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Vadose-zone Leaching and Saturated-zone Mixing Model in Heterogeneous Layers

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Abstract: A screening level model was developed for simulation of pollutant migration through the vadose-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. Using a simple mass-balance technique, the saturated-zone module estimates mixing of the vadose-zone leachate with 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. The model was verified by comparing to an analytical solution and laboratory soil column experiments. Three different soil sample sizes of Ottawa quartz sand and 480 ppm saline water as groundwater contamination were used to be validated successfully in a lab steady state soil column study. The graphical user interface based on the Microsoft Window function was added in the model so that input data preparation and output data visualization processes are automated.

Keywords: Vadose-zone; Saturated-zone; Leaching model; Groundwater; Modelling; GUI

1. INTRODUCTION

In the model, one-dimensional finite difference scheme was employed for solving the vadose-zone transport equation, and a mass-balance principle was used for the mixing calculation in the saturated aquifer underneath the soil columns. Although several computer codes (VLEACH, VLEACHSM 2.0, EPACML, etc.) are available which can incorporate the heterogeneity of the soil properties (e.g., volumetric water content and corresponding infiltration rate), often many sites do not have the same degree of sophistication in the field-measured data. Even when there is a site with a reasonable amount of field data, a screening level of estimation is often necessary before conducting a comprehensive simulation. Moreover, often times when the pollutant migrates vertically to the ground water, a quick estimation of this mixing is necessary. The Vadose-zone Leaching and Saturated-zone Mixing Model was designed to meet these needs by combining a one dimensional heterogeneous vadose zone transport model and saturated zone mixing model

In 1993, Ravi and Johnson [1993] developed one dimensional transport program called VLEACH, which handles only vertical migration of pollutant in a homogeneous soil column. Later, Lee [1996], Lee [1999], and Lee & In [2005] developed several versions of VLEACHSM by adding the liquid-phase dispersion, decay terms in the vadose zone, and the saturation zone mixing into VLEACH. This Vadose-zone Leaching and Saturated-zone Mixing Model is improved further by implementing the heterogeneous soil property and the Graphic User Interface (GUI). GUI relieves a user from tedious and error-prone processes of input data preparation and output visualization. In addition, the Vadose-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 soil column. Using a simple mass-balance technique, the saturated zone module estimates the concentration of contaminants by mixing of leachate from the vadose zone with groundwater. This module uses the effluent concentration at the bottom of the soil column, which is estimated from the vadose zone module. A complete mixing of the leachate with the groundwater is assumed.

2. Governing Equation and Boundary and Initial Conditions

2. 1 Vadose Zone Transport

Considering the three equilibrium phases of pollutants in an unsaturated soil column, its one-dimensional governing transport equation is expressed as follows:

$$\theta_w \frac{\partial C_w}{\partial t} + \theta_a \frac{\partial C_a}{\partial t} + \rho_b \frac{\partial C_s}{\partial t} =$$

$$\frac{\partial}{\partial z}\left(\theta_w D_w \frac{\partial C_w}{\partial z}\right) + \frac{\partial}{\partial z}\left(\theta_a D_a \frac{\partial C_a}{\partial z}\right) \qquad (1)$$

$$-\frac{\partial}{\partial z}(q_w C_w) - \mu_w \theta_w C_w - \mu_a \theta_a C_a - \mu_s \rho_b C_s$$

where, C_w = concentration of a contaminant in liquid (water) phase [mg/L], C_a = concentration of a contaminant in vapour (air) phase [mg/L], C_s = concentration of a contaminant in solid phase [mg/Kg], θ_w = volumetric water content (volume of water / total volume) [m³/m³], θ_a = air-filled porosity (volume of air / total volume) [m³/m³]. Note that the total porosity (n) equals the sum of the water filled porosity and the air filled porosity. q_w = water flow velocity (recharge rate) [m/yr]. The air flow velocity (q_a) is assumed to be zero in this study. D_w = dispersion coefficient for the liquid phase contaminant in the pore water [m²/yr]. D_a = gaseous phase diffusion coefficient in the pore air [m²/yr], μ_w = first order decay rate of a contaminant in water phase [l/yr], μ_a = first order decay rate of a contaminant in gaseous phase [l/yr], μ_s = first order decay rate of a contaminant in solid phase [l/yr]. For simplicity, it is assumed that $\mu_w = \mu_a = \mu_s = \mu$. ρ_b = bulk density of the soil [gr/cm³], z = vertical coordinate with positive being downward, t = time [yr].

Instantaneous equilibrium (partitioning) of the contaminant among the phases is assumed according to the following linear relationships:

Liquid-solid phase equilibrium is

$$C_s = K_d C_w \qquad (2)$$

Liquid-gas phase equilibrium is

$$C_a = H C_w \qquad (3)$$

where K_d [ml/g] is the distribution coefficient between the solid phase and liquid phase, and H [dimensionless] is 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] is the organic carbon-water partition coefficient and f_{oc} [g/g] is 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-m³/mol) using the following equation

$$H = \frac{K_H}{(RT)} \qquad (4)$$

where K_H [atm-m³/mol] is the dimensional form of Henry's Law constant, R is the universal gas constant (R = 8.2 × 10⁻⁵ atm-m³/mol·K), and T is the absolute temperature in Kelvin (ºK = 273.16 + ºC).

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

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

where α_L is the longitudinal dispersivity [feet] of the vadose 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]:

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

where D_{air} is the diffusion coefficient of the contaminant in the free air, and n is the total porosity of the soil.

By substituting equation (2) and (3) into (1), and neglecting the air flow velocity ($q_a = 0$), the governing transport equation can be simplified as:

$$\theta \frac{\partial C_w}{\partial t} = D \frac{\partial^2 C_w}{\partial t^2} +$$

$$\left(\frac{\partial D}{\partial z} - q_w\right)\frac{\partial C_w}{\partial z} - \left(\frac{\partial q_w}{\partial z} + \mu\theta\right)C_w \qquad (7)$$

where,

$$\theta = \theta_w + \theta_a H + \rho_b K_d$$

and

$$D \equiv \theta_w D_w + \theta_a D_a H$$

To solve the above equation (7), an initial condition is given as follows.

$$C_w(z,t)\big|_{t=0} = \frac{C_s(z,0)}{K_d} \qquad (K_d > 0) \qquad (8a)$$

$$C_w(z,t)\big|_{t=0} = C_w(z,0) \qquad (K_d = 0) \qquad (8b)$$

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 most common type of boundary condition to be applied at the top of the soil column is either the first type (Dirichlet's) or the third type (Cauchy's) of boundary condition as shown below.

$$C_w\big|_{z=0} = C_0(z=0)\exp(-\gamma t) \quad (t \le t_0) \quad (9a)$$

$$C_w\big|_{z=0} = 0 \quad (t > t_0) \tag{9b}$$

or

$$-D\frac{\partial C_w}{\partial z} + q_w C_w \bigg|_{z=0} =$$

$$q_w C_0 \exp(-\gamma t) \quad (t \le t_0) \tag{10a}$$

$$-D\frac{\partial C_w}{\partial z} + q_w C_w \bigg|_{z=0} = 0 \quad (t > t_0) \tag{10b}$$

where C_0 is the liquid phase solute concentration in the infiltration water, γ 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 continuos input.

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

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

In applying this boundary condition, equation (11) is actually implemented at a finite column length (i.e., $z \ne \infty$). 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:

$$M(z,t) = M_a + M_w + M_s =$$

$$[\theta_a H + \theta_w + \rho_b K_d]C_w = \theta C_w \tag{12}$$

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 (USPEA, [1989] and Summers *et al.*, [1980]):

$$C_{mix} = \frac{\left(C_{aq} q_{aq} A_{aq} + C_w q_w A_{soil}\right)}{\left(q_{aq} A_{aq} + q_w A_{soil}\right)} \tag{13}$$

where C_{aq} is the concentration of horizontal groundwater influx, q_{aq} is the Darcy velocity in the aquifer, A_{aq} is the cross-sectional aquifer area perpendicular to the groundwater flow direction, and A_{soil} is the cross-sectional area perpendicular to the vertical infiltration in the soil column. The aquifer area (A_{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: one from the effluent from the upgradient cell and the other from the leaching from the soil column.

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

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

where, H_d is the penetration depth [m], α_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 Vadose 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:

$$C_w \rightarrow \frac{\left(C_i^{k+1} + C_i^k\right)}{2},$$

$$\frac{\partial C_w}{\partial t} \rightarrow \frac{\left(C_i^{k+1} - C_i^k\right)}{\Delta t} \quad (15)$$

where Δt 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:

$$\left(-M_i + M_i' - N_i\right)C_{i-1}^{k+1} + \left(1 + 2M_i + N_i' + L_i\right)C_i^{k+1} +$$

$$\left(-M_i - M_i' + N_i\right)C_{i+1}^{k+1}$$

$$= \left(M_i - M_i' + N_i\right)C_{i-1}^{k} + \left(1 - 2M_i - N_i' - L_i\right)C_i^{k} +$$

$$\left(M_i + M_i' - N_i\right)C_{i+1}^{k} \quad (16)$$

where the dimensionless constants M_i, M_i', N_i, N_i', and L_i are: depth.

$$M_i \equiv \frac{\Delta t}{2(\Delta z)^2}\frac{1}{\theta_i}D_i ,$$

$$M_i' \equiv \frac{\Delta t}{2(\Delta z)^2}\frac{1}{\theta_i}\frac{D_{i+1} - D_{i-1}}{4}$$

$$N_i \equiv \frac{\Delta t}{4\Delta z}\frac{1}{\theta_i}q_i ,$$

$$N_i' \equiv \frac{\Delta t}{4\Delta z}\frac{1}{\theta_i}\left(q_{i+1} - q_{i-1}\right)$$

$$L_i \equiv \frac{\Delta t}{2}\mu \quad (17)$$

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

$$C_i^1 = \frac{\left(C_s^1\right)_i}{K_d} \quad (K_d > 0, \, 2 \leq i \leq \text{n-1}) \quad (18a)$$

or

$$C_i^1 = \left(C_w^1\right)_i \quad (K_d = 0, \, 2 \leq i \leq \text{n-1}) \quad (18b)$$

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

First Type Top Boundary Condition,

$$C_i^k = \frac{C_s^k(z = 0)}{K_d}\exp\left\{-\gamma(k-1)\Delta t\right\}$$

$$(k = 1, 2, \ldots)$$

$$C_s^k \neq 0, \quad \text{when } t \leq t_0$$

$$C_s^k = 0, \quad \text{when } t > t_0 \quad (19)$$

Third Type Top Boundary Condition,

$$C_1^{k+1} - \frac{\Psi'}{\Phi'}C_2^{k+1} =$$

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

where

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

$$\Psi^{'} = \frac{DM}{4(\Delta z)(M + N)},$$

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

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

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

$$\frac{C_n^{k+1} - C_{n-1}^{k+1}}{\Delta z} = 0 \qquad (21)$$

The above finite difference form of simultaneous equations are computer coded in C++ to solve for the value of C_1^{k+1} by 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 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, takes eigenvectors of the space-discretization operator, including the boundary conditions, as a basis for the representation of the spatial behaviour of the solution [Hirsch, 1989 & Ames, 1997]. Based on the von Neumann method, Crank-Nicolson scheme of finite difference equation can be derived as:

$$\Delta z < \frac{2D}{q_w} \qquad (22)$$

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

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 vadose zone, hydrodynamic dispersion of contaminant is inevitable. In addition, the air diffusion coefficients of selected organic compounds are must greater than zero.

Note that the numerical criteria presented in equations (22) and (23) are general guidelines and may not work for all situations. To ensure a stable result, the output should be checked thoroughly. If the results show any oscillation or negative concentration, a smaller time step and/or cell size than the values from the above equations should be tried. After having "stable" results, additional runs are recommended to check convergence of the simulation results. In this case, even a smaller time step and/or cell size can be used to check whether the solution converges or not.

3.3 Saturated Zone Mixing

Based on the mass balance principle of equation (13) and the mixed concentration are estimated as:

$$C_{mx1} = \frac{q_{w1} L_1 C_{w1} + q_{aq} \tilde{H}_{d1} C_{aq}}{q_{mx1} \tilde{H}_{d1}} \qquad (24a)$$

$$C_{mx2} = \frac{q_{w2} L_2 C_{w2} + q_{aq} \tilde{H}_{d1} C_{aq} + q_{mx1} \tilde{H}_{d1} C_{mx1}}{q_{mx1}(\tilde{H}_{d1} + \tilde{H}_{d2})} \qquad (24b)$$

where $C_{mx(i)}$ is the "Mixed Concentration in Groundwater".

4. PROGRAM EXECUTION

A hypothetical example with two soil columns is simulated to demonstrate the impact of soil heterogeneity [Press *et al.*, 1992].

4.1 Problem Descriptions

The example depicts two soil columns arranged perpendicular to the ground-water flow direction (see Figure 1). The vadose 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 Figure 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 1st type top boundary condition. The soil column 2 has 3rd

type top boundary condition. This assumed set of parameters are derived based on a filed geologic
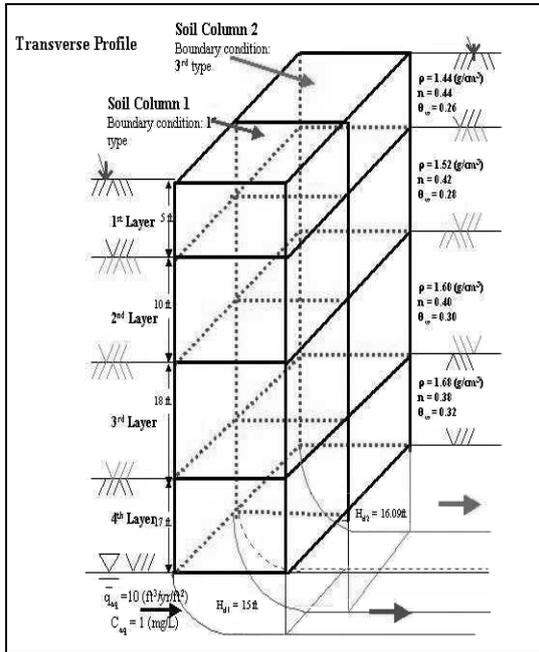

Figure 1. Profile example problem. Soil column 1 is the 1st type of boundary condition. Soil column 2 is the 3rd type boundary condition.

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 is kept constant (0.3048 $m^3/yr/m^2$) in order to keep water-filled porosity of each layer constant. Organic content f_{oc} is also kept constant (0.005 g/g).

4.2 Results

To demonstrate the soil heterogeneity effect in the column the homogeneous soil columns were simulated (Figures 2 and 3). Note that, for homogeneous soil, uniform values soil property were used (bulk density 1.6, total porosity $n = 0.4$, and water filled porosity = 0.3). In both Soil Column 1 and Soil Column 2, the effects of putting four different layers instead of one vertically homogenous layer are obvious as shown in Figures 4 and 5. The liquid-phase contaminant
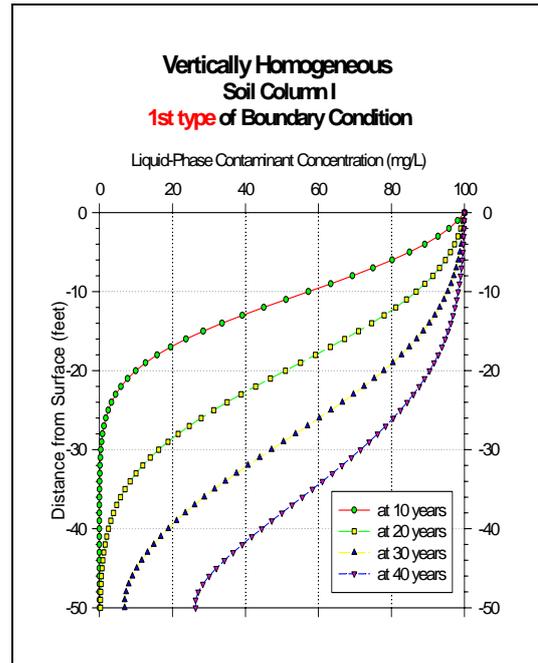

Figure 2. Simulation results of homogeneous soil column I, 1st type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft).
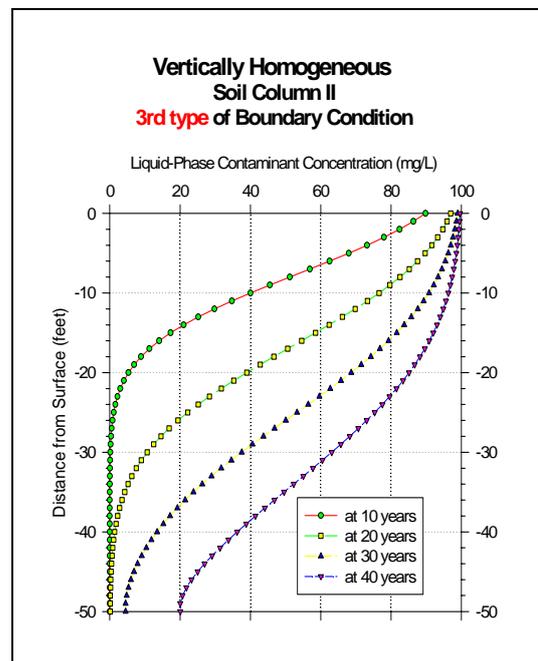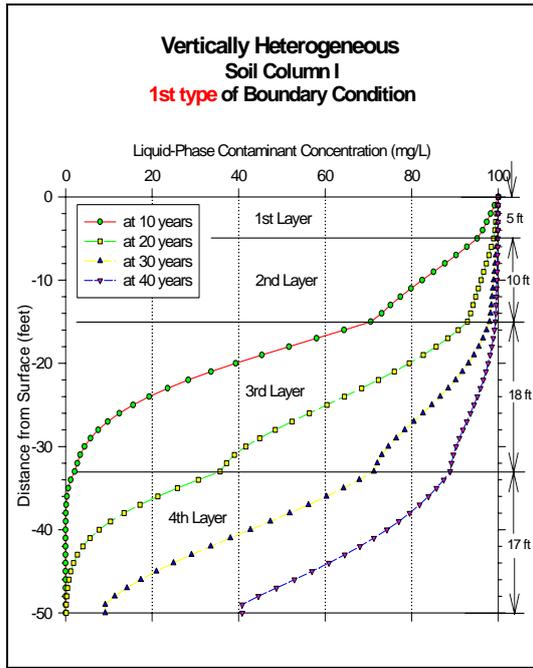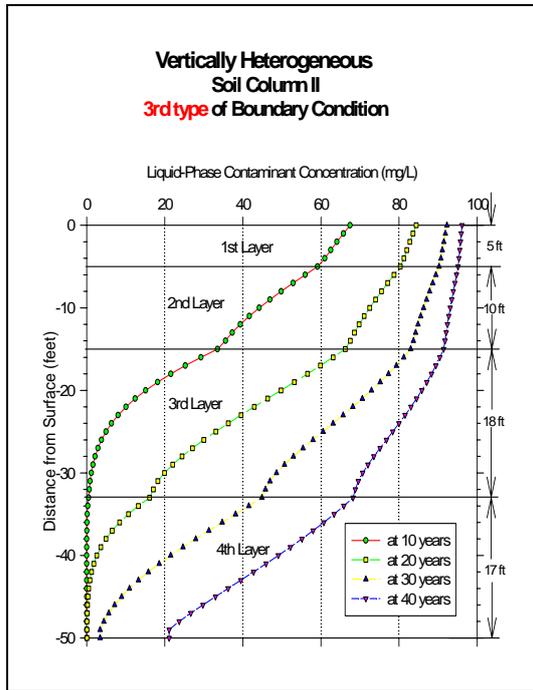

Figure 3. Simulation results of homogeneous soil column II, 3rd type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft).

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

Figure 5. Simulation results of heterogeneous soil column II, 3rd type of boundary condition at 10, 20, 30, and 40 years of liquid-phase contaminant concentration (mg/L) vs. depth from ground surface (ft).

went deeper at 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 ~ -1.524 m) and second (-1.524 ~ -4.572 m) layers are larger than the homogeneous value of 0.4. Also, since the total porosity and water-filled porosity values are the same with the homogenous ones for the third layer (-4.572 ~ -10.058 m), the inclination of contaminant profile mostly seems identical to the homogenous case. After 20 years, the contaminant penetrates deeper while keeping a similar profile, and after 30 years, contaminant completely reaches the bottom of the vadose zone, mixing into groundwater. The simulation results of mixed concentration at the ground water were shown in Figure 6.
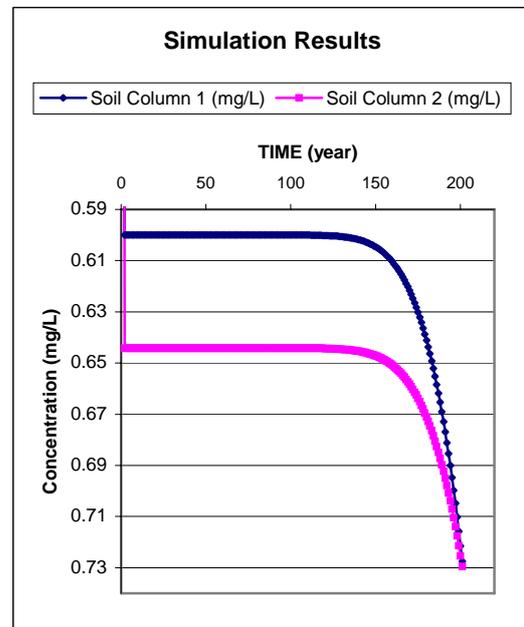

Figure 6. Simulation results of mixed concentration at the ground water.

A homogeneous soil column was simulated using Vadose-zone Leaching and Saturated-zone Mixing Model and the results were compared with an analytical transport solution [Cleary and Ungs, 1994] (see Figure 7). The close matched of the results indicates Vadose-zone Leaching and Saturated-zone Mixing Model program works correctly in homogeneous case. For the heterogeneous case, Vadose-zone Leaching and Saturated-zone Mixing Model results showed reasonable match with column experiment data [Cleary and Ungs, 1994], which is available

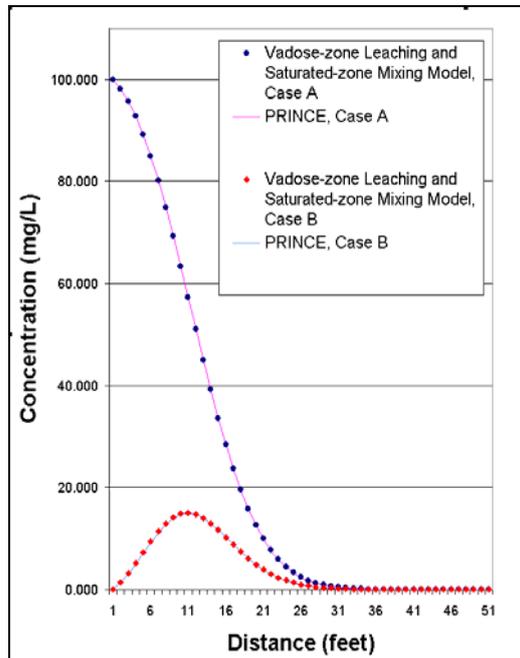through Internet (http://www.vadose.net) [Lee, 1999].

Figure 7. Verification of Vadose-zone Leaching and Saturated-zone Mixing Model with homogeneous soil column by comparing with an analytical solution (PRINCE) modified from Cleary and Ungs [1994].

5. SUMMARY

Vadose-zone Leaching and Saturated-zone Mixing Model was developed. This program can handle vertical heterogeneity of soil. The Graphic User Interface (GUI) used in Vadose-zone Leaching and Saturated-zone Mixing Model has made it easy to create the input data file and view the simulation results. Because Vadose-zone Leaching and Saturated-zone Mixing Model can deal with vertically heterogeneous soil columns which require more parameters to describe the soil properties of each layer, such an easy-to-use interface is even more important.

Verification of this program showed good match between Vadose-zone Leaching and Saturated-zone Mixing Model results and an analytical solution (for homogeneous column) and the soil column experiment (for heterogeneous soil). Development of Vadose-zone Leaching and Saturated-zone Mixing Model was funded partially by the United State Environmental Protection Agency.

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