Time-Splitting Methods for Advection-Diffusion-Reaction Equations Arising in Contaminant Transport

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Abbreviated Title Time-Splitting Methods

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Abstract: Two time-splitting methods for advection-diffusion-reaction problems are discussed and analyzed. Numerical results for the first approach applied to bioremediation of contaminants in groundwater are presented.

1. Introduction. In this paper we consider two time-splitting algorithms for solving systems of nonlinear advection-diffusion-reaction problems of the form:

$$\phi_i \frac{\partial c_i}{\partial t} - \nabla \cdot D(u) \nabla c_i + u \cdot \nabla c_i = (\tilde{c}_i - c_i)\tilde{q} + \phi R_i \mathcal{R}_i(c_1, c_2, \ldots, c_M), \quad x \in \Omega, \ t \in J, \tag{1.1}$$

$$(1.2) \qquad u \cdot n = D(u)\nabla c_i \cdot n = 0, \quad x \in \partial\Omega, \quad t \in J,$$

and

$$(1.3) \qquad c_i(x,0) = c_i^0(x), \quad x \in \Omega,$$

where $i = 1, 2, \ldots, M$, Ω is a bounded domain in $\mathbf{R}^d$, $J = [0, T]$. These types of equations arise in the modeling of contaminant transport in groundwater. In this context, c_i is the concentration of component i (e.g., contaminants, other nutrients, microorganisms, etc.); $\tilde{q} = \max(q, 0)$ is nonzero at source points only; u is the Darcy velocity; $D(u)$ is the hydrodynamic diffusion/dispersion tensor; $\phi_i = \phi R_i$; where ϕ is porosity and R_i is the retardation factor due to adsorption; and $\mathcal{R}_i$ are kinetic terms which account for biodegradation of contaminants, utilization of nutrients, and growth and decay of microorganisms. For $d = 2$, D generally has the form

$$(1.4) \qquad D(u) = D_m I + \frac{\alpha_l}{|u|}\begin{bmatrix} u_x^2 & u_x u_y \\ u_x u_y & u_y^2 \end{bmatrix} + \frac{\alpha_t}{|u|}\begin{bmatrix} u_y^2 & -u_x u_y \\ -u_x u_y & u_x^2 \end{bmatrix},$$

where D_m is the molecular diffusivity and α_l and α_t are the longitudinal and transverse dispersivities, respectively. Furthermore, in (1.1) $\tilde{c}$ is specified at injection wells and $\tilde{c} = c$ at production wells.

For convenience, we assume (1.1) is Ω-periodic; i.e., we assume all functions in (1.1) are spatially Ω-periodic. This is physically reasonable, since no-flow boundaries are generally treated by reflection, and because in general interior flow patterns are much more important than boundary effects. Thus, the no-flow boundary conditions above can be dropped.

This paper is divided into six additional sections. In the second section we briefly describe the importance of modeling the fate and transport of contaminants and the possible potential of biorestoration in cleanup strategies. In the third section we establish mathematical notation. The modified method of characteristics (MMOC) for linear advection problems with u assumed to be given is described in Section 4. In Section 5 a time-splitting method is formulated in which the advection-diffusion equations are treated by the MMOC and subsequently, reactions are computed using many small time steps. Theoretical error estimates for this algorithm are given, including a new result in which the dispersion tensor is assumed to be only postive semi-definite. This time-splitting algorithm has been used extensively to model contaminant transport and biodegradation in two and three spatial dimensions [2, 3, 4, 15, 16, 17]. Some numerical experiments of *in-situ* biorestoration are presented in Section 6. A new time-splitting in which the reactions are computed along characteristics is formulated and analyzed in Section 7.

2. Contaminant Transport and Biodegration. Groundwater contamination is of growing concern to the United States. Of the fifty largest cities in the United States, over thirty-four obtain their drinking water from groundwater. In addition groundwater supplies the needs of over 50% of the U. S. population. At the same time the growth of industrial wastes has risen from approximately 57 million metric tons in 1980 (with as little as 10% disposed over properly) to 265 million metic tons in 1989. These contaminants include inorganic chemicals such as arsenic, nitrate, fluoride, radium, and lead and organic chemicals such as gasoline, DDT, PCB, and detergents, and volatile organics such as chlorinated solvents (TCE), carbon tetrachloride, and vinyl chloride, biological matter, and radioactive compounds. Sources of groundwater contamination include land disposal of waste materials, water wells, sewage and waste water disposal systems, leaks and spills, oil, gas and mining activities, agricultural practices, chemical injection wells, and stormwater runoff.

One of the most promising restoration techniques is bioremediation. Here indigenous microflora are used to remove subsurface pollutants by biodegradation. Numerous laboratory and field studies have shown microbes degrade various compounds in both aerobic and anaerobic conditions and control contaminant movement; see [10, 11, 13, 14] and the references therein. *In-situ* biodegradation involves enhancement of these natural processes by introducing nutrients and dissolved oxygen into the system.

The biorestoration process involves transport of substrates, nutrients, and microorganisms, and interaction of components in the aqueous phase with the solid phase through adsorption and biodegradation. Many factors must be considered, such as the characteristics of the organisms, the primary substrate and nutrients, cometabolic substrates, contaminant concentration and location, available electron acceptors, and environmental conditions, including adsorption, ion exchange, the effect of colloidal particles, soil pH and mineral composition, temperature, rock heterogeneity, fractures, effective dispersivity, and changes in rock properties due to microbial activity. Much of the field and laboratory data used to estimate these effects is incomplete and/or inexact, and is scale-dependent.

Mathematical modeling serves as a valuable link between the experimental studies and scientific theory. It is especially useful in understanding the mechanisms of interactive transport, sorption, and biodegradation, analyzing the sensitivity and significance of various parameters in a model and providing insight into field data collection activities, and efficiently investigating various strategies for remediation and mitigation.

In Section 6 we briefly describe the governing equation of flow and transport with biodegradation in a saturated porous medium. For simplicity we assume linear sorption and aerobic conditions. More general conditions such as the Michaelis-Menten kinetics can be treated with the numerical techniques discussed in this paper.

The coupled nonlinear advection-diffusion-reaction system consists of M_s electron donors or substrates (contaminants) and M_n electron acceptors or nutrients and a system of M_x ordinary differential equations involving microbial mass. The flow is given by Darcy's Law and the continuity equation. Transport of microbes can also be treated if one assumes a system of advection-diffusion-reaction equations rather than a system of ordinary differential equations.

3. Notation and definitions. On Ω, let $L^2(\Omega)$ and $L^\infty(\Omega)$ denote the standard Banach spaces, with norms $||\cdot||$ and $||\cdot||_\infty$, respectively. Let $H^m(\Omega)$ and $W^m_\infty(\Omega)$ denote the standard Sobolev spaces, with norms $||\cdot||_m$, and $||\cdot||_{W^m_\infty}$, respectively. Let $(\cdot,\cdot)$ denote the L^2 inner product on Ω.

Let $[a,b] \subset [0,T]$ denote a time interval, $X = X(\Omega)$ a Banach or Sobolev space. To incorporate time dependence, we use the notation $L^p(a,b;X)$ and $||\cdot||_{L^p(a,b;X)}$ to denote the space and norm, respectively, of X-valued functions f with the map $t \to ||f(\cdot,t)||_X$ belonging to $L^p(a,b)$. If $[a,b] = [0,T]$, we simplify our notation and write $L^p(X)$ for $L^p(0,T;X)$.

Let $\Delta t = T/N^*$ for some positive integer N^*, $t^n = n\Delta t$, $n = 0,\dots,N^*$, and $f^n = f(t^n)$. In our time-splitting procedure we will also use a "small" time step Δt_s, with $\Delta t_s = \Delta t/N$, where N is another positive integer. Let $f^{j,n} = f(t^n + j\Delta t_s)$, $j = 0,\dots,N$.

Finally, let $\mathcal{M}_h$ denote a finite dimensional subspace of $H^1(\Omega)$ consisting of continuous piecewise polynomials of degree $\leq k$ on a quasi-uniform mesh of diameter $\leq h$.

4. Description of the method. Before defining our time-splitting procedure we comment on the MMOC applied to the single advection-diffusion equation

$$\phi_i \frac{\partial c_i}{\partial t} + u \cdot \nabla c_i - \nabla \cdot D \nabla c_i = 0, \tag{4.5}$$

where $\phi_i = \phi R_i$. We write

$$\phi_i \frac{\partial c_i}{\partial t} + u \cdot \nabla c_i$$

as a directional derivative. Let τ_i denote the unit vector in the direction (u, ϕ_i) in $\Omega \times J$ and set $\psi_i = \sqrt{|u|^2 + \phi_i^2}$. Then one obtains

$$\psi_i \frac{\partial c}{\partial \tau_i} - \nabla \cdot D \nabla c_i = 0. \tag{4.6}$$

Let

$$\check{x} = x - u_i \Delta t,$$

and

$$\check{c}(x) = c(\check{x}),$$

where $u_i = u/\phi_i$. Then

$$\psi_i \frac{\partial c_i}{\partial \tau_i} \approx \phi_i \frac{c_i^n - \check{c}_i^{n-1}}{\Delta t}.$$

For analysis of the MMOC procedure and application to problems in porous media the reader is referred to [7, 8, 9, 12].

In our error analysis, we will compare the approximate solution to a projection in $\mathcal{M}_h$. Let $\tilde{C}_i^n \in \mathcal{M}_h$ satisfy

$$\left(\phi_i \frac{\tilde{C}_i^n - \check{\tilde{C}}_i^{n-1}}{\Delta t}, \chi\right) + (D\nabla \tilde{C}_i^n, \nabla \chi) = (q(\tilde{c}_i^n - \tilde{C}_i^n), \chi) + (\mathcal{R}_i(c^n), \chi), \tag{4.7}$$

$$\chi \in \mathcal{M}_h, \quad n \geq 1, \quad i = 1, \ldots, M,$$

and

$$\tilde{C}_i^0(x) = C_i^e(x, 0). \tag{4.8}$$

Here, if D is uniformly positive definite, that is, $0 < D_* \leq D(u)$, $C_i^e(x,t)$ is the "elliptic projection" of $c(x,t)$ into the space $\mathcal{M}_h$, given by

$$(D\nabla C_i^e, \nabla \chi) + (C_i^e, \chi) + (\tilde{q} C_i^e, \chi) = (D\nabla c_i, \nabla \chi) + (c_i, \chi) + (\tilde{q} c_i, \chi), \quad \chi \in \mathcal{M}_h.$$

When D is positive semidefinite, C_i^e is the L^2 projection, given by

$$(C_i^e(\cdot, t) - c_i(\cdot, t), \chi) = 0, \quad \chi \in \mathcal{M}_h. \tag{4.9}$$

5. The first time-splitting approach. The time-split procedure for solving (1.1) is a collection of maps $C_i : \{t^0, t^1, \ldots, t^{N^*}\} \to \mathcal{M}_h$ defined by

$$\left(\phi_i \frac{C_i^n - \check{\bar{C}}_i^{N,n-1}}{\Delta t}, \chi\right) + (D\nabla C_i^n, \nabla\chi) = (q(\tilde{c}_i^n - C_i^n), \chi), \tag{5.10}$$
$$\chi \in \mathcal{M}_h, \quad n \geq 1, \quad i = 1, \ldots, M,$$

where

$$\bar{C}_i^0(x) = \tilde{C}_i^0(x). \tag{5.11}$$

We will discuss two different choices for the function $\bar{C}_i(x)$. The first choice, presented here and analyzed in [15], is based on solving the system of ordinary differential equations

$$c_t = \mathcal{R}(c), \quad c = (c_1, \ldots, c_M), \quad \mathcal{R} = (\mathcal{R}_1, \ldots, \mathcal{R}_M) \tag{5.12}$$

along lines of constant x. For simplicity, in this paper, we will assume the system (5.12) is approximated by explicit Euler; however, our analysis can be generalized to higher-order explicit or implicit techniques [15]. In this case, $\bar{C}^{N,n-1}$ is defined recursively by

$$\bar{C}_i^{0,n-1} = C_i^{n-1}, \tag{5.13}$$

and for $j = 1, \ldots, N$,

$$\bar{C}_i^{j,n-1} = \bar{C}_i^{j-1,n-1} + \Delta t_s \mathcal{R}_i(\bar{C}^{j-1,n-1}). \tag{5.14}$$

Note that (5.10) and (5.13)-(5.14) represent a time-splitting approach, where at the beginning of each time step, we first solve (5.12) using explicit Euler, and use the results of this step as the "initial condition" for the transport step, where advection and diffusion are approximated by the MMOC.

In [15], the following error estimate is derived for the method outlined above.

THEOREM 5.1. *Let c_i satisfy (1.1) and assume the data and c_i are sufficiently smooth, $0 \leq \phi_* \leq \phi_i$, $D(u)$ is uniformly positive definite, and $\mathcal{R}_i$ is Lipschitz continuous, $i = 1, \ldots, M$. Let $C_i \in \mathcal{M}_h$ be the approximation given by (5.10), (5.13)-(5.14). Then there exists a positive constant K, such that*

$$\max_n \|\phi_i^{1/2} C_i^n - c_i^n\| \leq K(\Delta t + h^{(r+1)}), \quad i = 1, \ldots, M, \tag{5.15}$$

where r is the maximum degree of polynomials in $\mathcal{M}_h$.

The smoothness assumptions on coefficients and data needed to prove Theorem 5.1 are given in [5, 15].

Theorem 5.1 can be modified to treat the case where $D(u)$ is positive semidefinite using the techniques in [5]; in this case the exponent on h is r. This rate is optimal for the case $r = 2$ [6].

6. Numerical results for first time-splitting approach. In this section, we present some two-dimensional results for a three-component system. Let $c_1 = S$ denote the concentration of contaminant, $c_2 = O$ the concentration of dissolved oxygen, and $c_3 = M$ the concentration of microorganisms. We will assume microorganisms are immobile, and that biodegradation is described by the Monod kinetic equations. Thus, the system of equations to be solved is

$$(6.16) \qquad \phi R_1 \frac{\partial S}{\partial t} - \nabla \cdot D(u)\nabla S + u \cdot \nabla S = (\tilde{c}_1 - S)\tilde{q} + \phi R_1 \mathcal{R}_1(S, O, M),$$

$$(6.17) \qquad \phi \frac{\partial O}{\partial t} - \nabla \cdot D(u)\nabla O + u \cdot \nabla O = (\tilde{c}_2 - O)\tilde{q} + \phi \mathcal{R}_2(S, O, M),$$

and

$$(6.18) \qquad \frac{\partial M}{\partial t} = \mathcal{R}_3(S, O, M),$$

where

$$(6.19) \qquad R_1(S, O, M) = -M \cdot k \cdot \left(\frac{S}{K_S + S}\right) \cdot \left(\frac{O}{K_O + O}\right)$$

$$(6.20) \qquad R_2(S, O, M) = -M \cdot k \cdot f \cdot \left(\frac{S}{K_S + S}\right) \cdot \left(\frac{O}{K_O + O}\right)$$

and

$$(6.21) \quad R_3(S, O, M) = M \cdot k \cdot Y \cdot \left(\frac{S}{K_S + S}\right) \cdot \left(\frac{O}{K_O + O}\right) + k_c \cdot Y \cdot OC - b \cdot M.$$

In these equations, k is the maximum substrate utilization rate per unit mass microorganisms, Y is the microbial yield coefficient, K_S is the substrate half saturation constant, K_O is the oxygen half saturation constant, b is the microbial decay rate, f is the ratio of oxygen to substrate consumed, k_c is the first order decay rate of natural organic carbon, and OC is the natural organic carbon concentration. Further details of this formulation and parameter selection can be found in [1].

In the simulations described below, we assumed a rectangular, heterogeneous aquifer of 2000 by 800 feet, with no-flow boundary conditions on the horizontal boundaries and pressure (inflow and outflow) boundary conditions on the vertical boundaries, with a pressure drop of 28 feet across the region. Figure 1 shows a contour plot of an initial substrate plume, which we generated numerically by introducing sources of contamination throughout the region, and assuming constant, nonzero background concentrations of dissolved oxygen and microorganisms. Figure 2 shows the amount of oxygen left in the system after five years of contaminant injection and biodegradation.

Next, we place a series of injection and production wells throughout the system which inject dissolved oxygen and produce a mixture of dissolved oxygen and substrate. The intent is to stimulate biodegradation and contain the contaminant plume. One problem of interest in determining biorestoration strategies is to determine "optimal" flow rates and well patterns. Here we have adopted a strategy based on graphically monitoring the contaminant plume movement, and changing well locations and flow rates at one year intervals. In Figure 3-5, we present the contaminant solution at three, five, and seven years. The well locations are superimposed on the plots, a cross denotes an injection well and a circle a production well. Note that after seven years, most of the contaminant has been removed.

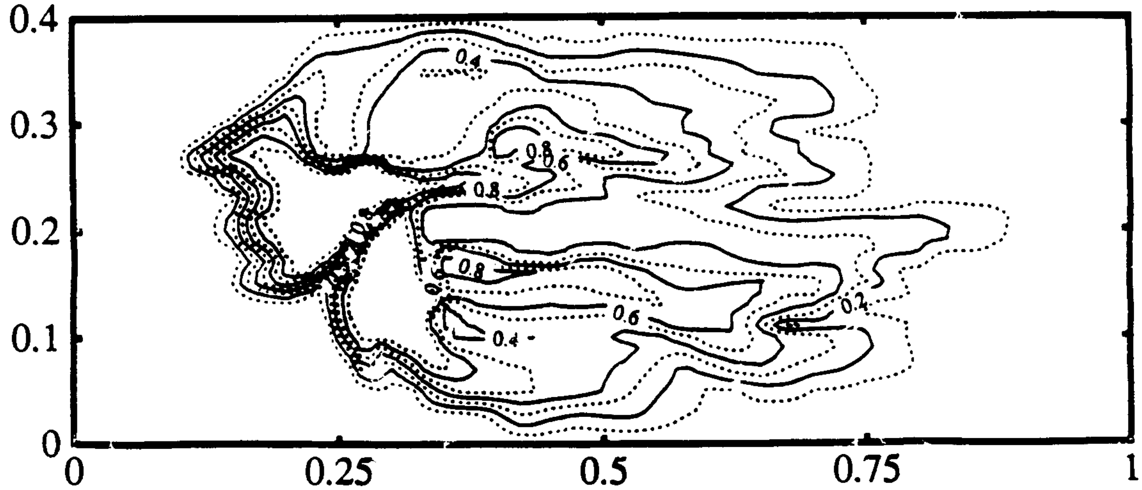

FIG. 1. *Initial substrate plume*

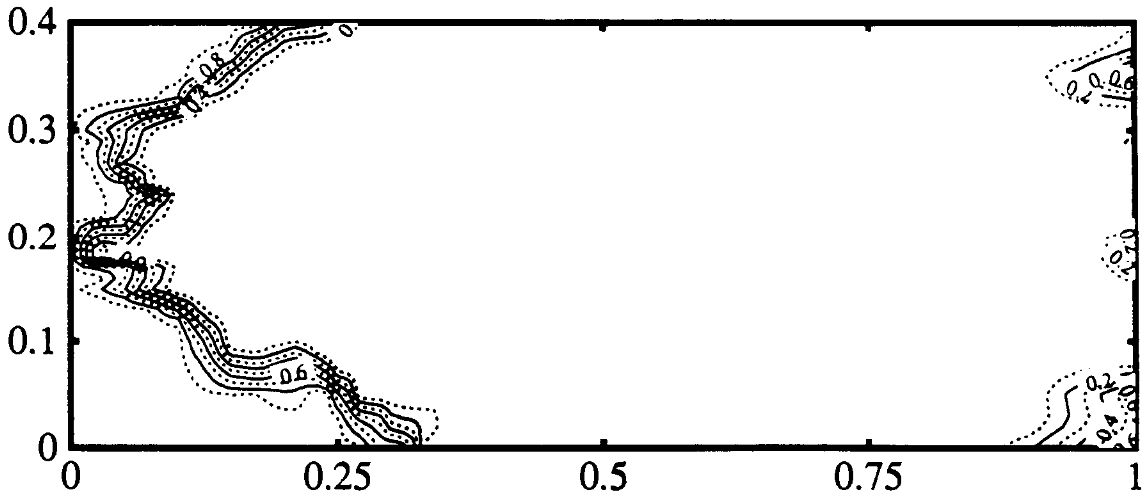

FIG. 2. *Initial oxygen plume*

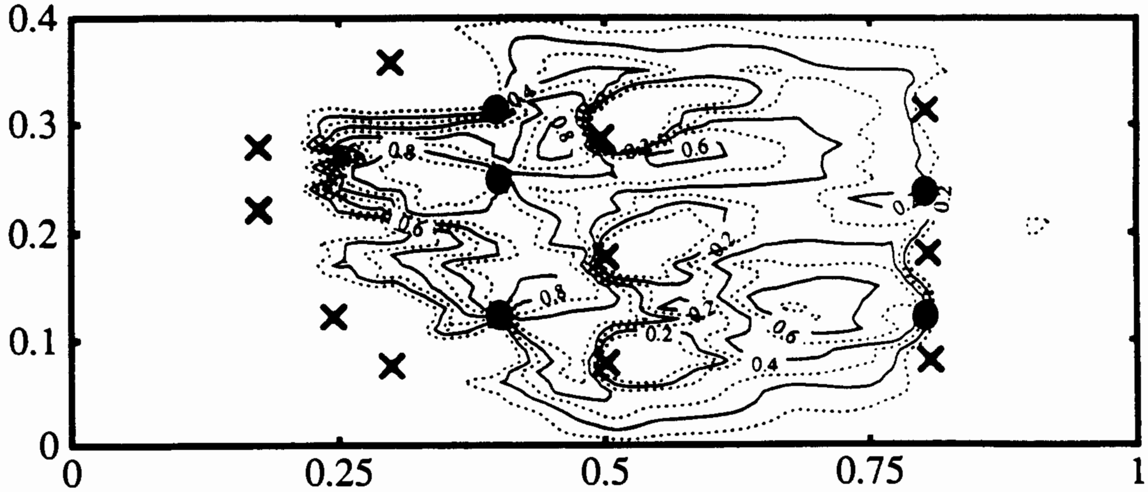

FIG. 3. *Substrate plume after 3 years of biorestoration*

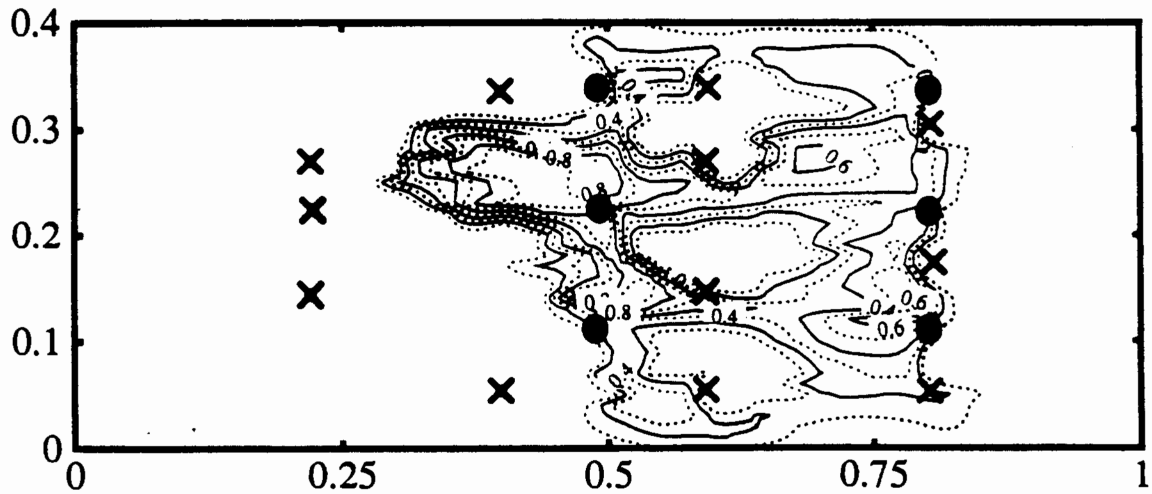

FIG. 4. *Substrate plume after 5 years of biorestoration*

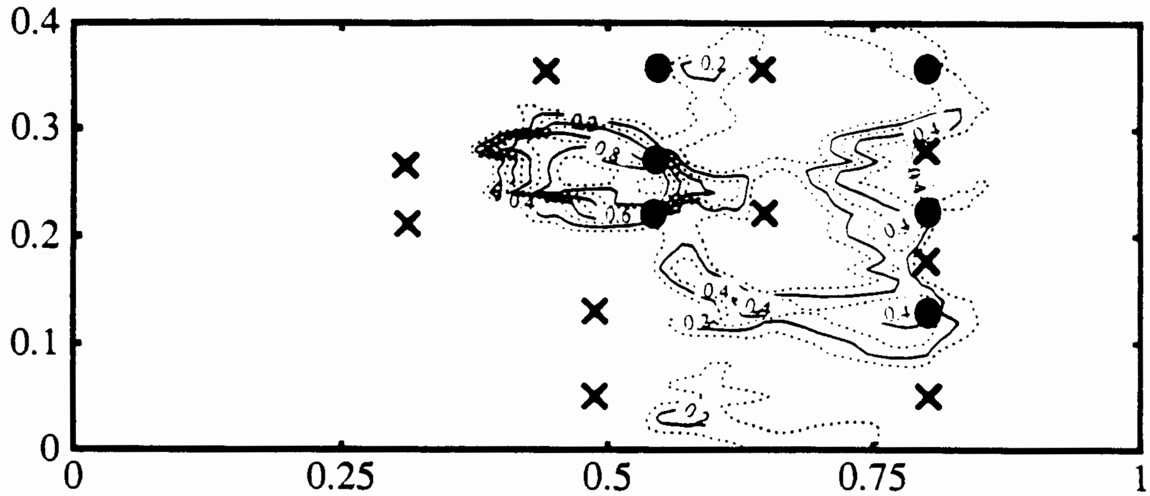

FIG. 5. *Substrate plume after 7 years of biorestoration*

The physical parameters used in the simulations were $\phi = 0.25$, $\alpha_l = 10$ feet, and $\alpha_t = 1$ foot. The flow rates varied between 20 and 40 square feet / day. The biodegradation parameters were $k = 0.17$/day, $k_c = 2.7 * 10^{-6}$, $OC = 750$, $F = 3.0$, $Y = 0.13$, $K_S = 0.13$ mg/l, $K_O = 0.1$ mg/l, and $b = .01$/day.

In the numerical simulator, different non-uniform grids were used with each change in the well pattern, with grids concentrated around the wells. A constant Δt of 4 days was assumed, and $\Delta t_s = .004$ days.

7. The second time-splitting approach. We now present another time-splitting approach. At each time step, we advect and react by solving

$$\psi_i \frac{\partial c_i^R}{\partial \tau_i} = \phi_i \mathcal{R}_i(c_1, c_2, \ldots, c_{i-1}, c_i^R, c_{i+1}, \ldots, c_M), \tag{7.22}$$

and the solution generated from this step is used as initial data for the equation

$$\phi_i \frac{\partial c_i^D}{\partial t} - \nabla \cdot D \nabla c_i^D = \tilde{q}(\tilde{c}_i - c_i^D). \tag{7.23}$$

That is, we advect and react along the characteristic direction . For problems with small diffusion, this approach should give a more accurate representation of the physics of the problem, since the reactions "move with the flow."

Let $\tau_{i,j}^{n-1}(x)$ denote the point $(x - u_i(N-j)\Delta t_s, t^{n-1})$, and $\tau_{i,j}^{j,n-1}(x) = (x - u_i(N - j)\Delta t_s, t^{n-1} + j\Delta t_s)$. Assume $C_1^{n-1}, \ldots, C_M^{n-1} \in \mathcal{M}_h$ are known. Define

$$\bar{C}_i^{0,n-1}(x) = C_i^{n-1}(\check{x}). \tag{7.24}$$

For $j = 0, \ldots, N-1$, set

$$\begin{aligned} \bar{C}_i^{j+1,n-1} &= \bar{C}_i^{j,n-1} + \Delta t_s \mathcal{R}_i(C_1(\tau_{i,j}^{n-1}), C_2(\tau_{i,j}^{n-1}), \ldots, C_{i-1}(\tau_{i,j}^{n-1}), \\ &\qquad \bar{C}_i^{j,n-1}, C_{i+1}(\tau_{i,j}^{n-1}), \ldots, C_M(\tau_{i,j}^{n-1})). \end{aligned} \tag{7.25}$$

Note that (7.24)-(7.26) is an explicit Euler approximation to (7.22). Next, we solve

$$(7.26)\quad \left(\phi_i \frac{C_i^n - \bar{C}_i^{N,n-1}}{\Delta t}, \chi\right) + (D\nabla C_i^n, \nabla\chi) = (\tilde{q}(\tilde{c}_i^n - C_i^n), \chi),$$
$$\chi \in \mathcal{M}_h, \quad n \geq 1, \quad i = 1, \ldots, M.$$

In the error analysis below, K will represent a generic constant, independent of discretization parameters.

In order to analyze (7.24)-(7.26), let $\zeta_i = C_i - \tilde{C}_i$, and $\xi_i = c_i - \tilde{C}_i$, where $\tilde{C}_i$ is given by (4.7) and (4.8). Then, subtract (4.7) from (7.26) and use (7.24) and (7.26) to obtain

$$(7.27)\quad \left(\phi_i \frac{\zeta_i^n - \check{\zeta}_i^{n-1}}{\Delta t}, \chi\right) + (D\nabla\zeta_i^n, \nabla\chi) + (\tilde{q}\zeta_i^n, \chi)$$
$$= (\phi_i(\bar{\mathcal{R}}_i{}^n - \mathcal{R}_i(c^n)), \chi), \quad \chi \in \mathcal{M}_h, \quad n \geq 1, \quad i = 1, \ldots, M,$$

where

$$(7.28)\quad \bar{\mathcal{R}}_i{}^n = \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t} \mathcal{R}_i(C_1(\tau_{i,j}^{n-1}), C_2(\tau_{i,j}^{n-1}), \ldots, C_{i-1}(\tau_{i,j}^{n-1}),$$
$$\bar{C}_i^{j,n-1}, C_{i+1}(\tau_{i,j}^{n-1}), \ldots, C_M(\tau_{i,j}^{n-1})).$$

Set $\chi = \zeta_i^n$ to obtain

$$(7.29)\quad \frac{||\phi_i^{1/2}\zeta_i^n||^2 - ||\phi_i^{1/2}\check{\zeta}_i^{n-1}||^2}{\Delta t} + ||D^{1/2}\nabla\zeta_i^n||^2 + ||q^{1/2}\zeta_i^n||^2$$
$$\leq |(\phi_i(\bar{\mathcal{R}}_i{}^n - \mathcal{R}_i(c^n)), \zeta_i^n)|.$$

In order to analyze the term on the right side of (7.29), assume a two component system ($M = 2$). Assume $i = 1$ and drop the subscript on $\mathcal{R}$ momentarily, and consider

$$(7.30)\quad \left|\phi_i \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t} \mathcal{R}(\bar{C}_1^{j,n-1}, C_2(\tau_{1,j}^{n-1})) - \mathcal{R}(c_1^n, c_2^n)\right|$$
$$\leq \phi_1 \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t} \Big\{|\mathcal{R}(\bar{C}_1^{j,n-1}, C_2(\tau_{1,j}^{n-1})) - \mathcal{R}(\bar{c}_1^{j,n-1}, C_2(\tau_{1,j}^{n-1}))|$$
$$+|\mathcal{R}(\bar{c}_1^{j,n-1}, C_2(\tau_{1,j}^{n-1})) - \mathcal{R}(\bar{c}_1^{j,n-1}, c_2(\tau_{1,j}^{n-1}))|$$
$$+|\mathcal{R}(\bar{c}_1^{j,n-1}, c_2(\tau_{1,j}^{n-1})) - \mathcal{R}(\bar{c}_1^{j,n-1}, c_2(\tau_{1,j}^{j,n-1}))|$$
$$+|\mathcal{R}(\bar{c}_1^{j,n-1}, c_2(\tau_{1,j}^{j,n-1})) - \mathcal{R}(c_1(\tau_{1,j}^{j,n-1}), c_2(\tau_{1,j}^{j,n-1}))|$$
$$+|\mathcal{R}(c_1(\tau_{1,j}^{j,n-1}), c_2(\tau_{1,j}^{j,n-1})) - \mathcal{R}(c_1^n, c_2^n)|\Big\}$$
$$\equiv |T_1| + |T_2| + |T_3| + |T_4| + |T_5|.$$

Here $\bar{c}_1^{0,n-1}(x) = c_1^{n-1}(\check{x})$, and for $j = 0, \ldots, N-1$,

$$(7.31)\quad \bar{c}_1^{j,n-1} = \bar{c}_1^{j-1,n-1} + \Delta t_s \mathcal{R}(\bar{c}_1^{j-1,n-1}, c_2(\tau_{1,j}^{j,n-1})).$$

Let L be the Lipschitz constant for $\mathcal{R}$. By (7.24), (7.26), and (7.31), we find

$$(7.32)\ |\bar{C}_1^{j,n-1} - \bar{c}_1^{j,n-1}| \leq |\bar{C}_1^{0,n-1} - \bar{c}_1^{0,n-1}| + L\sum_{k=0}^{j-1} \frac{\Delta t_s}{\Delta t}\left[|\bar{C}_1^{k,n-1} - \bar{c}_1^{k,n-1}| + |C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{k,n-1})|\right].$$

For the last term on the right side of (7.32) we note that

$$(7.33)\ \begin{aligned} |C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{k,n-1})| &\leq |C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{n-1})| + |c_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{k,n-1})| \\ &\leq |C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{n-1})| + Kj\Delta t_s \end{aligned}$$

by the definition of $\tau_{i,j}^{n-1}$ and $\tau_{i,j}^{j,n-1}$. Thus

$$(7.34)\ |\bar{C}_1^{j,n-1} - \bar{c}_1^{j,n-1}| \leq |\bar{C}_1^{0,n-1} - \bar{c}_1^{0,n-1}| + K\Delta t + L\sum_{k=0}^{j-1} \frac{\Delta t_s}{\Delta t}\left[|\bar{C}_1^{k,n-1} - \bar{c}_1^{k,n-1}| + |C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{k,n-1})|\right].$$

Applying this recursion relationship to each entry in the sum we find

$$(7.35)\ \begin{aligned} &|\bar{C}_1^{j,n-1} - \bar{c}_1^{j,n-1}| \\ &\leq K\left[|\bar{C}_1^{0,n-1} - \bar{c}_1^{0,n-1}| + \Delta t + \sum_{k=0}^{N-1} \frac{\Delta t_s}{\Delta t}|C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{n-1})|\right], \end{aligned}$$

where K is a sufficiently large constant which grows exponentially with N. Multiplying by $\frac{\Delta t_s}{\Delta t}$ and summing on j, $j = 0, \ldots, N-1$, we find

$$(7.36)\ |T_1| \leq K\phi_1\left[|C^{n-1}(\check{x}) - c^{n-1}(\check{x})| + \Delta t + \sum_{k=0}^{N-1} \frac{\Delta t_s}{\Delta t}|C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{n-1})|\right].$$

Similarly

$$(7.37)\qquad |T_2| \leq K\phi_1 \sum_{k=0}^{N-1} \frac{\Delta t_s}{\Delta t}|C_2(\tau_{1,k}^{n-1}) - c_2(\tau_{1,k}^{n-1})|.$$

By the definitions of $\tau_{1,j}^{n-1}$ and $\tau_{1,j}^{j,n-1}$,

$$(7.38)\qquad \begin{aligned} |T_3| &\leq L\phi_1 \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t}|c_2(\tau_{1,j}^{n-1}) - c_2(\tau_{1,j}^{j,n-1})| \\ &\leq K\sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t} j\Delta t_s \\ &\leq K\Delta t. \end{aligned}$$

For T_4, we have

$$(7.39)\qquad |T_4| \leq L\phi_1 \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t}|\bar{c}_1^{j,n-1} - c(\tau_{1,j}^{j,n-1})|.$$

Let c_1^R denote the solution to the initial value problem (7.22), with initial condition $c_1^R(0) = c_1^{n-1}(\check{x})$, then

$$(7.40) \qquad |\bar{c}_1^{j,n-1} - c(\tau_{1,j}^{j,n-1})| \le |\bar{c}_1^{j,n-1} - c_1^R(\tau_{1,j}^{j,n-1})| + |c_1^R(\tau_{1,j}^{j,n-1}) - c_1(\tau_{1,j}^{j,n-1})|.$$

The first term represents the error in applying Euler's method to (7.22), thus

$$(7.41) \qquad |\bar{c}_1^{j,n-1} - c_1^R(\tau_{1,j}^{j,n-1})| \le K\Delta t_s ||(c_1^R)_{\tau\tau}||_\infty.$$

Writing the differential equation (1.1) as

$$(7.42) \qquad \psi_1 \frac{\partial c_1}{\partial \tau_1} - \nabla \cdot D\nabla c_1 = \phi_1 \mathcal{R}(c_1, c_2),$$

and integrating (7.42) and (7.22) from $\tau_* = \tau_{1,0}^{n-1}$ to $\tau^* = \tau_{1,j}^{j,n-1}$, we find

$$\begin{aligned}(7.43) \qquad & |c_1^R(\tau_{1,j}^{j,n-1}) - c_1(\tau_{1,j}^{j,n-1})| \\ & \le \int_{\tau_*}^{\tau^*} |\psi_1^{-1}(\nabla \cdot D\nabla c_1)| d\tau + \int_{\tau_*}^{\tau^*} |\psi_1^{-1}\phi_1(\mathcal{R}(c_1^R, c_2) - \mathcal{R}(c_1, c_2))| d\tau \\ & \le K\left[\Delta t + \int_{\tau_*}^{\tau^*} |c_1^R - c_1| d\tau\right].\end{aligned}$$

Thus, by Gronwall's Lemma applied to (7.43), and (7.41),

$$(7.44) \qquad |T_4| \le K\Delta t.$$

Finally,

$$\begin{aligned}(7.45) \qquad |T_5| & \le L\left[|c_1(\tau_{1,j}^{j,n-1}) - c_1^n| + |c_2(\tau_{1,j}^{j,n-1}) - c_2^n|\right] \\ & \le K\Delta t.\end{aligned}$$

Combining the estimates for $T_1 - T_5$, we find

$$\begin{aligned}& \left|\phi_1 \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t} \mathcal{R}(\bar{C}_1^{j-1,n-1}(x), C_2(\tau_{1,j}^{n-1}(x)) - \mathcal{R}(c_1^n(x), c_2^n(x))\right| \\ & \quad \le K\left\{\Delta t + \left|\phi_1(C_1^{n-1}(\check{x}) - c_1^{n-1}(\check{x}))\right| + \phi_1 \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t} \left|C_2(\tau_{i,j}^{n-1}(x)) - c_2(\tau_{i,j}^{n-1}(x))\right|\right\} \\ & \quad \le K\left\{\Delta t + |\phi_1 \check{\zeta}_1^{n-1}(x)| + |\phi_1 \check{\xi}_1^{n-1}(x)| \right. \\ & \qquad \left. + \phi_1 \sum_{j=0}^{N-1} \frac{\Delta t_s}{\Delta t}\left[|\zeta_2^{n-1}(\tau_{i,j}^{n-1}(x))| + |\xi_2^{n-1}(\tau_{i,j}^{n-1}(x))|\right]\right\}.\end{aligned}$$

Under the assumption of no-flow boundary conditions, one can show [5]

$$||\check{f}|| \le ||f||$$

for any $L^2(\Omega)$ function f. Thus, assuming $\phi_2/\phi_1 \leq K$,

$$\begin{aligned}
|(\phi_1(\bar{\mathcal{R}_1}^n - \mathcal{R}_1(c^n)), \zeta_1^n)| &\leq K||\phi_1^{1/2}\bar{\mathcal{R}_1}^n - \mathcal{R}_1(c^n)||^2 + K||\phi_1^{1/2}\zeta_1^n||^2 \\
&\leq K\left[\Delta t^2 + ||\phi_1^{1/2}\zeta_1^{n-1}||^2 + ||\phi_1^{1/2}\xi_1^{n-1}||^2\right. \\
&\quad \left. + ||\phi_2^{1/2}\zeta_2^{n-1}||^2 + ||\phi_2^{1/2}\xi_2^{n-1}||^2\right] \\
&\quad + K||\phi_1^{1/2}\zeta_1^n||^2.
\end{aligned}$$

Substituting into (7.29) and rearranging some terms we find

$$\begin{aligned}
(7.46)\quad & \frac{||\phi_1^{1/2}\zeta_1^n||^2 - ||\phi_1^{1/2}\zeta_1^{n-1}||^2}{\Delta t} + ||D^{1/2}\nabla\zeta_1^n||^2 + ||q^{1/2}\zeta_1^n||^2 \\
&\leq \frac{||\phi_1^{1/2}\check{\zeta}_1^{n-1}||^2 - ||\phi_1^{1/2}\zeta_1^{n-1}||^2}{\Delta t} + K||\phi_1^{1/2}\zeta_1^n||^2 \\
&\quad + K\left[\Delta t^2 + ||\phi_1^{1/2}\zeta_1^{n-1}||^2 + ||\phi_1^{1/2}\xi_1^{n-1}||^2 + ||\phi_2^{1/2}\zeta_2^{n-1}||^2 + ||\phi_2^{1/2}\xi_2^{n-1}||^2\right].
\end{aligned}$$

By Lemma 3.1 in [5], we have

$$(7.47)\qquad \frac{||\phi_1^{1/2}\check{\zeta}_1^{n-1}||^2 - ||\phi_1^{1/2}\zeta_1^{n-1}||^2}{2\Delta t} \leq K||\phi_1^{1/2}\zeta_1^{n-1}||^2.$$

Applying this inequality to (7.46) for each component $i = 1, \ldots, M$, adding these equations together, multiplying the result by Δt and summing on n, and applying Gronwall's Lemma we find

$$(7.48)\qquad \max_n ||\phi_i^{1/2}\zeta_i^n|| \leq K\Delta t, \quad i = 1, \ldots, M.$$

Using arguments found in, e.g., [5], one can show

$$(7.49)\qquad \max_n ||\phi_i^{1/2}\xi_i^n|| \leq K(\Delta t + h^{r+1}).$$

Applying the triangle inequality we obtain the following result.

THEOREM 7.1. *Let c_i, $i = 1, \ldots, M$ satisfy (1.1) and assume the data and c_i are sufficiently smooth. Let $C_i \in \mathcal{M}_h$ be the approximation given by (7.24)-(7.26). Then there exists a positive constant K, such that*

$$(7.50)\qquad \max_n ||C_i^n - c_i^n|| \leq K(\Delta t + h^{r+l}), \quad i = 1, \ldots, M,$$

where r is the maximum degree of polynomials in $\mathcal{M}_h$, and $l = 1$ if $D(u)$ is uniformly positive definite, $l = 0$ if $D(u)$ is positive semi-definite.

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