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Solute transport in aquifers with evolving scale heterogeneity

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Abstract

Transport processes in groundwater systems with spatially heterogeneous properties often exhibit anomalous behavior. Using first-order approximations in velocity fluctuations we show that anomalous superdiffusive behavior may result if velocity fields are modeled as superpositions of random space functions with correlation structures consisting of linear combinations of short-range correlations. In particular, this corresponds to the superposition of independent random velocity fields with increasing integral scales proposed as model for evolving scale heterogeneity of natural porous media [Gelhar, L. W. Water Resour. Res. 22 (1986), 135S-145S]. Monte Carlo simulations of transport in such multi-scale fields support the theoretical results and demonstrate the approach to superdiffusive behavior as the number of superposed scales increases.

1 Lagrangian and Eulerian representations of diffusion in random velocity fields

Let $\{X_{i,t} = X_i(t), i = 1, 2, 3, t \geq 0\}$ be the advection-dispersion process with a space variable drift $\mathbf{V}(\mathbf{x})$, sample of a statistically homogeneous random

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velocity field, and a local diffusion coefficient D, assumed constant, described by the Itô equation

$$X_{i,t} = x_{i,0} + \int_0^t V_i(\mathbf{X}_{t'})dt' + W_{i,t}, \tag{1}$$

where $x_{i,0}$ is a deterministic initial position and W_i are the components of a Wiener process of mean zero and variance $\langle W_{i,t}^2 \rangle = 2Dt$.

The solution g of the Fokker-Planck equation

$$\partial_t g + \mathbf{u}\nabla g = D\nabla^2 g, \tag{2}$$

which corresponds to Green's function in Eulerian approaches to dispersion in random fields, is the density of the transition probability $P(d\mathbf{x}, t, \mathbf{x}_0) = g(\mathbf{x}, t|\mathbf{x}_0)d\mathbf{x}$ of the Itô process (1). Equation (2) is the starting point in Eulerian perturbation approaches [1, 7, 8].

The following three centered processes are useful to describe the effective and ensemble dispersion and the fluctuations of the center of mass: $X_{i,t}^{eff} = X_{i,t} - \langle X_{i,t} \rangle$, the process centered about the mean $\langle X_{i,t} \rangle$ of $X_{i,t}$ in single realizations of the random velocity field, $\langle X_{i,t}^{eff} \rangle = 0$, the process $X_{i,t}^{ens} = X_{i,t} - \overline{\langle X_{i,t} \rangle}$ centered about the average $\overline{\langle X_{i,t} \rangle}$ of $\langle X_{i,t} \rangle$ over the ensemble of realizations, $\overline{\langle X_{i,t}^{ens} \rangle} = 0$, and the fluctuation of the center of mass $X_{i,t}^{cm} = \langle X_{i,t} \rangle - \overline{\langle X_{i,t} \rangle}$, with $\overline{X_{i,t}^{cm}} = 0$. The half derivative of the variance of these tree processes defines, respectively, the **effective** and **ensemble** dispersion coefficients [1], and the **center of mass** coefficient [24],

$$\begin{aligned}
D_{ii}^{eff}(t) &= \frac{1}{2}\frac{dS(t)}{dt}, \quad S_{ii}(t) = \overline{\langle (X_t^{eff})^2 \rangle}, \\
D_{ii}^{ens}(t) &= \frac{1}{2}\frac{d\Sigma(t)}{dt}, \quad \Sigma_{ii}(t) = \overline{\langle (X_t^{ens})^2 \rangle}, \\
D_{ii}^{cm}(t) &= \frac{1}{2}\frac{dR(t)}{dt}, \quad R_{ii}(t) = \overline{\langle (X_t^{cm})^2 \rangle},
\end{aligned} \tag{3}$$

related by $D_{ii}^{cm}(t) = D^{ens}(t) - D^{eff}(t)$.

According to (1), the process $X_{i,t}^{ens}$ starting from $x_{i,0} = 0$ verifies

$$X_{i,t}^{ens} = \int_0^t u_i(\mathbf{X}_{t'})dt' + W_{i,t}, \tag{4}$$

where $u_i = V_i - \overline{\langle V_i \rangle}$. The variance of the process (4) is obtained as follows,

$$\begin{aligned}
\Sigma_{ii}(t) &= \int_0^t \int_0^t \overline{\langle u_i(\mathbf{X}_{t'})u_i(\mathbf{X}_{t''})\rangle} dt'dt'' + 2\int_0^t \overline{\langle u_i(\mathbf{X}_{t'})W_{i,t}\rangle} dt' + \langle W_{i,t}^2 \rangle \\
&= 2Dt + 2\int_0^t dt' \int_0^{t'} \overline{\langle u_i(\mathbf{X}_{t'})u_i(\mathbf{X}_{t''})\rangle} dt'',
\end{aligned}$$

where we used the statistical homogeneity of the velocity field, $\overline{\langle u_i \rangle} = 0$, and the independence of the Wiener process on velocity statistics, which, together, cancel the cross-correlation term. The process $X^{cm}_{i,t}$ verifies (4) with $u_i = \langle V_i \rangle - \overline{\langle V_i \rangle}$ and the second term set to zero and has the variance

$$R_{ii}(t) = 2 \int_0^t \int_0^t \overline{\langle u_i(\mathbf{X}_{t'}) \rangle \langle u_i(\mathbf{X}_{t''}) \rangle} dt' dt''.$$

Further, the definitions (3) yield

$$D^{ens}_{ii}(t) = D + \int_0^t \overline{\langle u_i(\mathbf{X}_t) u_i(\mathbf{X}_{t'}) \rangle} dt', \tag{5}$$

$$D^{cm}_{ii}(t) = \int_0^t \overline{\langle u_i(\mathbf{X}_t) \rangle \langle u_i(\mathbf{X}_{t'}) \rangle} dt', \tag{6}$$

$$D^{eff}_{ii}(t) = D^{ens}_{i,i}(t) - D^{cm}_{i,i}(t). \tag{7}$$

The first iteration of (4) about the un-perturbed problem consisting of diffusion with constant coefficient D in the mean flow field $U = \overline{\langle V_i \rangle}$, with trajectory $X^{(0)}_{i,t} = \delta_{j,1} U t + W_{i,t}$, leads to replacing $\mathbf{X}_t$ by $\mathbf{X}^{(o)}_t$ in (5) and (6). Then, (5) gives the following first-order approximation of the ensemble dispersion coefficient

$$\begin{aligned} D^{ens}_{ii}(t) - D &= \int_0^t \overline{\langle u_i(\mathbf{X}^{(0)}_t) u_i(\mathbf{X}^{(0)}_{t'}) \rangle} dt' \\ &= \int_0^t dt' \int \int \overline{u_i(\mathbf{x}) u_i(\mathbf{x}')} p(\mathbf{x}, t; \mathbf{x}', t') d\mathbf{x} d\mathbf{x}', \end{aligned} \tag{8}$$

where $p(\mathbf{x}, t; \mathbf{x}', t') = \langle \delta(\mathbf{x} - \mathbf{X}^{(0)}_t) \delta(\mathbf{x}' - \mathbf{X}^{(0)}_{t'}) \rangle$ is the Gaussian joint probability density of the process $X^{(0)}_{i,t}$ expressed with the aid of the Dirac δ-function [23]. Similarly, with $p(\mathbf{x}, t) = \langle \delta(\mathbf{x} - \mathbf{X}^{(0)}_t) \rangle$, (6) gives the first-order approximation of the center of mass coefficient

$$D^{cm}_{ii} = \int_0^t dt' \int \int \overline{u_i(\mathbf{x}) u_i(\mathbf{x}')} p(\mathbf{x}, t) p(\mathbf{x}', t') d\mathbf{x} d\mathbf{x}'. \tag{9}$$

Finally, $D^{eff}_{ii}(t)$ is obtained from (7). Together, (7-9) give the (stochastic) Lagrangian formulation of the first-order approximation equivalent to that obtained in the Eulerian perturbation approach [1, 7, 8].

Remark 1. As follows from (8-9), for a superposition $u_i(\mathbf{x}) = u^1_i(\mathbf{x}) + \cdots + u^n_i(\mathbf{x})$ of statistically independent velocity fields the dispersion coefficients estimated to the first-order are given by the sum of terms determined by the corresponding correlations $\overline{u^1_i(\mathbf{x}) u^1_i(\mathbf{x}')}, \cdots, \overline{u^n_i(\mathbf{x}) u^n_i(\mathbf{x}')}$.

2 Quantifying anomalous diffusion by memory terms

In the following, by X_t we denote one of the processes $X_{i,t}$, $X_{i,t}^{eff}$, $X_{i,t}^{ens}$, or $X_{i,t}^{cm}$. Further, we consider the uniform time partition $0 < \Delta < \cdots < k\Delta < \cdots < (K-1)\Delta < K\Delta$ and for $k > 0$ we define the increments $X_{k,k-1} = X_{t_k} - X_{t_{k-1}}$ and $s_{k,k-1} = (X_{k,k-1})^2$. For a sum of two increments $X_{k,0} = X_{k-1,0} + X_{k,k-1}$ we have

$$s_{k,0} = s_{k-1,0} + s_{k,k-1} + m_{k,k-1,0} \tag{10}$$

where $m_{k,k-1,0} = 2X_{k-1,0}X_{k,k-1}$. By iterating the binomial formula (10) one obtains:

$$\begin{aligned}
s_{1,0} &= s_{1,0} \\
s_{2,0} &= s_{1,0} + s_{2,1} + m_{2,1,0} \\
\cdots & \quad \cdots\cdots\cdots\cdots\cdots\cdots \\
s_{k,0} &= s_{k-1,0} + s_{k,k-1} + m_{k,k-1,0} \\
\cdots & \quad \cdots\cdots\cdots\cdots\cdots\cdots \\
s_{K-1,0} &= s_{K-2,0} + s_{K-1,K-2} + m_{K-1,K-2,0} \\
s_{K,0} &= s_{K-1,0} + s_{K,K-1} + m_{K,K-1,0}
\end{aligned}$$

Summing up these relations we get

$$s_{K,0} = \sum_{k=1}^{K} s_{k,k-1} + \sum_{k=1}^{K} m_{k,k-1,0}, \tag{11}$$

which, using the relation

$$m_{k,k-1,0} = 2X_{k-1,0}X_{k,k-1} = 2X_{k,k-1}\sum_{l=1}^{k-1} X_{l,l-1}$$

can be further expressed as

$$s_{K,0} = \sum_{k=1}^{K} s_{k,k-1} + 2\sum_{k=1}^{K}\sum_{l=1}^{k-1} X_{k,k-1}X_{l,l-1}. \tag{12}$$

For $X_t = X_t^{ens}$, the expectation $\Sigma_{k,0} = \overline{\langle s_{k,0}\rangle}$ of (10) verifies

$$\Sigma_{k,0} = \Sigma_{k-1,0} + \Sigma_{k,k-1} + M_{k,k-1,0}, \tag{13}$$

where $M_{k,k-1,0} = \overline{\langle m_{k,k-1,0}\rangle}$. For continuous time $t = k\Delta$, (13) becomes

$$\Sigma_{t,0} = \Sigma_{t-\Delta,0} + \Sigma_{t,t-\Delta} + M_{t,t-\Delta,0}. \tag{14}$$

According to the definition of the ensemble coefficient in (3), the half time derive of (14) yields

$$D^{ens}_{t,0} = D^{ens}_{t,t-\Delta} + MD^{ens}_{t,t-\Delta,0}. \tag{15}$$

The last term of (14), $M_{t,t-\Delta,0}$ is a **memory term** quantifying the departure from normal-diffusion behavior of the process X^{ens}_t [25, 22] and its half derivative $MD^{ens}_{t,t-\Delta,0} = (1/2)dM_{t,t-\Delta,0}/dt$ is a **memory dispersion coefficient**. Relations similar to (14-15) also hold for S, D^{eff}, and for R, D^{cm}. The expectation of the second term of Eq. (11),

$$CM_t = \sum_{k=1}^{t/\Delta} \overline{\langle m_{k\Delta,(k-1)\Delta,0}\rangle}, \tag{16}$$

is a cumulative memory term which, according to Eq. (12), contains a hierarchy of correlations between increments along paths of the transport process. Estimations of such cumulative memory terms by simulations of transport in single realizations of the random velocity field are presented in Ref. [29].

According to (15) and (5), the memory dispersion coefficient corresponding to the increment of the process X_t over the time interval τ has the following Lagrangian expression

$$\begin{aligned} MD^{ens}_{ii}(t,\tau,0) &= D^{ens}_{ii}(t,0) - D^{ens}_{ii}(t,\tau) \\ &= \int_0^{\tau} \overline{\langle u_i(\mathbf{X}_t)u_i(\mathbf{X}_{t'})\rangle} dt'. \end{aligned} \tag{17}$$

With (8), the first order approximation of (17) is given by

$$MD^{ens}_{ii}(t,\tau,0) = \int_0^{\tau} dt' \int\int \overline{u_i(\mathbf{x})u_i(\mathbf{x}')}p(\mathbf{x},t;\mathbf{x}',t')d\mathbf{x}d\mathbf{x}'. \tag{18}$$

Memory coefficients (18) as well as their time integrals (i.e. memory terms) were computed in [25] for short range correlations of the velocity field as well as for diffusion in perfectly stratified fields (model of Matheron and de Marsily). In [22], the time behavior of the memory terms for fractional Gaussian noise has been also analyzed. Jeon and Metzler [16] used correlations of increments to quantify the memory of two special anomalous diffusion processes (fractional Brownian motion and processes governed by fractional Langevin equation). Their correlations are in fact memory terms related to the variance of the process by Eq. (13). For instance, in case of fractional Brownian motion with variance $\Sigma_{t_2,t_1} = (t_2 - t_1)^{2H}$ and Hurst coefficient $0 < H < 1$, for two successive time intervals Δ, from (14) one obtains

$$M_{\Delta} = 2\overline{\langle X_{\Delta,0}X_{2\Delta,\Delta}\rangle} = \Sigma_{2\Delta,0} - \Sigma_{\Delta,0} - \Sigma_{2\Delta,\Delta} = (2^{2H}-2)\Delta^{2H}, \tag{19}$$

which is just Eq. (4.4) of Jeon and Metzler [16].

3 Explicit first-order results for transport in aquifers

Approximations of the flow and transport equations for small variance of the logarithm of the hydraulic conductivity K of the aquifer system lead to explicit functional dependencies of the dispersion coefficients on the covariance C_Y of the random field $Y = \ln K$ [14].

With the common convention for the Fourier transform

$$\hat{u}_j(\mathbf{k}) = \int u_j(\mathbf{x}) \exp(-i\mathbf{k}\cdot\mathbf{x}) d\mathbf{x}, \tag{20}$$

$$u_j(\mathbf{x}) = \frac{1}{(2\pi)^d} \int \hat{u}_j(\mathbf{k}) \exp(i\mathbf{k}\cdot\mathbf{x}) d\mathbf{k}, \quad j = 1 \cdots d,\ d = 2, 3, \tag{21}$$

the first-order approximation in σ_Y^2 yields (e.g., [14, 21, 7, 8])

$$\overline{\hat{u}_j(\mathbf{k})\hat{u}_j(\mathbf{k}')} = (2\pi)^d U^2 \delta(\mathbf{k}+\mathbf{k}') p_j^2(\mathbf{k}) \hat{C}_Y(\mathbf{k}), \tag{22}$$

where $p_j(\mathbf{k}) = (\delta_{j,1} - k_1 k_j / k^2)$ are projectors which ensure the incompressibility of the flow.

Remark 2. The Dirac function $\delta(\mathbf{k}+\mathbf{k}')$ which ensures the statistical homogeneity in (22) has to be changed into $\delta(\mathbf{k}-\mathbf{k}')$ to obtain a similar first-order relation for $\overline{\hat{u}_j(\mathbf{k})\hat{u}_j^*(\mathbf{k}')}$, where $\hat{u}_j^*$ denotes the complex conjugate [5, 6, 21]. Even though it leads to the same results, this relation render the computation more complicated.

Using the inverse Fourier transform (21) and (22), the Eulerian velocity correlation from (8) and (18) can be computed as follows

$$\begin{aligned} \overline{u_j(\mathbf{x})u_j(\mathbf{x}')} &= \frac{1}{(2\pi)^{2d}} \int\int \overline{\hat{u}_j(\mathbf{k})\hat{u}_j(\mathbf{k}')} \exp(i\mathbf{k}\cdot\mathbf{x}) \exp(i\mathbf{k}'\cdot\mathbf{x}') d\mathbf{k} d\mathbf{k}' \\ &= \frac{U^2}{(2\pi)^d} \int p_j^2(\mathbf{k}) \hat{C}_Y(\mathbf{k}) \exp(i\mathbf{k}\cdot\mathbf{x}) d\mathbf{k} \int \delta(\mathbf{k}+\mathbf{k}') \exp(i\mathbf{k}'\cdot\mathbf{x}') d\mathbf{k}' \\ &= \frac{U^2}{(2\pi)^d} \int p_j^2(\mathbf{k}) \hat{C}_Y(\mathbf{k}) \exp[i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')] d\mathbf{k}. \end{aligned} \tag{23}$$

With (23), the integrand of the time integral in (8) and (18) becomes

$$\begin{aligned} &\int\int \overline{u_j(\mathbf{x})u_j(\mathbf{x}')} p(\mathbf{x},t;\mathbf{x}',t') d\mathbf{x} d\mathbf{x}' = \\ &= \frac{U^2}{(2\pi)^d} \int p_j^2(\mathbf{k}) \hat{C}_Y(\mathbf{k}) d\mathbf{k} \int\int \exp[i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}') p(\mathbf{x},t;\mathbf{x}',t')] d\mathbf{x} d\mathbf{x}' \\ &= \frac{U^2}{(2\pi)^d} \int p_j^2(\mathbf{k}) \hat{C}_Y(\mathbf{k}) \exp[ik_1 U(t-t') - k^2 D(t-t')] d\mathbf{k}. \end{aligned} \tag{24}$$

The passage to the last line in (24) was achieved by using the expression

$$\int\int \exp[i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')]p(\mathbf{x},t;\mathbf{x}',t')d\mathbf{x}d\mathbf{x}' = \exp[ik_1U(t-t') - k^2D(t-t')]$$

of the characteristic function of the Gaussian increment $(\mathbf{X}_t^{(0)} - \mathbf{X}_{t'}^{(0)})$ of the un-perturbed process $X_{j,t}^{(0)} = \delta_{j,1}Ut + W_{j,t}$, $j = 1\cdots d$ (see e.g. [18]).

Remark 3. Changing the sign convention of the Fourier transform,

$$\hat{u}_j(\mathbf{k}) = \int u_j(\mathbf{x})\exp(i\mathbf{k}\cdot\mathbf{x})d\mathbf{x},$$

changes the sign of the first term of the characteristic function without altering the sign of the second term, $\exp[-ik_1U(t-t') - k^2D(t-t')]$ (see detailed computation of characteristic function for Gaussian random variables of Papoulis and Pillai [18], Example 5-28, p. 153]. However, changing the sign of the imaginary exponent is equivalent to changing U into $-U$ (this also changes the sign of p_j (see Gelhar and Axness [14], Eq. (28a)), but (24) depends only on p_j^2). Since second moments and dispersion coefficients do not depend on the sign of the mean flow, the expression (26) gives the same result irrespective of the sign of the imaginary exponent.

Inserting (24) into (8) one obtains the explicit expression of the advective contribution to the ensemble dispersion coefficient,

$$\begin{aligned}\delta\{D_{ii}^{ens}(t)\} &= D_{ii}^{ens}(t) - D\\ &= \frac{U^2}{(2\pi)^d}\int_0^t dt'\int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1U(t-t') - k^2D(t-t')]d\mathbf{k}. \end{aligned}\tag{25}$$

With the change of variable $t'' = t - t'$, (25) becomes

$$\begin{aligned}\delta\{D_{ii}^{ens}(t)\} &= -\frac{U^2}{(2\pi)^d}\int_t^0 dt''\int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1Ut'' - k^2Dt'']d\mathbf{k}\\ &= \frac{U^2}{(2\pi)^d}\int_0^t dt''\int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1Ut'' - k^2Dt'']d\mathbf{k}. \end{aligned}\tag{26}$$

Remark 4.1. The time derivative of (26) is identical with the result of Dagan ([5], Eq. (3.14); [6], Eq. (17)), and Schwarze et al. ([21], Eq. (13)). Dagan [6] derived the result by using the changed sign in the homogeneity condition (Remark 2) and opposite Fourier transform convention (Remark 3).

Remark 4.2. The opposite Fourier transform convention and the same homogeneity condition as in (22) change ik_1Ut'' into $-ik_1Ut''$ and let $-k^2Dt''$ unchanged (see Remark 3). This is the case in the derivation of the variance

Σ_{jj} by Fiori and Dagan [12] (Eq. (14)), which is exactly the integral of (25) from above, with ik_1Ut'' replaced by $-ik_1Ut''$. Dentz et al. [7] used the same conventions as Fiori and Dagan [12], (i.e. Eqs. (16) and (25) in [7]) and obtained the same result. The result given by their Eq. (40) with $\tilde{g}_0$ replaced by the characteristic function of the un-perturbed problem ([7], Eq. (25)), is just the relation (26) from above, with exponent ik_1Ut'' replaced by $-ik_1Ut''$.

The corresponding memory coefficient is similarly obtained by inserting (24) into (18),

$$MD_{ii}^{ens}(t,\tau,0) = \frac{U^2}{(2\pi)^d}\int_0^\tau dt' \int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1U(t-t')-k^2D(t-t')]d\mathbf{k}. \tag{27}$$

Making the change of variable $t'' = t - t'$ in (27), we see that the ensemble memory coefficients can be computed, after a simple change of the integration limits in the time integral, with the explicit expression (26) of the advective contributions $\delta\{D_{ii}^{ens}\}$ to the ensemble coefficients:

$$MD_{ii}^{ens}(t,\tau,0) = \frac{U^2}{(2\pi)^d}\int_{t-\tau}^{t} dt'' \int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1Ut'' - k^2Dt'']d\mathbf{k}. \tag{28}$$

Remark 5. The memory coefficient (27) can also be obtained within the Eulerian approach of Dentz et al. [7] if in Eq. (18) from [8] the initial concentration $\hat{\rho}$ is replaced by the perturbation solution for point source at time τ given in Eq. (26) from [7], i.e. $\hat{\rho}(k,\tau) = g(k,\tau)$. Then Eq. (18) from [8] gives $\hat{g}(k,t-\tau)$. Further, inserting $\hat{g}(k,t-\tau)$ in Eq. (14) from [8] and "expanding the logarithm consistently up to second order in the fluctuations" (as described at the beginning of Sect. 3.2 of [8]), should reproduce the expression (27) of the memory coefficient $MD^{ens}(t,\tau,0)$.

To derive the explicit form of the effective coefficients (7) we first have to compute the center of mass coefficients (9). Similarly to (24), we obtain

$$\begin{aligned}
&\int\int \overline{u_j(\mathbf{x})u_j(\mathbf{x}')}p(\mathbf{x},t)p(\mathbf{x}',t')d\mathbf{x}d\mathbf{x}' \\
&= \frac{U^2}{(2\pi)^d}\int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})d\mathbf{k}\int\int \exp[i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')p(\mathbf{x},t)p(\mathbf{x}',t')]d\mathbf{x}d\mathbf{x}' \\
&= \frac{U^2}{(2\pi)^d}\int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1U(t-t') - k^2D(t+t')]d\mathbf{k} \qquad (29)
\end{aligned}$$

The passage to the last line in (29) is now achieved through the expressions

$$\int\int \exp(i\mathbf{k}\cdot\mathbf{x})p(\mathbf{x},t)d\mathbf{x} = \exp[ik_1Ut - k^2Dt] \text{ and}$$

$$\int\int \exp(-i\mathbf{k}\cdot\mathbf{x}')p(\mathbf{x}',t')d\mathbf{x}' = \exp[-ik_1Ut' - k^2Dt']$$

of the characteristic functions of the Gaussian un-perturbed process $X_{j,t}^{(0)} = \delta_{j,1}Ut + W_{j,t},\ j = 1\cdots d$ [18], where we used the property pointed out in Remark 3. Inserting (29) into (9), we obtain the center of mass coefficient

$$D_{ii}^{cm}(t) = \frac{U^2}{(2\pi)^d}\int_0^t dt' \int p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1U(t-t') - k^2D(t+t')]d\mathbf{k}. \quad (30)$$

With the change of variable $t'' = t - t'$ (which implies $t + t' = 2t - t''$) the expression (30) of the center of mass coefficients becomes

$$\begin{aligned} &D_{ii}^{cm}(t)\\ &= -\frac{U^2}{(2\pi)^d}\int_t^0 dt'' \int \exp(-2k^2Dt)p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1Ut'' + k^2Dt'']d\mathbf{k}\\ &= \frac{U^2}{(2\pi)^d}\int_0^t dt'' \int \exp(-2k^2Dt)p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1Ut'' + k^2Dt'']d\mathbf{k}. \quad (31)\end{aligned}$$

The center of mass coefficients (30) and the advective contribution (26) to the ensemble dispersion coefficients determine the advective contributions to the effective coefficients according to (7):

$$\delta\{D_{ii}^{eff}(t)\} = D_{ii}^{ens}(t) - D_{i,i}^{cm}(t) - D = \delta\{D_{ii}^{ens}(t)\} - D_{i,i}^{cm}(t). \quad (32)$$

Remark 6. The ensemble contribution (26) and the center of mass coefficient (30) particularize to isotropic local diffusion coefficient the general relations M^- and M^+, respectively, derived by Dentz et al. ([7] Eq. (47)). We also note that (30) is the time derivative of the relation (15) of Fiori and Dagan [12], which, together with Remarks 4.1 and 4.2, proves the equivalence of Eulerian expressions for the ensemble and effective dispersion coefficients of Dentz et al. [7, 8] and the Lagragian expressions derived by Fiori and Dagan [12] and by Vanderborght [31] for the corresponding variances. We note here that the Eulerian approach has the important advantage of providing first-order approximations of the concentration fields (e.g., Eq. (26) in [7], on which the derivation of the dispersion coefficients is actually based).

The memory coefficient of the center of mass is readily obtained, similarly to (28), by a change of the integration limits in the expression (31) of the center of mass dispersion coefficients:

$$\begin{aligned} &MD_{ii}^{cm}(t,\tau,0)\\ &= \frac{U^2}{(2\pi)^d}\int_{t-\tau}^t dt'' \int \exp(-2k^2Dt)p_j^2(\mathbf{k})\hat{C}_Y(\mathbf{k})\exp[ik_1Ut'' + k^2Dt'']d\mathbf{k}. \quad (33)\end{aligned}$$

Finally, the memory coefficient of the effective dispersion is obtained from the relations (32), (31), and (28),

$$MD_{ii}^{eff}(t) = MD_{ii}^{ens}(t) - MD_{i,i}^{cm}(t). \tag{34}$$

Remark 7. The memory coefficients (28), (33), and (34) can be computed with the explicit expressions of the advective contribution to the ensemble dispersion coefficients (25) and of the center of mass coefficients (31) by changing the lower limit of the time integrals from 0 to $t - \tau$.

4 Explicit first-order results for power-law correlated log-K fields

The first-order results for log-K fields with finite correlation lengths can be further used to develop approaches for a superposition of scales [9, 20]. To proceed, let us consider the particular case of an isotropic power-law covariance of $Y = \ln K$,

$$C_Y^{PL}(\mathbf{x}) = \sigma_Y^2 z^{-\beta}, \text{ where } z = \left(1 + \frac{|\mathbf{x}|^2}{L^2}\right)^{\frac{1}{2}}, \ |\mathbf{x}|^2 = \sum_{j=1}^{d} x_j^2, \ \ 0 \le \beta \le 2, \tag{35}$$

where L is some length scale associated with the spatial extension of the system. Power-law covariances can be constructed by integrating covariances with continuously increasing finite integral scales λ ([15], p. 370, expression 3.478 (1.)) by the relation

$$\int_0^\infty \lambda^{\beta-1} \exp\left(-\mu\lambda^p\right) d\lambda = \frac{1}{p}\mu^{-\frac{\beta}{p}}\Gamma\left(\frac{\beta}{p}\right), \tag{36}$$

where $\mu > 0$ and $\beta > 0$. For $\mu = z^2$ and $p = 2$, (36) yields a superposition of Gaussian covariances:

$$\begin{aligned}
C_Y^{PL}(\mathbf{x})/\sigma_Y^2 &= z^{-\beta} = \frac{2}{\Gamma(\frac{\beta}{2})}\int_0^\infty \lambda^{\beta-1}\exp(-z^2\lambda^2)d\lambda \\
&= \frac{2}{\Gamma(\frac{\beta}{2})}\int_0^\infty \lambda^{\beta-1}\exp\left[-\left(1+\frac{|\mathbf{x}|^2}{L^2}\right)\lambda^2\right]d\lambda \\
&= \int_0^\infty d\Lambda(\lambda)\exp\left(-\frac{1}{2}\frac{|\mathbf{x}|^2}{l(\lambda)^2}\right),
\end{aligned} \tag{37}$$

where

$$d\Lambda(\lambda) = \frac{2}{\Gamma(\frac{\beta}{2})}\lambda^{\beta-1}\exp(-\lambda^2)d\lambda \quad \text{and} \quad l(\lambda) = \frac{L}{\lambda\sqrt{2}}. \tag{38}$$

The integrand in (37) has the from of the Gaussian C_Y considered by Dentz et al. [7, 8].

Redefining z in (35) as $z = (1 + |\mathbf{x}|/L)$ and choosing $\mu = z$ and $p = 1$, one constructs a power-law covariance via (36) as a superposition of exponential covariances:

$$\begin{aligned} C_Y^{PL}(\mathbf{x})/\sigma_Y^2 &= z^{-\beta} = \frac{1}{\Gamma(\beta)}\int_0^\infty \lambda^{\beta-1}\exp(-z\lambda)d\lambda \\ &= \frac{1}{\Gamma(\beta)}\int_0^\infty \lambda^{\beta-1}\exp\left[-\left(1+\frac{|\mathbf{x}|}{L}\right)\lambda\right]d\lambda \\ &= \int_0^\infty d\Lambda(\lambda)\exp\left(-\frac{|\mathbf{x}|}{l(\lambda)}\right), \end{aligned} \tag{39}$$

where

$$d\Lambda(\lambda) = \frac{1}{\Gamma(\beta)}\lambda^{\beta-1}\exp(-\lambda)d\lambda \quad \text{and} \quad l(\lambda) = \frac{L}{\lambda}. \tag{40}$$

The integrand in (39) is the isotropic version of the exponential C_Y considered by Fiori [10] and Vanderborght [31].

Remark 8. For $\beta = 1$ in either (37) or (39) one obtains the covariance of the "$1/z$ noise" as an integral of Gaussian or exponential covariances with continuous scale parameter λ. Further, by defining $z = |\mathbf{x}|/L$ the covariance $\sigma_Y^2 z^{-1}$ is simply given by integrating Gaussian covariances with respect to we get $d\Lambda(\lambda) = \frac{2}{\sqrt{\pi}}d\lambda$ in (37) or exponential covariances with respect to $d\Lambda(\lambda) = d\lambda$ in (39).This corresponds to the limit of infinite superposition of log-K fields with finite correlation scales.

By the linearity of the superposition relations (37), respectively (39), we have the general relations

$$C_Y^{PL}(\mathbf{x}) = \int_0^\infty d\Lambda(\lambda)C_Y(\mathbf{x},\lambda), \quad \widehat{C_Y^{PL}}(\mathbf{k}) = \int_0^\infty d\Lambda(\lambda)\widehat{C_Y}(\mathbf{k},\lambda) \tag{41}$$

By inserting (41) in (25), (30), and (7), we finally obtain the superposition relations

$$\delta\{D_{ii}^{PL,ens}\}(t) = \int_0^\infty d\Lambda(\lambda)\{D_{ii}^{ens}\}(t,\lambda)$$

$$\delta\{D_{ii}^{PL,eff}\}(t) = \int_0^\infty d\Lambda(\lambda)\{D_{ii}^{eff}\}(t,\lambda). \tag{42}$$

According to (42), the memory coefficients (28), (33), and (34) are given by integrals with respect to $d\Lambda(\lambda)$ of the coefficients for given λ derived in the previous section.

5 Numerical simulations of transport in aquifers with multiscale hydraulic conductivity

To investigate the behavior of several transport observables we conducted 256 global random walk (GRW) simulations of a two-dimensional isotropic diffusion, with coefficient $D = 0.01$ m^2/d which is a typical value for local dispersion coefficient in aquifers, in multiscale velocity fields. The latter, generated with the Kraichnan method, are first-order approximations of the flow equations for a lnK field consisting of a superposition of statistically independent lnK fields with isotropic exponential correlation and constant variance $\sigma_Y^2 = 0.1$.

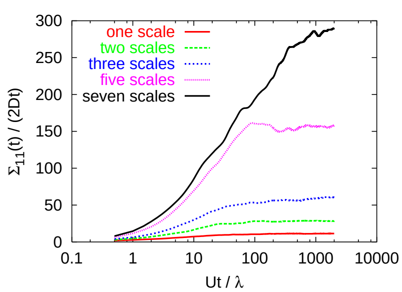

Figure 1: Longitudinal ensemble dispersion coefficients for log-K fields with increasing integral scales.

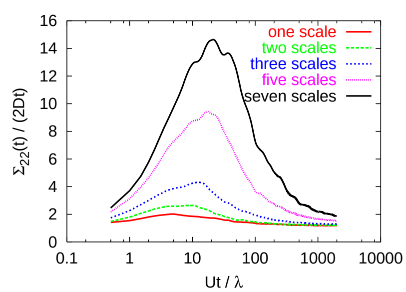

Figure 2: Transverse ensemble dispersion coefficients for log-K fields with increasing integral scales.

The integral scales (defined as integrals of the correlation function, not divided by the variance) of the lnK fields were increased with a constant step $\lambda = 1$ m, so that at the n-the step $\lambda_n = \lambda_{n-1} + n$, which results in $\lambda_n =$

$(n+1)n/2$. For instance, the superposition of 2 fields has the variance 0.2 and the integral scale $\lambda_2 = 3$ m and that of seven fields (the largest one investigated here) has the variance 0.7 and $\lambda_7 = 28$ m.

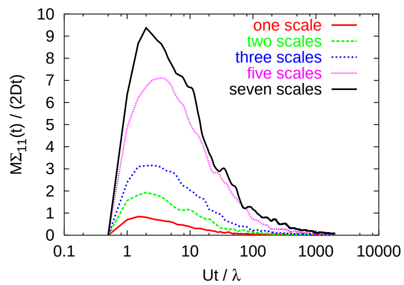

Figure 3: Longitudinal ensemble memory coefficients for log-K fields with increasing integral scales.

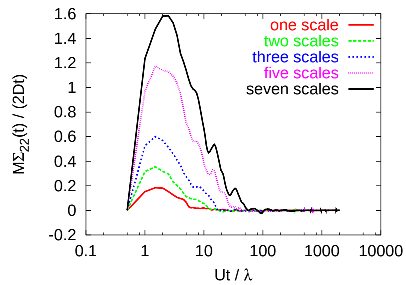

Figure 4: Transverse ensemble memory coefficients for log-K fields with increasing integral scales.

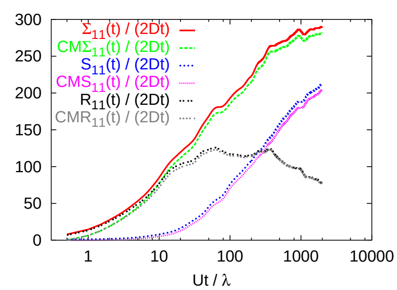

Figure 5: Cumulative longitudinal memory coefficients compared with the corresponding ensemble, effective and center of mass dispersion coefficients ($\lambda_7 = 28$ m).

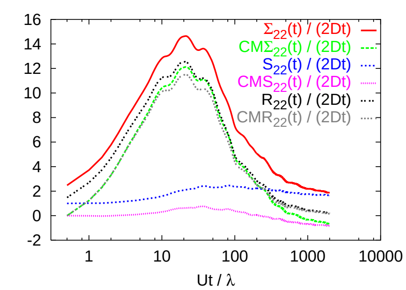

Figure 6: Cumulative transverse memory coefficients compared with the corresponding ensemble, effective and center of mass dispersion coefficients ($\lambda_7 = 28$ m).

The GRW algorithm has been developed by Vamoş et al. [30]. Implementation details and applications for transport in saturated aquifers can be found in [24, 19, 27, 23, 28]. In our GRW numerical simulations we estimate dispersion coefficients for processes starting from instantaneous point injections by the mean half-slope of the variances defined in (3). The ensemble dispersion coefficients $\Sigma_{ii}(t)/(2t)$ and the one-step memory dispersion coefficients $M_{ii}(t)/(2t)$, where $M_{ii}(t) = M_{ii}(t, t-\Delta, 0)$ (15), normalized by the local dispersion coefficient, are presented in Figs. 1-4. (See Ref. [26] for results on effective and center of mass coefficients.) The extinction of the memory coefficients (Figs. 3-4) coincides with the approach to a normal diffusive behavior with constant ensemble dispersion coefficients, shown in Figs. 1-2, in agreement with (15) and (17).

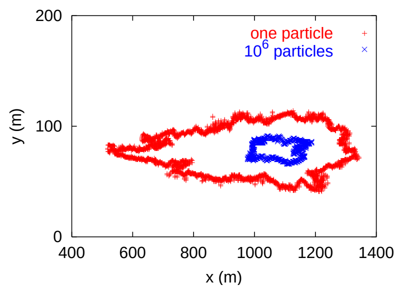

Figure 7: Solute plume at $t = 1000$ days ($\lambda_7 = 28$ m).

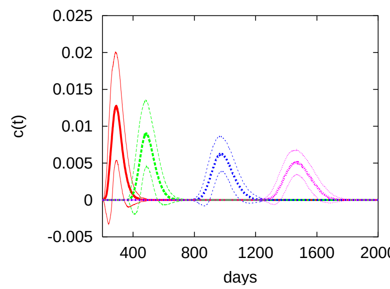

Figure 8: Breakthrough curve $c(t) \pm \sigma_c(t)$ at x =300, 500, 1000, and 1500 m ($\lambda_7 = 28$ m).

Figures 5 and 6 show the cumulative memory coefficients $CM(t)/(2t)$, with CM given by (16), compared with the corresponding ensemble, effective and center of mass coefficients, in case of the seven-scales velocity field. The cumulative memory coefficients are close to the corresponding dispersion coefficients, the small difference between the two coefficients being just the half slope of the average of the first term in (11). The latter is a dispersion coefficient describing the displacement of solute particles over one step Δ (in our case, the simulation time step). In case of ensemble dispersion Σ_{ii}, we can see that the average of (11) is a discrete Taylor formula approximating the ensemble coefficient (5). Thus, ensemble coefficients are mainly determined by correlations of increments of the process X_i^{ens}, corresponding to correlations of the Lagrangian velocity. This makes the difference from genuine diffusion processes (Brownian motions) where the increments are uncorrelated and the one-step dispersion defines the constant diffusion coefficient.

Figures 1 and 5 show that both the ensemble and the effective coefficients behave anomalous in a time windows which increase with the number of superposed scales. Anomalous diffusive behavior is also indicated by the asymmetry of the solute plume at 1000 days, presented in Fig. 7, and by the breakthrough curve $c(t)$ of the concentration spatially averaged over the vertical section of the simulation domain, recorded at increasing distances along the mean flow direction, shown in Fig. 8. (See Ref. [26] for other simulation results which indicate anomalous behavior).

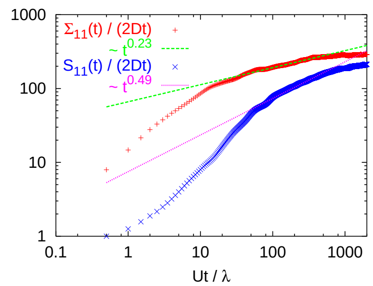

Figure 9: Power-law fit between 10 and 500 days of longitudinal ensemble and effective coefficients; standard fit-errors for power-law exponents of 0.58% and 0.46%, respectively ($\lambda_7 = 28$ m).

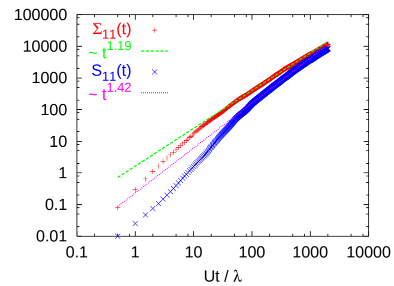

Figure 10: Power-law fit between 10 and 500 days of longitudinal ensemble and effective variances; standard fit-errors for power-law exponents of 0.14% and 0.08%, respectively ($\lambda_7 = 28$ m).

To further investigate the anomalous behavior induced by the multiscale structure of the velocity field, we fitted dispersion coefficients with power-law functions. Figures 9 and 10 indicate a power law behavior of the coefficients. We remark that ensemble and effective coefficients and variances have different time behavior, with a larger slope for the effective quantities. This seems to be at variance with the theoretical result for power-law correlated fields obtained by Fiori [11]. Therefore, we consider that the current numerical results do not allow us to draw a definite conclusion about the power law behavior of the dispersion coefficients.

As we have seen in the previous section, power-law covariances can be constructed by integrating covariances with continuously increasing integral scales. An infinite sum of equal-weight exponential correlations results in an $1/x$-type correlation (Remark 8; see also expression 3.478 (1.) in [15, p. 370]) Such a correlation leads to a $\sim (t \ln t - t)$ behavior of the ensemble variance Σ_{ii} [22]. Since this would be the theoretical infinite limit of our simulations, we also fitted the dispersion coefficients $\Sigma_{ii}/(2t)$ with $\ln t$ and the variances Σ_{ii} with $t \ln t - t$ (Figs. 11 and 12). At a first sight these fitting results are also acceptable. Nevertheless, a more detailed analysis is required to decide which time-behavior, the power-law one from Figs. 9-10 or the $\ln t$-type behavior from Figs. 11-12 provides the better fit with the results of our numerical experiment. An independent selection criterium would be the numerical evaluation of dispersion and memory terms derived in Section 3 for

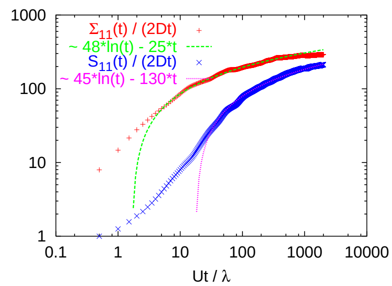

Figure 11: Fit of longitudinal ensemble and effective coefficients with $a \ln t - b$ between 10 and 500 days; standard fit-errors for the coefficients $\{a,\ b\}$ of $\{0.30\%,\ 3.16\%\}$ and $\{0.36\%,\ 0.68\%\}$, respectively ($\lambda_7 = 28$ m).

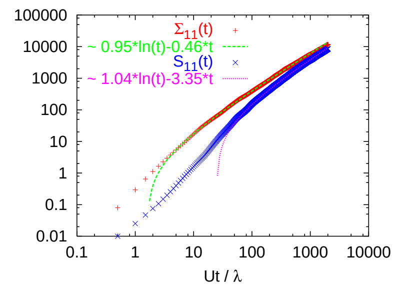

Figure 12: Fit of longitudinal ensemble and effective variances with $(at \ln t - bt)$ between 10 and 500 days; standard fit-errors for the coefficients $\{a,\ b\}$ of $\{0.70\%,\ 8.46\%\}$ and $\{0.22\%,\ 0.40\%\}$, respectively ($\lambda_7 = 28$ m).

an $1/x$-correlated log-K field, using the integral representation (39-40) derived in Section 4.

There is a long standing attempt to introduce power law correlations and anomalous dispersion in modeling groundwater transport by a rather abstract mathematical approach [4, 2, 9, 11, 3, 25, 22]. Nevertheless, a superposition of a finite number of scales seems to be a more natural assumption in many contamination scenarios for evolving scale groundwater systems [13, 17]. Numerical simulations of transport in multiscale velocity fields provide a predictive tool for such scenarios and may support the development of adequate models.

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