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# Unsaturated-Zone Leaching and Saturated-Zone Mixing Model in Heterogeneous Layers

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**Summary.** A screening level model was developed for simulation of pollutant migration through the unsaturated-zone and subsequent mixing within the saturated-zone. This one-dimensional finite difference model simulates the transport processes of liquid-phase advection, liquid- and vapor-phase dispersion, sorption, and decay of the contaminant. The model estimates contaminant concentration in the saturated-zone by using a simple mass balance technique for mixing of the unsaturated-zone leachate with the groundwater. The model can be a useful tool in making preliminary assessments of the potential impacts of contaminants in the subsurface. The model can handle vertical heterogeneity of the soil columns and non-uniform initial contaminant concentration. It was verified by comparing its simulation results to an analytical solution and laboratory soil column experiments. In addition the model was validated against laboratory experiments with three different soil sample sizes of Ottawa quartz sand and 480 ppm saline water as a groundwater contamination source.

**Key words:** unsaturated-zone, saturated-zone, leaching model, groundwater, modelling, GUI

## 1 Introduction

Although several computer codes, such as VLEACH, VLEACHSM 2.0, EPACML, etc. can incorporate the heterogeneity of the soil properties often many sites do not have the sufficient degree of details of field-measured data to allow the use of the existing models. Even when there is a site with a reasonable amount of field data, a rigorous parameter estimation and calibration work is often necessary before conducting a comprehensive simulation. This manuscript describes a newly developed model titled Unsaturated-Zone Leaching and Saturated-Zone Mixing Model. The model allows sufficient model simulations and estimation of contaminant concentration with a smaller degree of site sampling, analysis, and parameter estimation than the existing models. It accomplishes it by

employing a one-dimensional (1-D) finite difference scheme for solving the unsaturated-zone transport equation and uses a mass-balanced technique to estimate contaminant concentration in the saturated zone.

Ravi and Johnson (1993) developed a 1-D transport program, titled VLEACH, which handles only vertical migration of pollutant in a homogeneous soil column. Later, Lee (1996, 1999a), and Lee and In (2005) developed several versions of VLEACHSM by adding the liquid-phase dispersion, decay terms in the unsaturated zone, and the saturation zone mixing into VLEACH. This Unsaturated-Zone Leaching and Saturated-Zone Mixing Model is further improved by implementing the heterogeneous soil property and a Graphic User Interface. In addition, the Unsaturated-Zone Leaching and Saturated-Zone Mixing Model allows the specification of two different types (Dirichlet's and Cauchy's) of boundary conditions at the top of the soil column. Using a simple mass-balance technique, the saturated zone module estimates the concentration of contaminants by mixing of leachate from the unsaturated zone with groundwater.

## 2 Governing Equation and Boundary and Initial Conditions

### 2.1 Unsaturated Zone Transport

Considering the three equilibrium phases of pollutants in an unsaturated soil column, its 1-D governing transport equation can be expressed as follows:

$$\theta_w \frac{\partial C_w}{\partial t} + \theta_a \frac{\partial C_a}{\partial t} + \rho_b \frac{\partial C_s}{\partial t} = \frac{\partial}{\partial z} \left( \theta_w C_w \frac{\partial C_w}{\partial z} \right) + \frac{\partial}{\partial z} \left( \theta_a C_a \frac{\partial C_a}{\partial z} \right) - \frac{\partial}{\partial z} (q_w C_w) - \mu_w \theta_w C_w - \mu_a \theta_a C_a - \mu_s \rho_b C_s \quad (1)$$

where,  $C_w$  denotes the concentration of a contaminant in liquid (water) phase (mg/L),  $C_a$  denotes the concentration of a contaminant in vapor (air) phase (mg/L),  $C_s$  denotes the concentration of a contaminant in solid phase (mg/kg),  $\theta_w$  denotes the volumetric water content (volume of water/total volume) ( $\text{m}^3/\text{m}^3$ ),  $\theta_a$  denotes the air-filled porosity (volume of air/total volume) ( $\text{m}^3/\text{m}^3$ ),  $q_w$  denotes water flow velocity (recharge rate) (m/yr),  $D_w$  denotes dispersion coefficient for the liquid phase contaminant in the pore water ( $\text{m}^2/\text{yr}$ ).  $D_a$  denotes gaseous phase diffusion coefficient in the pore air ( $\text{m}^2/\text{yr}$ ),  $\mu_w$  denotes first order decay rate of a contaminant in water phase (1/yr),  $\mu_a$  denotes first order decay rate of a contaminant in gaseous phase (1/yr),  $\mu_s$  denotes first order decay rate of a contaminant in solid phase (1/yr),  $\rho_b$  denotes bulk density of the soil ( $\text{gr}/\text{cm}^3$ ),  $z$  denotes vertical coordinate with positive being downward, and  $t$  denotes elapsed time. Several assumptions were considered in the development of (1) as follow: The total porosity

( $n$ ) equals the sum of the water filled porosity the air filled porosity. The air flow velocity ( $q_a$ ) is assumed to be zero. For simplicity, it is also assumed that  $\mu_w = \mu_a = \mu_s = \mu$ . Instantaneous equilibrium (partitioning) of the contaminant among the phases according to the following linear relationships:

- Liquid-solid phase equilibrium is

$$C_s = K_d C_w . \tag{2}$$

- Liquid-gas phase equilibrium is

$$C_a = H C_w . \tag{3}$$

In (2) and (3),  $K_d$  (ml/g) denotes the distribution coefficient between the solid phase and liquid phase, and  $H$  (dimensionless) denotes the partition coefficient between the air phase and water phase. Using the empirical relationship,  $K_d$  can be expressed as  $K_d = K_{oc} \cdot f_{oc}$ , where  $K_{oc}$  (ml/g) denotes the organic carbon-water partition coefficient and  $f_{oc}$  (g/g) denotes the fraction organic carbon of the soil.

The dimensionless form of the Henry’s partition coefficient,  $H$ , can be determined from the more common form having the units of atmospheres-cubic meters per mole (atm-m<sup>3</sup>/mol) using the following equation

$$H = \frac{K_H}{RT} \tag{4}$$

where  $K_H$  (atm-m<sup>3</sup>/mol) denotes the dimensional form of Henry’s Law constant,  $R$  denotes the universal gas constant ( $R = 8.2 \times 10^{-5}$  atm-m<sup>3</sup>/molK), and  $T$  is the absolute temperature in Kelvin ( $^{\circ}\text{K} = 273.16 + ^{\circ}\text{C}$ ).

The dispersion coefficient in the unsaturated zone is regarded as a linear function of the pore water velocity as:

$$D_w = \alpha_L \left( \frac{q_w}{\theta_w} \right) \tag{5}$$

where  $\alpha_L$  is the longitudinal dispersivity (feet) of the unsaturated zone.

The gas phase diffusion coefficient ( $D_a$ ) in the porous medium is calculated by modifying the free air diffusion coefficient using the Millington model (1959):

$$D_a = D_{\text{air}} \frac{(n - \theta_w)^{7/3}}{n^2} \tag{6}$$

where  $D_{\text{air}}$  denotes the diffusion coefficient of the contaminant in the free air.

By substituting equation (2) and (3) into (1), the governing transport equation can be simplified as:

$$\theta \frac{\partial C_w}{\partial t} = D \frac{\partial^2 C_w}{\partial z^2} + \left( \frac{\partial D}{\partial z} - q_w \right) \frac{\partial C_w}{\partial z} - \left( \frac{\partial q_w}{\partial z} + \mu \theta \right) C_w \tag{7}$$

where  $\theta = \theta_w + \theta_a H + \rho_b K_d$  and  $D \equiv \theta_w D_w + \theta_a D_a H$ .

The initial conditions used in this work to solve equation (7) are given in the following equation

$$C_w(z, t)|_{t=0} = \begin{cases} C_s(z, 0)/K_d & \text{if } K_d > 0, \\ C_w(z, 0) & \text{if } K_d = 0, \end{cases} \quad (8)$$

where  $C_s(z, 0)$  is the initial solid-phase concentration specified by the user. When the distribution coefficient ( $K_d = K_{oc} \cdot f_{oc}$ ) is zero, liquid-phase concentration must be entered as an initial concentration to avoid the program run-time error (division by zero).

The following assumptions are made in the conceptualization of the unsaturated zone:

1. Linear isotherms describe the partitioning of the pollutant among the liquid, vapor, and solid phases with instantaneous equilibrium.
2. Liquid phase dispersion and vapor phase diffusion are combined as one term via the Henry's equilibrium constant.
3. Vapor-phase advection is assumed negligible compared to the infiltrating water velocity and omitted in this screening level model.
4. Contaminant decay is assumed by a first-order process using two constants; one constant for the contaminant decay in the soil, and another constant for the contaminant source reduction.
5. Flow and mixing processes in the capillary fringe is beyond the scope of this work and thus it also assumes that the capillary fringe depth is negligible with respect to the unsaturated column.

The most common types of boundary condition applied at the top of the soil column are either the first type (Dirichlet's) or the third type (Cauchy's). Equation (9) describes these boundary conditions.

$$C_w|_{z=0} = \begin{cases} C_0(z=0) \exp(-\gamma t) & \text{if } t \leq t_0 \\ 0 & \text{if } t > t_0 \end{cases} \quad (9)$$

or

$$-D \frac{\partial C_w}{\partial z} + q_w C_w \Big|_{z=0} = \begin{cases} q_w C_0 \exp(-\gamma t) & \text{if } t \leq t_0 \\ 0 & \text{if } t > t_0 \end{cases} \quad (10)$$

where  $C_0$  is the liquid phase solute concentration in the infiltration water,  $\gamma$  is the decay rate (1/yr) of the solute source due to either degradation or flushing by the infiltration, and  $t_0$  is the duration of solute release (yr) which can be selected to simulate either "slug" or continuous input.

At the bottom of the soil column, the second type boundary condition (Neuman's) is commonly applied

$$\frac{\partial C_w}{\partial z} = 0 \quad (z = \infty) \quad (11)$$

In applying this boundary condition, equation (11) is actually implemented at a finite column length (i.e.,  $z \neq \infty$ ). To reduce the finite length effect, dummy cells are added at the bottom of the soil column automatically in the numerical calculation in the model. After evaluation of  $C_w(z, t)$ , the total contaminant mass ( $M$ ) per unit volume of the soil is calculated as:

$$M(z, t) = M_a + M_w + M_s = [\theta_a H + \theta_w + \rho_b K_d] C_w = \theta C_w. \quad (12)$$

## 2.2 Saturated Zone Mixing

After estimating the liquid phase solute concentration ( $C_w$ ) at the bottom of the soil column, the mixed concentration in the aquifer can be calculated using a mass-balance technique as below (USEPA 1989, Summers et al. 1980):

$$C_{\text{mix}} = \frac{C_{\text{aq}} q_{\text{aq}} A_{\text{aq}} + C_w q_w A_{\text{soil}}}{q_{\text{aq}} A_{\text{aq}} + q_w A_{\text{soil}}} \quad (13)$$

where  $C_{\text{aq}}$  is the concentration of horizontal groundwater influx,  $q_{\text{aq}}$  is the Darcy velocity in the aquifer,  $A_{\text{aq}}$  is the cross-sectional aquifer area perpendicular to the groundwater flow direction, and  $A_{\text{soil}}$  is the cross-sectional area perpendicular to the vertical infiltration in the soil column. The aquifer area ( $A_{\text{aq}}$ ) is determined by multiplying the horizontal width of the soil column with the vertical solute penetration depth.

Procedure for the mixing calculation is different depending on the type of soil column arrangement. In the case of the transverse (right angle) arrangement, the mixing calculation is straight forward: simply apply equation (13) at the each mixing element underneath the soil columns. For the parallel arrangement case, however, the mixed concentration at the upgradient cell is considered as an influx concentration to the next cell. The mixing concentration at the next cell is estimated by reapplying the equation (13) using the two inflow concentrations.

The solute penetration depth is the mixing thickness of the contaminant in the aquifer beneath the unsaturated soil column. An estimation of the plume thickness in an aquifer can be made using the relationship below (USEPA 1990):

$$H_d = \sqrt{2\alpha_v L} + B \left[ 1 - \exp\left(\frac{-Lq_w}{q_{\text{aq}} B}\right) \right] \quad (14)$$

where,  $H_d$  is the penetration depth (m),  $\alpha_v$  is the transverse (vertical) dispersivity (m) of the aquifer,  $L$  is the horizontal length dimension of the waste (m), and  $B$  is the aquifer thickness (m). In equation (14) the first term represents the thickness of the plume due to vertical dispersion and the second term represents that due to displacement from infiltration water. When implementing this relationship, it is necessary to specify that in the event the computed value of  $H_d$  is greater than  $B$ , the penetration thickness,  $H_d$  is set equal to  $B$ .

### 3 Numerical Implementation

#### 3.1 Unsaturated Zone Leaching

The governing solute transport equation (7) is solved using the finite difference method. Differential equations dealing with liquid contaminant concentration  $C_w$  as a function of time and depth are converted into the finite difference equations dealing with the corresponding variable  $C_i^k$  centered on time between two time steps:

$$C_w \rightarrow \frac{C_i^{k+1} + C_i^k}{2}, \quad \frac{\partial C_w}{\partial t} \approx \frac{C_i^{k+1} - C_i^k}{\Delta t} \tag{15}$$

where  $\Delta t$  is the time increment, the subscript  $i$  refers to the discretized soil column cell and the superscript  $k$  refers to the time level. The subscript  $w$  is dropped for simplicity. Converting the other terms into finite difference form, the governing equation can be written as:

$$\begin{aligned} &(-M_i + M'_i - N_i)C_{i-1}^{k+1} + (1 + 2M_i + N'_i + L_i)C_i^{k+1} + (-M_i - M'_i + N_i)C_{i+1}^{k+1} \\ &= (M_i - M'_i + N_i)C_{i-1}^k + (1 - 2M_i - N'_i - L_i)C_i^k + (M_i + M'_i - N_i)C_{i+1}^k \end{aligned} \tag{16}$$

where the dimensionless constants  $M_i, M'_i, N_i, N'_i,$  and  $L_i$  are:

$$\begin{aligned} M_i &\equiv \frac{\Delta t}{2(\Delta z)^2} \frac{1}{\theta_i} D_i, & M'_i &\equiv \frac{\Delta t}{2(\Delta z)^2} \frac{1}{\theta_i} \frac{D_{i+1} - D_{i-1}}{4}, \\ N_i &\equiv \frac{\Delta t}{4\Delta z} \frac{1}{\theta_i} q_i, & N'_i &\equiv \frac{\Delta t}{4\Delta z} \frac{1}{\theta_i} (q_{i+1} - q_{i-1}), & L_i &\equiv \frac{\Delta t}{2} \mu. \end{aligned} \tag{17}$$

Similarly, the finite difference form of the initial condition for the liquid phase solute concentration is

$$C_i^1 = \begin{cases} (C_s^1)_i / K_d & \text{if } K_d > 0, \\ (C_w^1)_i & \text{if } K_d = 0, \end{cases} \quad 2 < i < n - 1. \tag{18}$$

The finite difference forms of the top boundary conditions for the soil column are:

- First Type Top Boundary Condition

$$\begin{aligned} C_i^k &= \frac{C_s^k(z = 0)}{K_d} \exp[-\gamma(k - 1)\Delta t], \\ C_s^k &\neq 0 \text{ if } t \leq t_0, \\ C_s^k &= 0 \text{ if } t > t_0, \end{aligned} \quad k = 1, 2, \dots \tag{19}$$

- Third Type Top Boundary Condition

$$C_1^{k+1} - \frac{\Psi'}{\Phi'} C_2^{k+1} = -\frac{\Omega'}{\Phi'} C_2^k + \frac{\Psi'}{\Phi'} C_2^k + \frac{q_w C_0}{\Phi'} \exp(-\gamma t) \tag{20}$$

where

$$\Phi' = \frac{D(2M + L + 1)}{4(\Delta z)(M + N)} + \frac{q_w}{2}, \quad \Psi' = \frac{DM}{4(\Delta z)(M + N)},$$

$$\Omega' = \frac{D(2M + L - 1)}{4(\Delta z)(M + N)} + \frac{q_w}{2},$$

and  $M, N, L$  were defined in equation (17).

The second type bottom boundary condition is used in this model as follows:

$$\frac{C_n^{k+1} - C_{n-1}^{k+1}}{\Delta z} = 0. \tag{21}$$

This work incorporates the development of a C++ computer code to solve for the values of  $C_1^{k+1}$  using the above finite difference form of simultaneous equations by employment of the Thomas algorithm (Press et al. 1992).

### 3.2 Numerical Stability

Often, the efficiency of a numerical technique is limited due to the instability, oscillation, and mass-balance problems. Several methods have been proposed to determine the stability criteria of finite difference calculation (e.g., Fourier expansion method, matrix method, and other, (Hirsch 1989)). The Fourier expansion method, developed by von Neumann, relies on a Fourier decomposition of the numerical solution in space while neglecting boundary conditions. It provides necessary conditions for stability of constant coefficient problems regardless of the type of boundary condition (Mitchel and Griffiths 1980). The matrix method, however, analyzes the eigenvectors of the space-discretization operator, including the boundary conditions, as a basis for the representation of the spatial behavior of the solution (Hirsch 1989, Ames 1997). Based on the von Neumann method, Crank–Nicolson scheme of finite difference equation can be optimize with:

$$\Delta z < \frac{2D}{q_w}, \tag{22}$$

$$\Delta t < \frac{2\theta(\Delta z)^2}{\sqrt{(2D)^2 - (q_w \Delta z)^2}}. \tag{23}$$

According to the stability criteria expressed in equations (22) and (23), it is clear that the combined dispersion coefficient ( $D$ ) must be greater than zero. In natural soil conditions, it is rare to have a value of  $D$  as zero or close to zero. Specifically, if there is downward infiltration in the unsaturated zone, hydrodynamic dispersion of contaminant is inevitable. In addition, the air diffusion coefficients of selected organic compounds must also be greater than zero.

### 3.3 Saturated Zone Mixing

Based on the mass balance principle of equation (13), the mixed solute concentrations are estimated as:

$$\begin{aligned} C_{mx1} &= \frac{q_{w1}L_1C_{w1} + q_{aq}\tilde{H}_{d1}C_{aq}}{q_{mx1}\tilde{H}_{d1}}, \\ C_{mx2} &= \frac{q_{w2}L_2C_{w2} + q_{aq}\tilde{H}_{d1}C_{aq} + q_{mx1}\tilde{H}_{d1}C_{mx1}}{q_{mx1}(\tilde{H}_{d1} + \tilde{H}_{d2})}, \end{aligned} \quad (24)$$

where  $C_{mx(i)}$  is the ‘‘Mixed Concentration in Groundwater’’ and  $\tilde{H}$  is the adjusted penetration depth for each column.

## 4 Program Execution

The hypothetical example with two soil columns of Press et al. (1992) is simulated herein to demonstrate the impact of soil heterogeneity on the spread on the contaminant in the soil.

### 4.1 Problem Descriptions

The example depicts two soil columns arranged perpendicular to the groundwater flow direction (see Fig. 1). The unsaturated zone soil is divided into four soil layers whose total porosity decreases (from 0.44 to 0.38) and water-filled porosity increases (from 0.26 to 0.32) along with the depth shown in Fig. 1. The bulk density is adjusted according to the total porosity change (contribution from the water content change is disregarded). The soil column 1 has first type top boundary condition. The soil column 2 has third type top boundary condition. This assumed set of parameters are derived based on a filed geologic situation where the total porosity of soil decreases along with the depth due to gravitational pressure while the soil becomes wetter (water-filled porosity increases) along with the depth because water sinks down to the lower layers. Recharge rate  $q_w$  is kept constant ( $0.3048 \text{ m}^3/\text{yr}/\text{m}^2$ ) in order to keep water-filled porosity of each layer constant. Organic content  $f_{oc}$  is also kept constant ( $0.005 \text{ g/g}$ ).

### 4.2 Results

Simulation of a homogeneous soil columns were conducted (Figs. 2 and 3) in order to demonstrate the soil heterogeneity effect on the dispersion of the contaminant. The uniform homogeneous soil properties that were used in the simulations were bulk density of 1.6, total porosity of 0.4, and water filled porosity of 0.3. In both Soil Column 1 and Soil Column 2, the effects of having



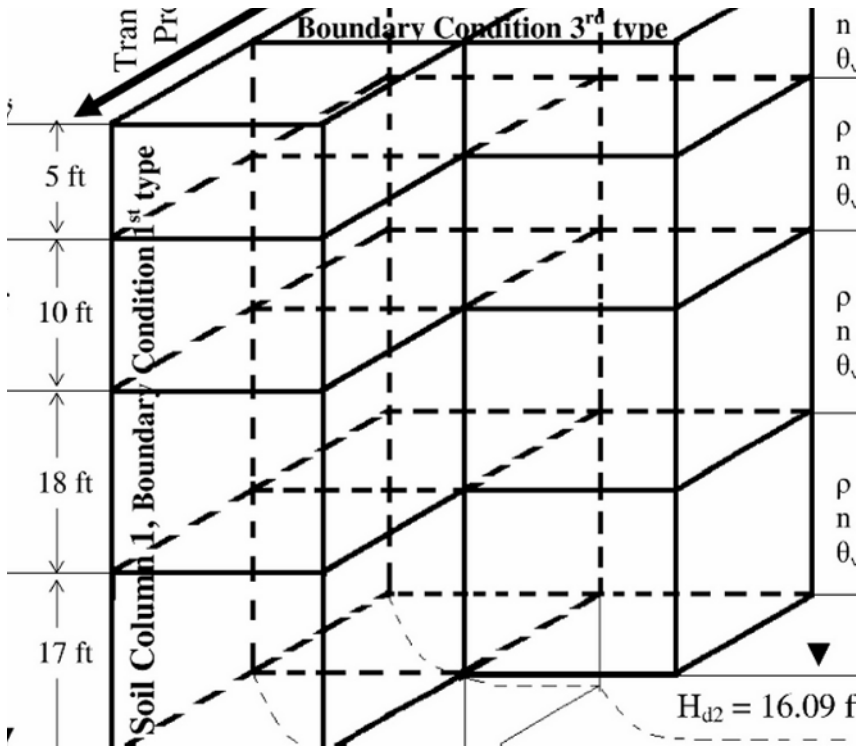
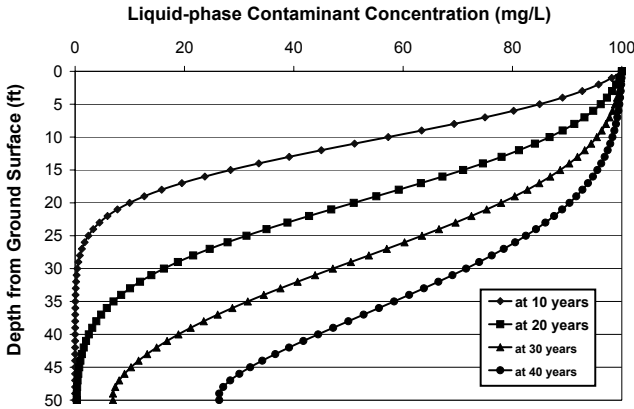


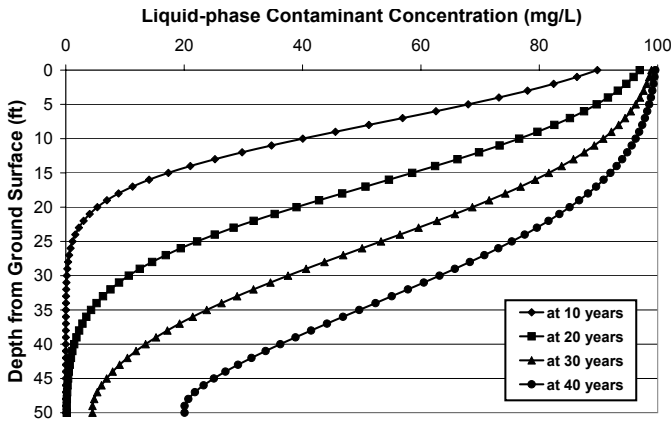
Fig. 1. Profile example problem. Soil column 1 is the first type of boundary condition. Soil column 2 is the third type boundary condition

four different layers instead of one vertically homogenous layer are obvious as shown in Figs. 4 and 5. The liquid-phase contaminant leached deeper in the heterogeneous soil after the same period of time (71 mg/L at  $-4.572$  m in Soil Column 1 at 10 years compared with 28 mg/L for the homogenous case). That is because the total porosity values for the first ( $0 \sim -1.524$  m) and second ( $-1.524 \sim -4.572$  m) layers are larger than the homogeneous value of 0.4. Also, since the total porosity and water-filled porosity values in the third layer ( $-4.572 \sim -10.058$  m) were the same for both the homogenous and the heterogeneous soils, the inclination of the contaminant profile seems to be identical in both case. After 20 years, the contaminant penetrated deeper while keeping a similar profile. After 30 years, the contaminant completely reached the bottom of the unsaturated zone and mixed with the groundwater. Figure 6 depicts the simulation results of mixing the leachate with the ground water.

Figure 7 illustrates a comparison between predicted results of the Unsaturated-zone Leaching and Saturated-zone Mixing Model for simulation of a homogeneous soil column with an analytical transport solution of Cleary and

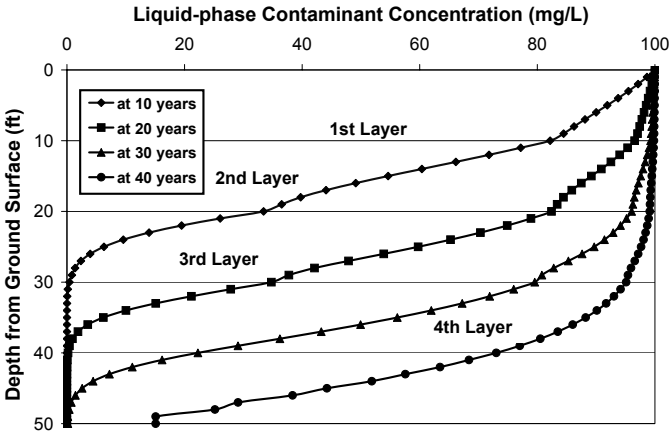


**Fig. 2.** Simulation results of homogeneous soil column 1, first type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft)

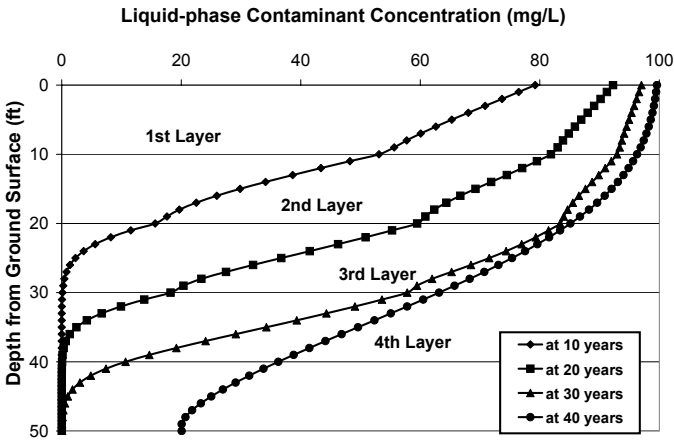


**Fig. 3.** Simulation results of homogeneous soil column 2, third type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft)

Ungs (1994). The close matched of the results indicates that the Unsaturated-zone Leaching and Saturated-zone Mixing Model program works correctly in homogeneous case. For the heterogeneous case, Unsaturated-zone Leaching and Saturated-zone Mixing Model results showed reasonable match with column experiment data (Cleary and Ungs 1994), which is available through Internet (<http://www.vadose.net>) (Lee 1999b).



**Fig. 4.** Simulation results of heterogeneous soil column 1, first type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft)



**Fig. 5.** Simulation results of heterogeneous soil column 2, third type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft)

## 5 Summary

Unsaturated-zone Leaching and Saturated-zone Mixing Model was developed. This computer code can handle vertical heterogeneity of soil. The Graphic User Interface used in Unsaturated-zone Leaching and Saturated-zone Mixing Model has made it easy to create the input data file and view the simulation results. Additional benefits stems from the ability to handle the many additional parameters that are needed for vertically heterogeneous soil columns,

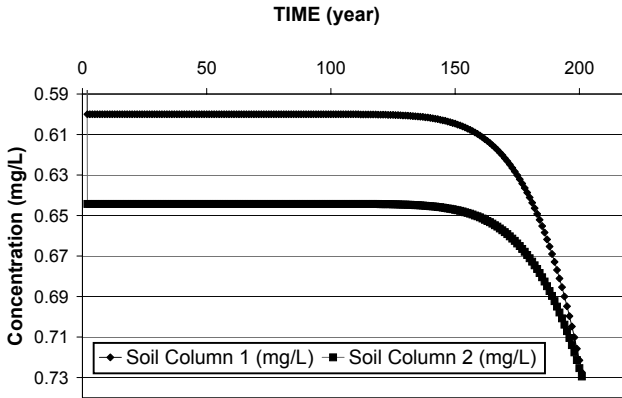


Fig. 6. Simulation results of mixed concentration at the ground water

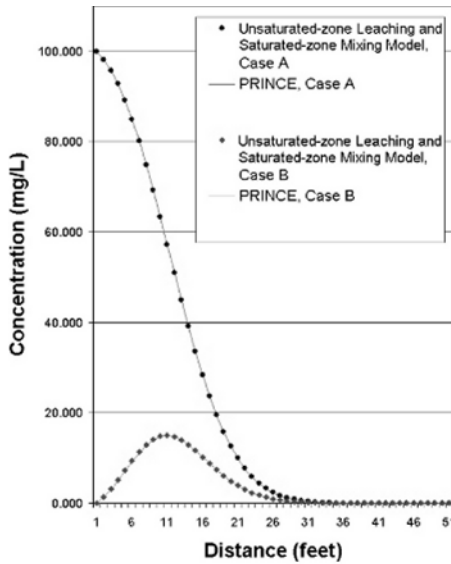


Fig. 7. Verification of Unsaturated-zone Leaching and Saturated-zone Mixing Model with homogeneous soil column by comparing with an analytical solution (PRINCE) modified from Cleary and Ungs (1994). This figure is not referred to in the text!

where a set of parameters is needed to describe the soil properties of each layer.

Verification of the computer code resulted with good match between Unsaturated-zone Leaching and Saturated-zone Mixing Model predictions and an analytical solution (for homogeneous column) and the soil column experiment (for heterogeneous soil). Development of Unsaturated-zone Leaching

and Saturated-zone Mixing Model was funded partially by the United State Environmental Protection Agency.

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