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Comment on "Frequency-dependent dispersion in porous media"

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In a recent paper, Valdès-Parada and Alvarez-Ramirez [Phys. Rev. E **84**, 031201 (2011)] used the technique of volume averaging to derive a "frequency-dependent" dispersion tensor, $\mathbf{D}_\gamma^*$, the goal of which is to describe solute transport in porous media undergoing periodic processes. We describe two issues related to this dispersion tensor. First, we demonstrate that the definition of $\mathbf{D}_\gamma^*$ is erroneous and derive a corrected version, $\mathbf{D}_\gamma^{*c}$. With this modification, the approach of Valdès-Parada and Alvarez-Ramirez becomes strictly equivalent to the one devised by Moyne [Adv. Water Res. **20**, 63 (1997)]. Second, we show that the term "frequency-dependent dispersion" is misleading because $\mathbf{D}_\gamma^*$ and $\mathbf{D}_\gamma^{*c}$ do not depend on the process operating frequency, χ. The study carried out by Valdès-Parada and Alvarez-Ramirez represents a spectral analysis of the relaxation of $\mathbf{D}_\gamma^*$ towards its steady-state, independent of any periodic operation or excitation.

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I. INTRODUCTION

The investigation of dynamic dispersion in porous media has a long history. It was first explored by addressing issues with the early time regime and unsteady flows in Taylor's tube problem [1]. One of the simplest models that was proposed, e.g., in Ref. [2], is an advection-dispersion equation with a time-dependent longitudinal dispersion coefficient, $\mathsf{D}_\gamma^{*s}(t)$,

$$\partial_t \langle c_{A\gamma}\rangle^\gamma + \langle v_\gamma\rangle^\gamma \partial_x \langle c_{A\gamma}\rangle^\gamma = \mathsf{D}_\gamma^{*s}(t)\partial_x^2 \langle c_{A\gamma}\rangle^\gamma, \tag{1}$$

where $\langle\cdot\rangle^\gamma$ is the cross-sectional average over the fluid phase (γ). Here, and throughout this publication, volume fractions have been assumed constant and disappear from the analysis.

This approach was criticized by a number of authors, including Sir Geoffrey Taylor himself. As he pointed out in Ref. [3], and as discussed later on in Ref. [4], "*it seems therefore that no physical meaning can be attached to the use of equations in which the coefficient of diffusion varies with the time of diffusion, even though the formulae produced by their use do represent adequately the concentrations in particular cases*." To circumvent this problem, a delay-diffusion description was developed (see, for instance, Refs. [4,5]),

$$\begin{aligned}&\partial_t \langle c_{A\gamma}\rangle^\gamma + \langle v_\gamma\rangle^\gamma \partial_x \langle c_{A\gamma}\rangle^\gamma \\ &\quad= \int_0^t \partial_\tau \mathsf{D}_\gamma^{*c}(\tau)\partial_x^2 \langle c_{A\gamma}\rangle^\gamma (t-\tau)d\tau \\ &\quad= \partial_t \mathsf{D}_\gamma^{*c} \star \partial_x^2 \langle c_{A\gamma}\rangle^\gamma (t),\end{aligned} \tag{2}$$

based on the introduction of a memory function, i.e., a time convolution, denoted here by $\star$. In Eq. (2), the time derivative used in the integrand is fundamental because it ensures that $\mathsf{D}_\gamma^{*c}(t)$ conforms with essential requirements associated with a notion of dynamic dispersion. One of these requirements is that Eq. (2) should degenerate into Taylor's dispersion equation in the limit of long times, i.e., when macroscopic times are significantly larger than characteristic times associated with the relaxation of $\mathsf{D}_\gamma^{*c}(t)$ towards Taylor's dispersion coefficient, $\mathsf{D}_\gamma^{*c}(\infty)$. Roughly speaking, this asymptotic behavior corresponds to passing $\mathsf{D}_\gamma^{*c}(t)$ to the limit $u(t)\mathsf{D}_\gamma^{*c}(\infty)$ in the convolution product, where $u(t)$ is the unit step function. The result of this operation leads to the following expression for the right-hand side of Eq. (2):

$$\begin{aligned}&\int_0^t \mathsf{D}_\gamma^{*c}(\infty)\partial_\tau u(\tau)\partial_x^2 \langle c_{A\gamma}\rangle^\gamma (t-\tau)d\tau \\ &\quad= \mathsf{D}_\gamma^{*c}(\infty)\int_0^t \delta(\tau)\partial_x^2 \langle c_{A\gamma}\rangle^\gamma (t-\tau)d\tau \\ &\quad= \mathsf{D}_\gamma^{*c}(\infty)\partial_x^2 \langle c_{A\gamma}\rangle^\gamma (t),\end{aligned} \tag{3}$$

where $\delta(t)$ is the Dirac distribution. Here, we see that the time derivative is indispensable because it yields the correct asymptotic result, Eq. (3), obtained by Taylor in Ref. [1]. Note that a rigorous analysis of convergence, although an interesting problem, is beyond the scope of this Comment.

By formulating their model in the Laplace and frequency domains rather than in the time domain, Valdès-Parada and Alvarez-Ramirez [6] obscure physical interpretation and comparison with previous works. The term dynamic dispersion, as used in Ref. [6], suggests a notion similar to D_γ^{*s}. However, applying an inverse Laplace transform to Eq. (14) in Ref. [6] yields

$$\begin{aligned}&\partial_t \langle c_{A\gamma}\rangle^\gamma + \nabla\cdot(\langle \mathbf{v}_\gamma\rangle^\gamma \langle c_{A\gamma}\rangle^\gamma) \\ &\quad= \nabla\cdot\left[\int_0^t \mathbf{D}_\gamma^*(\tau)\cdot\nabla\langle c_{A\gamma}\rangle^\gamma (t-\tau)d\tau\right] \\ &\quad= \nabla\cdot[\mathbf{D}_\gamma^*\cdot\star\nabla\langle c_{A\gamma}\rangle^\gamma (t)],\end{aligned} \tag{4}$$

which hints at a connection with the convolution formulation, Eq. (2), and $\mathsf{D}_\gamma^{*c}(t)$. In this expression, the tensor, $\mathbf{D}_\gamma^*(t)$, corresponding to Eq. (15) in Ref. [6], can be written in the time domain as

$$\mathbf{D}_\gamma^*(t) = \mathscr{D}_\gamma\left(\mathbf{I}\delta(t) + \frac{1}{\mathscr{V}_\gamma}\int_{A_{\gamma\kappa}} \mathbf{n}_{\gamma\kappa}\mathbf{b}_\gamma(\mathbf{r},t)dA\right) - \langle\tilde{\mathbf{v}}_\gamma\mathbf{b}_\gamma(\mathbf{r},t)\rangle^\gamma, \tag{5}$$

where $\mathbf{b}_\gamma$ solves

$$\partial_t \mathbf{b}_\gamma(\mathbf{r},t) + \mathbf{v}_\gamma \cdot \nabla \mathbf{b}_\gamma(\mathbf{r},t) + \tilde{\mathbf{v}}_\gamma \delta(t) = \mathscr{D}_\gamma \nabla^2 \mathbf{b}_\gamma(\mathbf{r},t), \quad \text{in the phase } (\gamma), \tag{6}$$

with

$$-\mathbf{n}_{\gamma\kappa} \cdot \nabla \mathbf{b}_\gamma(\mathbf{r},t) = \mathbf{n}_{\gamma\kappa}\delta(t), \quad \text{on } A_{\gamma\kappa}. \tag{7}$$

To ensure uniqueness of the solution (see details in [6]), we prescribe the zero initial condition, $\mathbf{b}_\gamma(\mathbf{r},t<0)=\mathbf{0}$, the average constraint, $\langle \mathbf{b}_\gamma(\mathbf{r},t)\rangle^\gamma = \mathbf{0}$, and local periodicity.

Equations (4) and (5) are not wrong and can be used to describe solute transport. However, $\mathbf{D}^*_\gamma(t)$, Eq. (5), cannot be interpreted as a dispersion tensor because Eq. (4) neglects the time derivative used with the convolution in Eq. (2). Therefore, it exhibits features that are inconsistent with the notion of dispersion:

(1) $\mathsf{D}^*_{\gamma,ij}$ have dimensions of $[(\text{length})^2 \times (\text{time})^{-2}]$, instead of $[(\text{length})^2 \times (\text{time})^{-1}]$.

(2) $\mathbf{D}^*_\gamma$ does not yield the correct asymptotic behavior. Considering Eq. (5), we remark that $\mathbf{D}^*_\gamma$ may be approximated by $\mathbf{D}^{*c}_\gamma(\infty)\delta(t)$ in the long-time limit, but not by $\mathbf{D}^{*c}_\gamma(\infty)u(t)$.

(3) $\mathsf{D}^*_{\gamma,ij}$ are distributions, not functions, and $\mathbf{D}^*_\gamma$ is not necessarily positive-semidefinite. Indeed, the identity tensor on the right-hand side of Eq. (5) is weighted by the Dirac distribution, $\delta(t)$. Therefore, diagonal components $\mathsf{D}^*_{\gamma,ii}$ will be *negative* in the early times of the diffusive regime, when the tensor describing tortuosity effects, $\frac{1}{\mathscr{V}_\gamma}\int_{A_{\gamma\kappa}} \mathbf{n}_{\gamma\kappa}\mathbf{b}_\gamma(\mathbf{r},t)dA$, is dominant.

It follows that a three-dimensional analog of Eq. (2) should be used instead of Eq. (4), and reads

$$\partial_t \langle c_{A\gamma}\rangle^\gamma + \nabla\cdot(\langle \mathbf{v}_\gamma\rangle^\gamma \langle c_{A\gamma}\rangle^\gamma) = \nabla\cdot\left[\partial_t \mathbf{D}^{*c}_\gamma \cdot\star \nabla \langle c_{A\gamma}\rangle^\gamma(t)\right], \tag{8}$$

where $\mathbf{D}^{*c}_\gamma(t)$ is the correct transient dispersion tensor. This idea was used by Moyne [7] to develop a two-equation model of transient dispersion. Note that all these models are based on the assumption that spatial memory effects can be neglected, which may be inaccurate if the nonlocal spatial and temporal effects are strongly coupled. To model such effects, a variety of techniques may be used, including nonlocal formulations involving spatial convolutions (see Ref. [8]), direct numerical computation of the transport equations at the pore scale, and higher-order theories.

In addition to this problem in the definition of the dispersion tensor, there is an issue related to the notion of frequency used in Ref. [6]. In theory, $\mathbf{D}^{*c}_\gamma(t)$ and $\mathbf{D}^*_\gamma(t)$ may depend on the frequency, χ, of external periodic operations. For example, Smith in Ref. [5] considered the case of an oscillatory flow with a frequency-dependent velocity, $\mathbf{v}_\gamma(t,\chi)$. In this case, the dispersion coefficient, $\mathsf{D}^{*c}_\gamma(t,\chi)$, depends on the frequency of the excitation signal, $\mathbf{v}_\gamma(t,\chi)$. More generally, $\mathbf{D}^{*c}_\gamma(t)$ will exhibit frequency dependence if at least one of the parameters oscillates in the initial boundary value problem given by Eqs. (13) in Ref. [6] and Eqs. (6) and (7) herein. In such cases, either the velocity field, $\mathbf{v}_\gamma$, the molecular diffusion, $\mathscr{D}_\gamma$, or the position of the interface, $A_{\gamma\kappa}$, would need to vary in time following a periodic pattern.

Such frequency dependence is incompatible with the hypotheses made in Ref. [6]. Indeed, $\mathscr{D}_\gamma$ is a constant scalar; $A_{\gamma\kappa}$ is static; and the flow is implicitly steady. This is obvious in Eq. (3) in Ref. [6] where the Laplace transform does not apply to the velocity field. Consequently, the model is not as general as suggested in the Introduction of [6] and is in fact limited to the description of periodic discharges of solute at the macroscopic level. In this case, $\mathbf{D}^{*c}_\gamma(t)$ has been implicitly decoupled from macroscale source terms during upscaling and is not frequency dependent.

In our opinion, this is the result of a misinterpretation in the frequency analysis. In Eq (8), $\mathbf{D}^{*c}_\gamma(t)$ is a transient parameter and its relaxation can be decomposed into a spectrum of frequencies, ω, via Fourier or Laplace transforms. However, the result of this analysis is independent of the frequency of the solute discharge, which will affect the behavior of the average concentration $\langle c_{A\gamma}\rangle^\gamma$ only through macroscale boundary conditions. Hence, Figs. 4 and 6 in Ref. [6] must be understood as the spectral decomposition of $\mathbf{D}^*_\gamma(t)$ relaxing towards its nonperiodic steady state, not as the frequency analysis of the response of the dispersion tensor to periodic excitations. In addition, Eqs. (6) and (14) in Ref. [6] need to be treated with great care because they describe spatial variations for a *fixed value of Fourier's frequency, ω,* as illustrated in Ref. [6], Sec. VI. Many problems would involve time-dependent boundary conditions characterized by a spectrum of frequencies for which the impact of the convolution would be emphasized.

II. CORRECTION TO THE DISPERSION TENSOR IN THE CASE OF PERIODIC SOLUTE DISCHARGE

In this section, we show that, in the case of periodic macroscale solute discharge, Eq. (8) and $\mathbf{D}^{*c}_\gamma(t)$ can be readily obtained using the results in Ref. [6]. Our analysis is based on the following relationship between $\mathbf{D}^{*c}_\gamma(t)$ and $\mathbf{D}^*_\gamma(t)$ [see Eqs. (4) and (8)]:

$$\mathbf{D}^*_\gamma(t) = \partial_t \mathbf{D}^{*c}_\gamma(t). \tag{9}$$

Further, consider the unilateral Laplace transform defined by $\bar{c}_{A\gamma}(s) = \int_0^\infty e^{-st} c_{A\gamma} dt$. Following [6], we simplify expressions in the Laplace domain by imposing zero initial conditions. With this constraint, we have

$$\bar{\mathbf{D}}^{*c}_\gamma(s) = \frac{1}{s}\bar{\mathbf{D}}^*_\gamma(s). \tag{10}$$

Applying the Laplace transform to Eq. (5) and then using Eq. (10) yields

$$\bar{\mathbf{D}}^{*c}_\gamma(s) = \mathscr{D}_\gamma\left(\mathbf{I}\frac{1}{s} + \frac{1}{\mathscr{V}_\gamma}\int_{A_{\gamma\kappa}} \mathbf{n}_{\gamma\kappa}\frac{\bar{\mathbf{b}}_\gamma(\mathbf{r},s)}{s}dA\right) - \left\langle \tilde{\mathbf{v}}_\gamma \frac{\bar{\mathbf{b}}_\gamma(\mathbf{r},s)}{s}\right\rangle^\gamma. \tag{11}$$

Further, we define $\bar{\mathbf{B}}_\gamma$ by

$$\bar{\mathbf{B}}_\gamma(\mathbf{r},s) = \frac{\bar{\mathbf{b}}_\gamma(\mathbf{r},s)}{s}. \tag{12}$$

Using Eqs. (13) in Ref. [6] leads to the following differential equation for $\tilde{\mathbf{B}}_\gamma(\mathbf{r},s)$:

$$s\tilde{\mathbf{B}}_\gamma(\mathbf{r},s)+\mathbf{v}_\gamma\cdot\boldsymbol{\nabla}\tilde{\mathbf{B}}_\gamma(\mathbf{r},s)+\frac{1}{s}\tilde{\mathbf{v}}_\gamma = \mathscr{D}_\gamma\boldsymbol{\nabla}^2\tilde{\mathbf{B}}_\gamma(\mathbf{r},s),\quad \text{in the phase } (\gamma), \tag{13}$$

with the boundary condition

$$-\mathbf{n}_{\gamma\kappa}\cdot\boldsymbol{\nabla}\tilde{\mathbf{B}}_\gamma(\mathbf{r},s)=\frac{1}{s}\mathbf{n}_{\gamma\kappa},\quad \text{on } A_{\gamma\kappa}, \tag{14}$$

and local periodicity. We also have the average constraint $\langle\tilde{\mathbf{B}}_\gamma\rangle^\gamma=\mathbf{0}$.

Returning to the time domain, the so-called closure given by Eq. (12) in Ref. [6] becomes

$$\tilde{c}_{A\gamma}=\partial_t\,\mathbf{B}_\gamma(\mathbf{r},t)\cdot\star\boldsymbol{\nabla}\langle c_{A\gamma}\rangle^\gamma=\partial_t B_{\gamma,i}*\partial_i\langle c_{A\gamma}\rangle^\gamma, \tag{15}$$

which is reminiscent of the closure proposed in Ref. [7] for the two-equation heat transfer problem. Here, $\mathbf{B}_\gamma(\mathbf{r},t)$ solves the following initial boundary value problem:

$$\partial_t\mathbf{B}_\gamma(\mathbf{r},t)+\mathbf{v}_\gamma\cdot\boldsymbol{\nabla}\mathbf{B}_\gamma(\mathbf{r},t)+\tilde{\mathbf{v}}_\gamma u(t) = \mathscr{D}_\gamma\boldsymbol{\nabla}^2\mathbf{B}_\gamma(\mathbf{r},t),\quad \text{in the phase } (\gamma), \tag{16}$$

with the boundary condition

$$-\mathbf{n}_{\gamma\kappa}\cdot\boldsymbol{\nabla}\mathbf{B}_\gamma(\mathbf{r},t)=\mathbf{n}_{\gamma\kappa}u(t),\quad \text{on } A_{\gamma\kappa}, \tag{17}$$

and local periodicity. We also have the initial condition, $\mathbf{B}_\gamma(\mathbf{r},t<0)=\mathbf{0}$, and the average constraint, $\langle\mathbf{B}_\gamma\rangle^\gamma=\mathbf{0}$. Again, we emphasize that the solution of this problem *does not depend on the frequency* of any source term associated with $\langle c_{A\gamma}\rangle^\gamma$, and that $\mathbf{B}_\gamma(\mathbf{r},t)$, solution of an inhomogeneous linear advection-diffusion equation, will relax towards a nonperiodic steady-state field $\mathbf{B}_\gamma(\mathbf{r},\infty)$ in the asymptotic regime.

In the time domain, the dispersion tensor, $\mathbf{D}^{*c}_\gamma(t)$, reads

$$\mathbf{D}^{*c}_\gamma(t)=\mathscr{D}_\gamma\left(\mathbf{I}u(t)+\frac{1}{\mathscr{V}_\gamma}\int_{A_{\gamma\kappa}}\mathbf{n}_{\gamma\kappa}\mathbf{B}_\gamma(\mathbf{r},t)dA\right) -\langle\tilde{\mathbf{v}}_\gamma\mathbf{B}_\gamma(\mathbf{r},t)\rangle^\gamma. \tag{18}$$

The traditional steady-state dispersion tensor can be expressed as

$$\mathbf{D}^{*c}_\gamma(\infty)=\mathscr{D}_\gamma\left(\mathbf{I}+\frac{1}{\mathscr{V}_\gamma}\int_{A_{\gamma\kappa}}\mathbf{n}_{\gamma\kappa}\mathbf{B}_\gamma(\mathbf{r},\infty)dA\right)-\langle\tilde{\mathbf{v}}_\gamma\mathbf{B}_\gamma(\mathbf{r},\infty)\rangle^\gamma. \tag{19}$$

III. NUMERICAL RESULTS

To illustrate the temporal behavior of this transient dispersion tensor, we use COMSOL MULTIPHYSICS 4.2 to solve numerically the following nondimensionalized closure problem for the periodic unit cell described in [6] with $\varepsilon_\gamma=0.56$:

$$\partial_\tau\mathbf{B}'_\gamma+\mathbf{v}'_\gamma\cdot\boldsymbol{\nabla}_{\mathbf{r}'}\mathbf{B}'_\gamma+\tilde{\mathbf{v}}'_\gamma=\frac{1}{\mathrm{Pe}}\nabla^2_{\mathbf{r}'}\mathbf{B}'_\gamma,\quad \text{in the phase } (\gamma), \tag{20}$$

with $\mathbf{B}'_\gamma=\mathbf{B}_\gamma/(l_\gamma-l_\kappa)$, $\mathbf{r}'=\mathbf{r}/(l_\gamma-l_\kappa)$, $\tau=\sqrt{\langle\mathbf{v}_\gamma\rangle^\gamma\cdot\langle\mathbf{v}_\gamma\rangle^\gamma}\,t/(l_\gamma-l_\kappa)$, $\mathrm{Pe}=\sqrt{\langle\mathbf{v}_\gamma\rangle^\gamma\cdot\langle\mathbf{v}_\gamma\rangle^\gamma}(l_\gamma-l_\kappa)/\mathscr{D}_\gamma$, and $\mathbf{v}'_\gamma=\mathbf{v}_\gamma/\sqrt{\langle\mathbf{v}_\gamma\rangle^\gamma\cdot\langle\mathbf{v}_\gamma\rangle^\gamma}$. Boundary conditions are

$$-\mathbf{n}_{\gamma\kappa}\cdot\boldsymbol{\nabla}_{\mathbf{r}'}\mathbf{B}'_\gamma=\mathbf{n}_{\gamma\kappa}u(\tau),\quad \text{on } A_{\gamma\kappa}, \tag{21}$$

and local periodicity. We also have the initial condition, $\mathbf{B}'_\gamma(\mathbf{r}',\tau<0)=\mathbf{0}$, and the average constraint, $\langle\mathbf{B}'_\gamma\rangle^\gamma=\mathbf{0}$.

Similarly, the flow was obtained by imposing a pressure gradient and solving periodic Stokes equations. The longitudinal dispersion coefficient, $\mathsf{D}^{*c}_{\gamma,xx}(t)/\mathscr{D}_\gamma$, was computed using Eq. (18). Results for different values of the Péclet number, Pe, are plotted in Fig. 1. The dispersion coefficient clearly relaxes towards its steady state, $\mathsf{D}^{*c}_{\gamma,xx}(\infty)$. Figure 6(a) in Ref. [6] shows a very similar behavior for the longitudinal dispersion with a reflection symmetry that indicates the time-frequency relationship $\omega\sim t^{-1}$. This shows that the analysis carried out in Ref. [6] describes the relaxation of $\mathbf{D}^*_\gamma(t)$ *in the frequency domain*.

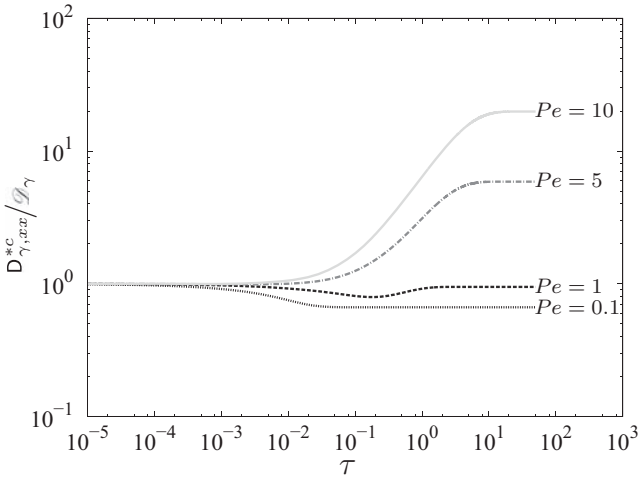


FIG. 1. Logarithmic plots illustrating the temporal behavior of $\mathsf{D}^{*c}_{\gamma,xx}/\mathscr{D}_\gamma$ [implicitly weighted by $u(\tau)$] for the unit cell presented in Ref. [6] and $\varepsilon_\gamma=0.56$. Results were computed using COMSOL MULTIPHYSICS 4.2. In the short-time regime, $\mathsf{D}^{*c}_{\gamma,xx}/\mathscr{D}_\gamma$ tends towards unity because we have assumed zero initial conditions for $\mathbf{B}'_\gamma(\mathbf{r},t)$. In the long-time regime, $\mathsf{D}^{*c}_{\gamma,xx}/\mathscr{D}_\gamma<1$ when tortuosity effects dominate dispersion effects and $\mathsf{D}^{*c}_{\gamma,xx}/\mathscr{D}_\gamma>1$ when dispersion effects dominate tortuosity effects.

This application also highlights that time-dependent problems must be treated with care. In general, formulations similar to Eq. (8) should be used, especially when boundary conditions are characterized by a single relatively large frequency or by a spectrum involving large frequencies, or when small-time relaxation phenomena are of interest. On the other hand, if relevant time-scale constraints are satisfied, the asymptotic version of Eq. (8) may be used.

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