

Evaluation of the first-order approximations for transport in heterogeneous media

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Longitudinal dispersion coefficients in given realizations of the transport computed by two currently used approximations of the first-order in velocity variance are compared with accurate global random walk simulations. The comparisons are performed for the same ensemble of realizations of the Darcy velocity field, approximated by a quasiperiodic random field, for lognormal hydraulic conductivity with small variance and finite correlation lengths. The results show that at finite times of about one dispersion timescale, the mean coefficient is underestimated by $\approx 20\%$, and the fluctuations are overestimated by $\approx 80\%$. At larger times the errors decrease monotonously, and the first-order approximations yield fairly good predictions for the mean and the fluctuations of the dispersion coefficient.

1. Introduction

[2] In this technical note, by “first-order approximations” we denote asymptotic expansions of the solutions of the transport equations, for given velocity realizations, which approximate second-order moments of the concentration or dispersion coefficients at the same order of magnitude as the velocity variance. These first-order approaches to transport in saturated porous formations can be classified as “Eulerian perturbation methods” (EPM) and “Langevin iteration methods” (LIM).

[3] The EPM method considered here derives the approximations by a perturbation approach applied to the solution of the partial differential equation for the concentration field $c(\mathbf{x}, t)$ [Bouchaud and Georges, 1990; Dentz et al., 2003; Eberhard, 2004]. For constant porosity and nonreactive transport, c obeys the advection-dispersion equation

$$\partial_t c + \mathbf{V} \nabla c = D_0 \nabla^2 c. \quad (1)$$

For the sake of comparisons, we consider in (1) a constant local dispersion coefficient D_0 and a stationary velocity field $\mathbf{V}$ which is a realization of a statistically homogeneous random space function. The unperturbed problem in EPM methods is obtained by replacing $\mathbf{V}$ in (1) by the constant mean of the velocity field. Systematic EPM expansions are obtained by iterations of an integral equation for Fourier transformed concentration equivalent to (1) [Bouchaud and Georges, 1990, section 4.2.2]. Here we consider the ap-

proximation of the dispersion coefficients for a single realization of the velocity field given by the first EPM iteration of Eberhard [2004].

[4] LIM methods use the Langevin equation for the trajectories of the transport process which is equivalent to the Eulerian equation (1) [see, e.g., Bouchaud and Georges, 1990, section 2.3]. In the following we consider a point-like instantaneous source located at the origin of the coordinate system. For a given realization of the velocity field, the equivalent description is given by the ensemble of all trajectories starting from $\mathbf{X}(0) = 0$, which solve the integral form of the Langevin equation

$$X_i(t) = \int_0^t V_i(\mathbf{X}(t')) dt' + \int_0^t dW_i(t'), \quad (2)$$

where $i = 1, \dots, d$ and d is the space dimension. The second integral in (2) is a Wiener process with zero mean and variance $2D_0 t$.

[5] Following Eberhard [2004], the LIM method can be formulated as an approximation of (2) given by

$$X_i^{(0)}(t) = (Ut)\delta_{i1} + \int_0^t dW_i(t'), \quad (3)$$

$$X_i^{(1)}(t) = (Ut)\delta_{i1} + \int_0^t u_i(\mathbf{X}^{(0)}(t')) dt' + \int_0^t dW_i(t'), \quad (4)$$

where U is the constant mean velocity, $u_i = V_i - U\delta_{i1}$ are the random velocity fluctuations and the 1 axis of the coordinate system is oriented in the direction of the mean flow. It is obvious that the first iteration of the Langevin equation (4) is the approximation obtained “by leaving the ‘Brownian motion’ component as part of the zeroth-order term” which gives the particle displacement covariance (3.14) of Dagan [1987]. This formula is a key tool for investigations in the spirit of Dagan’s theory of transport

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[Fiori and Dagan, 2000]. It is also noteworthy to remark that (3) and (4) represent the first step in the Picard method of successive approximations, as used for instance in numerical schemes for solving the Langevin equation [Carvalho *et al.*, 2005] or in analytical approximations for the Lagrangian equation of motion in random fields [Phythian, 1975].

[6] The Langevin equation (3) describes a diffusion in the mean velocity field $\mathbf{U}$ and is strictly equivalent to the unperturbed problem of the EPM method. Both EPM and LIM approximate ensemble averaged dispersion coefficients up to the order of the velocity variance. It is expected therefore that EPM and LIM, having in common the reference to the same unperturbed problem yield equivalent first-order approximations.

[7] The validity of the first-order stochastic theories of flow and transport in porous media has mainly been investigated in the case of vanishing local dispersion [see, e.g., Chin and Wang, 1992]. For nonvanishing local dispersion, the existing comparisons between EPM solution for transport in Gaussian velocity fields and particle tracking simulations, using approximations by quasiperiodic random functions of the Gaussian field, show large disagreement, mostly on the longitudinal dispersion coefficient [Dentz *et al.*, 2003]. What has not yet been done is an evaluation of the predictions provided by the first-order theories for the fluctuations of the dispersion coefficients. The latter is crucial in assessing the reliability of first-order theory for investigations on the self-averaging properties of the transport. Here, we disjoint the transport and flow problems. Using the same ensemble of velocity realizations, we evaluate the EPM and LIM approximations for the mean and the fluctuations of the longitudinal dispersion coefficient by comparisons with a numerical method which was independently validated.

2. Dispersion Quantities

[8] In the following by X we denote the longitudinal trajectory component described by (2). The longitudinal dispersion of the solute in a given realization of the transport is described by the second-order central moment

$$S(t) = \langle [X(t) - \langle X(t) \rangle_{\mathbf{w}}]^2 \rangle_{\mathbf{w}}, \quad (5)$$

where the subscript $\mathbf{w}$ denotes the average over the realizations of the Wiener process. The dispersion coefficients $D = \frac{1}{2}S/t$ and $D_1 = \frac{1}{2}dS/dt$ have the meaning of a diffusion constant when the transport in heterogeneous media has “normal” (or “Fickian”) diffusion behavior at large times, i.e., $D = D_1$ and $S = 2tD$ [Bouchaud and Georges, 1990]. Other dispersion quantities are provided by the second central moment of the ensemble averaged concentration

$$\Sigma(t) = \langle [X(t) - \langle X(t) \rangle_{\mathbf{wv}}]^2 \rangle_{\mathbf{wv}}, \quad (6)$$

where the subscript $\mathbf{v}$ stands for an additional ensemble average, and by the corresponding “average” dispersion coefficients $D^{av} = \frac{1}{2}\Sigma/t$ and $D_1^{av} = \frac{1}{2}d\Sigma/dt$. We note that the difference between (6) and the ensemble average of (5) gives the variance of the center of mass of the plume [Suci

et al., 2006, equation (8)]. The coefficient D^{av} characterizes the envelope of the diffusion fronts in given realizations and is in general larger than the ensemble average $\langle D \rangle_{\mathbf{v}}$ of the single realization dispersion coefficient [see, e.g., Bouchaud and Georges, 1990, section 2.1, and references therein]. The ensemble average predictor of the dispersive flux $\mathbf{J} = \mathbf{V}c - D_0\nabla c$ corresponding to processes described by (1) is nonlocal in space-time and non-Fickian. Even if localized forms can be derived under some restrictive conditions, the localized dispersion coefficients still depend on space and time [Morales-Casique *et al.*, 2006a]. Therefore, at finite times the dispersion coefficients can merely be used as convenient representations of the second moments (5) and (6).

[9] With the transformations $X = \hat{X}\lambda$, $t = \hat{t}\lambda/U$, $V = U + \varepsilon\hat{u}U$ and $W = \hat{W}(D_0\lambda/U)^{1/2}$, the nondimensional form of (2) for the longitudinal displacement reads

$$\hat{X}(\hat{t}) = \hat{t} + \varepsilon \int_0^{\hat{t}} \hat{u}(\hat{t}') d\hat{t}' + Pe^{-1/2} \int_0^{\hat{t}} d\hat{W}(\hat{t}'),$$

where $Pe = U\lambda/D_0$ is the Péclet number. The small parameter ε which describes the velocity fluctuations can be estimated by the relative standard deviation of the longitudinal velocity $\varepsilon \simeq \sigma_u/U$. Since for the typical advection dominated transport investigated here $\varepsilon = 0.19$ and $Pe^{-1/2} = 0.1$ (see next section), we have the following order of magnitude relation $\varepsilon = \mathcal{O}(Pe^{-1/2})$. In a consistent expansion of $\hat{X}$ for $\varepsilon \rightarrow 0$, the leading term cannot contain contributions of order higher than ε^0 . Thus (3) has to be replaced by $X^{(0)}(t) = Ut$ and the first iteration (4) yields a diffusion process in a deterministic time-dependent velocity field $U + u(Ut)$, with variance $S(t) = \langle [X^{(1)}(t) - \langle X^{(1)}(t) \rangle_{\mathbf{w}}]^2 \rangle_{\mathbf{w}} = 2D_0t$. It follows that for all realizations $D = \frac{1}{2}S/t = D_0$. However, the consistent first order approximation captures the enhanced dispersion caused by velocity fluctuations through the moment (6) of the ensemble averaged concentration. Owing to the independence between dispersive and advective displacements, the corresponding average longitudinal coefficient $D_1^{av} = \frac{1}{2}d\Sigma/dt$ is readily shown to be

$$D_1^{av}(t) = D_0 + \int_0^t r(Ut') dt', \quad (7)$$

where $r(Ut) = \langle u(0)u(Ut) \rangle_{\mathbf{v}}$ is the Eulerian correlation of the longitudinal velocity component. The coefficient (7) is the output of the earlier “first-order” approximation of Dagan [1984], investigated numerically (for $D_0 = 0$) by Chin and Wang [1992].

3. Evaluation Method

[10] Our approach is based on an approximation of Darcy flow by quasiperiodic random fields, which has the advantage to result in explicit EPM and LIM expressions for the effective coefficients in single realizations of the transport and to allow large-scale numerical simulations at reasonable computational costs. This approximation, in the form of the Kraichnan routine, was already successfully used in numerical investigations on large-scale behavior of the dispersion coefficients [Dentz *et al.*, 2003; Eberhard, 2004]. The

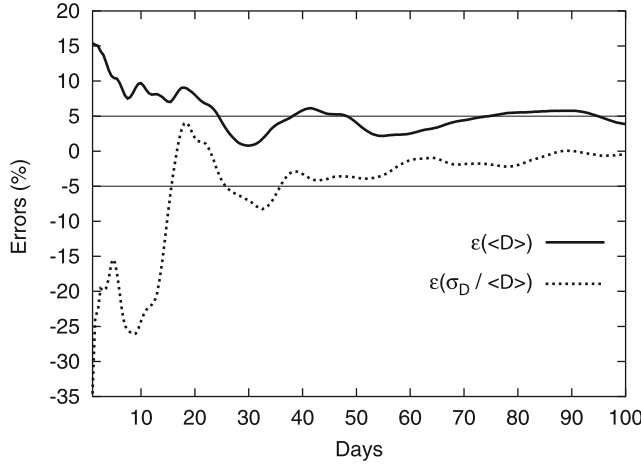


Figure 1. Error estimations for the GRW computations.

incompressible Darcy flow for log hydraulic conductivity with small variance σ^2 and Gauss-shaped isotropic correlation with correlation length λ is approximated in the order σ by a superposition of N_p periodic modes

$$V_i(\mathbf{x}) = U\delta_{i1} + U\sigma\sqrt{\frac{2}{N_p}} \sum_{l=1}^{N_p} p_l(\mathbf{q}_l) \cos(\mathbf{q}_l \cdot \mathbf{x} + \alpha_l). \quad (8)$$

The wave vectors $\mathbf{q}_l$ are independently normally distributed random variables with zero mean and variance λ^{-2} , and the phases α_l are uniformly distributed in the interval $[0, 2\pi]$. The functions p_l are projectors which ensure the incompressibility of the flow. For $N_p \rightarrow \infty$, V_i tends to a Gaussian velocity field [Kraichnan, 1970].

[11] For the evaluation of the first-order approximations we consider the two-dimensional transport problem for fixed local dispersion coefficient, $D_0 = 0.01$ m²/d, and an ensemble of 512 velocity realizations characterized by the parameters $\sigma^2 = 0.1$, $\lambda = 1$ m, $U = 1$ m/d, and $N_p = 6400$. The estimated standard deviation of the longitudinal velocity component (8) is $\sigma_u = 0.19$ m/d. For the small value $\sigma^2 = 0.1$ the Darcy velocity field can be accurately described by a Gaussian distribution (as indicated, for example, by Figure 20 of Morales-Casique *et al.* [2006b]) and the results based on the Kraichnan procedure can be relevant for real cases. It should also be noted that accuracy for larger σ^2 and/or Péclet numbers demands higher-order approximations, as plentifully demonstrated by recent investigations on the iterative expansions of the exact nonlocal equations for the ensemble average and the variance of the concentration [Morales-Casique *et al.*, 2006a, 2006b].

[12] The evaluation is performed by comparisons for the same ensemble of transport realizations between first-order approximations and numerical simulations by the “global random walk” (GRW) algorithm, implemented in the so-called “reduced fluctuations” version, see [Vamoş *et al.*, 2003]. Statistically stable simulations of transport in each realization of the velocity field are obtained by releasing $N = 10^{10}$ particles at the origin of the grid [Suciu *et al.*, 2006]. In order to simulate the advective-dispersive transport in unbounded domains, the grid, with constant step of 0.1 m, is chosen to be larger than the maximum extension of the plume. The restriction to the two-dimensional case enables us to

perform accurate GRW simulations over thousands of correlation lengths, which are necessary to describe the convergence toward the macrodispersion limit. We evaluate numerically, for 512 realizations (8) of the velocity, the explicit EPM and LIM expressions for D_1 given respectively in Appendices A1 and B1 of Eberhard [2004], adapted for two-dimensional problems. Comparisons are done between the approximated coefficient $D = \frac{1}{t} \int_0^t D_1(t') dt'$ and the coefficient D given by GRW simulations.

[13] Before we address the evaluation of EPM and LIM, we give an evaluation of the GRW procedure. This is done by comparisons using a recently developed method [Suciu *et al.*, 2005] called “biased global random walk” (BGRW). Unlike GRW and particle tracking procedure, BGRW simulates the advective displacements by a bias in the probability associated with the diffusive jumps of the particles. Since in a single computation step the particles move only to the nearest grid sites, BGRW has no overshooting errors. However, as BGRW requires larger computing resources, GRW is more suitable for large-scale computations.

[14] The overshooting errors in GRW procedure are estimated by the percentage relative error $\varepsilon(\beta) = 100[\beta(\text{GRW}) - \beta(\text{BGRW})]/\beta(\text{BGRW})$, where β stands, respectively, for the average $\langle D \rangle_v$ of the dispersion coefficient over the ensemble of velocity realizations and for the relative standard deviation $\sigma_D / \langle D \rangle_v$. The comparison with BGRW was limited to 100 days, $N_p = 64$ modes in the Kraichnan routine and 256 realizations. The results presented in Figure 1 show that after about 35 days the mean as well as the fluctuations are reproduced in GRW procedure with errors of the order of 5% (indicated by horizontal lines in Figure 1).

[15] The ensemble mean dispersion coefficients $\langle D \rangle_v$ given by EPM and LIM are compared to the GRW results in Figure 2 and the corresponding sample-to-sample fluctuations $\sigma_D / \langle D \rangle_v$ are presented in Figure 3. Additionally, by curves labeled EPM₁ and LIM₁, we represented in Figures 2 and 3 the means and the fluctuations of the primary data D_1 given by the two approximations. The consistent first-order approximation of the average coefficient (7) was computed by numerical integration of the Eulerian velocity correla-

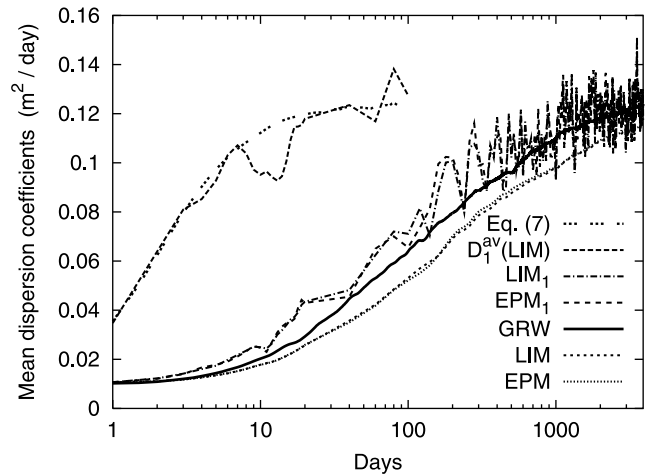


Figure 2. Comparisons for the mean dispersion coefficients given by different first-order approximations and by the GRW simulations.

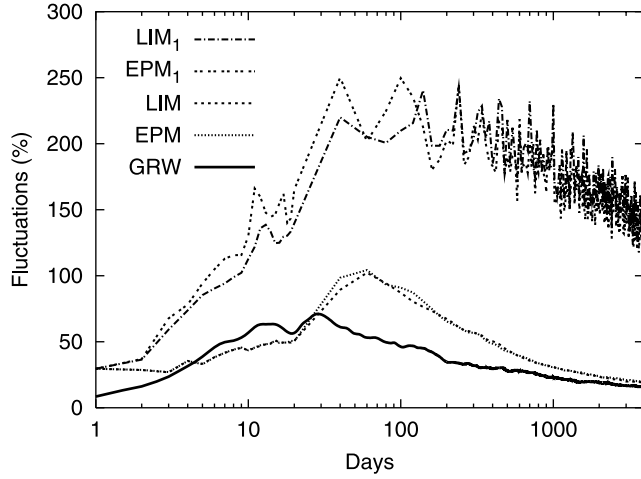


Figure 3. Comparison between the fluctuations of the dispersion coefficient given by the different first-order approximations and GRW simulations.

tion, estimated as follows. First, the correlations are computed for ensembles of 512 velocity realizations generated by (8) on straight lines starting from all grid points in a slab with dimensions $\lambda \times 100\lambda$, oriented across the mean flow. Then, in order to obtain a better estimation for the theoretical statistical homogeneous Gaussian field, the correlation was estimated by an average over all initial grid points. The coefficient (7) is also represented in Figure 2 and compared to D_1^{av} (LIM), which represents the average coefficient $\frac{1}{2}d\Sigma/dt$ derived by LIM method, using (3), (4) and (6).

4. Interpretation of the Results

[16] The numerical results for LIM and EPM, presented in Figures 2 and 3, are very similar, indicating that the two methods give equivalent asymptotic expansions in velocity fluctuations. The output D_1 of LIM and EPM behaves noisily and shows much larger sample-to-sample fluctuations than its time average D . Excepting the noise, the coefficient D_1^{av} (LIM) is practically identical with the average coefficient (7) given by the consistent first-order expansion (Figure 2). This is another indication for the robustness of the first-order approximation of *Dagan* [1984] for the second moments (6) of the ensemble averaged concentration, which completes the conclusion drawn by *Chin and Wang* [1992] for the purely advective transport. Since the average coefficients D_1^{av} highly underestimate the traveltime necessary to approach the macrodispersive behavior (see Figure 2), the moments (5) and the associated coefficients D and D_1 have to be used to describe a typical solute plume [*Bouchaud and Georges*, 1990, section 2.1]. As we have seen in the section 2 above, the consistent expansion truncated at the first order in velocity fluctuations does not capture the behavior of the typical dispersion coefficient D and its fluctuations. By leaving the effect of the local dispersion in the zeroth-order term, the LIM and EPM methods avoid the complexity of higher-order approximations and provide useful analytical expressions for the effective coefficients in given realizations.

[17] The deviations of the first-order approximations from the results given by simulations are significantly larger

than the error estimations for GRW presented in Figure 1. This is clearly shown by Figure 4, where the relative errors with respect to GRW simulations, for the mean and the fluctuations computed by LIM, are shown in the same percentage representation as in Figure 1. For instance, at 100 days which corresponds to one dispersion timescale λ^2/D_0 , the mean coefficient is underestimated by approximately 20% and the fluctuations are overestimated by 80%. For large times the errors decrease and approach the overshooting error estimations for GRW (horizontal lines in Figure 4).

[18] The to some extent similar two-dimensional simulations of *Dentz et al.* [2003] (performed by tracking ~ 100 particles in 2500 realizations of Kraichnan fields with $N_p = 64$ modes, for $Pe = 100$ and $\sigma^2 = 1$) indicate that the EPM analytical approximation for Gaussian velocity fields underestimates the mean longitudinal dispersion coefficient $\langle D_1 \rangle_v$ at one dispersion timescale by about 200%. As expected, the higher variance of the log hydraulic conductivity field results in larger velocity fluctuations and deteriorates the accuracy of the approximation. However, the large deviations from simulations can also be explained by trapping phenomena related to closed streamlines in the velocity field generated by the Kraichnan method [*Dentz et al.*, 2003].

5. Conclusions

[19] The evaluation presented in this technical note is based on statistically stable GRW simulations (obtained with $N = 10^{10}$ particles in one realization), for the parameters $Pe = 100$ and $\sigma^2 = 0.1$ which minimize possible artifacts in such two-dimensional numerical simulations, and uses 512 Kraichnan velocity realizations with $N_p = 6400$ periodic modes which ensure reliable simulations of transport in Gaussian fields [*Suciu et al.*, 2006, sections A2, B1, and B2]. Moreover, the LIM and EPM approximations are computed for the same ensemble of Kraichnan velocities and the GRW procedure itself is also independently evaluated. Under these conditions, the differences shown in Figures 2–4 have to be caused by the first-order approximations of the transport equations. The approximation errors given in Figure 4 provide useful minimal estimations

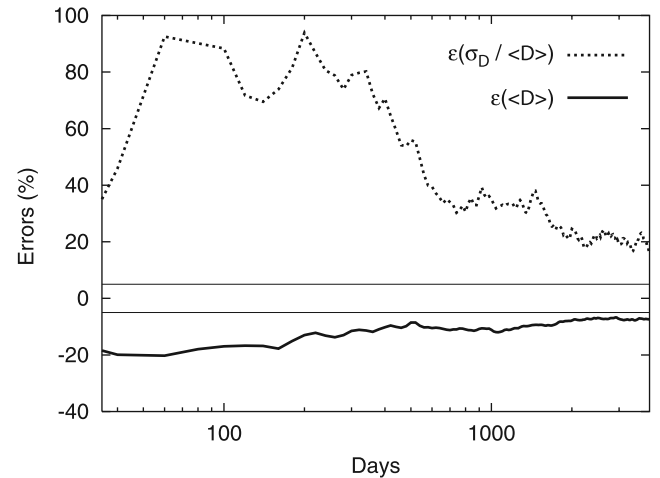


Figure 4. Error estimations for the mean and the fluctuations of the dispersion coefficient computed by the first-order approximations.

for larger σ^2 and/or Pe , when the accuracy is expected to be worse [Dentz et al., 2003; Morales-Casique et al., 2006b]. For moderate variability of the hydraulic conductivity these approximations exhibit reasonable predictions for times larger than the dispersion scale. The convergence of the errors for the mean value and the fluctuations of the longitudinal coefficient to the precision of the numerical method further indicates that the LIM and EPM approximations are mainly useful in investigations on large-scale behavior and self-averaging properties of the transport.

[20] On the basis of this evaluation the LIM method has been used to investigate the performance of the Kraichnan routine in more detail. It has been found that the fluctuations of the dispersion coefficients of transport in Kraichnan fields grow logarithmically, after a time that increases with the number of periodic modes. However, using a number of modes of the order of the total simulation time and hundreds of realizations, the simulations based on the Kraichnan method reproduce quite well the self-averaging behavior of transport in Gaussian fields with finite correlation range (J. Eberhard et al., On the self-averaging of dispersion for transport in random media, submitted to Journal of Physics A: Mathematical and General, 2006).

[21] Insofar as the advection-dispersion local-scale process, described by (1) or (2), is an acceptable description for real world contamination scenarios, the approximations evaluated here could be very useful in applications. EPM and LIM provide dispersion quantities D and D_1 equivalent to second moments of the plume, without solving the transport equations and regardless the Fickian or non-Fickian behavior of the process. The best fit of theoretical and field-scale measured quantities allows, in principle, the estimation of the parameters σ^2 and λ . Dagan [1984] determined σ^2 and λ by a best fit of the time averaged coefficient (7) and the coefficient $\frac{1}{2}S/t$ measured in the Borden tracer test. Since, as shown in Figure 2, the coefficient (7) overestimates the time behavior of typical solute plumes at early times, the results obtained in this way ($\sigma^2 = 0.19$ and $\lambda = 1.4$ m for horizontal correlation scale) underestimate the range of parameters obtained by different methods ($\sigma^2 = 0.25 \div 0.39$ and $\lambda = 2.7 \div 11.6$ m) [Rajaram and Gelhar, 1991]. By comparing the measured slope of the longitudinal second moment with the EPM theoretical expression of $\langle D_1 \rangle_v$, Dentz et al. [2000] found a good agreement, when parameters relevant for Borden site were used. The evaluation of first-order approximations presented in this note suggests an improvement of the parameter identification by using the mean slope $D = \frac{1}{2}S/t$ instead of the local one, $D_1 = \frac{1}{2}dS/dt$. The former is affected by much smaller fluctuations, as shown in Figure 3, and is therefore more representative for single realizations such as a field experiment.

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