On the Use of Large Time Steps with ELLAM for Transport with Kinetic Reactions Over Heterogeneous Domains

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An Eulerian Lagrangian localized adjoint method (ELLAM) is considered for the resolution of advection-dominated transport problems in porous media. Contrary to standard Eulerian methods, ELLAM can use large time steps because the advection term is approximated accurately without any CFL restriction. However, it is shown in this article that special care must be taken for the approximation of the dispersive and reactive terms when large time steps are used over heterogeneous domains. An alternative procedure is proposed. It is based on an equivalent dispersion coefficient or an equivalent reaction rate when different zones are encountered during the tracking. Numerical experiments are performed with variable dispersion or variable reaction rates over space (including nonlinearity). When classical ELLAM require numerous time steps to handle heterogeneity, the alternative procedure is shown to perform with the same accuracy in a single time step.
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Introduction

In this work, the Eulerian-Lagrangian localized adjoint method (ELLAM) is considered for the resolution of advection dispersion reaction equations (ADRE) with a dominant advection term. The ELLAM is an improved method of characteristics that guaranties mass conservation and treats general boundary conditions naturally in its formulation.[1,2] Recently, ELLAM has been largely used for multidimensional advection dispersion transport problems.[1,3–6] For the sake of simplicity, the problem is handled here in 1D but could be easily extended to multidimensional transport.

Even though the ELLAM is powerful, it may suffer however from nonphysical oscillations[4,7] and/or numerical diffusion.[8,9] The spurious oscillations of ELLAM have been successfully avoided by using: (i) the moving grid procedure for one-dimensional problems,[2] and (ii) strategic integration points for unstructured 2D triangular meshes.[6] On the other hand, the numerical diffusion of ELLAM used with several time steps can be avoided by handling the same characteristics for all time steps and by interpolating only the concentration variations due to the dispersion mechanism.[9]

The ELLAM uses space-time test functions defined as the solutions to the local adjoint operator (see later). With ELLAM, large time steps could be used to solve the transport problem since the advection term is approximated accurately without any CFL restriction. Usually, the dispersion and reaction terms are discretized in the same way as with

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standard Eulerian methods, for instance with one step-backward Euler approximation in time and any finite-volume, or finite-element scheme in space (see e.g., Russell and Celia,[1] Binning and Celia,[3] Younes et al.[6]). The present work points out eventual discrepancies and how to overcome them, when calculations of the dispersion and reaction terms are performed with large time steps over heterogeneous media. Usually, these discrepancies are not encountered with Eulerian approaches because the latter have to deal with small time steps for accuracy and/or stability reasons. With ELLAM, it is legitimate to draw benefits from the capability to solve accurately the advection term with very few time steps. This work proposes an alternative procedure using equivalent dispersion coefficient and equivalent linear/nonlinear reaction rates when different zones are encountered along transport pathways. Actually, this method monitors and then averages all dispersion-reaction heterogeneities encountered by a particle along its characteristic during a given time step. Numerical experiments are performed and show the efficiency of this procedure while keeping very few time steps for the calculations.

The Eulerian Lagrangian Localized Adjoint Method for ADE

One-dimensional transport in a homogeneous medium can be depicted by the following advection dispersion equation (ADE):

$$\frac{\partial C}{\partial t}+\frac{\partial}{\partial x}\left(VC-D\frac{\partial C}{\partial x}\right)=0 \qquad (1)$$

C is the solute concentration [ML^{-3}], V the fluid velocity [LT^{-1}], and D [L^{2}T^{-1}] the dispersion coefficient. Equation 1 is defined in the space-time domain $\Omega_{xt}=[0,l]\times[0,T]$. For the sake of simplicity, only the following initial and boundary conditions are considered:

$$C(x,0)=C_0(x),\ C(0,t)=\overline{C}_e(t) \quad \text{and} \quad \left(-D\frac{\partial C}{\partial x}\right)_{x=l}=0$$

The spatial domain $\Omega=[0,l]$ is partitioned with a uniform discretization at the step Δx. Time is divided into discrete and equal steps Δt.

The variational formulation of Eq. 1, using space-time test functions $\omega=\omega(x,t)$ is written as:

$$\int_0^T\int_\Omega\left[\frac{\partial C}{\partial t}\omega+\frac{\partial(VC)}{\partial x}\omega-\frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right)\omega\right]dxdt\ =0 \qquad (2)$$

The ELLAM selects test functions $\omega(x,t)$ constant along the characteristics:

$$\frac{d\omega}{dt}=0\ \text{i.e.,}\ \frac{\partial\omega}{\partial t}+V\frac{\partial\omega}{\partial x}\ =0 \qquad (3)$$

As done in Russell and Celia,[1] the functions $\omega(x,t)$ are strictly defined at time $t=t^{n+1}$ with $\omega_i(x,t^{n+1})$ a piecewise linear function in space (chapeau function). Thus, assuming a null dispersive flux at the inflow boundary, using the Green's theorem and Expression (3) [see Binning and Celia,[3] for details] allows to rewrite (2) in the form:

$$\int_\Omega C^{n+1}\omega_i^{n+1}dx+\int_{t^n}^{t^{n+1}}\int_\Omega D\frac{\partial C}{\partial x}.\frac{\partial\omega_i^{n+1}}{\partial x}dx-V\int_{t^n}^{t^{n+1}}\left[\overline{C_e}(t)\omega_i\right]_{x=0}dt =\int_\Omega C^n\omega_i^n(x)dx \qquad (4)$$

The concentration C is defined within a finite element framework as a combination of piecewise linear basis functions: $C=\sum_i C_i\omega_i$. The first integral in the left-hand-side of (4) is the standard finite element mass integral (accumulation term in Ω). The boundary integral (in $x=0$) and the integral at the old time level t^n are evaluated by a numerical integration with a forward-tracking approach. Discrete quadrature points are regularly distributed over the fixed grid at t^n and tracked from t^n to t^{n+1}, where the evaluation of $\omega_i(x,t^{n+1})$ is straightforward.[1]

For interior nodes, except the dispersion term discussed later, the discretization of (4) by using a mass lumping procedure is written as in Younes et al.[9]:

$$\Delta xC_i^{n+1}+\int_{t^n}^{t^{n+1}}\int_\Omega D\frac{\partial C}{\partial x}.\frac{\partial\omega_i^{n+1}}{\partial x}dx=V\overline{C}_e^{n+1}\sum_{q=1}^{N_t}\omega_i(0,t^q)W_e^q +\sum_A\left[\sum_{p=1}^{N_A}(C^n(x^p,t^n)\omega_i(x^p,t^n)W_A^p)\right] \qquad (5)$$

N_t is the number of integration points along the time axis between t^n and t^{n+1}, each point $(0,t^q)$ being assigned with a weight W_e^q. N_A is the number of spatial integration points for each cell A, each point (x^p,t^n) having a weight W_A^p. To evaluate $\omega_i(x^p,t^n)$ in (5), the location in space x^p at time t^n is tracked forward to $\tilde{x}^p$ at t^{n+1} and the relation $\omega_i(x^p,t^n)=\omega_i(\tilde{x}^p,t^{n+1})$ is used (ω_i is fully defined by analytical expressions at t^{n+1}). In the same way, each point $(0,t^q)$ is tracked to $(\tilde{x},t^{n+1})$ with $\tilde{x}$ the location reached at t^{n+1} when starting from 0 at t^q.

The Dispersion Integral Over Heterogeneous Domains

The equivalent dispersion coefficient

The dispersion integral in (5) is generally evaluated by using a one-step backward Euler approximation in time as done with a standard finite element method:

$$\int_{t^n}^{t^{n+1}}\int_\Omega D\frac{\partial C}{\partial x}.\frac{\partial\omega_i^{n+1}}{\partial x}dx\approx\Delta t\ D_i^{n+1}\left(\frac{-C_{i-1}^{n+1}+2C_i^{n+1}-C_{i+1}^{n+1}}{\Delta x}\right) \qquad (6)$$

Compared with the Crank-Nicholson scheme, the one-step backward Euler approximation generates a first-order error in time. But this temporal error which only affects dispersion (since ELLAMs belong to the so-called characteristic methods free of truncation errors from the advection term) is not

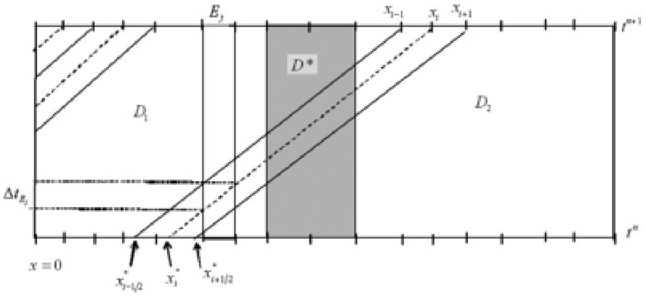

Figure 1. Tracking over a heterogeneous domain.

significant and the scheme is highly popular in engineering practice.[10]

Classical Eulerian approaches to the ADE often handle small time steps for either accuracy (implicit schemes) or stability (explicit schemes) reasons. Therefore, the standard discretization (6) is sufficient to account for mass fluxes exchanged by dispersion within Δt. This approximation may fail however with ELLAM and large time steps over heterogeneous domains. This is simply because D_i^{n+1}, which is the local dispersion at the point reached by a particle at t^{n+1}, is not representative of the dispersion experienced along the characteristic between t^n and t^{n+1}.

For the purpose of illustration, consider a heterogeneous domain with three different dispersion coefficients (D_1, D^*, and D_2) as shown in Figure 1. It is supposed that with a large time step Δt, the three zones are crossed during the tracking. The evaluation of the dispersion integral in (6) assumes that dispersion occurs with the coefficient D_2 during the whole time step and thus overlooks the coefficients D_1 and D^*. To overcome this problem, the approximation (6) is replaced by the following one:

$$\int_{t^n}^{t^{n+1}} \int_{\Omega} D \frac{\partial C}{\partial x} \cdot \frac{\partial \omega_i^{n+1}}{\partial x} dx \approx \Delta t \; D_i^{eq} \left(\frac{-C_{i-1}^{n+1} + 2C_i^{n+1} - C_{i+1}^{n+1}}{\Delta x} \right) \tag{7}$$

with D_i^{eq} the equivalent dispersion coefficient during Δt. To evaluate this coefficient, the dispersion coefficients D^{E_j} for all crossed element E_j between t^n and t^{n+1} are monitored and stored, as well as the duration Δt_{E_j} to cross each element E_j. The equivalent dispersion coefficient D_i^{eq} is then calculated as:

$$D_i^{\text{eq}} = \frac{\sum\limits_j \Delta t_{E_j} D^{E_j}}{\sum\limits_j \Delta t_{E_j}} \tag{8}$$

The justification of (8) is quite simple. Basically, Gaussian diffusion (or dispersion) is a Markovian process stating that solute particles move randomly by independent successive jumps. Assume, in one dimension for the sake of simplicity, that a particle moves from x_0 to x_n by successive jumps (random variables) $J_{i \to i+1}$ between locations x_i, x_{i+1}. Assume now, for a set of particles that the random variables $J_{i \to i+1}$ are associated with variances $\sigma_{i,i+1}^2$ (of length of jumps). The total trajectory for a single particle can be written as the sum of independent random variables: $\sum_{i=0}^{n-1} J_{i \to i+1}$. It is well known that a variable adding independent random variables has a variance equal to the sum of the variances, i.e., $\sigma^2 = \sum_{i=0}^{n-1} \sigma_{i,i+1}^2$. Knowing that the variance of diffusive (dispersive) jumps can be written as $2D_{i,i+1}\Delta t$ for jumps at constant time step Δt, it is obvious that the equivalent dispersion will obey the following relationship: $2D_{\text{eq}} \sum_{i=0}^{n-1} \Delta t = 2 \sum_{i=0}^{n-1} D_{i,i+1} \Delta t \Rightarrow D_{\text{eq}} = \left(\sum_{i=0}^{n-1} D_{i,i+1} \right) \Big/ n$ which is the arithmetic mean of the experienced diffusion (dispersion) coefficients. This reasoning applied to heterogeneous zones crossed by the characteristics of ELLAMs yields the Expression (8). It can be also raised that the reasoning could apply to covariance of motion of particles along different directions in space. Thus, for 2D or 3D problems with full dispersion tensors, the arithmetic mean would hold for each component of the tensor.

Application to the advection dispersion equation

The following example simulates the motion of a concentration front in a heterogeneous porous medium of length $l = 1500$ m. The domain is initially free of solute and boundary conditions are $C(0,t) = 1$ g/l and $\partial C(l,t)/\partial x = 0$. The domain is discretized at the constant space step $\Delta x = 10$ m and the simulation time is $T = 500$ days. Although ELLAM performs well with varying velocity,[2,6,9] for the sake of simplicity the fluid velocity is set at a constant value $V = 1$ m/day inside the whole domain. This domain is made of three juxtaposed zones of length $l_1 = 450$ m, $l_2 = 600$ m, and $l_3 = 450$ m with dispersion coefficients $D_1 = 0.1$ m^2/day, $D_2 = 0.01$ m^2/day, and $D_3 = 0.1$ m^2/day, respectively. The problem is simulated for both small time steps ($\Delta t = 1$ day) and a single large one ($\Delta t = 500$ days) and by using ELLAM with the standard approximation (6) or the new formulation in (7). The results are plotted in Figure 2. The standard approximation (6) fails to give the right solution when used with large time steps on heterogeneous domain. On the other hand, the solution obtained by the modified ELLAM scheme (7) is accurate for small and large time steps.

Transport with Kinetic Reactions

The ELLAM was shown to be highly accurate for the simulation of reactive transport problems with kinetic effects when combined with the sequential noniterative approach (SNIA). As shown by Younes and Fahs,[11] SNIA-ELLAM also allows for large time steps over homogeneous domains without loss of accuracy. With SNIA, the ADE is solved first to get the transported concentrations. These concentrations are then used as initial conditions for the resolution of the reaction equations with the parameters of the reaction rates taken equal to that of the arrival element.

In the same way as for dispersion discussed previously, taking the arrival parameters can be flawed with large time steps over chemically heterogeneous domains. Again, an alternative procedure is developed by calculating equivalent reaction rates from local values of the zones crossed during the tracking. For a large majority of kinetically controlled reactions involving, e.g., the species C, the kinetics can be written as:

$$\frac{\partial C}{\partial t} = -\lambda(C, C_k)C + F\ (C_k) \tag{9}$$

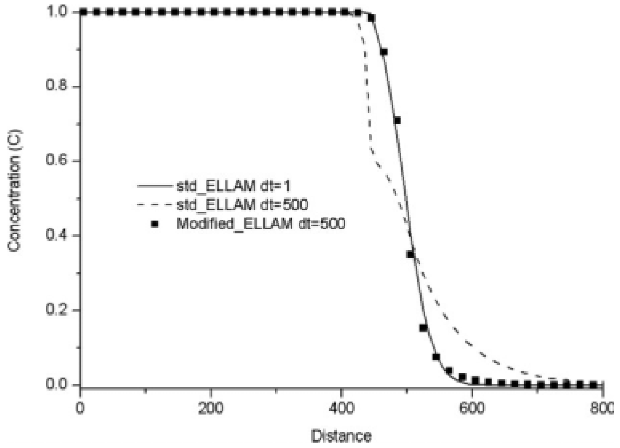

Figure 2. Concentration profiles at time t = 500 days for advection and heterogeneous dispersion.

Solutions obtained with the standard and modified ELLAM schemes for large and small time steps (the small time step calculation with modified ELLAM is not reported since fully merged with the standard ELLAM solution).

$\lambda(C, C_k)$ [T^{-1}] is the kinetic rate which may depend on C and C_k ($k = 1 \ldots s$), i.e., the other species interacting with C, F (C_k) [$ML^{-3}T^{-1}$] is a kinetically controlled source term producing the species C from the other species C_k. Assume now that a particle (or equivalently a function ω_i) "belongs" to species C and may "pass" to any other species C_k due to reaction. Whatever the complexity of the term F (C_k), the conditional probability to stay in C less than the duration t knowing the particle was initially in C is ruled by the equation $\partial C/\partial t = -\lambda C$. This is simply because for a single particle initially in C, all the other terms C_k are null as well as the source term F. Thus, according to the obvious solution to the differential equation ruling C, the conditional probability would write:

$$\partial C/\partial t = -\lambda C \Rightarrow C(t) = C(0)\exp(-\lambda t) \Rightarrow P_C(t/ \text{ in } C \text{ at } 0) = 1 - \exp(-\lambda t) \qquad (10)$$

Suppose now that the particle has to move through several successive elements E_j with mean resting times $\Delta t_{E_j} = t_j - t_{j-1}$. The conditional probability to stay in the species C during Δt_{E_j} in each element E_j knowing the particle was in C at t_{j-1} would be ruled by the relation $C(t_j) = C(t_{j-1})\exp(-\lambda_j \Delta t_{E_j})$. Thus, for a total time $\Delta t = \sum_j \Delta t_{E_j}$, assuming that conditional probabilities in E_j are independent, the probability to stay in the species C would be ruled by:

$$C(\Delta t) = C(0)\exp(-\lambda^{\mathrm{eq}}\Delta t) = C(0)\prod_j \exp(-\lambda_j \Delta t_{E_j}) = C(0)\exp\left(\sum_j \lambda_j \Delta t_{E_j}\right) \qquad (11)$$

It is obvious from Expression (11) that for a time step $\Delta t = \sum_j \Delta t_{E_j}$, the equivalent kinetic rate can be written as:

$$\lambda^{\mathrm{eq}} = \frac{\sum_j \Delta t_{E_j} \lambda_j}{\sum_j \Delta t_{E_j}} \qquad (12)$$

which is again the arithmetic mean of local values weighted by mean resting times in elements E_j.

Application to transport with linear kinetic reactions

In this example, it is dealt with transport of a couple of species in solution C_1 and C_2 with a linear kinetic reaction between the species. Transport equations can be written as:

$$\begin{aligned} \frac{\partial C_1}{\partial t} + \frac{\partial}{\partial x}\left(VC_1 - D\frac{\partial C_1}{\partial x}\right) &= -\lambda C_1 + \mu C_2 \\ \frac{\partial C_2}{\partial t} + \frac{\partial}{\partial x}\left(VC_2 - D\frac{\partial C_2}{\partial x}\right) &= \lambda C_1 - \mu C_2 \end{aligned} \qquad (13)$$

with λ and μ [T^{-1}] the first-order reaction rates that may vary within the domain. Let us consider a domain of length $l = 150$ m discretized at the constant space step $\Delta x = 1$ m. Boundary conditions are $C_1(0, t) = 3$ g/l, $C_2(0, t) = 5$ g/l, and

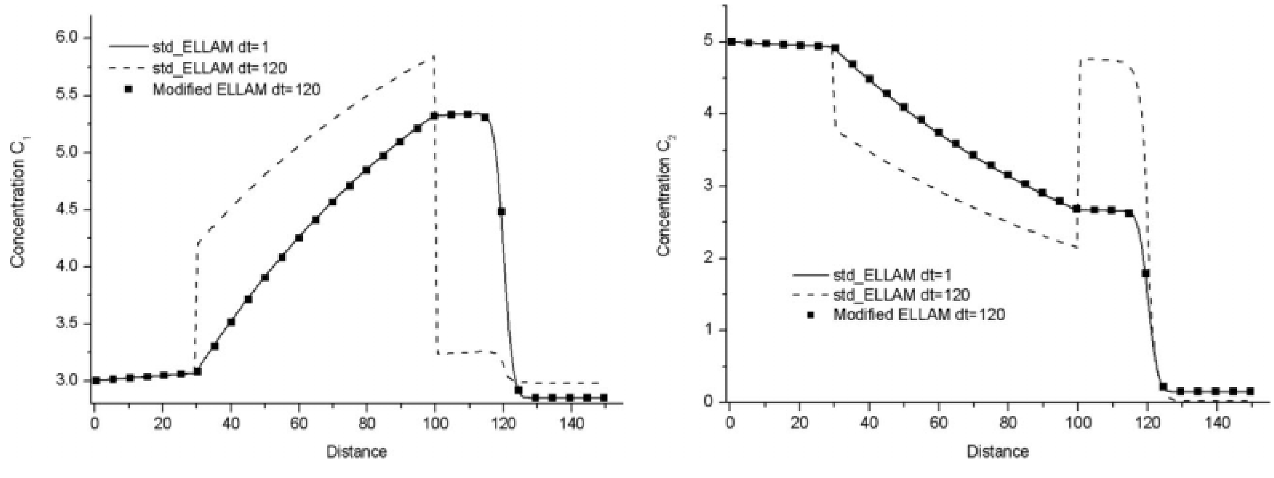

Figure 3. Transport with linear kinetic reaction between two species C_1 and C_2 in solution.

Concentration profiles at time t = 120 days for standard and modified ELLAM with small and large time steps (small time step calculation with modified ELLAM is not reported since merged with results from standard ELLAM solution).

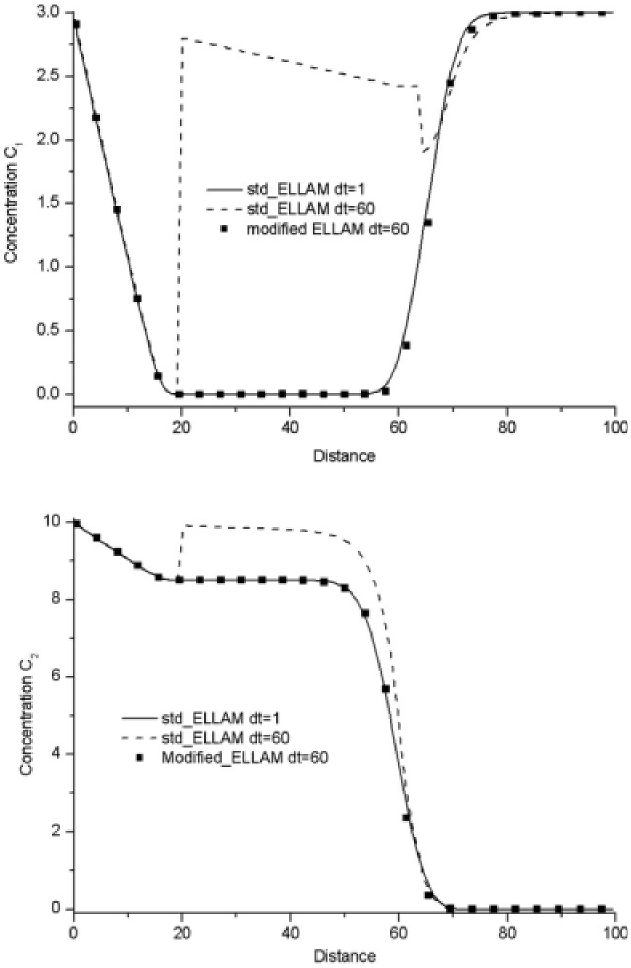

Figure 4. Transport with nonlinear aerobic biodegradation (concentration of bacteria C_B varying in the domain).

Concentration profiles at time $t = 60$ days for standard and modified ELLAM with small and large time steps (small time step calculation with modified ELLAM is not reported since merged with results from standard ELLAM solution).

$(\partial C/\partial x|_{x=l} = 0)$ at the outflow boundary. The domain is initially free of any solute, the simulation time is $T = 120$ days, fluid velocity and dispersion coefficient are $V = 1$ m/day and $D = 0.02$ m^2/day, respectively. The domain is decomposed in three zones of length $l_1 = 30$ m, $l_2 = 70$ m, and $l_3 = 50$ m with the following set of reaction rates: $\lambda_1 = 5 \times 10^{-5}$ day^{-1}, $\mu_1 = 5 \times 10^{-4}$ day^{-1}, $\lambda_2 = 10^{-3}$ day^{-1}, $\mu_2 = 10^{-2}$ day^{-1}, $\lambda_3 = 5 \times 10^{-5}$ day^{-1}, $\mu_3 = 5 \times 10^{-4}$ day^{-1}.

Results of the standard and the modified ELLAM schemes with large ($\Delta t = 120$ days) and small time steps ($\Delta t = 1$ day) are reported in Figure 3. As for the case of heterogeneous dispersion, the standard approach gives accurate results only when small time steps are used. The modified approach gives the same accurate results for both small and large time steps.

Application to transport with nonlinear kinetic reactions

It is dealt with the nonlinear aerobic biodegradation transport problem given in Celia et al.[12] This problem represents the transport of an organic contaminant and dissolved oxygen in the presence of stationary biological populations. The set of transport equations is written as follows:

$$\begin{cases} \dfrac{\partial C_1}{\partial t} + \dfrac{\partial (V_1 C_1)}{\partial x} - \dfrac{\partial}{\partial x}\left(D_1 \dfrac{\partial C_1}{\partial x}\right) + \left(\dfrac{\mu_1 C_B}{K_h^1 + C_1}\right)\delta_1 C_1 \\ \qquad\qquad = -\kappa_{12}\left(\dfrac{\mu_2 C_B}{K_h^2 + C_2}\right) C_2 \delta_2 \\ \dfrac{\partial C_2}{\partial t} + \dfrac{\partial (V_2 C_2)}{\partial x} - \dfrac{\partial}{\partial x}\left(D_2 \dfrac{\partial C_2}{\partial x}\right) + \left(\dfrac{\mu_2 C_B}{K_h^2 + C_2}\right)\delta_2 C_2 \\ \qquad\qquad = -\kappa_{21}\left(\dfrac{\mu_1 C_B}{K_h^1 + C_1}\right) C_1 \delta_1 \end{cases} \tag{14}$$

C_α is the concentration of species α $(\alpha = 1, 2)$, μ_α [T^{-1}] is the maximum uptake rate for species α, K_h^α [ML^{-3}] is the so-called half-saturation constant for species α, $\kappa_{\alpha\beta}$ [dimensionless] denotes the yield ratio for species α when species β is limiting the reaction, δ_α equals one if species α is limiting and zero otherwise, C_B is the concentration of the stationary bacterial population. The parameter values are assigned as

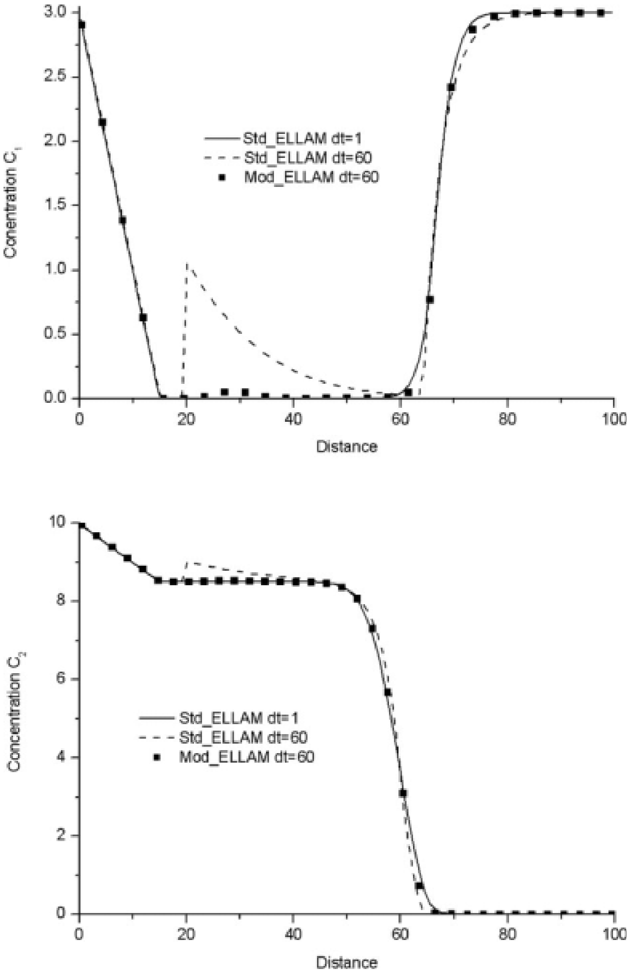

Figure 5. Transport with nonlinear aerobic biodegradation (K_h^1 and K_h^2 varying in the domain).

Concentration profiles at time $t = 60$ days for standard and modified ELLAM with small and large time steps (small time step calculation with modified ELLAM is not reported since merged with results from standard ELLAM solution).

follows: $\mu_1 = \mu_2 = 1.0$ day^{-1}, $K_h^1 = K_h^2 = 0.1$ mg/L, $\kappa_{12} = 2.0, \kappa_{21} = 0.5$, $V_1 = V_2 = 1$ m/day, $D_1 = D_2 = 0.2$ m^2/day. Initial conditions are $C_1(x,0) = 3.0$ mg/L mg/L, $C_2(x,0) = 0$. Dirichlet conditions are specified at the inflow boundary $C_1(0,t) = 3.0$ mg/L, $C_2(0,t) = 10.0$ mg/L, whereas homogeneous Neumann conditions are prescribed at the outflow boundary $(\partial C_\alpha/\partial x|_{x=l} = 0; \alpha = 1,2)$. The one-dimensional domain of size $l = 100$ m is discretized at the constant space step $\Delta x = 1$ m and the simulation time is $T = 60$ days. The microbial concentration is varying in the domain: $C_{B1} = 0.2$ mg/L for the first zone of length $l_1 = 20$ m, $C_{B2} = 0.01$ mg/L for the second zone of length $l_2 = 50$ m, and $C_{B3} = 0.2$ mg/L for the third zone of length $l_3 = 30$ m.

Results with small time steps ($\Delta t = 1$ day) and a single large time step ($\Delta t = 60$ days) for the standard and the modified approaches are plotted in Figure 4. Again, these results show the strength of the modified ELLAM method since it provides accurate results with both small and large time steps whereas standard ELLAM fails with large time steps.

Finally, the effects of nonlinear variations are illustrated by making to vary the parameters K_h^1 and K_h^2 in the denominator of Eq. 14. K_h^1 and K_h^2 are taken as follows: $K_h^1|_1 = K_h^2|_1 = K_h^1|_3 = K_h^2|_3 = 0.01$ in the first and third zone, $K_h^1|_2 = K_h^2|_2 = 2$ in the second zone. Results with small time steps ($\Delta t = 1$ day) and a single large time step ($\Delta t = 60$ days) with the standard and the modified approaches are plotted in Figure 5. It clearly shows the ability of the modified ELLAM to handle heterogeneity with large time steps even in the case of nonlinear depending parameters.

Conclusion

The ELLAM allows for large time steps because the advection term can be accurately approximated without any CFL restriction. Usually, the dispersion and reaction terms are discretized using a one-step backward Euler approximation in time which may be flawed for heterogeneous transport. In that case, the approximation can overlook the local dispersion and/or reaction rates experienced along a characteristic during large time steps. An alternative procedure based on the derivation of an equivalent dispersion (or reaction) coefficient is developed. It is shown that for both dispersion and reaction rates, the equivalent parameter is the arithmetic mean of local values weighted by the mean time (advection time) spent under the influence of the local parameter. Numerical experiments show the efficiency of this procedure for domains with heterogeneous dispersion and heterogeneous linear or nonlinear reactions. Even though the topic is not discussed in the article, heterogeneity of dispersion and kinetic rates can be mixed within the same transport scenario. Since dispersion and reactions in solution are independent mechanisms, calculating equivalent parameters makes no matter of mixing. The principle of weighted arithmetic mean (see earlier) still holds for both mechanisms. In the end, the enviable capability of ELLAM to handle transport with large time steps is extended to the case of heterogeneous media.

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