Three-dimensional simulation of organic transport with aerobic biodegradation

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ABSTRACT A fully three-dimensional model for the simulation of advective-dispersive transport of organic contaminants with aerobic biodegradation is presented. The physical basis of the model is the coupled transport of an organic solute and dissolved oxygen, both interacting with a stationary microbial population. The numerics are based on a newly-developed time integration scheme for the finite element equations, and a conjugate gradient solver is used in the solution. The level of efficiency obtained with this scheme is sufficient to make the 3D approach viable for practical problems. Preliminary results indicate that the dimensionality effect, compared to an equivalent 2D simulation, may be profound under oxygen-limiting conditions.

INTRODUCTION

Aerobic biodegradation is an important mechanism for the remediation of aquifers contaminated by dissolved organic substances. This type of contamination is often initiated by an accidental spill of some organic substance such as an industrial solvent or gasoline, which percolates into the ground, leaving zones of residual suspended in the pore structure. The residual then forms a long-lasting source of dissolved-phase contaminants. The mechanics of this process have been described in detail by Schwille (1988). Because organic contaminants can be highly toxic at very low concentrations, they form a serious threat to the quality of our groundwater resources. Biodegradation often offers the only effective means of removal.

The process of biodegradation involving a dissolved organic contaminant, dissolved oxygen, and a microbial population has been discussed by, among others, MacQuarrie et al. (1989), who developed a successful numerical model for the simulation of biodegradation in two dimensions. This model was subsequently applied by MacQuarrie and Sudicky (1989) in a detailed sensitivity analysis involving homogeneous as well as heterogeneous media.

In natural groundwater systems, the localized nature of the source will nearly always cause the dissolved organic contaminants to spread out and migrate in three dimensions. An example is the organic plume at the Gloucester landfill near Ottawa (Jackson et al., 1985). When local-source plumes are simulated, the dimensionality effect can be profound, as was shown by Burnett and Frind (1987). Unfortunately, fully three-dimensional advective-dispersive transport models tend to be computationally demanding; with several interacting constituents the cost may become excessive. For this reason, the conventional approach is to neglect one of the transverse dispersion components and simulate the plume in 2D. A practical 3D biodegradation model for application to large-scale problems has, to our knowledge, not yet appeared in the literature.

In this paper, we give a brief description of a newly-developed fully 3D biodegradation model. The model is based on a finite element algorithm designed for the simulation of coupled non-linear transport of several chemical species in solution. The algorithm uses a unique time-integration scheme that produces a symmetric coefficient matrix for the advection-dispersion
equation (Leismann and Frind, 1989). A highly efficient symmetric conjugate gradient solver is used in the solution. The same symmetric-matrix algorithm was also applied in a large-scale simulation of the coupled transport of nitrate and sulfate in a watertable aquifer near Hannover, Germany (Frind et al., 1989).

THEORY

The governing equations for the coupled advective-dispersive transport of an organic solute in an oxygenated groundwater system, according to MacQuarrie et al. (1989), are formed by combining the advection-dispersion equation for each species with the dual-Monod equation describing the decay or growth of the species. The equations for the organic contaminant and the dissolved oxygen are, respectively,

\[
\frac{\partial}{\partial x_i} \left( \frac{D_{ij}}{R_S} \frac{\partial S}{\partial x_j} \right) - v_i \frac{\partial S}{\partial x_i} - M \frac{k}{R_S} \left( \frac{S}{K_S + S} \right) \left( \frac{A}{K_A + A} \right) = \frac{\partial S}{\partial t}
\]

and

\[
\frac{\partial}{\partial x_i} \left( \frac{D_{ij}}{R_A} \frac{\partial A}{\partial x_j} \right) - v_i \frac{\partial A}{\partial x_i} - XM \frac{k}{R_A} \left( \frac{S}{K_S + S} \right) \left( \frac{A}{K_A + A} \right) = \frac{\partial A}{\partial t}
\]

where \( x_i \) are the spatial coordinates, \( t \) is time, \( v_i \) is the average interstitial velocity, \( k \) is the maximum rate of substrate (organic) utilization, \( S \) and \( A \) are the organic and the electron acceptor (oxygen) concentrations respectively, \( K_S \) and \( K_A \) are the organic and oxygen half-utilization rates, \( X \) is the ratio of oxygen to organic consumed, \( M \) is the total microbial mass, and \( R_S \) is the solute retardation factor. The latter is defined as \( R_S = 1 + K_d \rho_b / \eta \) with \( \rho_b \) being the bulk mass density, \( \eta \) the porosity, and \( K_d \) the distribution coefficient for linear partitioning. The organic decay term (term 3 in eq. 1) is divided by \( R_S \), implying that sorbed organics are not available as substrate for microbial consumption. The dissolved oxygen is assumed to be unretarded \((R_A = 1)\). The principal components of the dispersion tensor \( D_{ij} \) assumed to be the same for both organic and oxygen, are defined as

\[
D_{xx} = \alpha_L v + D_d \quad \quad \quad \quad D_{yy} = \alpha_TH v + D_d \quad \quad \quad \quad D_{zz} = \alpha_TV v + D_d
\]

where \( \alpha_L \), \( \alpha_TH \), and \( \alpha_TV \) are the longitudinal, transverse horizontal, and transverse vertical dispersivities, respectively, and \( D_d \) is the diffusion coefficient. A more general form of \( D_{ij} \) is given by Burnett and Frind (1987).

The governing equation for microbe transport can be written in the same form as (1) or (2). However, noting that the microbe populations form essentially stationary colonies, with minimal migration (MacQuarrie et al., 1989), we will neglect the mobile component of the microbe population and use instead a simple exponential growth/decay equation of the form

\[
YMk \left( \frac{S}{K_S + S} \right) \left( \frac{A}{K_A + A} \right) \quad - \quad bM = \frac{\partial M}{\partial t}
\]

where \( Y \) is the microbial yield per unit of substrate (organic) consumed, and
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b is the first-order microbial decay coefficient. The above assumes that the microbial population dynamics are unaffected by temperature, pH, and other factors. Equations (1), (2), and (3) are non-linear and coupled.

The boundary and initial conditions to (1) and (2) are of the usual Dirichlet and Cauchy type described in standard text books. Equation (3) requires only an initial value for solution.

NUMERICAL SOLUTION

We base the numerical solution of the governing equations on a unique time-weighting scheme for the advection-dispersion equation that produces a symmetric coefficient matrix, while providing second-order accuracy in time (Leismann and Frind, 1989). This is achieved by placement of the advective term at the old time level in the time marching scheme, by introduction of a compensating artificial diffusion term, and by the optimal time weighting of all terms in the numerical equations. A symmetric conjugate gradient matrix solver (Schmid and Braess, 1988) is used to solve the numerical equations. The high level of efficiency thus obtained is crucial to the success of the 3D scheme.

The time-discretized form of equations (1) and (2) can be written as

\[
\left( \frac{\partial^2 c^{n+1}}{\partial x_i \partial x_j} - \frac{v_i}{R} \frac{\partial c^{n+1}}{\partial x_i} - \theta \lambda c^{n+1} - \frac{c^{n+1}}{\Delta t} \right)
\]

\[
= - \left( \frac{\partial^2 c^n}{\partial x_i \partial x_j} + \frac{v_i}{R} \frac{\partial c^n}{\partial x_i} - \theta \lambda c^n - \frac{c^n}{\Delta t} \right)
\]

(4)

where \(c\) stands for either \(S\) or \(A\) and \(R\) stands for either \(R_S\) or \(R_A\). The decay coefficient \(\lambda\) in (4) is defined as

\[
\lambda \equiv \lambda_S = M \frac{k}{R_S} \left( 1 \frac{A}{K_S + A} \right)
\]

(5a)

for the organic solute, and

\[
\lambda \equiv \lambda_A = XM \frac{k}{R_A} \left( 1 \frac{S}{K_S + S} \frac{1}{K_A + A} \right)
\]

(5b)

for the dissolved oxygen. The time derivative \(\partial c/\partial t\) in (4) has been replaced by a finite difference approximation, with \(n+1\) and \(n\) being the new and old time levels respectively, and \(\Delta t\) being the time step. The artificial diffusion term (Leismann and Frind, 1989) is defined as \(D_{ij}^* = v_i v_j \Delta t/2\), and the factor \(\theta\) represents the individual time weighting of the terms in the equation, with \(\theta = 1 - \theta\). The optimal time weighting factors are \(\theta_d = 1\), \(\theta_a = 1/2\), \(\theta_v = 0\), and \(\theta_y = 1/2\). With this weighting, the advective term vanishes from the \(n+1\) time level and the coefficient matrix becomes symmetric. The resulting numerical scheme is second-order accurate with respect to the advective term and behaves essentially like the fully second-order accurate Crank-Nicolson scheme.

Since the microbial growth equation (3) represents an initial value problem, the solution at the new time level \(n+1\) can be written directly in terms
of the value at the old time level \( n \). The solution is

\[
M^{n+1} = M^n e^{\lambda M \Delta t}
\]  

(6)

where

\[
\lambda \equiv \lambda_M = Yk \left( \frac{S}{K_S + S} \right) \left( \frac{A}{K_A + A} \right) - b
\]

(7)

Equation (6) is coupled to equations (4) through the microbial mass in the decay terms in (5). The non-linearity in the governing equations exists at two levels, first at the level of the individual equations for organic and oxygen, and second in the coupling between all three equations. We will handle both non-linearity levels at the same time by moving the non-linear term \( \theta A \lambda c^{n+1} \) in (4) to the right-hand side of the equations, thus linearizing the left-hand side. Equations (4) are then solved within a single iteration loop.

The microbial mass \( M \) in equations (5), which couples equations (4) and (6), can be calculated as an average value over the time step, consistent with the overall centered time weighting. The exact average is given by

\[
M = \frac{M_{t_0}}{\Delta t} \int_{t_0}^{t_0 + \Delta t} e^{\lambda_M (t - t_0)} \, dt = \frac{M_{t_0}}{\lambda_M \Delta t} \left[ e^{\lambda_M \Delta t} - 1 \right]
\]

(8)

where \( t_0 \) is the current time at the beginning of the time step, and \( M_{t_0} \) is the corresponding total microbial mass. The microbial mass is updated immediately during each iteration on the basis of the current average organic and substrate mass. This simultaneous updating of both the decay and the growth terms is essential for producing rapid convergence. After convergence is obtained for the organic and oxygen concentrations for the time step, the microbe concentration at the new time level is calculated directly from (6).

A standard Galerkin finite element formulation with isoparametric block elements (Frind and Verge, 1978) is used for the spatial integration. Other element types are also possible.

The grid resolution is governed in the flow direction by the grid Peclet number constraint \( P = v \Delta l / D \leq 2 \), where \( v \) is the magnitude of the velocity, \( \Delta l \) is the maximum grid spacing in the flow direction, and \( D \) is the longitudinal dispersion coefficient. The constraint in the transverse direction is given by the need for an adequate resolution of the concentration profiles. The time step \( \Delta t \) is controlled through the Courant number constraint \( C = v \Delta t / \Delta l \leq 1 \). Adherence to these constraints produces a solution that is essentially second-order accurate.

ILLUSTRATIVE EXAMPLE: THE EFFECT OF DIMENSIONALITY

A test problem was designed to demonstrate the favourable computational aspects of the numerical scheme proposed here for the efficient solution of large 3D problems, and to show the importance of dimensionality in the representation of the biodegradation process. For example, it has been established that oxygen availability is a critical factor for the aerobic degradation process to proceed (Barker et al., 1987; MacQuarrie and Sudicky, 1989; Borden and Bedient, 1986). However, if the problem is simulated in the vertical plane only, as in most existing models, the influence of oxygen dispersal
into the organic plume in the horizontal transverse direction would be lost. This is an important consideration since most sandy aquifers tend to be more dispersive in this direction than in the vertical transverse direction.

The test problem is comprised of the homogeneous aquifer shown in Fig. 1, with dimensions $X_L = 45$ m, $Y_L = 16$ m, and $Z_L = 6$ m. The groundwater velocity is a uniform 0.09 m/d in the $x$-direction. The organic source is represented by a 2 m cube centered at $x = 4$ m, $y = 0$, $z = 3$ m, which is assumed to be placed into the aquifer instantaneously at time $t = 0$. Initially, the domain outside the source zone is free of any dissolved organic and contains a background oxygen concentration of 3500 $\mu$g/L. The initial source concentrations are 3000 $\mu$g/L of organic solute and 1000 $\mu$g/L of oxygen.

![Figure 1](image)

*Figure 1* Three-dimensional aquifer with organic source (symmetric about $y = 0$).

The physical and biochemical transport parameter values are given in Table 1. The organic parameters are identical to those obtained by MacQuarrie and Sudicky (1989) from a lab column experiment involving the hydrocarbon toluene. The values of the dual-Monod kinetic parameters suggest the chemical is degraded relatively easily. For simplicity, the diffusion coefficient is assumed to be the same for organic and oxygen. Since mechanical dispersion dominates in the transverse horizontal direction, this assumption will not affect the dimensionality comparison.

The domain was discretized into a grid consisting of $45 \times 16 \times 30$ elements in the $x$, $y$, and $z$-directions respectively, for a total of 21,600 elements ($46 \times 17 \times 31 = 24,242$ nodes). Because of symmetry, only that half of the domain with $y \geq 0$ is simulated. In order to demonstrate the effects of dimensionality, a 2D simulation was also performed by using a one-element vertical slice from this grid, with a thickness such that the initial organic mass is identical to that of the 3D case. A time step of $\Delta t = 2$ days ($\Delta t = 1$ day up to 10 days) was used in both cases.

Figure 2 compares organic concentration profiles along the plume centreline, $y = 0$, $z = Z_L/2$, obtained from the 3D and 2D simulations at 50, 100, 200 and 300 days. At $t = 50$ days and prior, very little difference exists between the 3D and 2D results. This is because at early time, the effect of outward horizontal transverse dispersion of the organic, or inward horizontal transverse dispersion of the oxygen, has not yet fully developed so as to affect the concentrations of the plume interior. However, at later times, the horizontal transverse dispersion mechanism becomes non-negligible such that the 3D peak organic concentration values are reduced by about 20 percent compared to the 2D values. After 200 days, the peak organic concentration has declined by about a factor of 20 from its initial value to 155 $\mu$g/L.

The corresponding oxygen depletion profiles along the plume centreline
Table 1 3D Biodegradation Model Input Parameters

Dispersion Parameters:
- Longitudinal dispersivity \( (\alpha_L) \): 0.810 m
- Transverse horizontal dispersivity \( (\alpha_{TH}) \): 0.056 m
- Transverse vertical dispersivity \( (\alpha_{TV}) \): 0.005 m
- Diffusion coefficient \( (D_d) \): 0.52 \times 10^{-5} \text{ m}^2/\text{day}

Organic Parameters:
- Initial source concentration: 3000 \text{ \mu g/L}
- Background concentration: 0.0 \text{ \mu g/L}
- Retardation factor \( (R_S) \): 1.4
- Maximum utilization rate \( (k) \): 0.493 \text{ day}^{-1}
- Half-utilization constant \( (K_S) \): 655 \text{ \mu g/L}

Oxygen Parameters:
- Initial source concentration: 1000 \text{ \mu g/L}
- Background concentration: 3500 \text{ \mu g/L}
- Retardation factor \( (R_A) \): 1.0
- Oxygen/organic utilization ratio \( (X) \): 3.13
- Half-utilization constant \( (K_A) \): 100 \text{ \mu g/L}

Microbe Parameters:
- Initial source concentration: 0.23 \text{ \mu g/(1000cc)}
- Background concentration: 0.23 \text{ \mu g/(1000cc)}
- Microbial yield coefficient \( (Y) \): 0.426
- Decay coefficient \( (b) \): 0.76 \times 10^{-12} \text{ day}^{-1}

are provided in Figure 3. Because the oxygen utilization ratio is 3.13 for this problem, the oxygen depletion plume is much larger than the organic plume. The oxygen concentration decreases at first, reaching a minimum at 100 days, and increases thereafter. The rapid early decrease in oxygen concentration reflects the increase in microbial activity taking place at that time. Once again, little difference exists between the 2D and 3D simulations at early time, but the discrepancy at later time becomes quite pronounced. For example, at 200 days the maximum oxygen depletion in the 2D plume is about 50 percent greater than that in the 3D plume. In terms of net oxygen availability, the 3D model predicts about 1500 \text{ \mu g/L} at 200 days, while the 2D model predicts only about 400 \text{ \mu g/L}.

The discrepancy in the oxygen concentrations due to dimensionality can have profound effects on the predictive reliability of the biodegradation simulation. The greater oxygen depletion predicted by the 2D approach would tend to give the appearance of a greater slowing of the biodegradation process than what may actually occur. In the example, the 3D simulation suggests that aerobic conditions prevail at all times, while the 2D simulation predicts a valley of almost complete oxygen depletion along the plume centreline during the 100 to 200 day period. With the given physical and biochemical parameters, the biodegradation process eventually goes to completion for both the 2D and 3D cases because oxygen is replenished by diffusion from outlying areas in the aquifer and by upstream advection, but this process proceeds more effectively in the 3D case. If the background oxygen concentration were lower, the differences due to dimensionality would become much more pronounced.

Both the organic and oxygen plumes are slightly skewed, with tailing in
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Figure 2 Profiles of organic solute along centre of plume, 3D and 2D simulations.

Figure 3 Profiles of dissolved oxygen along centre of plume, 3D and 2D simulations.

the direction of flow, due to oxygen being supplied by advection from upstream. The centre of mass of the oxygen depletion plume appears retarded with respect to the advective advance; this is not due to phase partitioning, but to the utilization of oxygen in the biodegradation process.

In terms of computational effort, the 3D simulation with over 24,000 nodes, and 155 time steps taken to 300 days, required 126 CPU minutes on the CRAY X-MP/24. Using a convergence tolerance of 0.1 $\mu g/L$, the organic-oxygen iteration at each time step converged within 5-8 iterations during the early time period, and 1-2 iterations during the later time period. Independently of that, the preconditioned conjugate gradient solver always converged to the solution within 1-2 iterations.

CONCLUSION

The transport of biodegradable organic contaminants can be economically simulated in three dimensions. The 3D simulation provides a more realistic
prediction than a conventional 2D simulation because the mechanism of oxygen replenishment, which is essential to the maintenance of the aerobic process, is more correctly represented in 3D. As a result, the 3D model will generally predict higher oxygen concentrations than a 2D model with the same set of parameters. Although under the conditions used in the example, the net effect on the organic is not substantial, different background conditions or biochemical parameters can easily lead to longer periods of oxygen depletion, and hence will produce a larger dimensionality effect. This aspect will be investigated in further analyses.

With a CPU time of just over 2 hours on the CRAY for the fully 3D simulation to 300 days, the 3D approach is definitely viable and affordable. Regardless of the availability of 3D data in practical situations, the inclusion of the third dimension can always be expected to give a more valid representation of the physical processes.

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