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Finite element modeling of in-situ air sparging for groundwater remediation

Krishna R. Reddy & Jinnan Zhou
Department of Civil and Materials Engineering, University of Illinois at Chicago, Ill., USA

ABSTRACT: Mathematical models are useful to serve as tools in the design of optimal remediation systems. This paper describes the development and numerical implementation of a mathematical model to simulate air sparging process for remediating saturated soils and groundwater contaminated with volatile organic compounds (VOCs). The air sparging process involves injection of air below the area of contamination to facilitate partitioning of the dissolved and sorbed VOCs into the gas phase and to enhance the aerobic biodegradation of the VOCs. The major contaminant transport and transformation processes that occur during air sparging operations include advection, diffusion, dispersion, adsorption-desorption, volatilization, biodegradation, and dissolution. The model incorporates these processes on three phases, namely water, air and VOC. Ultimately, the model is intended to be used for designing the optimal air sparging systems that are efficient, economical and safe.

1 INTRODUCTION

In-situ air sparging is a developing remediation technique that has significant potential for use at VOC-contaminated saturated soils and groundwater. This technique consists of injecting air below the contaminated area to partition the dissolved, sorbed and free phase VOCs into the gas phase and to enhance the aerobic biodegradation of the VOCs. Because of buoyancy effect, the VOCs in gas phase are transported by air to the vadose zone where they are removed and subsequently treated by a soil vapor extraction system. A detailed description of the air sparging process and an overview of the past investigations on the air sparging are given by Reddy et al. (1995).

The VOCs exist in dissolved phase, adsorbed to the soil solids, and also exist in free phase. Different mass transport and transformation processes affect the fate of these VOCs during air sparging operations which include volatilization, desorption, dissolution, and biodegradation. These interphase mass transfer or transformation processes are also dependent on the advective-dispersive characteristics of the groundwater. The advection, hydrodynamic dispersion, and sorption have been studied by numerous investigators and are straightforward to include in mathematical modeling. However, the understanding of different processes that occur on microscale during air sparging is primitive. A few models which were developed exclusively for air sparging systems are very simplified and do not accurately account for the different mechanisms and contaminant interactions which occur during the air sparging operation (Reddy et al., 1995). Therefore, the development of a rigorous mathematical model which accurately simulates the air sparging process has been initiated at the University of Illinois at Chicago (UIC).

2 MATHEMATICAL MODEL

The model incorporates: (1) the volatilization, dissolution, and biodegradation processes at the microscale, (2) advection, hydrodynamic dispersion at the macroscale, and (3) the interphase mass transfer processes, including sorption-desorption, and diffusion at both scales. The model considers three phases: water, air, and organic chemical species (VOCs).

Assuming the validity of Darcy's law and linear adsorption isotherm, the mathematical description of the average movement of a species α in phase f is given by Abriola and Pinder (1985) as follows:

$$\frac{\partial}{\partial t}[\rho_f \epsilon_{\alpha f}(nS_f + K_{\alpha f,d}\rho_b)] + \nabla\bullet[\rho_f \epsilon_{\alpha f} v_f] - \nabla\bullet[nS_f D_{\alpha f}\nabla(\rho_f \epsilon_{\alpha f})] - r_{\alpha f} - q_{\alpha f} = 0 \qquad (1)$$

$$v_f = -\frac{kk_{rf}}{\mu_f}(\nabla p_f + \rho_{mf} g \nabla z \qquad (2)$$

where ρ_f is the molar density of phase f, $\epsilon_{\alpha f}$ is the species mole fraction, v_f is the Darcy velocity vector, $D_{\alpha f}$ is the dispersion tensor, n is the porosity, S_f is the phase saturation, $r_{\alpha f}$ is the interphase transfer of species α to or from phase f, $q_{\alpha f}$ is the sources and/or sinks of species α to phase f, $K_{\alpha f,d}$ is the partitioning coefficient of species α between soil phase and f phase, ρ_b is the bulk mass density of the soil phase, k is the intrinsic permeability tensor, k_{rf} is the relative permeability, μ_f is the viscosity, p is the phase pressure, g is gravitational acceleration, and ρ_{mf} is the mass density.

Equations (1) and (2) are subject to the following constraints:

$$\sum_{\alpha=1}^{N} \epsilon_{\alpha f} = 1.0 \qquad (3)$$

$$\sum_{f=1}^{3} S_f = 1.0 \qquad (4)$$

The dispersion tensor $D_{\alpha f}$ is defined by Bear (1972) as:

$$D_{\alpha fij} = \alpha_T |v| \delta_{ij} + (\alpha_L - \alpha_T)\frac{v_i v_j}{|v|} - D^m_{\alpha f}\tau_f \qquad (5)$$

where α_L is the longitudinal dispersivity, α_T is the transverse dispersivity, δ_{ij} is Kronecker delta, $|v|$ is the absolute value of the Darcy velocity, $D_{\alpha f}^m$ is the coefficient of molecular diffusivity, and τ_f is the tortuosity. The tortuosity in a partially saturated porous media is defined by Millington and Quirk (1961) as follows:

$$\tau_f = \frac{n_f^{7/3}}{n^2} \qquad (6)$$

where n_f is the phase f filled porosity, and n is the total porosity.

The capillary pressure between phases is given by:

$$p_{gw} = p_g - p_w \qquad (7)$$

$$p_{go} = p_g - p_o \qquad (8)$$

$$p_{ow} = p_o - p_w \qquad (9)$$

where p_{gw} is the capillary pressure between the gas (g) and water (w) phases, p_{go} is the capillary pressure between the gas (g) and organic (o) phases, and p_{ow} is the capillary pressure between the organic (o) and the water (w) phases.

The interphase mass transfer of the organic species occurs by sorption-desorption, dissolution, and volatilization. The sorption-desorption is accounted for in Eqn. (1). Based on gas-transfer principles (Matter-Muller et al. 1981; Roberts et al. 1984), the relationship between the concentrations of organic species in the water and gas phases is:

$$C_g = C_w H_c\left\{1 - \exp\left[-\frac{K_{La}V_w}{Q_g H_c}\left(\frac{t}{t_r}\right)\right]\right\} \qquad (10)$$

where C_g is the gas-phase concentration, C_w is the water-phase concentration, H_c is Henry's constant (dimensionless), K_{La} is the overall water-phase mass-transfer coefficient, V_w is the water volume, Q_g is the gas flow rate, t_r is the total retention time of the air from the injection well screen to the free water surface, and t is the retention time of the air rising through water. By assuming that the change of C_w during the retention time t_r is negligible, the following is obtained from Eqn. (10):

$$\frac{\partial C_g}{\partial t} = -\frac{K_{La}V_w}{Q_g H_c t_r}(H_c C_w - C_g) \qquad (11)$$

The dissolution of the VOCs from the free phase into the water phase is expressed by the linear-driving force model as given below (Powers et al., 1991):

$$F_\alpha^{0w} = -K_{\alpha o}^{ow}(C_{\alpha w} - C_{\alpha s}) \qquad (12)$$

where F^{0w}_α is the flux of species α between the two phases, $K^{0w}_{\alpha o}$ is the effective mass transfer coefficient of species α between the free phase and the water phase, $C_{\alpha w}$ is the concentration of species α in water phase, and $C_{\alpha s}$ is the equilibrium concentration of species α in the water phase.

The volatilization of VOCs from the free phase into the air is expressed by the following equation (Hines and Maddox, 1985):

$$F_\alpha^{og} = -K_{\alpha g}^{og}(C_{\alpha g} - C_{\alpha gm}) \qquad (13)$$

where F^{og}_{α} is the flux of species α between the free phase and the gas phase, $K^{0g}_{\alpha g}$ is the effective mass transfer coefficient of species α between the free phase and the gas phase, $C_{\alpha g}$ is the concentration of species α in the gas phase, and $C_{\alpha gm}$ is the equilibrium concentration of species α in the gas phase.

The dual Monod formulation (Borden and Bedient 1986; McQuarrie and Sudicky 1990) is used to include biodegradation as a sink term:

$$q_{ow}^{bio}=-k_u C_{mt}\left(\frac{\varepsilon_{ow}}{K_o+\varepsilon_{ow}}\right)\left(\frac{\varepsilon_{aw}}{K_a+\varepsilon_{aw}}\right) \quad (14)$$

$$q_{aw}^{bio}=-k_u F C_{mt}\left(\frac{\varepsilon_{ow}}{K_o+\varepsilon_{ow}}\right)\left(\frac{\varepsilon_{aw}}{K_a+\varepsilon_{aw}}\right) \quad (15)$$

$$q_{m}^{bio}=-k_u Y C_{mt}\left(\frac{\varepsilon_{ow}}{K_o+\varepsilon_{ow}}\right)\left(\frac{\varepsilon_{aw}}{K_a+\varepsilon_{aw}}\right) \quad (16)$$

where q_{ow}^{bio} is rate of biodegradation of the organic compound, C_{mt} is total molar concentration of microorganisms, k_u is maximum organic utilization rate, ϵ_{ow} is the mole fraction of organic compound in the water phase, ϵ_{aw} is mole fraction of the electron acceptor (oxygen) in the water phase, K_o is the organic compound half-saturation constant, K_a is the electron acceptor half-saturation constant, q_{aw}^{bio} is the rate of consumption of oxygen in the water phase, F is the ratio of moles of oxygen consumed to moles of organic compound utilized, q_m^{bio} is the rate of production of microorganisms, and Y is the ratio of moles of microorganisms consumed to moles of organic compound biodegraded (also known as the microbial yield coefficient). Mass concentrations of microorganisms and Y are converted to mole fractions using an arbitrary molecular weight and molar density assigned to the microorganisms.

If a linear adsorption isotherm is used to represent the distribution of the microorganisms between the soil and the water phases, and it is assumed that the microorganisms are present only in the soil and the water phases, C_{mt} is given by:

$$C_{mt}=\rho_w \varepsilon_{mw}(nS_w+K_{mw,d}\rho_b) \quad (17)$$

where ρ_w is the molar density of water, ϵ_{mw} is the mole fraction of microorganisms in the water phases, $K_{mw,d}$ is linear adsorption coefficient of microorganisms in the soil and water phases.

By introducing the above interphase mass transfer and sink terms into Eqn. (1), the following equations are obtained:

For water phase:

$$\frac{\partial}{\partial t}[\rho_w \epsilon_{\alpha w}(nS_w+K_{\alpha w,d}\rho_b)]+\frac{\partial}{\partial x_i}(\rho_w \epsilon_{\alpha w} v_w) -\frac{\partial}{\partial x_i}\left(nS_w D_{\alpha w}\frac{\partial(\rho_w \epsilon_{\alpha w})}{\partial x_j}\right) +K_{\alpha o}^{ow}(C_{\alpha w}-C_{\alpha s})-\frac{K_{La}V_w}{Q_g H_{c\alpha} t_r}(H_{c\alpha}C_{\alpha w}-C_{\alpha g}) -k_u C_{mt}\left(\frac{\varepsilon_{\alpha w}}{K_o+\varepsilon_{\alpha w}}\right)\left(\frac{\varepsilon_{aw}}{K_a+\varepsilon_{aw}}\right)=0 \quad (18)$$

By expanding the advective and the mass accumulation terms and using the continuity equation for water flow,

$$-\frac{\partial(v_w)}{\partial x_i}=\frac{\partial}{\partial t}(nS_w) \quad (19)$$

assuming that the term containing time derivative of $k_{\alpha,d}\rho_b$ is negligible and converting to concentrations, the following equation is obtained:

$$(nS_w+K_{\alpha w,d}\rho_b)\frac{\partial C_{\alpha w}}{\partial t}+v_w\frac{\partial C_{\alpha w}}{\partial x_i} -\frac{\partial}{\partial x_i}\left(nS_w D_{\alpha w}\frac{\partial C_{\alpha w}}{\partial x_j}\right)-\frac{K_{La}V_w}{Q_g H_{c\alpha} t_r}(H_{c\alpha}C_{\alpha w}-C_{\alpha g}) +K_{\alpha o}^{ow}(C_{\alpha w}-C_{\alpha s})-k_u C_{mt}\left(\frac{\varepsilon_{ow}}{K_o+\varepsilon_{ow}}\right)\left(\frac{\varepsilon_{aw}}{K_a+\varepsilon_{aw}}\right)=0 \quad (20)$$

For gas phase:

$$(nS_g)\frac{\partial C_{\alpha g}}{\partial t}+v_g\frac{\partial C_{\alpha g}}{\partial x_i}-\frac{\partial}{\partial x_i}\left(nS_g D_{\alpha g}\frac{\partial C_{\alpha g}}{\partial x_j}\right) +\frac{K_{La}V_w}{Q_g H_{c\alpha} t_r}(H_{c\alpha}C_{\alpha w}-C_{\alpha g})+qC_{\alpha g} +K_{\alpha g}^{og}(C_{g\alpha}-C_{gm\alpha})=0 \quad (21)$$

where q is the air flow source.

For free phase VOCs:

$$(nS_o)\frac{\partial C_{\alpha o}}{\partial t}+v_o\frac{\partial C_{\alpha o}}{\partial x_i}-\frac{\partial}{\partial x_i}\left(nS_o D_{\alpha o}\frac{\partial C_{\alpha o}}{\partial x_j}\right) -K_{\alpha g}^{og}(C_{g\alpha}-C_{gm\alpha})-K_{\alpha o}^{ow}(C_{\alpha w}-C_{\alpha s})=0 \quad (22)$$

3 NUMERICAL IMPLEMENTATION

The upstream-weighted residual finite element presented by Huyakorn and Nilkuha (1979) is used to discretize the transport equations, and the function C is represented by a trial function of the following type:

$$\overline{C}(x_i,t)=N_J(x_i)C_J(t) \qquad J=1,2,\ldots,n \tag{23}$$

where $N_J(x_i)$ and $C_J(t)$ are the basis functions and nodal values of concentration at time t and n is the number of nodes in the finite element network. To weigh the spatial derivative terms of the equations (20) to (22), asymmetric weighing functions W_I are used. The weighted residual integral transport equations are given as:

<u>Water phase:</u>

$$\int_R W_I\left[v_w\frac{\partial\overline{C_{\alpha w}}}{\partial x_i}-\frac{\partial}{\partial x_i}\left(nS_wD_{\alpha w}\frac{\partial\overline{C_{\alpha w}}}{\partial x_j}\right)\right]dR$$
$$+\int_R N_I[(nS_w+K_{\alpha w,d}\rho_b)\frac{\partial\overline{C_{\alpha w}}}{\partial t}$$
$$-\frac{K_{La}V_w}{Q_gH_{c\alpha}t_r}(H_{c\alpha}\overline{C_{\alpha w}}-\overline{C_{\alpha g}})]dR+\int_R N_I[K_{\alpha o}^{ow}(\overline{C_{\alpha w}}-C_{\alpha s})$$
$$-k_uC_{mt}\left(\frac{\overline{C_{\alpha w}}}{K_o+\overline{C_{\alpha w}}}\right)\left(\frac{\overline{C_{aw}}}{K_a+\overline{C_{aw}}}\right)]dR=0 \tag{24}$$

Applying Green's Theorem to the second derivative term one obtains:

$$\int_R[W_Iv_w\frac{\partial N_J}{\partial x_i}+nS_wD_{\alpha w}\frac{\partial W_I}{\partial x_i}\frac{\partial N_J}{\partial x_j}]C_{J_{\alpha w}}dR$$
$$+\int_R N_I[-N_Jk_uC_{mt}\left(\frac{C_{J_{\alpha w}}}{K_o+N_JC_{J_{\alpha w}}}\right)\left(\frac{C_{J_{aw}}}{K_a+N_JC_{J_{aw}}}\right)$$
$$+K_{\alpha o}^{ow}(N_JC_{J_{\alpha w}}-C_{\alpha s})+N_J(nS_w+K_{\alpha w,d}\rho_b)\frac{\partial C_{J_{\alpha w}}}{\partial t}$$
$$-\frac{K_{La}V_w}{Q_gH_{c\alpha}t_r}(N_JH_{c\alpha}C_{J_{\alpha w}}-C_{J_{\alpha g}})]dR$$
$$-\int_B W_I(nS_wD_{\alpha w}\frac{\partial C_{J_{\alpha w}}}{\partial x_j})n_idB=0 \tag{25}$$

Now, let

$$E_{IJ}=\sum_e E_{IJ}^e=\sum_e\int_{R^e}[W_IV_w\frac{\partial N_J}{\partial x_i}+nS_wD_{\alpha w}\frac{\partial W_I}{\partial x_i}\frac{\partial N_J}{\partial x_j}]dR \tag{26}$$

$$B_{IJ}^*=\sum_e B_{IJ}^{*e}=\sum_e\int_{R^e}N_I[-N_J\frac{K_{La}V_w}{Q_gt_r}+N_JK_{\alpha o}^{ow}$$
$$-N_Jk_uC_{mt}\left(\frac{1}{K_o+N_JC_{J_{\alpha w}}}\right)\left(\frac{C_{J_{aw}}}{K_a+N_JC_{J_{aw}}}\right)]dR \tag{27}$$

We have:

$$(E_{IJ}+B_{IJ}^*)C_{J_{\alpha w}}+B_{IJ}\frac{dC_{J_{\alpha w}}}{dt}=F_I \tag{30}$$

$$B_{IJ}=\sum_e B_{IJ}^e=\sum_e\int_{R^e}(nS_w+k_{\alpha w,d}\rho_b)N_IN_JdR \tag{28}$$

$$F_I=\sum_e F_I^e=\sum_e[\int_{R^e}N_I[-\frac{K_{La}V_w}{Q_gH_{c\alpha}t_r}C_{J_{\alpha g}}+K_{\alpha o}^{ow}C_{\alpha s}]dR$$
$$+\int_{B^e}W_I(nS_wD_{\alpha w}\frac{\partial C_{J_{\alpha w}}}{\partial x_j}n_i)dB] \tag{29}$$

Similarly for gas phase:

$$E_{IJ}=\sum_e E_{IJ}^e=\int_{R^e}[W_IV_g\frac{\partial N_J}{\partial x_i}+nS_gD_{\alpha g}\frac{\partial W_I}{\partial x_i}\frac{\partial N_J}{\partial x_j}]dR \tag{31}$$

$$F_I=\sum_e F_I^e=\sum_e[\int_{R^e}N_I[-\frac{K_{La}V_w}{Q_gH_{c\alpha}t_r}C_{J_{\alpha w}}$$
$$-K_{\alpha g}^{og}C_{\alpha gm}]dR+\int_{B^e}W_InS_gD_{\alpha g}\frac{\partial C_{J_{\alpha g}}}{\partial x_j}n_idB] \tag{32}$$

$$B_{IJ}=\sum_e B_{IJ}^e=\sum_e\int_{R^e}(nS_g)N_IN_JdR \tag{33}$$

$$B_{IJ}^*=\sum_e B_{IJ}^{*e}=\sum_e\int_{R^e}N_I[N_J\frac{K_{La}V_w}{Q_gt_r}+N_JK_{\alpha g}^{og}+N_Jq]dR \tag{34}$$

We again have:

$$(E_{IJ}+B_{IJ}^*)C_{J_{\alpha g}}+B_{IJ}\frac{dC_{J_{\alpha g}}}{dt}=F_I \tag{35}$$

We can write the equations for free phase VOCs in the same way:

$$E_{IJ}=\sum_e E_{IJ}^e=\int_{R^e}[W_IV_o\frac{\partial N_J}{\partial x_i}+nS_oD_{\alpha o}\frac{\partial W_I}{\partial x_i}\frac{\partial N_J}{\partial x_j}]dR \tag{36}$$

$$F_I=\sum_e F_I^e=\sum_e[\int_{R^e}N_I[K_{\alpha g}^{og}C_{\alpha gm}-K_{\alpha o}^{ow}C_{\alpha s}]dR$$
$$+\int_{B^e}W_InS_oD_{\alpha o}\frac{\partial C_{J_{\alpha o}}}{\partial x_j}n_idB] \tag{37}$$

$$B_{IJ}=\sum_e B_{IJ}^e=\sum_e \int_{R^e} (nS_o)N_I N_J dR \tag{38}$$

$$B_{IJ}^*=\sum_e B_{IJ}^{*e}=\sum_e \int_{R^e} N_I[-N_J K_{\alpha g}^{og}-N_J K_{\alpha o}^{og}]dR \tag{39}$$

$$(E_{IJ}+B_{IJ}^*)C_{J_{\alpha o}}+B_{IJ}\frac{dC_{J_{\alpha o}}}{dt}=F_I \tag{40}$$

Time integration of equations (30), (35) and (40) is performed using a general finite difference method which leads to the following form:

$$[\omega(E_{IJ}+B_{IJ}^*)+\frac{B_{IJ}}{\Delta t_k}]C_J^{k+1}=(\omega-1)(E_{IJ}+B_{IJ}^*)C_J^k$$

$$+\frac{B_{IJ}}{\Delta t_k}C_J^k+\omega F_I^{k+1}+(1-\omega)F_I^k \tag{41}$$

where ω is the time weighing factor.

4 SUMMARY

A rigorous mathematical model is described to simulate air sparging process for remediating groundwater contaminated with VOCs. The model incorporates different contaminant transport and transformation processes including volatilization, dissolution, and biodegradation. The numerical implementation of the model using the finite element approach is explained. The model is currently being calibrated based on the laboratory aquifer simulation tests and further calibrated and verified based on a field air sparging pilot tests. The model is useful to optimize the number and spacing of injection and extraction wells, air injection pressures and flow rates, and to assess the long-term performance of the air sparging systems.

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