between the soil particles. This remaining oil may persist for long periods of time, slowly dissolving into the water and moving in the water phase through advection and dispersion. In the unsaturated zone, residual oil as well as oil dissolved in the water phase may also volatilize into the soil gas phase. In the absence of significant pressure and temperature gradients in the soil gas phase, vapors less dense than air may rise to the ground surface, while those more dense than air may sink to the capillary fringe, leading to increased contamination of the saturated zone. Once in the groundwater system, estimates of oil plume migration over time is necessary in order to design an efficient and effective remediation program. Groundwater modeling serves as a quick and efficient tool in setting up the appropriate remediation program. In many regulatory jurisdictions the use of the liquid phase contaminant in an environmental field setting is prohibited, and numerical modeling is therefore often the only practical alternative in studying the field-scale behaviour of these compounds (Kim and Corapcioglu, 2003).

A number of multiphase flow models in the contaminant hydrology literature have been presented. Faust (1985) presented an isothermal two-dimensional finite difference simulator. It describes the simultaneous flow of water and NAPL under saturated and unsaturated conditions. Abriola and Pinder (1985) formulated a one-dimensional finite difference model which included immiscible organic flow, water flow, and equilibrium inter-phase transfer between the immiscible organic phase, the water phase, and a static gas phase as cited by Sleep and Sykes (1989). Faust et al. (1989) developed model that might be used to three-dimensional two-phase transient flow system based on finite difference formulation. The governing equations were cast in terms of non-wetting fluid (NAPL) pressure and water saturation. Similarly, Parker et al. and Kuppusamy et al. as cited by (Suk, 2003) developed a two-dimensional multiphase flow simulator involving three immiscible fluids: namely, air, water and NAPL with the assumption of constant air phase pressure. Kaluarachchi and Parker (1989) applied a two-dimensional finite element model named MOFA-2D for three phases, multicomponent, isothermal flow and transport by allowing for interphase mass exchange but assuming gas phase pressure gradients are negligible.

The present study is aimed to develop a verified multiphase, one-dimensional, finite-difference numerical simulator which tracks the percentage of LNAPL saturation as well as the lateral and vertical position of the LNAPL plume in the subsurface at the specified times with different types of boundary conditions. The present model is formulated in terms of two primary unknowns: wetting

phase pressure and non-wetting phase pressure. It is tested on a simple hypothetical problem adopted by Kaluarachchi and Parker (1989).

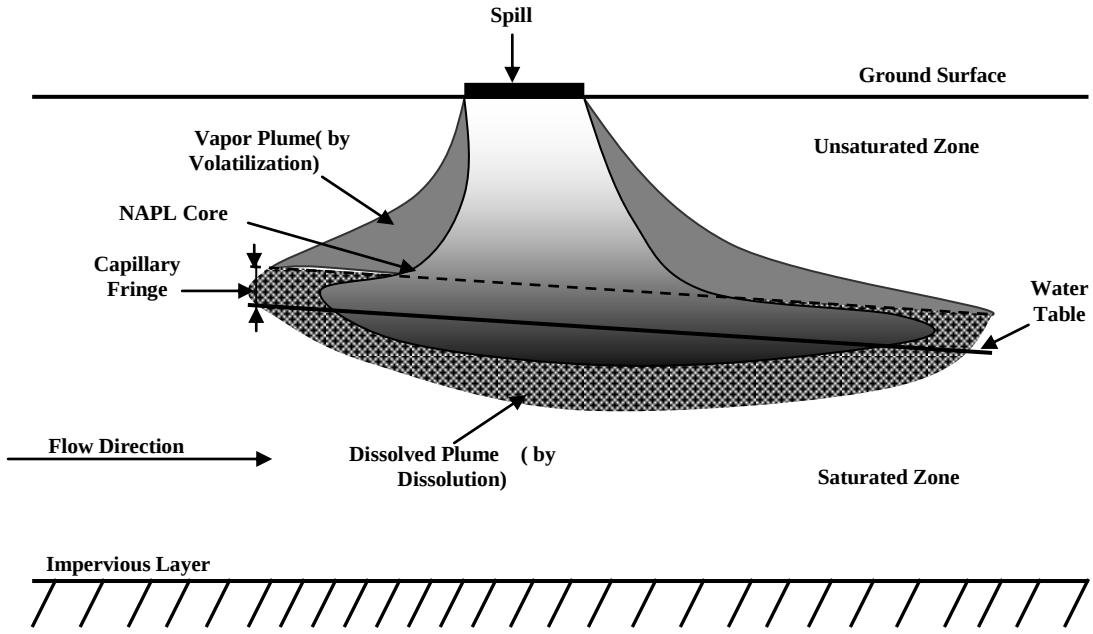

Fig.1: Conceptualization representation of LNAPL migration and contamination of the subsurface.

GOVERNING EQUATIONS

The unsaturated zone is a multiphase system, consisting of at least three phases: a solid phase of the soil matrix, a gaseous phase and the water phase. Additional phase may also be present such as a separate phase organic liquid. In the air/oil/water system of the vadose zone, oil is the wetting phase with respect to air on the surface of the water enveloping the soil grains and the water is the wetting phase with respect to oil on the soil grain surfaces. The movement of a non-aqueous phase liquid through the unsaturated-saturated zone may be represented mathematically as a case of two-phase flow because the air phase equation can be eliminated by the assumption that the air phase remains constant essentially at atmospheric pressure and consequently the pressure gradients in the air phase are negligible (Faust et al., 1989). Also, assuming that there is no component partitioning between liquid phases (Kueper and Frind, 1991).The mass balance equation for each of the fluid phase in cartesian coordinates can be written as (Bear, 1972): -

$$-\frac{\partial}{\partial x_i}\left(\rho_f \cdot q_f\right)+Q_f=\frac{\partial}{\partial t}\left(\phi \cdot \rho_f \cdot S_f\right) \tag{1}$$

Where f Subscript denoting the phase the equation applies. In the present study, f will refer to water and oil unless otherwise noted, ϕ porosity of the medium, ρ_f density of phase f [M][L^{-3}], S_f saturation of phase f [L^3/L^3], q_f volumetric flux (or Darcy flux) of phase f [L][T^{-1}], Q_f source or sink of phase f and t is the time [T]. Darcy's law is an empirical relationship that describes the relation between the flux and the individual phase pressure. Since its discovery last century it has been derived from the momentum balance equations (Kueper and Frind, 1991).

$$q_f = -\frac{k_{ij} \cdot k_{rf}}{\mu_f}\left(\frac{\partial \mathrm{P}_f}{\partial x_j} + \rho_f \cdot g \frac{\partial z}{\partial x_j}\right) \tag{2}$$

Where k_{ij} is the intrinsic permeability tensor of the medium [L^2], k_{rf} is the relative permeability of the phase f which is a function to either water (wetting phase) or oil (non-wetting phase). The relative permeability is a non-linear function of saturation. It ranges in value from 0 when the fluid is not present, to 1 when the fluid is presented, μ_f dynamic viscosity of fluid f [M][L^{-1}][T^{-1}], P_f fluid pressure of phase f [M][L^{-1}] [T^{-2}], g acceleration due to gravity vector [L][T^{-2}]. Darcy's law can be written equivalently in the form of the pressure head as below:

$$q_f = -\mathrm{K}_{fij}\left(\frac{\partial h_f}{\partial x_j} + \frac{\rho_f}{\rho_w} \cdot \frac{\partial z}{\partial x_j}\right) \tag{3}$$

With $\frac{\partial z}{\partial x_j}$ is a unit gravitational vector, where z is elevation and t is time. The volumetric flux can be thought of as the volume of fluid f passing through a unit area of porous medium in a unit time. This variable is the natural one when making mass fluid balance arguments. By substituting (eq.(3) into eq.(1)), assuming that the fluid and the porous media are incompressible, ignore the source-sink term, and according to Kaluarachchi and Parker (1989), the coordinate system is oriented with the conductivity tensor, or otherwise that off-diagonal components may be disregarded, so that $\mathrm{K}_{fsij} = 0$ for $i \neq j$. the resulting equation can be represented by: -

$$\frac{\partial}{\partial x}\left[\mathrm{K}_f\left(\frac{\partial h_f}{\partial x} + \frac{\rho_f}{\rho_w}\frac{\partial z}{\partial x}\right)\right] = \phi \frac{\partial S_f}{\partial t} \tag{4}$$

ONE-DIMENSIONAL NUMERICAL SOLUTION

Eq.4 will be re-written as two one-dimensional equations, one for water phase and the other for oil phase. Both of which are expressed in terms of the phase pressure head as below:-

$$\frac{\partial}{\partial z}\left[\mathrm{K}_w\left(\frac{\partial h_w}{\partial z} + 1\right)\right] = C_w \frac{\partial h_w}{\partial t} \tag{5}$$

$$\frac{\partial}{\partial z}\left[\mathrm{K}_o\left(\frac{\partial h_o}{\partial z} + \rho_{ro}\right)\right] = C_o \frac{\partial h_o}{\partial t} \tag{6}$$

Where z is positive upward vertical coordinate [L], $\mathrm{K}_w = \mathrm{K}_{ws} k_{rw}$ and $\mathrm{K}_o = \mathrm{K}_{os} k_{ro} = \frac{\mathrm{K}_{ws} k_{ro}}{\mu_{ro}}$; μ_{ro} is the ratio of oil to water viscosity, ρ_{ro} is the ratio of oil to water density and C is the specific fluid capacity, It is defined by $C_w = \phi \frac{\partial S_w}{\partial h_w}$ & $C_o = \phi \frac{\partial S_o}{\partial h_o}$.

However, in the present study, the two-pressure forms of the flow equations are cast in terms of the two fluid pressure heads. The equations in this form are a direct statement of conservation of mass. There are two principle difficulties associated with solving the governing equations (eqs.(5) &(6)). The first is that these equations are first order partial differential equations with respect to time and second order with space. Further, they are nonlinear differential equations because **C** and **K** are nonlinear functions of capillary pressure heads of two fluids. The second difficulty in solving these equations lies in the linearization procedure and the method for dealing with the coupling between the

equations. However, the solution techniques used here consisted of a finite-difference approximation (implicit method) of the differential equations, the Newton-Raphson with a Taylor series expansion to treat the nonlinearities, and direct matrix solution (Gauss-Elimination method). These techniques result in the following equations:-

$$a_{fi}^{k}\delta_{fi-1}^{k+1}+b_{fi}^{k}\delta_{fi}^{k+1}+c_{fi}^{k}\delta_{fi+1}^{k+1}=-r_{fi}^{k} \tag{7}$$

Where:-

$$a_{fi}^{k}=\left[-R1\left(\mathrm{K}_{fi-1}^{k}+h_{fi-1}^{k}\frac{\partial \mathrm{K}_{fi-1}^{k}}{\partial h_{fi-1}^{k}}\right)-R2\rho_{rf}\frac{\partial \mathrm{K}_{fi-1}^{k}}{\partial h_{fi-1}^{k}}\right]$$

$$b_{fi}^{k}=\left[C_{fi,j}^{k}+\left(h_{fi,j}^{k}-h_{fi,j}^{n}\right)\frac{\partial C_{fi,j}^{k}}{\partial h_{fi,j}^{k}}\right]+\left[R1\mathrm{K}_{fi-1,j}^{k}\right]+\left[R11\left(\mathrm{K}_{fi,j}^{k}+h_{fi,j}^{k}\frac{\partial \mathrm{K}_{fi,j}^{k}}{\partial h_{fi,j}^{k}}\right)\right]+\left[R2\rho_{rf}\frac{\partial \mathrm{K}_{fi,j}^{k}}{\partial h_{fi,j}^{k}}\right]$$

$$c_{fi}^{k}=-R11\mathrm{K}_{fi}^{k}$$

$$r_{fi}^{K}=C_{fi}^{k}\left(h_{fi}^{k}-h_{fi}^{n}\right)-R1\mathrm{K}_{fi-1}^{k}h_{fi-1}^{k}+R1\mathrm{K}_{fi-1}^{k}h_{fi}^{k}+R11\mathrm{K}_{fi}^{k}h_{fi}^{k}-R11\mathrm{K}_{fi}^{k}h_{fi+1}^{k}-R2\rho_{rf}\mathrm{K}_{fi-1}^{k}+R2\rho_{rf}\mathrm{K}_{fi}^{k}$$

$$R1=\frac{\Delta t}{\Delta z_i\left(z_{i-1}-z_i\right)}\quad,\quad R11=\frac{\Delta t}{\Delta z_i\left(z_i-z_{i+1}\right)}\quad\&\quad R2=\frac{\Delta t}{\Delta z_i}.$$

Where r_{fi}^{k} is the residual in node (i) at iteration k. At each time step the solution is iterated until the pressure head converges. During each iteration, the equations are solved to find the pressure heads at the new iteration level. Thus, the nonlinear coefficients (i.e. C & K) are evaluated using the pressure heads at the old iteration level thus linearizing the equations. The increment in pressure head at each iteration level must be added to the tried solution at iteration k, this increment is termed as $\delta^{n+1,k+1}$, to produce the solution to the next iteration, $k+1$ and next time step, $n+1$. Fig.2(a) shows unsaturated-saturated zone and Fig.2(b) shows the discretized domain in the z-direction with the time.

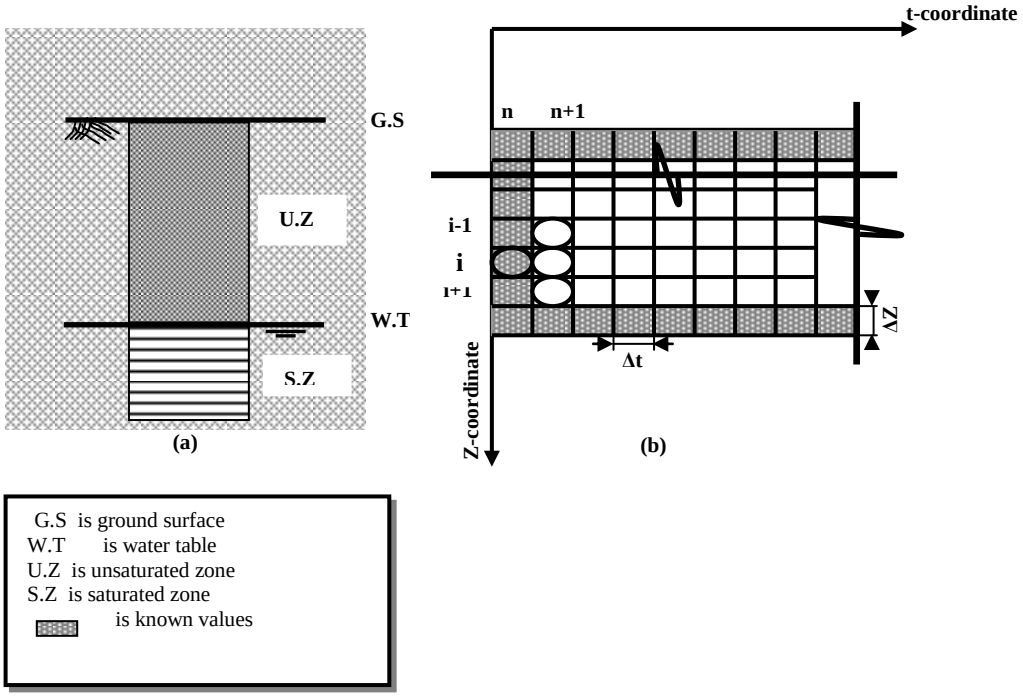

Fig.2: (a) The common situation to the virtual solution column, (b) image to the solution domain.

The coefficients a, b and c in eq. (7) are associated with δ_{i-1}^{k+1} , δ_i^{k+1} & δ_{i+1}^{k+1}, respectively, and in general it is formed as:-

$$A^k \delta^{k+1} = -r^k \tag{8}$$

Where A is the coefficient matrix for the linearized system. For each node (i), there is one linear equation in three variables δ_{i-1}^{k+1} , δ_i^{k+1} & δ_{i+1}^{k+1}. The collection of equations for each nodal solution leads to have a global tri-diagonal coefficient matrix (its bandwidth equal to three) to whole solution domain. For example, in a ten nodded domain, the soil column is subdivided into ten vertical grids. Thus, there will be ten equations that form the whole grids. These equations can be written, after application of the boundary and initial conditions, in matrix notation as:-

$$\begin{bmatrix} b_1 & c_1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ a_2 & b_2 & c_2 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & a_3 & b_3 & c_3 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & a_4 & b_4 & c_4 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & a_5 & b_5 & c_5 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & a_6 & b_6 & c_6 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & a_7 & b_7 & c_7 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & a_8 & b_8 & c_8 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & a_9 & b_9 & c_9 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & a_{10} & b_{10} \end{bmatrix}_k \begin{bmatrix} \delta_1 \\ \delta_2 \\ \delta_3 \\ \delta_4 \\ \delta_5 \\ \delta_6 \\ \delta_7 \\ \delta_8 \\ \delta_9 \\ \delta_{10} \end{bmatrix}_{k+1} = -\begin{Bmatrix} r_1 \\ r_2 \\ r_3 \\ r_4 \\ r_5 \\ r_6 \\ r_7 \\ r_8 \\ r_9 \\ r_{10} \end{Bmatrix}_k$$

This system is solved for δ and then the algorithm enters the next iteration with new values of h_f is evaluated as follows:-

$$h_{fi}^{k+1} = h_{fi}^k + \omega_f^{k+1} \delta_{fi}^{k+1} \tag{9}$$

Where ω_f^{k+1} is a damping parameter as mentioned by Cooley (1983). During each iteration, nodal pressure heads need to be updated for the next iteration. With highly nonlinear flow problems, the updating method introduced by Cooley (1983) which introduces an optimal relaxation scheme, which accounts for the maximum convergence error for entire mesh, is used in the present study in conjunction with Newton–Raphson scheme. For clarification, this method is briefly described as follows:-

Let e_f^{k+1} be the largest in absolute value of the δ_i^{k+1} values for all i. Then

Step(1):

$$s_f = \frac{e_f^{k+1}}{\omega_f^k e_f^k} \quad \text{for} \quad k > 0 \tag{10}$$

$$s_f = 1 \quad \text{for} \quad k = 0$$

Step(2):

$$\omega_f^* = \frac{3 + s_f}{3 + |s_f|} \quad \text{for} \quad s_f \geq -1 \tag{11}$$

$$\omega_f^* = \frac{1}{2|s_f|} \quad \text{for} \quad s_f < -1$$

Step(3):

$$\omega_f^{k+1} = \omega_f^* \quad \text{for} \quad \omega_f^* \left| e_f^{k+1} \right| \le e_{f\max} \tag{12}$$

$$\omega_f^{k+1} = \frac{e_{f\max}}{\left| e_f^{k+1} \right|} \quad \text{for} \quad \omega_f^* \left| e_f^{k+1} \right| > e_{f\max}$$

Where $e_{f\max}$ is the maximum allowable change in the fluid pressure head, h_f, during any iteration. This value is chosen beforehand. The convergence criterion used in the present study for a given phase f (=o,w) is as follows:-

$$\frac{\max.\left|\delta_{fi}^{k+1}\right|}{\max.\left|h_{fi}^{k} + \delta_{fi}^{k+1}\right|} \le \varepsilon \tag{١٣}$$

Where ε is a small number termed the convergence tolerance. A typical convergence criterion for pressure head is 0.001 or less.

When the Global finite difference approximation is founded by summing the node equations, the boundary terms must be accounted. Two types of boundary condition are considered. For Type-1 (Dirichlet), or fixed head boundary condition, the finite difference equation at boundary node (i) is replaced by $\delta_{fi,j}^{k+1} = zero$ or $h_{fi,j}^{k+1} = given$. The other form of boundary condition is Type-2 (Neumann), or specified fluid flux condition. A flux boundary condition is incorporated into the global approximation at boundary node (i) by using the discretized finite difference form of continuity equation in conjunction with Darcy's law.

CONSTITUTIVE RELATIONSHIPS

Eqs.(5) and (6) describe the flow conditions in the subsurface system. There are several unknowns in these equations. In order to close the system, constitutive relationships that relate the unknowns must be specified. These relationships can be written in a variety of ways that result in different variables becoming the dependent variables for the system.

The most common choices for dependent variables are the individual phase pressures and the phase saturations. The constitutive relationships that will be used in the present study are the pressure–saturation and relative permeability–saturation relationships. The fluid saturation is a function of the difference between the pressure of the two fluids in the porous medium, the pressure difference is called the capillary pressure , $P_{ow} = P_o - P_w$, or capillary pressure head $h_{ow} = h_o - h_w$.

Many different functional forms have been proposed to describe the pressure–saturation and relative permeability–saturation relationships. They are generally empirical relations. However, constitutive relationships used in the present study to describe three phase fluid relative permeabilities and saturations as functions of fluid heads described by (Parker et. al., 1987) which is based on (Van Genuchten's model, 1980).The following relationships will be needed to complete the description of a multiphase flow through the porous media: -

$$S_w + S_o + S_a = 1 \tag{14}$$

$$S_t = S_w + S_o \tag{15}$$

$$\bar{S}_w = \frac{S_w - S_r}{1 - S_r} \tag{16}$$

$$\bar{S}_t = \frac{S_t - S_r}{1 - S_r} \tag{17}$$

$$\bar{S}_w = \left[1 + (\alpha \cdot \beta_{ow} \cdot h_{ow})^n\right]^{-m} \qquad h_o > h_o^{cr.} \tag{18}$$

$$\bar{S}_w = \left[1 + (\alpha \cdot h_{aw})^n\right]^{-m} \qquad h_o \le h_o^{cr.} \tag{19}$$

$$h_o^{cr.} = \beta_{ow} h_w \Big/ (\beta_{ao} + \beta_{ow}) \tag{20}$$

$$\bar{S}_t = \left[1 + (\alpha \cdot \beta_{ao} \cdot h_{ao})^n\right]^{-m} \tag{21}$$

$$k_{rw} = \bar{S}_w^{1/2}\left[1 - \left(1 - \bar{S}_w^{1/m}\right)^m\right]^2 \tag{22}$$

$$k_{ro} = (S_t - S_w)^{1/2}\left[\left(1 - \bar{S}_w^{1/m}\right)^m - \left(1 - \bar{S}_t^{1/m}\right)^m\right]^2 \tag{23}$$

Where $h_o^{cr.}$ critical oil pressure head, [L], h_{ow} oil-water capillary pressure head $(= h_o - h_w)$, [L], h_{aw} air-water capillary pressure head $(= h_a - h_w)$, [L], h_{ao} air-oil capillary pressure head $(= h_a - h_o)$, [L], S_w water saturation, S_a air saturation, S_t total liquid saturation, $\bar{S}_w$ effective water saturation, $\bar{S}_t$ effective total liquid saturation, k_{rw} relative permeability of water, k_{ro} relative permeability of oil, S_r residual or irreducible saturation of water phase. Here α, n and $m\left(=1-1/n\right)$ are Van Genutchten's soil parameters, β_{ao} & β_{ow} are fluid-dependent scaling coefficients.

NUMERICAL RESULTS IN ONE DIMENSION AND DISCUSSION

A theoretical work presented by Kaluarachchi and Parker (1989) used as a verification to the present numerical model. They used Galerkin's finite element method for modeling of one-dimensional infiltration and redistribution of oil in a uniform soil profile. Also, they used a number of methods to determine the capacity terms related to oil and water phases. These methods are the chord-slope scheme and equilibrium scheme.

The problem analyzed here corresponds to a vertical soil column 100 cm long with an oil-free initial condition in equilibrium with a water table located 75 cm below the top surface. The simulation was achieved in two stages. The first stage is the infiltration stage, in which oil was allowed to infiltrate into the column under water equivalent oil pressure head of 3 cm until a total of 5 cm^3/cm^2 of oil had accumulated. The second stage is the redistribution stage, in which the source of oil is cutoff and oil is allowed to redistribute up to 100 hours. A schematic diagram of the problem is illustrated in Fig.3. The boundary conditions and system's parameters, i.e. fluid and soil properties which were used in this simulation are summarized in Tables (1) and (2) respectively.

The initial condition of zero oil saturation was achieved by fixing the initial oil pressure head at each node to the critical oil pressure head, $h_o^{cr.}$ which is defined in eq. (20). The water saturation distribution above the water table that is used in this simulation is initially at capillary equilibrium. This distribution is illustrated in Fig.4. The finite difference mesh consists of 100 grids with a uniform spacing of 1 cm and the time step varied between 0.00001 to 0.01 hours.

As pointed out by Kaluarachchi and Parker (1989), a jump condition in the water saturation versus air-water capillary pressure head function, $S_w(h_{aw})$, will occur during the transition from a two-phase air-water system to a three-phase air-oil-water system. To avoid numerical problems associated with this jump condition, a phase updating scheme is adopted at the end of each time step to index whether the node is a two or three phase system (i.e. oil is absent or present). Once a three-phase condition occurs, reversion to a two-phase condition is not allowed. The criterion for a node to change from the two to the three phase system is that $h_o > h_o^{cr.}$. It is important to note that the part of the domain remaining as an air-water system and consequently the capacity and relative permeability terms related to the oil phase C_o and k_{ro} will become zero and the oil flow equation solution reduce to the identity 0=0. To avoid this problem, minimum cutoff values of capacity and relative permeability terms related to the oil phase should be taken as $C_o = 10^{-6}$ and $k_{ro} = 10^{-6}$ as recommended by Kaluarachchi and Parker (1989).

The duration of the infiltration stage this is calculated from the present model was approximately 0.085 hours. While this value was 0.09 hours as calculated by MOFAT-2D model. There is a good agreement between these values. The total liquid saturation distribution at the end of the infiltration stage as computed from the present model are compared with those calculated by MOFAT-2D model by using chord-slope scheme and there is a good agreement between them as shown in Fig.4. The water saturation distribution at the end of the infiltration stage was identical to initial condition.

Saturation distributions at the end of 100 hours of redistribution are illustrated in Fig.5 and Fig.5. These results are compared with those of results MOFAT-2D model by using chord-slope scheme and equilibrium condition scheme, and, as shown in these figures, there is a good agreement between these results. Equilibrium distributions were calculated from hydrostatics for an oil volume of 5 cm subject to the imposed boundary conditions. The equilibrium condition was defined by Kaluarachchi and Parker (1989) as the fluid distributions at which the total head gradient of both phases with respect to elevation approaches zero. Kaluarachchi and Parker (1989) pointed out that convergence of chord-slope results toward the equilibrium results provides a verification check for the MOFAT-2D numerical model. It is to be noted, however, that whereas the equilibrium distribution indicates no oil above a depth of 43.0 cm, the MOFAT-2D numerical model, and the present model predict an average oil saturation of (7-8)% remaining above this depth even though oil velocities at 100 hours are practically zero.

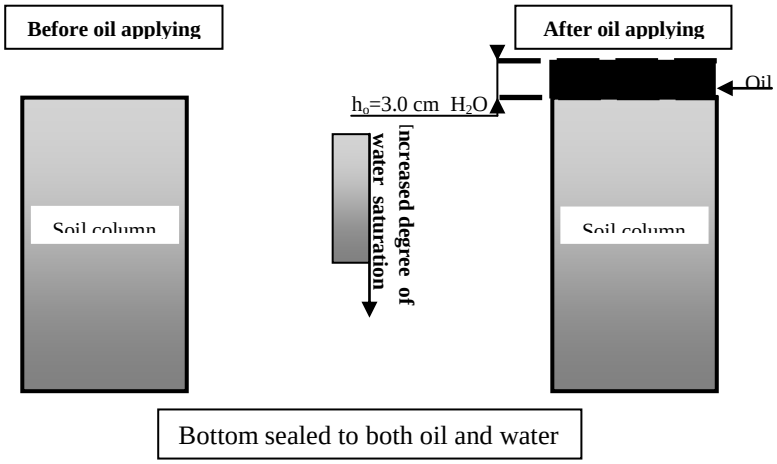

Fig.3: A **schematic diagram for the analyzed problem.**

Table1: Boundary conditions which are used for verification.

Stage	Phase	Boundary conditions	
		Upper boundary	lower boundary
Infiltration	Water	Zero flux	Constant head= 25.0 cm
	Oil	Constant head= 3.0 cm	Zero flux
Redistribution	Water	Zero flux	Constant head= 25.0 cm
	Oil	Zero flux	Zero flux

Table 2: Soil and fluid properties that are used for first verification.

Parameter	n	α	S_r	K_{sw}	β_{ao}	β_{ow}	ρ_{ro}	μ_{ro}	ϕ
Value	3.25	0.05	0.00	50.00	1.80	2.25	0.80	2.00	0.40

All units are given in centimeters and hours.

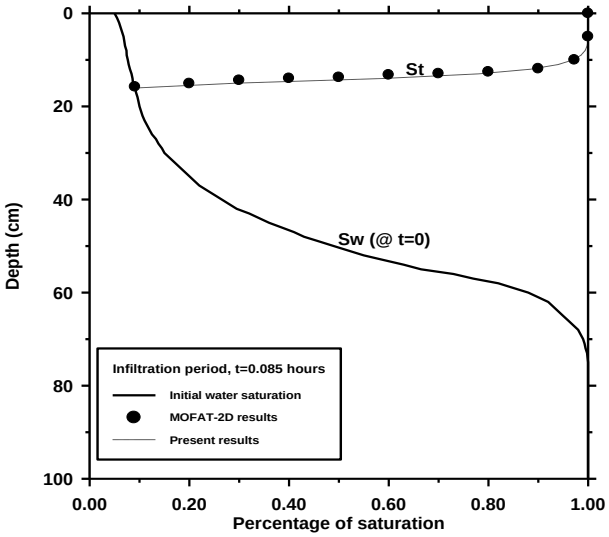

Fig.4: Distribution of initial water saturation and total liquid saturation at the end of the infiltration period.

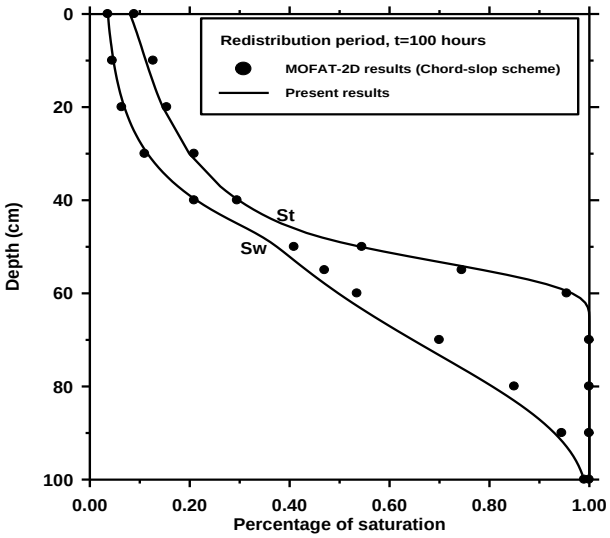

Fig.5: Distribution of water and total liquid saturation at the end of the redistribution period.

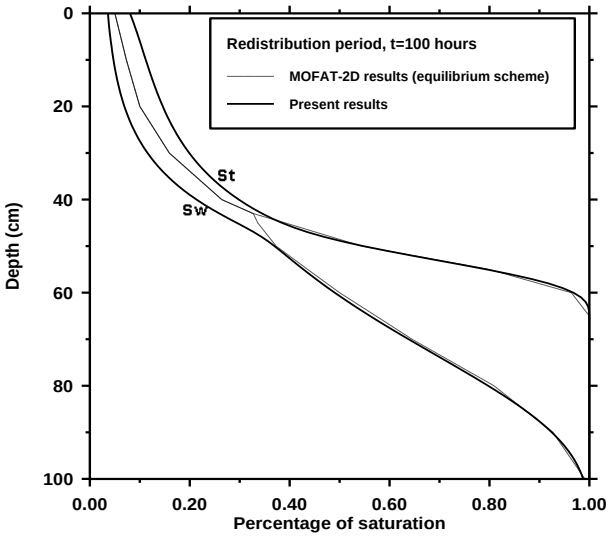

Fig.6: Distribution of water and total liquid saturation at the end of the redistribution period.

The mass balance associated with the above results shows an error in the oil phase during the infiltration stage is largest at the early time (up to 1%) and reduce to less than 0.0005% later times (Fig.7). The present mass balance results can be compared to an analogous lumped finite element solution in MOFAT-2D using the h-based form of the governing equations. The mass balance error associated with results of MOFAT-2D is greater than 1% at any time during the infiltration stage. This is because the finite element approximation may suffer from oscillatory solutions. Such oscillations are not present in any finite difference solutions. Because the only difference between the two solution producers as disscused by Celia et al. (1990) is the treatment of the time derivative term, these results imply that diagonalized time matrices are to be preferred. Thus the unsaturated flow equation is one that benefits from mass lumping in finite element approximation.

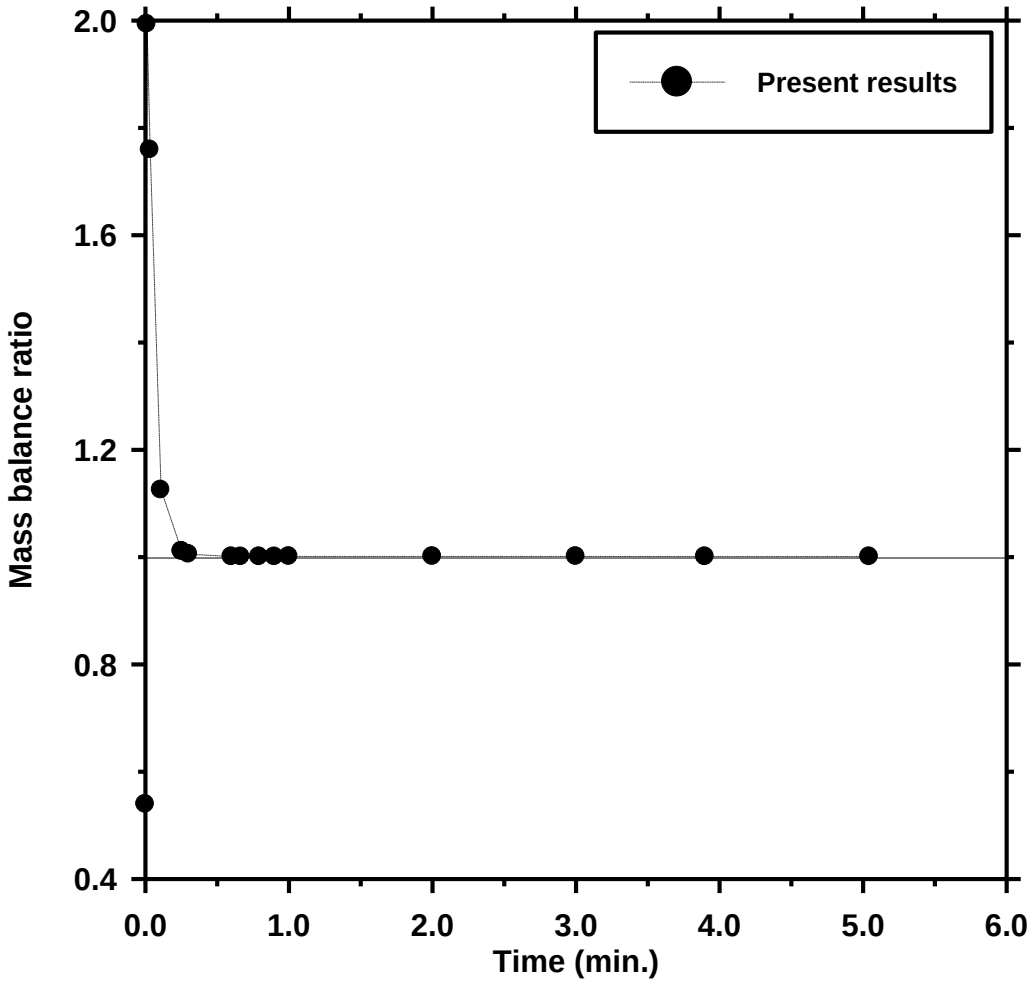

Fig.7: Mass balance of oil as a function of time, using the h-based form of equations.

MODEL IMPLEMENTATION

A computer program written in DIGITAL VISUAL FORTRAN (Version 5.0) was developed to implement the model described above. Inherent in any subsurface modeling algorithms are assumptions and limitations. The major assumptions include:- the pressure in the air phase is constant and equal to atmospheric pressure, both water and NAPL viscosities and densities are pressure independent, relative permeability of water is a function of water saturation, relative permeability of NAPL is a function of air and water saturations, capillary pressure is a function of water saturation, air saturation is a function of NAPL pressure, Darcy's equation for multiphase flow is valid, intrinsic permeability is a function of space and there is no inter-phase mass transfer (i.e ; the NAPL is truly immiscible in water).

The major limitations include:- the model can not treat highly pressurized systems in which the viscosity and density of the three phases are a function of pressure, fractured systems are not treated, transport of dissolved NAPL is not treated.

CONCLUSIONS

The following conclusions can be deduced :-

(**1**) The numerical solution based on the potential form of the governing equations with techniques consisted of Implicit Finite Difference, Newton-Raphson and Gauss-Elimination schemes showed to be an efficient procedure in solving one- dimensional water and LNAPL flow through the unsaturated-saturated zone in three fluid phase's system.
(**2**) The maximum LNAPL saturation occurred below the source of the contaminant during LNAPL infiltration. During redistribution, the LNAPL saturation had a maximum value at the advancing of the LNAPL infiltration front.
3) Mass balance associated with the results presented above show that the finite element approximation may suffer from oscillatory solutions. Such oscillations are not present in any finite difference solutions.

REFERENCES

Bear, J., "Dynamics of fluids in porous media". Elsevier, New York, 1972.

Cary, J.W., J.F. McBride, and C.S. Simmons, "Observations of water and oil infiltration into soil: Some simulation challenges". Water Resources Research, 25(1), 73-80, 1989.

Celia, M.A., E.T. Bouloutas, and R.L. Zarba, "A general mass-conservation numerical solution for the unsaturated flow equation". Water Resources Research, 26(7), 1483-1496, 1990.

Cooley, R.L., "Some new procedures for numerical solution of variably saturated flow problems". Water Resources Research, 19(5), 1271-1285, 1983.

El-Kadi, A.I., "Applicability of sharp-interface models for NAPL transport: 1. infiltration". Ground Water, 30(6), 849-856, 1992.

Faust, C.R., "Transport of immiscible fluids, within and below the unsaturated zone : A numerical model". Water Resources Research, 21(4), 587-596, 1985.

Faust, C., J. Guswa, and J. Mercer, "Simulation of three-dimensional flow of immiscible fluids within and below unsaturated zone". Water Resources Research, 25(12), 2449-2464, 1989.

Kaluarachchi, J.J., and J.C. Parker, "An efficient finite element method for modeling multiphase flow". Water Resources Research, 25, 43-54, 1989.

Kim, J., and M.Y. Corapcioglu, "Modeling dissolution and volatilization of LNAPL sources migrating on the groundwater table". J. of Contaminant Hydrology, 65, 137-158, 2003.

Kueper, B., and E. Frind, "Two-phase flow in heterogeneous porous media: 1. Model development". Water Resources Research, 27(6), 1049-1057, 1991.

Kueper, B., and E. Frind, "Two-phase flow in heterogeneous porous media: 1. Model application". Water Resources Research, 27(6), 1059-1070, 1991.

Parker, J. C., and R. J. Lenhard, "A Model for hysteretic constitutive relations governing multiphase flow: 1. saturation-pressure relations". Water Resources Research, 23(12), 2187-2196, 1987.

Parker, J. C., R. J. Lenhard, and T. Kuppusamy, "A parametric model for constitutive properties governing multiphase flow in porous media". Water Resources Research, 23(4), 618-624, 1987.

Marsman, A., "The influence of water percolation on flow of light non-aqueous phase liquid in soil". PH.D. Thesis, Wageningen University, Wageningen 2002.

Sleep, B.E., and J.F. Sykes, "Modeling the transport of volatile organics in variable saturated media". Water Resources Research, 25(1), 81-92, 1989.

Suk, H., "Development of 2- and 3-D simulator for three phase flow with general initial and boundary conditions on the fractional flow approach". PH.D. Thesis, Pennsylvania State University, 2003.

Van Genuchten, M., "A closed-form equation for predicting the hydraulic conductivity of unsaturated soils''. Soil Sci. Soc. Am. J.,44,892-898,1980.

SYMBOLS

A	Coefficient matrix	$[M^0 L^0 T^0]$
C_f	Specific fluid capacity	$[M^0 L^0 T^0]$
$e_{f\max}$	Maximum change in the fluid pressure head	[L]
g	Acceleration due to gravity vector	$[L T^{-2}]$
h_a	Air pressure head	[L]
h_{ao}	Air-oil capillary pressure head	[L]
h_{aw}	Air-water capillary pressure head	[L]
h_o	Oil pressure head	[L]
h_{ow}	Oil-water capillary pressure head	[L]
$h_o^{cr.}$	Critical oil pressure head	[L]
h_w	Water pressure head	[L]
i	Grid identification in Z coordinates	$[M^0 L^0 T^0]$
K	Hydraulic conductivity	$[L T^{-1}]$
K_{fs}	The conductivity when the medium is saturated with fluid f	$[L T^{-1}]$
k_r	Relative hydraulic conductivity	$[M^0 L^0 T^0]$
k	The intrinsic permeability tensor of the medium in eq. (2)	$[M^0 L^0 T^0]$
k	Iteration index	$[M^0 L^0 T^0]$
n	Time step identification (if it is superscript)	$[M^0 L^0 T^0]$
non	The n^{th} grid identification	$[M^0 L^0 T^0]$
nor	The number of rows	$[M^0 L^0 T^0]$
n, m	Van Genuchten's soil parameters	$[M^0 L^0 T^0]$
P_f	Fluid pressure of phase f	$[M L^{-1} T^{-2}]$
Q_f	Source or sink of phase f	$[M L^{-3} T^{-1}]$
q	Volumetric flux (or Darcy's flux)	$[L T^{-1}]$
r	The residual due to approximation	$[M^0 L^0 T^0]$
S_a	Degree of air saturation	%
S_o	Degree of oil saturation	%
S_r	Degree of residual wetting fluid saturation	%
S_t	Degree of total liquid saturation	%

S_w	Degree of water saturation	%
$\overline{S}_t$	Degree of effective total liquid saturation	%
$\overline{S}_w$	Degree of effective water saturation	%
t	Time coordinate	[T]

Greek symbols

α	Van Genuchten's soil parameter	$[L^{-1}]$
β_{ij}	Fluid-dependent scaling coefficient	$[M^0 L^0 T^0]$
δ	The difference between the approximation and exact solution	[L]
ε	Convergence tolerance	[L]
μ_f	Dynamic viscosity of fluid f	$[M\ L^{-1} T^{-1}]$
μ_{ro}	Ratio of oil to water viscosity	$[M^0 L^0 T^0]$
ρ_f	Density of phase f	$[M L^{-3}]$
ρ_{ro}	Ratio of oil to water density	$[M^0 L^0 T^0]$
ρ_w	Density of water at standard temperature and pressure	$[M L^{-3}]$
ϕ	Porosity of the medium	$[L^3 L^{-3}]$
ω	Damping parameter	$[M^0 L^0 T^0]$
Δt	Time step size	[T]
ΔZ	Vertical increment in Z-direction	[L]