Reply:

The principal criticism made by M. J. Simpson in the Letter to the Editor is that splitting errors can be significant with SNIA for dispersion-dominated kinetic reactive transport problems.

This criticism is out of the objectives of the article

In the article "Direct and Split Operator Approaches with ELLAM for reactive transport Equations," one is interested on advection dominated transport problems.[1] These problems can be encountered in many field situations, especially for small-scale simulations. For these problems, hyperbolic advective part of the partial differential equation becomes dominant and standard numerical methods, such as finite elements or finite volumes, generate solution with numerical diffusion and/or nonphysical oscillations.

The Eulerian-Lagrangian localized adjoint method (ELLAM) is an interesting alternative to standard methods. However, almost all developed ELLAMs suffer from non physical oscillations and/or numerical diffusion.[2] A new scheme, which removes nonphysical oscillations and numerical diffusion whatever the time step used, was obtained by combining a finite volume ELLAM with a moving grid procedure.[3] Therefore, the main objective of the article is not to study splitting errors for dispersive kinetic reactive transport problems, but to show the performances of the new ELLAM with both SNIA and DSA for advection dominated reactive (kinetic and equilibrium) transport problems. This objective is recalled many times in the article:

In the abstract:

- "In this work, we combine the moving mesh Eulerian Lagrangian localized adjoint method (ELLAM), with the direct substitution approach (DSA), and the sequential noniterative approach (SNIA) to accurately solve advection dominated problems including chemical reactions" (page 1, Lines 2–5)
- Performances of ELLAM are evaluated in comparing SNIA_ELLAM and DSA_ELLAM to SNIA_FE and DSA_FE (page 1, Lines 6-7)

In the introduction:

- In this work, we combine the moving mesh ELLAM with SNIA and DSA to solve accurately advection-dominated reactive transport problems. Five test cases are set up for comparing SNIA_ELLAM and DSA_ELLAM to SNIA_FE and DSA_FE. (page 2, last paragraph of the introduction)

In the section: kinetic reactions

- In this section, SNIA_ELLAM and DSA_ELLAM are used to simulate advection dominated kinetic reactive transport problems. Performances of ELLAM with SNIA and DSA are assessed in comparing SNIA_ELLAM and DSA_ELLAM to SNIA_FE and DSA_FE for the cases of linear kinetic, nonlinear kinetic and interphase mass transfer problems. (page 3, last paragraph)

In the section: equilibrium reactions

- In this section, SNIA_ELLAM and DSA_ELLAM are used to simulate advection-dominated transport coupled with sorption reactions at equilibrium. Performances of ELLAM with SNIA and DSA are assessed in comparing SNIA_ELLAM and DSA_ELLAM with SNIA_FE and DSA_FE for both linear and nonlinear equilibrium. (page 7, first paragraph of the section Results for equilibrium reactions)

Results of the article show:[1]

For advection-dominated reactive transport with kinetic reactions:

- SNIA_ELLAM gives less numerical diffusion than SNIA_FE;
- SNIA_ELLAM avoids mass balance errors present in the OS algorithm for problems with continuous mass injection at the inlet boundary;
- SNIA_ELLAM is highly accurate and efficient compared to DSA_ELLAM since chemistry can be solved accurately with specific methods, and small time steps (see Table 1 where SNIA_ELLAM and DSA_ELLAM are compared for the biodegradation problem).

For advection-dominated reactive transport with equilibrium reactions:

- SNIA_ELLAM induces numerical diffusion proportional to Δt, but SNIA_ELLAM is less diffusive than SNIA_FE;
- DSA_ELLAM is more accurate and more efficient than DSA_FE;
- DSA_ELLAM avoids the numerical diffusion and is more accurate than SNIA_ELLAM.

Numerical results with large diffusion and reaction coefficients

In the Letter to the Editor, we are criticized for taking small values for D, K and α. As explained before, this is out of the objective of the article, since we were interested on advection-dominated reactive transport problems. However, we give here results for the three studied transport problems with kinetic reactions (linear kinetic, nonlinear kinetic and interphase mass transfer) using larger coefficients in order to evaluate the internal error.[4] Results show that this error is not significant for all numerical experiments even with very large dispersive and reactive coefficients.

First-order decay problem

$$\frac{\partial(RC)}{\partial t}+\frac{\partial(qC)}{\partial x}+KC-\frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right)=0 \tag{1}$$

This problem is simulated with larger D and K.

The simulation time is $T = 400$ days, $\Delta x = 20$ m and $l = 1000$ m. We use the following parameters: $q = 20$ m/day, $R = 1$, $D = 20$ m^2/day (old value $D = 0.01$ m^2/day) and $K = 0.2$ day^{-1} (old value $K = 0.002$ day^{-1}).

SNIA_ELLAM_dt05 and SNIA_ELLAM_dt20 results are obtained with, respectively a small time step of 0.5 day and a large time step of 20 days.

Figure 1 shows that SNIA_ELLAM avoids OS errors. Indeed, for linear reactions only boundary errors occur.[4] These errors are avoided with SNIA_ELLAM.[1]

Aerobic biodegradation transport problem

The system of transport equations is given by

$$\begin{cases}\dfrac{\partial C_1}{\partial t}+\dfrac{\partial(q_1C_1)}{\partial x}-\dfrac{\partial}{\partial x}\left(D_1\dfrac{\partial C_1}{\partial x}\right)\\ \qquad +K_1(C_1,C_B)C_1=f_1(C_2,C_B)\\ \dfrac{\partial C_2}{\partial t}+\dfrac{\partial(q_2C_2)}{\partial x}-\dfrac{\partial}{\partial x}\left(D_2\dfrac{\partial C_2}{\partial x}\right)\\ \qquad +K_2(C_2,C_B)C_2=f_2(C_1,C_B)\end{cases} \tag{2}$$

The nonlinear reactions and source terms are

$$\begin{aligned}K_1(C_1,C_B)&=\left(\frac{\mu_1C_B}{K_h^1+C_1}\right)\delta_1,\\ K_2(C_2,C_B)&=\left(\frac{\mu_2C_B}{K_h^2+C_2}\right)\delta_2\end{aligned} \tag{3}$$

$$\begin{aligned}f_1(C_2,C_B)&=-\kappa_{12}\left(\frac{\mu_2C_B}{K_h^2+C_2}\right)C_2\delta_2\\ f_2(C_1,C_B)&=-\kappa_{21}\left(\frac{\mu_1C_B}{K_h^1+C_1}\right)C_1\delta_1\end{aligned}$$

For this problem, we use larger coefficients K_h^1, K_h^2, C_B and D. Parameter values are assigned as follows: $\mu_1 = \mu_2 = 1.0$, day^{-1}, K_h^1, $K_h^2 = 1$, mg/L (old values 0.1 mg/L), $\kappa_{12} < 20$, $\kappa_{21} = 0.5$, $q_1 = q_2 = 0.8$ m/day, $D = 5.0$ m^2/day (old value 0.2 m^2/day). The fixed microbial concentration is $C_B(x,t) = 5.0$ mg/L (old value 0.2 mg/L).

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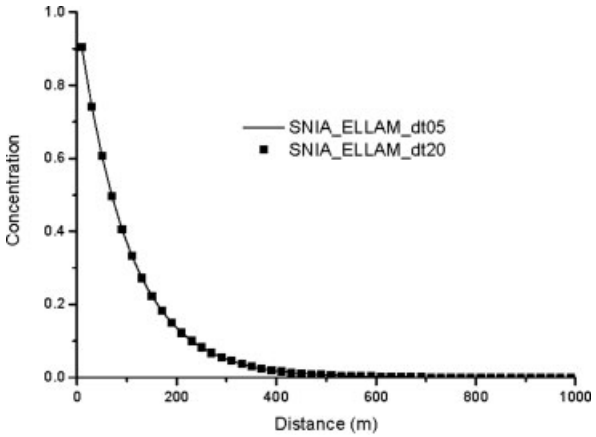

Figure 1. Results of SNIA_ELLAM for transport with first-order decay.

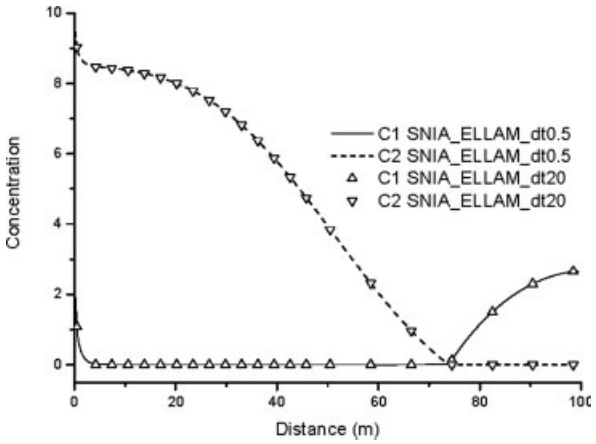

Figure 2. Results of SNIA_ELLAM for the aerobic biodegradation transport problem.

The one-dimensional (1-D) domain of size m is $l = 100$ m is discretized with a constant space step of $\Delta x = 1$ m, and the simulation time is $T = 60$ days.

SNIA_ELLAM_dt05 and SNIA_ELLAM_dt20 results are obtained with, respectively a small time step of 0.5 day, and a large time step of 20 days.

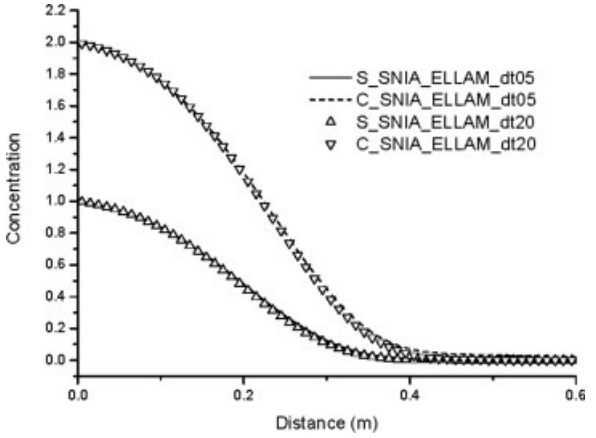

Figure 3. Results of SNIA_ELLAM for the interphase mass transfer problem.

Results of Figure 2 show that the internal error is small even for this case where D/V $\Delta x = 5$ and $C_B\ \Delta t = 100$.

Interphase mass transfer

Contrarily to previous case, this problem contains an heterogeneous kinetic reaction (in which the reactants are present in several phases). The governing equations with adsorption are

$$\frac{\partial C}{\partial t} + \frac{\partial S}{\partial t} + \frac{\partial (qC)}{\partial x} - \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) = 0$$

$$\frac{\partial S}{\partial t} = \alpha\left[kC^{nf} - S\right]$$

Simulations are performed with larger D,α and k. Parameters are assigned as follows: $q = 5.10^{-4}$ m/day, $D = 2.510^{-5}$ m^2/day (old value 5×10^{-8} m^2/day) $\alpha = 0.1$ (old value 0.001), $k = 2$ (old value 1) and $nf = 0.7$. The 1-D domain of size $l = 1$ m is discretized with a uniform mesh of $\Delta x = 0.01$m. The simulation time is $T = 1{,}000$ days.

SNIA_ELLAM_dt05 and SNIA_ELLAM_dt20 results are obtained with, respectively a small time step of 0.5 day and a large time step of 20 days.

Results of Figure 3 show that the internal error is small for this case where $D/(V\ \Delta x) = 5$ and $\alpha\Delta t = 2$.

To sum up, the internal error[4] of SNIA_ELLAM is very small for the performed numerical experiments even with large dispersive and reactive coefficients. Recall that for physical problems these coefficients cannot be so large. The internal error exists algebraically, but is much smaller than the rest of errors. For example, this internal error can be neglected compared to SNIA_ELLAM error for transport with equilibrium reactions (see Figure 8 in Younes and Fahs[1]).

Literature Cited

1. Younes A, Fahs M. Direct and split operator approaches with ELLAM for reactive transport equations. *AIChE J*. 2007;53:2161–2169.
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