(Short Communication)

# Simulations for Groundwater Remediation Using Three Remedial Techniques

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(Received July 6, 2000; Accepted December 29, 2000)

#### ABSTRACT

A three-dimensional (3D) solute transport model coupled with a 3D groundwater flow model is employed to simulate groundwater remediation when using natural attenuation, the pump-and-treat method, and the funnel-and-gate system. A hypothetical site is assumed to be contaminated by petroleum chemicals, where the highest concentration is 1000 mg/L. Under different dissolved oxygen concentrations, the simulation results demonstrate that both the pump-and-treat method and the funnel-and-gate system exhibit a very high level of efficiency in aquifer restoration.

Key Works: groundwater remediation, natural attenuation, pump-and-treat method, funnel-and-gate system

## I. Introduction

During recent decades, leakage from gasoline storage tanks and pipelines has allowed petroleum-derived hydrocarbons to contaminate soil and groundwater at thousands of sites in the U.S.A. Groundwater contamination due to petroleum chemicals has also attracted public attention in Taiwan recently. These highly concentrated petroleum organic chemicals may return to the life circle of human beings via groundwater flow. More and more scientists and engineers find this issue to be of great research interest, and efforts are being made to study remedial techniques for dealing with such chemicals.

The remediation of contaminated groundwater is a complicated, time-consuming and expensive task. In-situ remediation techniques, including the pump-and-treat method, the funnel-and-gate system, and natural attenuation, are employed in the treatment of contaminated groundwater. For the remediation of dissolved phase contaminant plumes, the pump-and-treat method is one of the most commonly used and most successful remediation techniques. The conventional pump-and-treat method is based on a simple concept wherein the contaminated groundwater is extracted from the subsurface and treated using an on-site treatment system. However, at many contaminated sites, the pump-and-treat method requires decades of costly operation to achieve the desired levels of cleanup (Haley et al., 1991). Mackay and Cherry (1989) indicated that when the system was operated for a long period, its limitations become apparent, resulting in inefficiency. An 'Installation Restoration Program' was implemented to deal

with contaminated groundwater due to a leaking underground tank at the Vance Air Force Base, located in Enid, Oklahoma, U.S.A. Five alternatives, which included no action, hydraulic containment or well points, intercept trenches or French drains, an in-situ physical treatment system, and in-situ bioremediation, were considered for groundwater remediation.

As a possible alternative to the pump-and-treat method, 'passive' in-situ treatment systems had also been examined in some studies (Burris and Cherry, 1992; Gillham and Burris, 1992; Hatfield et al., 1992). One passive technology receiving strong interest is the funnel-and-gate system (Starr and Cherry, 1994; Christensen and Hatfield, 1994; Sedivy et al., 1999). This system consists of a series of slurry-wall wings (funnel) that capture and divert a contaminant plume through a permeable treatment zone (gate) installed in the subsurface (Bedient et al., 1999). The treatment zone can be designed with activated carbon or zero-valent metal to treat organic contaminants or can be designed to slowly bleed oxygen and nutrients to allow in-situ biodegradation. At sites where the groundwater flow field is heterogeneous, the reactor of the funnel-and-gate system can be placed in the more permeable portions of the aquifer to enhance remediation.

National attenuation, also known as intrinsic or passive bioremediation, has also attracted much interest in recent years. It is considered a remediation option for both polluted soil and groundwater aquifers because it is cheaper and causes only minimal disturbance to the site. According to the U.S. EPA Office of Research and Development (ORD) and the Office of Solid Waste and Emergency Response (OSWER), natural attenuation is defined as the biodegradation, dispersion, dilution, sorption, volatilization, or biochemical stabilization of contaminants to reduce pollutant toxicity, mobility or volume effectively to levels that are protective of human health and the ecosystem. This technique requires extensive and thorough site characterization and monitoring to ensure that the natural attenuation processes continue to provide adequate risk protection.

Many simulation models, e.g., BIOPLUME II and 3DFATMIC (3-Dimensional subsurface Flow, fAte and Transport of MIcrobes and Chemicals), have been developed in recent years and employed as a predictive or management tool to simulate aquifer remediation at hypothetical or real-world sites. Both models combine the groundwater flow equation with the solute-transport equation. The BIOPLUME II model (Bedient et al., 1999) is a modified version of a two-dimensional transport model that is known as the method of characteristics (MOC) model and was developed by the U.S. Geological Survey (Konikow and Bredehoeft, 1978). The MOC model utilizes an alternating-direction implicit procedure to obtain a finite-difference approximation using the groundwater flow equation, and uses the method of characteristics to solve the solute-transport equation. 3DFATMIC, developed by Yeh and Cheng (1998), is a three-dimensional (3D) finite-element model which describes coupling flow and solute transport modules, and is capable of simulating the migration and fate of microbes and organic contaminants.

Bedient et al. (1999) conducted model analysis using BIOPLUME II to demonstrate how the model can be used to design bioremediation systems. In their study, three scenarios with different amounts of injected oxygen were tested to enhance biodegradation while the pump-and-treat method was used to restore the contaminated aquifer. These scenarios were also adopted in this study to conduct model simulation using 3DFATMIC. In the first scenario considered, no oxygen is injected at the field site, and biodegradation is due only to a background oxygen concentration of 3 mg/L; the purpose is to restore the aquifer simply by means of natural attenuation. In the second scenario, oxygen at a concentration of 20 mg/ L is injected throughout the injection and pumping period. In the third, oxygen at a concentration of 40 mg/L is injected into the wells. Note that the rate of injection of dissolved oxygen at various concentrations into all injection wells is always the same, i.e., 1 gpm (5.45  $\text{m}^3/\text{day}$ ). In practice, the concentration of saturated dissolved oxygen in water is about 10 mg/L (Sun et al., 1998). It should be noted that, the possible effect of bubble formation and pore clogging in the aquifer due to over-saturation of liquid oxygen was neglected in this study. Also, an additional case with a dissolved oxygen concentration of 10 mg/L in this shallow aquifer was simulated using the pump-and-treat method. In addition, use of the funnel-and-gate system for remediation of aquifer contamination was simulated. This technique is a good source control alternative for aquifer restoration. The results obtained in this

study may serve as a useful reference when assessing in-situ remedial strategies for groundwater contamination caused by petroleum organic chemicals.

## **II. Model Implementation**

3DFATMIC can simulate the migration and fate of seven components, which include three microbial populations, oxygen, nitrate, nutrients, and organic contaminants present in saturated or unsaturated aquifers. Each component has its own transport equation to represent its movement and fate during migration in the subsurface. The numerical algorithms for solving these partial differential equations use the Galerkin finite element method to approximate the flow equation and use the Eulerian-Lagrangian method, adapted zooming, and a peak capturing algorithm (LEZOOMPC) to solve each transport equation. LEZOOMPC can completely eliminate peak clipping, spurious oscillation, and numerical diffusion due to high levels of advection.

#### 1. Subsurface Flow Equation

The governing equation for flow, which describes the flow of a variable-density fluid, is basically Richard's equation. Based on the continuity of the fluid, the continuity of the solid, the consolidation of the media, and the equation of state, one can obtain the stating equation as

$$\frac{\rho}{\rho_w}\frac{d\theta}{dh}\frac{\partial h}{\partial t} = \nabla[K_s K_r (\nabla h + \frac{\rho}{\rho_w} \nabla Z)] + \frac{\rho^*}{\rho_w} q(or - \frac{\rho}{\rho_w} q),$$
(1)

where  $\rho$  is the density,  $\rho_w$  is the referenced density, *h* is the referenced pressure,  $\theta$  is the moisture content,  $K_s$  is the saturated hydraulic conductivity tensor,  $K_r$  is the relative hydraulic conductivity or relative permeability, *Z* is the potential head, *q* is the source and/or sink, and  $\rho^*$  is the density of the injected fluid.

#### 2. Transport Equations

Define the terms, r1, r2, r31, and r32, which represent the kinetics of biodegradation for microbes 1, 2, and 3, respectively, as

$$r1 = \frac{C_s}{K_{so}^1 + C_s} \frac{C_o}{K_o^1 + C_o} \frac{C_p}{K_{po}^1 + C_p},$$
 (2)

$$r2 = \frac{C_s}{K_{sn}^2 + C_s} \frac{C_n}{K_n^2 + C_o} \frac{C_p}{K_{pn}^2 + C_p},$$
 (3)

$$r31 = \frac{C_s}{K_{so}^3 + C_s} \frac{C_o}{K_o^3 + C_o} \frac{C_p}{K_{po}^3 + C_p},$$
 (4)

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$$r32 = \frac{C_s}{K_{sn}^3 + C_s} \frac{C_n}{K_n^3 + C_o} \frac{C_p}{K_{pn}^3 + C_p},$$
 (5)

where  $C_s$ ,  $C_o$ ,  $C_n$ , and  $C_p$  are the concentrations of the substrate, oxygen, nitrates, and nutrients, respectively;  $K_{so}^1$ ,  $K_o^1$ , and  $K_{po}^1$ are the retarded substrate, oxygen, and nutrient saturation constants under aerobic conditions, respectively;  $K_{sn}^2$ ,  $K_n^2$ , and  $K_{pn}^2$ are the retarded substrate, nitrate, and nutrient saturation constants under anaerobic conditions, respectively;  $K_{so}^3$ ,  $K_{sn}^3$ ,  $K_o^3$ ,  $K_n^3$ ,  $K_{po}^3$ , and  $K_{pn}^3$  are the retarded saturation constants under both aerobic and anaerobic conditions, respectively.

Transport of four components, the carbonaceous substrates, oxygen, nitrates and nutrients, in the bulk pore fluid is expressed by an advection-dispersion equation that combines source/sink terms accounting for biodegradation. The carbonaceous substrate is the pollutant, and both the oxygen and nitrate play the role of electron acceptors under aerobic and anaerobic conditions, respectively, in the subsurface flow system.

The transport equation for the substrate can then be expressed as

$$\begin{aligned} &(\theta + \rho_b K_{ds}) \frac{\partial C_s}{\partial t} + V \nabla C_s \\ &= \nabla \theta D \nabla C_s - \Lambda_s (\theta + \rho_b K_{ds}) C_s + q_{in} C_{sin} \\ &+ \left[ \frac{\rho_w}{\rho} V \nabla (\frac{\rho}{\rho_w}) - \frac{\rho^*}{\rho} q_{in} \right] C_s - \left[ (\theta + \rho_b K_{d1}) C_1 \right] \left\{ \frac{\mu_o^1}{Y_o^1} [r1] \right\} \\ &- \left[ (\theta + \rho_b K_{d2}) C_2 \right] \left\{ \frac{\mu_o^2}{Y_o^2} [r2] \right\} \\ &- \left[ (\theta + \rho_b K_{d3}) C_3 \right] \left\{ \frac{\mu_o^3}{Y_o^3} [r31] + \frac{\mu_a^3}{Y_a^3} [r32] I(C_o) \right\}, \end{aligned}$$
(6)

where  $\rho_b$  is the bulk density of the medium, *V* is the Darcy velocity, *D* is the dispersion coefficient tensor,  $\Lambda_s$  is the distribution coefficient of the dissolved substrate,  $K_{ds}$  is the transform rate constant, and  $q_{in}$  is the source rate of water;  $\mu_o^1$  and  $\mu_o^3$  are the maximum specific oxygen-base growth rate for microbe 1 and microbe 3, respectively;  $Y_o^1$  and  $Y_o^3$  are the yield coefficients for microbe 1 and microbe 3 utilizing oxygen, respectively;  $\mu_n^2$  is the maximum specific nitrate-based growth rate for microbe 2 utilizing nitrates;  $Y_n^2$  is the yield coefficient for microbe 2 utilizing nitrates. The last term on the right-hand side of Eq. (6) is the inhibition function (Widdowson *et al.*, 1998).

The transport equation for oxygen is

$$\begin{split} &(\theta + \rho_b K_{do}) \frac{\partial C_o}{\partial t} + V \nabla C_o \\ &= \nabla \theta D \nabla C_o - \Lambda_o (\theta + \rho_b K_{do}) C_o + q_{in} C_{oin} \end{split}$$

$$+ \left[\frac{\rho_{w}}{\rho}V\nabla(\frac{\rho}{\rho_{w}}) - \frac{\rho^{*}}{\rho}q_{in}\right]C_{o} - \left[(\theta + \rho_{b}K_{d1})C_{1}\right]$$

$$\cdot \left\{\gamma_{o}^{1}\mu_{o}^{1}[r1] + \alpha_{o}^{1}\lambda_{o}^{1}\left[\frac{C_{o}}{\Gamma_{o}^{1} + C_{o}}\right]\right\} - \left[(\theta + \rho_{b}K_{d3})C_{3}\right]$$

$$\cdot \left\{\gamma_{o}^{3}\mu_{o}^{3}[r31] + \alpha_{o}^{3}\lambda_{o}^{3}\left[\frac{C_{o}}{\Gamma_{o}^{3} + C_{o}}\right]\right\},$$
(7)

where  $\gamma_o^1$  and  $\gamma_o^3$  are the oxygen use coefficients for synthesis by microbe 1 and microbe 3, respectively;  $\alpha_o^1$  and  $\alpha_o^3$  are the oxygen use coefficients for energy of microbe 1 and microbe 3, respectively;  $\Gamma_o^1$  and  $\Gamma_o^3$  are the oxygen saturation constants for decay of microbe 1 and microbe 3, respectively;  $\lambda_o^1$  and  $\lambda_o^3$  are the microbial decay constants of microbe 1 and microbe 3, respectively.

The transport equations for nitrates and nutrients are similar in form to Eq. (7). The transport equation for microbes is

$$\begin{aligned} (\theta + \rho_b K_{di}) \frac{\partial C_i}{\partial t} + V \nabla C_i \\ &= \nabla \theta D \nabla C_i - \Lambda_i (\theta + \rho_b K_{di}) C_i + q_{in} C_{iin} \\ &+ \left[ \frac{\rho_w}{\rho} V \nabla (\frac{\rho}{\rho_w}) - \frac{\rho^*}{\rho} q_{in} \right] C_i + a \left[ (\theta + \rho_b K_{d1}) C_1 \right] \\ &\cdot \left\{ \mu_o^1 [r1] - \lambda_o^1 \right\} + b \left[ (\theta + \rho_b K_{d2}) C_2 \right] \left\{ \mu_n^2 [r] - \lambda_n^2 \right\} \\ &+ c \left[ (\theta + \rho_b K_{d3}) C_3 \right] \left\{ \left[ \mu_o^3 [r31] - \lambda_o^3 \right] \\ &+ \left[ \mu_n^3 [r32] I(C_o) - \lambda_n^3 I(C_o) \right] \right\}, \end{aligned}$$

where i = 1, 2, 3. For microbe 1, a = 1; b, c = 0; and the flow is under aerobic conditions. For microbe 2, b = 1; a, c = 0; and the flow is under anaerobic conditions. For microbe 3, c = 1; a, b = 0; and the flow is under both aerobic and anaerobic conditions. More detailed descriptions of those transport equations can be found in Yeh and Cheng (1998).

### III. Site and Model Information

The study site and its hydrogeological conditions were essentially adopted from Bedient *et al.* (1999). The two dimensional model domain was divided into a  $22 \times 22$  element mesh with a grid size of 15 m × 15 m to represent a shallow aquifer system. The thickness of the aquifer was 3.048 m, the bulk density was 1000 kg/m<sup>3</sup>, the effective porosity was 0.3, and hydraulic gradient was 0.001. The aquifer hydraulic conductivity was 5.267 m/day, the longitudinal dispersivity was 3.48 m, and the transverse dispersivity was 0.9 m. The aquifer was remediated for 2 years (730 days) using the pumpand-treat method with an equal flow rate of 5.45 m<sup>3</sup>/day for each well. Figure 1 shows the groundwater flow direction,

Table 1. Biodegradation Parameters and Hydrogeology

Variable	Value/unit
Growth rate, $\mu_o^{-1}$	2.10E-01 day <sup>-1</sup>
Yield coefficient, $Y_o^1$	4.26E-01 kg/kg
Microbial decay constant, $\lambda_o^{-1}$	0 1/day
Substrate saturation constant, $K_{so}^{1}$	6.54E-02 kg/m <sup>3</sup>
Oxygen saturation constant, $K_o^{-1}$	1.00E-02 kg/m <sup>3</sup>
Distribution coefficient of substrate, $K_{ds}$	0.4 m <sup>3</sup> /mg
Distribution coefficient of microbe, $K_{d1}$	1000 m <sup>3</sup> /mg

Source: Yeh and Cheng (1998).

Note: the superscript 1 represents microbe 1.





Fig. 1. (a) Location of injection wells and production wells. (b) Initial plume concentrations.

[Data from: Bedient et al. (1999).]

the existing concentration distribution of the plume, and the locations of the three injection wells and the three pumping wells, which were located upstream and downstream of the plume, respectively. The funnel-and-gate system configuration was designed to have a 90 degree apex angle with a funnel length of 85 m and a gate length of 15 m in order to produce a large composite capture zone (Starr and Cherry, 1994). Since the material of the gate was more permeable than that of the aquifer formation, the gate conductivity was chosen to be one order of magnitude larger than that of the aquifer, that is, 52.67 m/day. Since the funnel was composed of an impervious material, a funnel conductivity of  $8.64 \times 10^{-16}$  m/day was employed.

The contaminants studied were assumed to be petroleum chemicals. Therefore, biodegradation would occur under aerobic conditions with oxygen and the contaminants being the principal substrates. Accordingly, microbe 1 with a mass of  $1.77 \times 10^{-4}$  kg/m<sup>3</sup>, using oxygen-based respiration, was considered in the simulation of biodegradation. The relevant biodegradation parameters are given in Table 1 for aquifer bioremediation using 3DFATMIC as the simulation model.

## **IV. Simulation Results**

Results obtained using 3DFATMIC are plotted in Fig. 2. They show the extent of the contaminant plume when pumping occurred without enhanced biodegradation. It can be observed that the highest contaminant concentration after 730 days of pumping is 23 mg/L below the original highest concentration of 1000 mg/L. After 10 mg/L oxygen amendment for two years, the results illustrated in Fig. 3 indicate that the maximum concentration is 18 mg/L. Figure 4 shows the plume after continuous injection of 20 mg/L oxygen for 730 days. The maximum contaminant concentration in this case is 12 mg/L. When 40 mg/L oxygen is continuously injected, the resulting plume after a simulation time of 730 days as shown in Fig. 5 is only slightly lower than that shown in Fig. 4; the maximum concentration, however, is only 7 mg/L.

The overall extent of biodegradation after 2 years is summed up in Fig. 6, which shows contaminant concentrations across the centerline (from top to bottom) of the resultant plumes. Results indicate that the maximum concentration of the plume simulated using the 3DFATMIC model is slightly higher than that obtained using the BIOPLUME II model. Differences among those results may be mainly due to the following two factors. First, the models have different numerical methods. The BIOPLUME II model employs the finite



Fig. 2. Plume concentration after two years with no oxygen amendment.

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Fig. 3. Plume concentration after two years with 10 mg/L oxygen injected.



Fig. 4. Plume concentration after two years with 20 mg/L oxygen injected.

difference method and the method of characteristics, while the 3DFATMIC model uses the finite element method and the LEZOOMPC algorithm. The different numerical methods lead to somewhat different results. Second, the models use different biodegradation kinetics and parameters to model bioremediation. 3DFATMIC adopts the Monod formula to represent degradation kinetics; on the other hand, the BIOPLUME II model simply uses a factor or ratio between the oxygen consumption and contaminant to represent biodegradation. Nevertheless, these simulation results and comparisons the validity of 3DFATMIC model and the use of biodegradation parameters in 3DFATMIC.

The 3DFATMIC model was also used to simulate the case in which pumping and injection wells are not used; i.e., natural attenuation is the cleanup method used to remediate the contaminated site. Note that a background oxygen concentration of 3 mg/L was assumed throughout the whole



Fig. 5. Plume concentration after two years with 40 mg/L oxygen injected.



Fig. 6. Maximum contaminant concentrations along the plume centerline after 730 days in the simulation. The black line and dashed line show the simulation results for the BIOPLUME II model and 3DFATMIC model, respectively.



Fig. 7. Concentration distribution of petroleum chemicals under natural attenuation after 730 days. The background oxygen concentration is assumed to be 3 mg/L.



Fig. 8. Predicted concentrations versus time using natural attenuation.

Table 2.Comparison of Maximum Contaminant Concentration When<br/>Different Remediation Techniques Were Employed along with<br/>3DFATMIC for a Period of 730 Days

Remediation technique	Oxygen concentration (mg/L)			
	3	10	20	40
Natural attenuation	450			
Pump-and-treat	23	18	12	7
Funnel-and-gate	30	9.8	2	1

Note: the symbol --- represents no data.



Fig. 9. Concentration distribution of petroleum chemicals in an aquifer remediated using the funnel-and-gate system. The oxygen concentration in the gate is (a) 3 mg/L; (b)10 mg/L; (c) 20 mg/L; (d) 40 mg/L.

domain. The results based on a simulation time of 730 days are presented in Fig. 7. The highest contaminant concentration at 730 days is 450 mg/L. The simulation results plotted in Fig. 8 indicate that the system takes about 10 years to reduce the concentration level to 200 mg/L; however, this concentration still exceeds the EPA's effluent standards (100 mg/L). Therefore, the rate of contaminant degradation is extremely slow when natural attenuation is used to remediate the site. Figure 8, which shows the characteristic growth of microorganisms in exponential and declining growth phases, reveals a two-stage decay curve with sharp decay and slow decay portions (Benefield and Randall, 1972).

If the funnel-and-gate system is employed to remediate an aquifer, the concentration distribution should be different from those observed when the pump-and-treat method is used. Four oxygen concentrations of 0, 10, 20, and 40 mg/L were provided in the gate (reactor) of the funnel-and-gate system. The simulation results obtained using 3DFATMIC based on 730 days as shown in Fig. 9 indicate that the highest chemical concentrations downstream of the gate corresponding to various injected oxygen concentrations are 30, 9.8, 2, and 1 mg/L, respectively.

The maximum concentrations based on 2 years of remediation are listed in Table 2. The results obtained under a background oxygen concentration indicate that the highest contaminant plume decreases from 1000 mg/L to 450 mg/L when natural attenuation is employed, to 30 mg/L when the funnel-and-gate system is used, and to 23 mg/L when the pump-and-treat method is employed. It is evident that both the pump-and-treat method and the funnel-and-gate system are efficient ways to remediate such a contaminated site if the time needed to clean up the aquifer is crucial.

Moreover, under oxygen concentration of 10, 20, and 40 mg/L released from the gate, the funnel-and-gate system produces the lowest contaminant concentration among the three techniques.

## V. Summary and Conclusions

The groundwater system of a hypothetical site contaminated by petroleum chemicals with a plume having the concentration up to 1000 ppm has been studied. Four different oxygen concentrations, i.e., 3, 10, 20, and 40 mg/L, were considered when modeling aquifer bioremediation using the 3DFATMIC model. The background oxygen concentration in aquifer was assumed to be 3 mg/L. Oxygen concentrations of 10, 20, and 40 mg/L were used either as the injected concentration in the pump-and-treat method or as the concentration released from the gate in the funnel-and-gate system for bioremediation. The simulation results demonstrate that the rate of contaminant degradation is extremely slow when natural attenuation is used to remediate the site. On the other hand, both the pump-and-treat method and the funnel-and-gate system can efficiently achieve aquifer restoration. It is obvious that the 3DFATMIC model can provide useful results when in situ bioremedial strategies for dealing with groundwater contamination are assessed.

#### Acknowledgment

This study was supported by the National Science Council, R.O.C., under contract NSC 88-2211-E-009-003.

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# 三種地下水復育系統之模擬

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## 摘要

本文取用P.B. Bedient等人1999年書中的一個地下水受到油脂類有機物污染場址,其最高濃度達1000 mg/L。利用 一個三維污染物含生物性分解的傳輸模式結合地下水流模式3DFATMIC程式,模擬、分析藉由自然衰減、抽取處理系 統及漏斗組門系統三種方法,整治該場址的地下水。在不同的溶氧條件下,模擬結果顯示,抽取處理系統及漏斗阻門 系統皆能有效地復育受油脂類有機物污染場址。