Solute transport in aquifers with evolving scale heterogeneity

by

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No. 346 2011
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Abstract

A superposition of independent log-K fields with increasing integral scales has been proposed by many authors [13, 4, 9, 17, 3] as model for evolving scale heterogeneity of natural porous media. Here, we investigate the anomalous transport in multi-scale fields by first-order approximations to transport equations and numerical Monte Carlo simulations.

1 Lagrangian and Eulerian models of diffusion in random fields and their relationship

Let \( \{X_{i,t} = X_i(t), i = 1, 2, 3, t \geq 0\} \) be the advection-dispersion process with a space variable drift \( V(x) \) which is a sample of a random velocity field, and a local diffusion coefficient \( D \) which is assumed constant, described by the Itô equation

\[
X_{i,t} = x_{i,0} + \int_0^t V_i(X_{t'})dt' + W_{i,t},
\]

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

The solution \( g \) of the Fokker-Planck equation

\[
\partial_t g + u\nabla g = D\nabla^2 g,
\]

which corresponds to the Green’s function in Eulerian approaches to dispersion in random fields, is the density of the transition probability \( P(dx,t|x_0) = g(x,t|x_0)dx \) of the Itô process (1). Equation (2) is the starting point of the perturbation approach of [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: \( X_{i,t}^{\text{eff}} = X_{i,t} - \langle X_{i,t} \rangle \), the process centered about the mean \( \langle X_{i,t} \rangle \) of \( X_{i,t} \) in single realizations of the random velocity field, \( X_{i,t}^{\text{eff}} = 0 \), the process \( X_{i,t}^{\text{ens}} = X_{i,t} - \langle X_{i,t} \rangle \) centered about the average \( \langle X_{i,t} \rangle \) of \( X_{i,t} \) over the ensemble of realizations, \( \langle X_{i,t}^{\text{ens}} \rangle = 0 \),...
and the fluctuation of the center of mass \( X_{i,t}^{cn} = \langle X_{i,t} \rangle - \langle X_{i,t}^{0} \rangle \), with \( X_{i,t}^{0} = 0 \). Associated to these processes, one defines \([1]\):

\[
S_{ii}(t) = \langle (X_{i,t}^{eff})^2 \rangle, \quad D_{ii}^{eff}(t) = \frac{1}{2} \frac{dS(t)}{dt}, \text{ effective dispersion coefficients}
\]

\[
\Sigma_{ii}(t)_{ii} = \langle (X_{i,t}^{ens})^2 \rangle, \quad D_{ii}^{ens}(t) = \frac{1}{2} \frac{d\Sigma(t)}{dt}, \text{ ensemble dispersion coefficients}
\]

\[
R_{ii}(t) = \langle (X_{i,t}^{cn})^2 \rangle, \quad D_{ii}^{cn}(t) = \frac{1}{2} \frac{dR(t)}{dt} = D^{ens} - D^{eff}, \text{ center of mass dispersion coefficients}
\]

According to (1), for a process starting at \( x_{i,0} = 0 \) we have

\[
X_{i,t} = \int_{0}^{t} u_{i}(X_{k}) dt' + W_{i,t}, \quad (4)
\]

where \( u_{i} = V_{i} - \langle V_{i} \rangle \) for \( X_{i,t} = X_{i,t}^{eff} \) and \( u_{i} = V_{i} - \langle V_{i} \rangle \) for \( X_{i,t} = X_{i,t}^{ens} \). From (4), the variance of \( X_{i,t} = X_{i,t}^{ens} \) is obtained as follows,

\[
\Sigma_{ii}(t)_{ii} = \int_{0}^{t} \int_{0}^{t} \langle u_{i}(X_{k})u_{i}(X_{k'}) \rangle dt'' dt' + 2 \int_{0}^{t} \langle u_{i}(X_{k}) W_{i,t} \rangle dt' + \langle W_{i}^2 \rangle
\]

\[
= \int_{0}^{t} \int_{0}^{t} \langle u_{i}(X_{k})u_{i}(X_{k'}) \rangle dt'' dt' + 2 \int_{0}^{t} \langle u_{i}(X_{k}) W_{i,t} \rangle dt' + \langle W_{i}^2 \rangle
\]

\[
= 2Dt + 2 \int_{0}^{t} dt' \int_{0}^{t} \langle u_{i}(X_{k})u_{i}(X_{k'}) \rangle dt''
\]

where we used \( \langle u_{i} \rangle = 0 \) and the independence of the Wiener process on velocity statistics, which, together, cancel the middle term. Further, the definitions (3) yield

\[
D_{ii}^{ens}(t) = D + \int_{0}^{t} \langle u_{i}(X_{k})u_{i}(X_{k'}) \rangle dt', \quad (5)
\]

\[
D_{ii}^{cn}(t) = \int_{0}^{t} \langle u_{i}(X_{k}) \rangle \langle u_{i}(X_{k'}) \rangle dt', \quad (6)
\]

\[
D_{ii}^{eff}(t) = D_{ii}^{ens}(t) - D_{ii}^{cn}(t). \quad (7)
\]

The first iteration of (4) about the un-perturbed problem consisting of diffusion with constant coefficient \( D \) in the mean flow field \( U = \langle V_{i} \rangle \), with trajectory \( X_{i,t}^{0} = \delta_{i,1}Ut + W_{i,t} \), yields:

\[
D_{ii}^{ens}(t) - D = \int_{0}^{t} \langle u_{i}(X_{k}^{0})u_{i}(X_{k'}^{0}) \rangle dt'
\]

\[
= \int_{0}^{t} dt' \int \langle u_{i}(x)\delta(x - X_{k}^{0})u_{i}(x')\delta(x' - X_{k'}^{0}) \rangle dx dx'
\]

\[
= \int_{0}^{t} dt' \int \langle u_{i}(x)u_{i}(x') \rangle p(x, t; x', t') dx dx', \quad (8)
\]

where \( p(x, t; x', t') = \langle \delta(x - X_{k}^{0})\delta(x' - X_{k'}^{0}) \rangle \) is the Gaussian joint probability density of the process \( X_{i,t}^{0} \). Similarly,

\[
D_{ii}^{cn} = \int_{0}^{t} dt' \int \langle u_{i}(x)u_{i}(x') \rangle p(x, t)p(x', t') dx dx'
\]

Further, \( D_{ii}^{eff}(t) \) is obtained from (7). Together, (7-9) are the equivalent (stochastic) Lagrangian formulation of the first-order approximation which, in an Eulerian frame, has been presented in \([1, 7, 8]\).

**Remark 1.** As follows from (8-9), if \( u_{i}(x) \) is a superposition of independent velocity fields with different integral scales, then the dispersion coefficients estimated to the first-order are given by the sum of the coefficients associated to the terms of the velocity superposition.
2 Quantifying anomalous diffusion by memory terms of the ensemble, effective, and center of mass dispersion coefficients

In the following, by \( X_t \) we denote one of the components of \( X_{i,t} \) as well as those of the processes \( X_{i,t}^{eff} \), \( X_{i,t}^{ens} \) and \( X_{i,t}^{cm} \). 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

\[
s_{k,0} = s_{k-1,0} + s_{k,k-1} + m_{k,k-1,0}
\]

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

\[
\begin{align*}
s_{1,0} & = s_{1,0} \\
s_{2,0} & = s_{1,0} + s_{2,1,0} \\
\vdots & \\
s_{k,0} & = s_{k-1,0} + s_{k,k-1} + m_{k,k-1,0} \\
\vdots & \\
s_{K,0} & = s_{K-2,0} + s_{K-1,K-2} + m_{K-1,K-2,0} \\
\end{align*}
\]

Summing up these relations we get

\[
s_{K,0} = \sum_{k=1}^{K} s_{k,k-1} + \sum_{k=1}^{K} m_{k,k-1,0},
\]

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

\[
s_{K,0} = \sum_{k=1}^{K} s_{k,k-1} + 2 \sum_{k=1}^{K-1} \sum_{l=1}^{k} X_{k-1,0}X_{l,l-1}.
\]

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

\[
\Sigma_{k,0} = \Sigma_{k-1,0} + \Sigma_{k,k-1} + M_{k,k-1,0}.
\]

For continuous time \( t = k\Delta \), (13) becomes

\[
\Sigma_{t,0} = \Sigma_{t-\Delta,0} + \Sigma_{t,t-\Delta} + M_{t,t-\Delta,0}.
\]

According to the definition of the ensemble coefficient in (3), the half time derive of (13) yields

\[
D_{t,0}^{ens} = D_{t,\Delta}^{ens} + M D_{t,\Delta,0}^{ens}.
\]

The last term of (14) \( M_{t,t-\Delta,0} = \langle m_{t,t-\Delta,0} \rangle \) is a memory term quantifying the departure from normal-diffusion behavior of the process \( X_t^{ens} \) [23, 21] and its half derivative \( M D_{t,\Delta,0}^{ens} = (1/2)dM_{t,t-\Delta,0}/dt \) 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),

\[
CM_t = \sum_{k=1}^{t/\Delta} \langle m_{k\Delta,(k-1)\Delta,0} \rangle,
\]
is a cumulated memory term which, according to Eq. (12), contains a hierarchy of correlations between increments along paths of the transport process. Such cumulated memory terms, on paths of length $3\Delta$ were estimated from a “global random walk” (GRW) simulation of transport in a single realization of the velocity field by [24].

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

$$MD^{ens}_{ii}(t, \tau, 0) = D^{ens}_{ii}(t, 0) - D^{ens}_{ii}(t, \tau)$$

$$= \int_0^t \langle u_i(X_t)u_i(X_{t'}) \rangle dt' - \int_0^\tau \langle u_i(X_t)u_i(X_{t'}) \rangle dt'$$

$$= \int_0^\tau \langle u_i(X_t)u_i(X_{t'}) \rangle dt'$$

(17)

The first order approximation also yields

$$MD^{ens}_{ii}(t, \tau, 0) = \int_0^\tau dt' \int \int u_i(x)u_i(x')p(x, t; x', t')dx dx'$$

(18)

Memory coefficients (18) as well as their time integrals (i.e. memory terms) were computed in [23] for short range correlations of the velocity field and for diffusion in perfectly stratified fields (model of Matheron and de Marsily). In [21], the time behavior of the memory terms for fractional Gaussian noise has been also analyzed. Jeon and Metzler [16] used increments correlations 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 with 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 $\Delta$, from (14) one obtains

$$M_\Delta = 2\{X_{\Delta,0}X_{2\Delta,\Delta}\} = \Sigma_{2\Delta,0} - \Sigma_{\Delta,0} - \Sigma_{2\Delta,\Delta} = (2^{2H} - 2)\Delta^{2H},$$

(19)

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

3 Explicit first-order results for given statistics of the log-K field

The ensemble coefficient (8) and the memory coefficient (18) can be further expresses as function of the covariance $C_Y$ of the log-hydraulic conductivity $Y = \ln K$ in the frame of approximation of flow and transport equations to the first-order in the variance $\sigma^2_{lnK}$. With the common convention for the Fourier transform

$$\hat{u}_j(k) = \int u_j(x)exp(-ik \cdot x)dx,$$

$$u_j(x) = \frac{1}{(2\pi)^d} \int \hat{u}_j(k)exp(ik \cdot x)dk,$$

(20)

(21)

the first-order approximation yields (e.g., [20, 7, 8])

$$\hat{u}_j(k)\hat{u}_j(k') = (2\pi)^d U^2 \delta(k + k')p_j^2(k)C_Y(k),$$

(22)

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

Remark 2. The Dirac function $\delta(k + k')$ which ensures the statistical homogeneity in (22) has to be changed into $\delta(k - k')$ to obtain a similar first-order relation for $\hat{u}_j(k)\hat{u}_j^*(k')$, where $\hat{u}_j^*$ denotes the complex conjugate [5, 6, 20]. Even though it leads to the same results, this relation render the computation a bit tedious.
Using the inverse Fourier transform (21) and (22), the Eulerian velocity correlation from (8) and (18) can be computed as follows

\[
\overline{u_j(x)u_j(x')} = \frac{1}{(2\pi)^{2d}} \int \int \hat{u}_j(k) \hat{u}_j(k') \exp(i k \cdot x) \exp(i k' \cdot x') dk dk' \\
= \frac{U^2}{(2\pi)^d} \int p_j^2(k) \hat{C}_Y(k) \exp(i k \cdot x) \int \delta(k + k') \exp(i k' \cdot x') dk' \\
= \frac{U^2}{(2\pi)^d} \int p_j^2(k) \hat{C}_Y(k) \exp[i k \cdot (x - x')] dk
\]

(23)

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

\[
\int \int \overline{u_j(x)u_j(x')} p(x, t; x', t') dx dx' = \\
= \frac{U^2}{(2\pi)^d} \int p_j^2(k) \hat{C}_Y(k) dk \int \int \exp[i k \cdot (x - x')] p(x, t; x', t')] dx dx' \\
= \frac{U^2}{(2\pi)^d} \int p_j^2(k) \hat{C}_Y(k) \exp[i k_1 U(t - t') - k^2 D(t - t')] dk
\]

(24)

The passage to the last line in (24) was achieved through the relation

\[
\int \int \exp[i k \cdot (x - x')] p(x, t; x', t') dx dx' = \exp[i k_1 U(t - t') - k^2 D(t - t')]
\]

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

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

\[
\hat{u}_j(k) = \int u_j(x) \exp(i k \cdot x) dx,
\]

changes the sign of the first term of the characteristic function without altering the sign of the second term, \(\exp[-i k_1 U(t - t') - k^2 D(t - t')]\) [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) in (8) one obtains the explicit expression of the advective contribution to the ensemble dispersion coefficient,

\[
\delta\{D_{ii}^{ens}(t)\} = D_{ii}^{ens}(t) - D \\
= \frac{U^2}{(2\pi)^d} \int_0^t dt' \int p_j^2(k) \hat{C}_Y(k) \exp[i k_1 U(t - t') - k^2 D(t - t')] dk.
\]

(25)

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

\[
\delta\{D_{ii}^{ens}(t)\} = -\frac{U^2}{(2\pi)^d} \int_0^t dt'' \int p_j^2(k) \hat{C}_Y(k) \exp[i k_1 U t'' - k^2 D t''] dk
\]

\[
= \frac{U^2}{(2\pi)^d} \int_0^t dt'' \int p_j^2(k) \hat{C}_Y(k) \exp[i k_1 U t'' - k^2 D t''] dk.
\]

(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. ([20], 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 \( ik_1 Ut'' \) into \(-ik_1 Ut''\) and let \(-k^2 Dt''\) unchanged (see Remark 3). This is the case in the derivation of the variance \( \Sigma_{jj} \) by Fiori and Dagan [12] (Eq. (14)), which is exactly the integral of (25) from above, with \( ik_1 Ut'' \) replaced by \(-ik_1 Ut''\). 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. Their result is given by Eq. (40) with \( \hat{g}_0 \) replaced by the characteristic function of the un-perturbed problem ([7], Eq. (25)), which is just the relation (26) from above, with exponent \( ik_1 Ut'' \) replaced by \(-ik_1 Ut''\).

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

\[
MD^{ens}_{ii}(t, \tau, 0) = \frac{U^2}{(2\pi)^d} \int_0^\tau dt' \int p_j^2(k) \hat{C}_Y(k) \exp[ik_1 U(t - t') - k^2 D(t - t')]dk.  \tag{27}
\]

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 \( \delta \{ D^{ens}_{ii} \} \) to the ensemble coefficients:

\[
MD^{ens}_{ii}(t, \tau, 0) = \frac{U^2}{(2\pi)^d} \int_{t - \tau}^t dt'' \int p_j^2(k) \hat{C}_Y(k) \exp[ik_1 U(t'') - k^2 D(t'')]dk.  \tag{28}
\]

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 \( \tau \) given in Eq. (26) from [7], i.e. \( \hat{\rho}(k, \tau) = \hat{g}(k, \tau) \). 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 \( MD^{ens}_{ii}(t, \tau, 0) \).

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

\[
\int \int u_j(x)u_j(x')p(x, t)p(x', t')dx dx' = \frac{U^2}{(2\pi)^d} \int p_j^2(k) \hat{C}_Y(k)dk \int \int \exp[ik \cdot (x - x')]p(x, t)p(x', t')]dx dx' = \frac{U^2}{(2\pi)^d} \int p_j^2(k) \hat{C}_Y(k) \exp[ik_1 U(t - t'] - k^2 D(t + t')]dk.  \tag{29}
\]

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

\[
\int \int \exp(i k \cdot x)p(x, t)dx = \exp[ik_1 Ut - k^2 Dt],
\]

\[
\int \int \exp(-i k \cdot x')p(x', t')dx' = \exp[-ik_1 Ut' - k^2 Dt']
\]

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

\[
D^{cm}_{ii}(t) = \frac{U^2}{(2\pi)^d} \int_0^t dt'' \int p_j^2(k) \hat{C}_Y(k) \exp[ik_1 U(t - t') - k^2 D(t + t')]dk.  \tag{30}
\]

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

\[
D^{cm}_{ii}(t) = -\frac{U^2}{(2\pi)^d} \int_0^t dt'' \int \exp(-2k^2 Dt)p_j^2(k) \hat{C}_Y(k) \exp[ik_1 U(t'') + k^2 D(t'')]d^dk
\]

\[
= \frac{U^2}{(2\pi)^d} \int_0^t dt'' \int \exp(-2k^2 Dt)p_j^2(k) \hat{C}_Y(k) \exp[ik_1 U(t'') + k^2 D(t'')]d^dk.  \tag{31}
\]
The center of mass coefficients (30) and the ensemble contribution (26) determine the advective contributions to the effective coefficients according to (7):

\[
\delta \{ D_{tt}^{\text{eff}} (t) \} = D_{tt}^{\text{ens}} (t) - D_{tt}^{\text{cm}} (t) - D = \delta \{ D_{tt}^{\text{ens}} (t) \} - D_{tt}^{\text{cm}} (t).
\]

**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 of Dentz et al. [7, 8] and the Lagrangian expressions derived by Fiori and Dagan [12] and by Vanderborght [26] for the ensemble and effective dispersion coefficients, respectively, variances. Nevertheless, 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 in actually based).

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

\[
MD_{tt}^{\text{cm}} (t, \tau, 0) = \frac{U^2}{(2\pi)^d} \int_{t-\tau}^{t} dt'' \int \exp(-2k^2Dt') p_j^2(k) \tilde{C}_Y(k) \exp[i k_1 Ut'' + k^2 D t''] dk.
\]

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

\[
MD_{tt}^{\text{eff}} (t) = MD_{tt}^{\text{ens}} (t) - MD_{tt}^{\text{cm}} (t).
\]

**Remark 7.** The memory coefficients (28), (33), and (34) can be computed with the explicit expressions of the dispersion coefficients derived in either Eulerian frames ([1, 7, 8] or by Lagrangian approaches [10, 12, 26]. To achieve that, we only have to replace the lower limit by \( t - \tau \) in the time integrals from these expressions.

### 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, 19]. To proceed, let us consider the particular case of an isotropic power-law covariance of \( Y = \ln K \),

\[
C_{Y}^{PL} (\mathbf{x}) = \sigma_Y^2 \frac{z^{-\beta}}{L^2}, \text{where} \quad z = \left( 1 + \frac{\| \mathbf{x} \|^2}{L^2} \right)^{\frac{1}{2}}, \| \mathbf{x} \|^2 = \sum_{j=1}^{d} x_j^2, \text{and} \quad 0 \leq \beta \leq 2,
\]

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 \( \lambda \) ([15], p. 370, expression 3.478 (1.)) by the relation

\[
\int_{0}^{\infty} \lambda^{\beta-1} \exp(-\mu \lambda^p) \, d\lambda = \frac{1}{\mu} \mu^{\frac{\beta}{p}} \Gamma \left( \frac{\beta}{p} \right).
\]
where \( \mu > 0 \) and \( \beta > 0 \). For \( \mu = z^2 \) and \( p = 2 \), (36) yields a superposition of Gaussian covariances:

\[
C_{PY}^{PL}(x)/\sigma_Y^2 = z^{-\beta} = \frac{2}{\Gamma(\frac{3}{2})} \int_0^\infty \lambda^{\frac{3}{2}-1} \exp(-z^2 \lambda^2) d\lambda
\]

\[
= \frac{2}{\Gamma(\frac{3}{2})} \int_0^\infty \lambda^{\beta-1} \exp \left(- \left(1 + \frac{|x|^2}{L^2}\right) \lambda^2 \right) d\lambda
\]

\[
= \frac{2}{\Gamma(\frac{3}{2})} \int_0^\infty \lambda^{\beta-1} \exp(-\lambda^2) \exp \left(-\frac{1}{2} \frac{|x|^2}{L^2} \lambda^2 \right) d\lambda
\]

\[
= \int_0^\infty d\Lambda(\lambda) \exp \left(-\frac{1}{2} \frac{|x|^2}{l(\lambda)^2} \right), \tag{37}
\]

where

\[
\int_0^\infty d\Lambda(\lambda) = \frac{2}{\Gamma(\frac{3}{2})} \int_0^\infty \lambda^{\beta-1} \exp(-\lambda^2) d\lambda \quad \text{and} \quad l(\lambda) = \frac{L}{\sqrt{2} \lambda}. \tag{38}
\]

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

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

\[
C_{PY}^{PL}(x)/\sigma_Y^2 = z^{-\beta} = \frac{1}{\Gamma(\beta)} \int_0^\infty \lambda^{\beta-1} \exp(-z \lambda) d\lambda
\]

\[
= \frac{1}{\Gamma(\beta)} \int_0^\infty \lambda^{\beta-1} \exp \left(- \left(1 + \frac{|x|}{L}\right) \lambda \right) d\lambda
\]

\[
= \frac{1}{\Gamma(\beta)} \int_0^\infty \lambda^{\beta-1} \exp(-\lambda) \exp \left(-\frac{|x|}{L} \lambda \right) d\lambda
\]

\[
= \int_0^\infty d\Lambda(\lambda) \exp \left(-\frac{|x|}{l(\lambda)} \right), \tag{39}
\]

where

\[
\int_0^\infty d\Lambda(\lambda) = \frac{1}{\Gamma(\beta)} \int_0^\infty \lambda^{\beta-1} \exp(-\lambda) d\lambda \quad \text{and} \quad l(\lambda) = \frac{L}{\lambda}. \tag{40}
\]

The integrand in (39) is the isotropic version of the exponential \( C_Y \) considered by Fiori [10] and Vanderborght [26].

For \( \beta = 1 \) in (37) (or in (39)) one obtains the covariance of the “1/z noise” as an integral of Gaussian (or exponential) covariances with continuous scale parameter \( \lambda \). Further, by defining \( z = |x|/L \) in (39) one obtains \( d\Lambda(\lambda) = d\lambda \) and (37), respectively (39), gives the covariance of the 1/z noise as a simple integral of Gaussian, respectively exponential, modes. This corresponds to the limit of
infinite superposition of log-K fields with finite correlation scales, like those considered in the numerical investigations presented in the following.

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

\[ C^P_L(x) = \int_0^\infty d\Lambda(\lambda) C_Y(x, \lambda), \quad \hat{C}^P_L(k) = \int_0^\infty d\Lambda(\lambda) \hat{C}_Y(k, \lambda) \]  

(41)

By inserting (41) in (24) and (5), we finally obtain the superposition relations

\[ \delta\{D^P_L,\text{ens}\}(t) = \int_0^\infty d\Lambda(\lambda) \{D^\text{ens}_i\}(t, \lambda), \quad \delta\{D^P_L,\text{eff}\}(t) = \int_0^\infty d\Lambda(\lambda) \{D^\text{eff}_i\}(t, \lambda). \]  

(42)

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

5 Numerical simulations of transport in aquifers with multiscale hydraulic conductivity

To investigate numerically the behavior of several transport observables we computed an ensemble of 256 GRW simulations of isotropic diffusion, with coefficient \( D = 0.01 \text{ m/d} \) which is a typical value for local dispersion coefficient, 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 with isotropic exponential correlation and constant variance 0.1. 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 of 1m, so that the superposition of 2 fields has the variance 0.2 and integral scale \( \lambda = 3 \text{ m} \) and that of seven fields (the largest one investigated here) has the variance 0.7 and \( \lambda = 28 \text{ m} \). In our GRW numerical simulations it is more convenient to estimate dispersion coefficients for processes starting from instantaneous point injections by the mean half-slope, \( \Sigma_{ii}(t)/(2t), S_{ii}(t)/(2t), R_{ii}(t)/(2t) \). The GRW algorithm is presented in detail by Vamoș et al. [25] and its implementation for transport in saturated aquifers is described in reference [22]. The effective, ensemble, and center of mass coefficients and the corresponding one-step memory dispersion coefficients \( M_{ii}(t)/(2t) \), where \( M_{ii}(t) = M_{ii}(t, t - \Delta, 0) \), normalized by the local dispersion coefficient, are presented in Figs. 1-12. The extinction of the memory coefficients (Figs. 7-12) coincides with the approach to a normal diffusive behavior, in agreement with (15) and (17).

Figures 13 and 14 show the cumulated 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 cumulated 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 \( \Delta \) (which in our case is also the simulation time step). In case of ensemble dispersion \( \Sigma_{ii} \), 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 the increment correlations or by correlations of the Lagrangian velocity at different moments. 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 15 and 16 show the ensemble dispersion coefficients for one time-step increments of the process \( X^\text{ens}_{i;t} \), normalized by the local dispersion coefficient as well. According to (5), \( \Sigma_{ii}(t, t - \Delta) \) are given by the sum between the local dispersion coefficient and the integral of the Lagrangian velocity over a time step \( \Delta \) and they increase with the integral scale.
The asymmetry of the solute plume at 1000 days, presented in Fig. 17, is more pronounced than that recorded at 2000 days (Fig. 18). The asymmetry of the plume indicates that many solute particles move slower than the center of mass and remain behind the plume’s front. The explanation is, very likely, the persistence of negative velocity fluctuations, rather than the increase of fluctuations’ magnitude (note that the variance of the lnK field is only 0.7). This agrees with the anomalous behavior of the dispersion coefficients over the first 1000 days (Figs. 1 and 3) and with expected approach to a Fickian regime in the limit of very large times, ensured by the finiteness of the integral scale [23].

The breakthrough curves (Figs. 19-20) are also strongly asymmetric at early stages and become more and more symmetrical as the process approaches the Fickian regime. Ensembles of simulated breakthrough curves are available and can be used to compute means and variances of the travel times (Figs. 21-22). Complete probability distributions of breakthrough concentration or of the arrival times at reference planes can be readily obtained from the ensemble of 256 transport simulations.

The concentration at the center of mass, integrated over the transverse dimension of the plume decreases, and the corresponding coefficient of variation increases, with the increase of the integral scale (Figs. 23-24). One remarks a strong non-Gaussian behavior in seven-scales case (compare black and red lines). More information on effect of multiscale fields provide the spatial profiles of the cross-section mean concentration and representations based on them presented in Figs. 25-26. Here one observes small asymmetry of the concentration profiles, and mainly that of their standard deviation, for multiscale fields in the pre-asymptotic regime, which diminishes at larger travel distances. More evident is the asymmetry shown by Figs. 27-28. There we represented the asymmetry of the longitudinal dispersion coefficient computed as follows. First we decomposed the effective coefficient in single realizations as a sum of cross-section contributions

\[
D_{11}^{eff}(t) = \int \int (x_1 \cdot \langle x_1 \rangle)^2 p(x_1, x_2, t) dx_1 dx_2
\]

\[
= \int dx_1 \int (x_1 \cdot \langle x_1 \rangle)^2 p(x_1, x_2, t) dx_2
\]

\[
= \int d_{11}^{eff}(x_1, t) dx_1
\]

The ensemble averages \(d_{11}^{eff}(x_1, t)\) and the corresponding standard deviations are presented in Fig. 27 and 28, respectively. Here, again, one can see that asymmetry persists in pre-asymptotic regime and increases with the integral scale of the velocity field. Further, we quantified the asymmetry by

\[
\int_{x_1 < \langle x_1 \rangle} d_{11}^{eff}(x_1, t) dx_1 / \int_{x_1 > \langle x_1 \rangle} d_{11}^{eff}(x_1, t) dx_1.
\]

The asymmetry coefficient (44) is presented in Fig. 29 and the corresponding effective coefficient (43) (averaged over realizations) in Fig. 30. Fig. 29 supplies another evidence of the relation between integral scale and asymmetry, which manifests in pre-asymptotic regime. (The large positive asymmetry at 2000 days in seven-scales case is very likely an artifact due to numerical trapping of particles in two-dimensional GRW simulations [22]).

To further quantify the influence of multiscale velocity, we fitted dispersion and memory coefficients with power-law functions. The corresponding lines in log-log plots from Figs. 31-32 fit the behavior of the dispersion coefficients, variances, and memory terms in time windows proportional to the integral scale, of 15, 25, 35, 55, and 300 days for single-, two-, three-, five-, and seven-scale fields, respectively. The increase of time window with integral scale is consistent with theoretical and numerical results obtained in the past [9, 3]. Figures 33 and 34 show that ensemble and effective coefficients and variances have different time behavior, with larger slope of the effective quantities. This seems to be at variance with the theoretical result for power-law correlated fields obtained by Fiori [11].

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]. 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...
multiscale transport may be a bridge to theoretical modeling by fractional Brownian motion (fBm) and fractional Gaussian noise (fGn) models. For instance, the super-diffusive behavior shown in Figs. 31-34 can be modeled as a fBm and the fGn generating it can be readily obtained by two time derivatives, which reduce the power-law exponents by 2 [23, 21, 16].

As we discussed in the previous Section, power-law covariances can be constructed by integrating covariances with continuously increasing integral scales [15]. An infinite sum of equal-weight exponential correlations results in an \(1/x\)-type correlation (according to [15], p. 370, expression 3.478 (1.); see also the discussion after Eq. (40) in Section 4). Such correlation leads to a \((t \ln t - t)\) behavior of the ensemble variance \(\Sigma_{ij}\) [21]. Since this would be the theoretical infinite limit of our simulations, we also fitted dispersion coefficients and variances with \(\ln t\) and \(t \ln t - t\) respectively (Figures 35-36). At a first sight the fitting results are also acceptable. Nevertheless, a more detailed analysis is required to decide which time-behavior, the power-law one from Figs. 33-34 or the \(\ln t\)-type behavior from Figs. 35-36 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 an \(1/x\)-correlated log-K field, using the integral representation (39-40) derived in Section 4.

References


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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.

Figure 3: Longitudinal effective dispersion coefficients for log-K fields with increasing integral scales.

Figure 4: Transverse effective dispersion coefficients for log-K fields with increasing integral scales.

Figure 5: Longitudinal center of mass dispersion coefficients for log-K fields with increasing integral scales.

Figure 6: Transverse center of mass dispersion coefficients for log-K fields with increasing integral scales.
Figure 7: Longitudinal ensemble memory coefficients for log-K fields with increasing integral scales.

Figure 8: Transverse ensemble memory coefficients for log-K fields with increasing integral scales.

Figure 9: Longitudinal effective memory coefficients for log-K fields with increasing integral scales.

Figure 10: Transverse effective memory coefficients for log-K fields with increasing integral scales.

Figure 11: Longitudinal center of mass memory coefficients for log-K fields with increasing integral scales.

Figure 12: Transverse center of mass memory coefficients for log-K fields with increasing integral scales.
Figure 13: Cumulated longitudinal memory coefficients compared with the corresponding ensemble, effective and center of mass dispersion coefficients for the seven-scales log-K field (integral scale = 28 m).

Figure 14: Cumulated transverse memory coefficients compared with the corresponding ensemble, effective and center of mass dispersion coefficients for the seven-scales log-K field (integral scale = 28 m).

Figure 15: Longitudinal ensemble coefficients of the one-step increments for fields with increasing integral scales.

Figure 16: Transverse ensemble coefficients of the one-step increments for fields with increasing integral scales.

Figure 17: Solute plume at $t = 1000$ days for transport in the seven-scales log-K field.

Figure 18: Solute plume at $t = 2000$ days for transport in the seven-scales log-K field.
Figure 19: Breakthrough curve $c(t) \pm \sigma_c(t)$ at $x = 5, 50, 100$ m for transport in seven-scales log-K field.

Figure 20: Breakthrough curve $c(t) \pm \sigma_c(t)$ at $x = 300, 500, 1000, 1500$ m for transport in seven-scales log-K field.

Figure 21: Mean travel-time for breakthrough curves recorded at $x = 5, 50, 100, 300, 500, 1000, 1500$ m. Effective mean travel-times and corresponding standard deviations (error-bars) are computed by averaging over the ensemble of 256 transport simulations the single realization time-average mean travel times.

Figure 22: Travel-time variance for breakthrough curves recorded at $x = 5, 50, 100, 300, 500, 1000, 1500$ m. Effective travel-time variances and their standard deviations (error-bars) are computed by averaging over the ensemble of 256 simulations the single realization time-average travel-time variances.

Figure 23: Ensemble averaged concentrations at the plume center of mass for increasing integral scales.

Figure 24: Concentration coefficients of variation at the plume center of mass for increasing integral scales.
Figure 25: Cross section space-averaged concentration at the center of mass recorded at \( t = 500, 1000, 1500, 2000 \) days.

Figure 26: Standard deviation of cross section averaged concentration at the center of mass computed at \( t = 500, 1000, 1500, 2000 \) days.

Figure 27: Decomposition (43) of the longitudinal effective dispersion coefficient computed at \( t = 500, 1000, 1500, 2000 \) days.

Figure 28: Standard deviation of the decomposition of the longitudinal effective dispersion coefficient computed at \( t = 500, 1000, 1500, 2000 \) days.

Figure 29: Asymmetry (44) of the longitudinal effective dispersion coefficient computed at \( t = 500, 1000, 1500, 2000 \) days.

Figure 30: Longitudinal effective coefficient computed at \( t = 500, 1000, 1500, 2000 \) days.
Figure 31: Power-law fit of longitudinal ensemble variances for increasing scales.

Figure 32: Power-law fit of longitudinal ensemble memory terms for increasing scales.

Figure 33: 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.

Figure 34: 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.

Figure 35: 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.

Figure 36: 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.
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