

Memory effects induced by dependence on initial conditions and ergodicity of transport in heterogeneous media

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For transport in statistically homogeneous random velocity fields with properties that are routinely assumed in stochastic groundwater models, the one-particle dispersion (i.e., second central moment of the ensemble average concentration for a point source) is a “memory-free” quantity independent of initial conditions. Nonergodic behavior of large initial plumes, as manifest in deviations of actual solute dispersion from one-particle dispersion, is associated with a “memory term” consisting of correlations between initial positions and displacements of solute molecules. Reliable numerical experiments show that increasing the source dimensions has two opposite effects: it reduces the uncertainty related to the randomness of center of mass, but, at the same time, it yields large memory terms. The memory effects increase with the source dimension and depend on its shape and orientation. Large narrow sources oriented transverse to the mean flow direction yield ergodic behavior with respect to the one-particle dispersion of the longitudinal dispersion and nonergodic behavior of the transverse dispersion, whereas for large longitudinal sources, the longitudinal dispersion behaves nonergodically, and the transverse dispersion behaves ergodically. Such memory effects are significantly large over hundreds of heterogeneity scales and should therefore be considered in practical applications, for instance, calibration of model parameters, forecasting, and identification of the contaminant source.

1. Introduction

[2] The transport in heterogeneous media is conveniently characterized by the second central spatial moment tensor of the concentration field s_{lm} ($l, m = 1, 2, 3$). As well as providing a measure for the spatial extension of the solute plume, the dependence on time t of the second moment is commonly used to investigate whether the transport is diffusive, i.e., $s_{lm} \sim t$ [Sposito and Dagan, 1994]. Since it can be estimated, by either analytical approximations or numerical simulations, without solving the transport equations [Suci et al., 2006b; Eberhard et al., 2007] this quantity is particularly useful in investigations on pre-asymptotic transport regime, for which generally there are no close form solutions [see, e.g., Morales-Casique et al., 2006].

[3] A frequently used approach considers a local dispersion process and models the heterogeneity of the velocity at larger scales by a space random function. Let X_l be the l component of the trajectory of a solute molecule. To simplify matters, we consider only the diagonal components ll of the various second moments. For a fixed realization of the velocity, the second moment s_{ll} of the actual concentration is given by the dispersion of the molecules at a given time

$$s_{ll} = \langle [X_l - \langle X_l \rangle_{DX_0}]^2 \rangle_{DX_0}. \quad (1)$$

The subscripts D and X_0 in (1) denote respectively the average over the realizations of the local dispersion and the space average with respect to the initial distribution of molecules.

[4] To emphasize the role of initial conditions, we consider displacements $\tilde{X}_l = X_l - X_{0l}$ relative to initial positions X_{0l} which introduced in (1) yield

$$s_{ll} = S_{ll}(0) + \langle [\tilde{X}_l - \langle \tilde{X}_l \rangle_{DX_0}]^2 \rangle_{DX_0} + m_{ll}, \quad (2)$$

where $S_{ll}(0) = \langle [X_{0l} - \langle X_{0l} \rangle_{X_0}]^2 \rangle_{X_0}$ is the deterministic initial second moment. The last term in (2),

$$m_{ll} = 2 \langle [X_{0l} - \langle X_{0l} \rangle_{X_0}] \langle \tilde{X}_l \rangle_D \rangle_{X_0}, \quad (3)$$

describes spatial correlations between relative displacements on trajectories and initial positions and, therefore, it has been called “memory term” [Suciu and Vamoş, 2007; Suciu *et al.*, 2007b]. The implications of such terms for predicting solute dispersion were first investigated by Sposito and Dagan [1994], in the context of deterministic advective transport. A decade later, Fiori and Jancović [2005] simulated the time behavior of m_{II} , for advective transport as well, and concluded that a representative transverse dispersivity cannot be inferred from experiments done for large transverse sources. Recent numerical investigations which considered both advection and local dispersion [Suciu *et al.*, 2006a] also indicated that the transverse dispersion for large transverse sources significantly differs from theoretical “ergodic” results. This paper aims at highlighting relationships between initial conditions, memory terms, and the nonergodic behavior of the pre-asymptotic dispersion. Our study is based on “global random walk” (GRW) numerical simulations [Vamoş *et al.*, 2003], for different initial conditions, which complete previous ones presented by Suciu *et al.* [2006a].

2. Second Moments and Memory Terms

[5] Another equivalent expression of dispersion (1), which now highlights the randomness of the center of mass, is

$$s_{II} = \sigma_{II} - r_{II} \quad (4)$$

$$\sigma_{II} = \langle [X_I - \langle X_I \rangle_{DX_0V}]^2 \rangle_{DX_0} \quad (5)$$

$$r_{II} = [\langle X_I \rangle_{DX_0} - \langle X_I \rangle_{DX_0V}]^2, \quad (6)$$

where $\langle X_I \rangle_{DX_0V}$ is the average over the ensemble of realizations of the random velocity field (hereafter indicated by a subscript V) of the center of mass. The ensemble average of (4) is the well known identity [Kitanidis, 1988; Le Doussal and Machta, 1989; Naff *et al.*, 1998; Suciu *et al.*, 2006a] which relates the expected second moment $S_{II} = \langle s_{II} \rangle_V$ to the second moment of the mean concentration $\Sigma_{II} = \langle \sigma_{II} \rangle_V$ and the variance of the center of mass $R_{II} = \langle r_{II} \rangle_V$:

$$S_{II} = \Sigma_{II} - R_{II}. \quad (7)$$

[6] Assuming all necessary joint measurability conditions which allow permutations of averages [Zirbel, 2001] leads to

$$\Sigma_{II} = S_{II}(0) + \langle X_{II} \rangle_{X_0} + M_{II} + Q_{II}, \quad (8)$$

where $X_{II} = \langle [\tilde{X}_I - \langle \tilde{X}_I \rangle_{DV}]^2 \rangle_{DV}$ is the one-particle dispersion (defined by averaging with respect to D and V for a fixed initial position), $M_{II} = \langle m_{II} \rangle_V$ is the mean memory term, and $Q_{II} = \langle [\langle \tilde{X}_I \rangle_{DV} - \langle \tilde{X}_I \rangle_{DX_0V}]^2 \rangle_{X_0}$ is the spatial variance (computed by averages over X_0) of the one-particle center of mass $\langle \tilde{X}_I \rangle_{DV}$ [Suciu *et al.*, 2007b; Suciu and Vamoş, 2007].

[7] The terms of (8) depend, via the trajectory equation, on Lagrangian velocity field $\mathbf{V}^L(\mathbf{X}_0, t) = \mathbf{V}(\mathbf{X}(t; \mathbf{X}_0))$, which consists of observations of the random Eulerian velocity field $\mathbf{V}$ at random locations on the trajectory. If the Lagrangian field is statistically homogeneous the one-particle center of mass $\langle \tilde{X}_I \rangle_{DV}$ and dispersion X_{II} are independent of X_0 . Then M_{II} and Q_{II} vanish and (7)–(8) takes on the simpler form [see, e.g., Dagan, 1990]

$$S_{II} = S_{II}(0) + X_{II} - R_{II}. \quad (9)$$

[8] The homogeneity of $\mathbf{V}^L$ holds under conditions routinely assumed in stochastic modeling which essentially consist of statistical homogeneity of the Eulerian field, continuity of the velocity samples, and existence of unique solutions of trajectory equations. The equality of Lagrangian and Eulerian mean velocities requires stronger conditions. Depending on whether dispersion processes are modeled as spatially correlated noises or as Brownian motions, these are the flow’s incompressibility or both incompressibility and sample continuity of first derivatives of $\mathbf{V}$ [Zirbel, 2001]. Implications of velocity properties for a transport model based on Itô equation, explicit expressions for dispersion, and first-order approximations are presented at length in the preprint by N. Suciu *et al.* (Dependence on initial conditions, memory effects, and ergodicity of transport in heterogeneous media, 2008, Institute of Applied Mathematics, Friedrich-Alexander University Erlangen-Nuremberg, available at <http://www.am.uni-erlangen.de/de/preprints2000.html>), on which this paper is based.

[9] The one-particle quantities entering (8) are ensemble averages of the first two moments of plumes starting from point sources located at $\mathbf{x}_0$ which can be calculated from the ensemble average of the transition probability defined by [see, e.g., van Kampen, 1981] $p(\mathbf{x}, t|\mathbf{x}_0) = \langle \delta[\mathbf{x} - \mathbf{X}(t; \mathbf{x}_0)] \rangle_D$. Since the ensemble average $\langle p \rangle_V$ depends on velocity statistics through the moments of the Lagrangian velocity $\mathbf{V}^L$, it is translation invariant for homogeneous $\mathbf{V}^L$ [Suciu *et al.*, 2008] (see also the preprint by Suciu *et al.*, 2008). This property is helpful in renormalized series expansions for dispersion coefficients [Phythian and Curtis, 1978] and has been used by Dentz *et al.* [2000] to show the independence of the ensemble coefficients $\frac{1}{2}d\Sigma_{II}/dt$ from the initial concentration distribution.

[10] Homogeneous random fields with finite correlation range are ergodic: space averages of the velocity are unbiased estimators strongly convergent to the ensemble mean velocity [Chilès and Delfiner, 1999]. There is also a numerical evidence that space averages with respect to the location of the source of the concentration moments resulted from simulations of diffusion in Gaussian fields converge to their ensemble averages [Suciu *et al.*, 2007a]. For plumes with initial dimensions larger than the heterogeneity scale [Dagan, 1990] one can therefore assume that, according to (6), $r_{II} \approx R_{II} \approx 0$. When ergodic behavior prevails, it follows from (9) that the one-particle dispersion X_{II} provides an idealized description of the expected second moment,

$$S_{II} \approx S_{II}(0) + X_{II}. \quad (10)$$

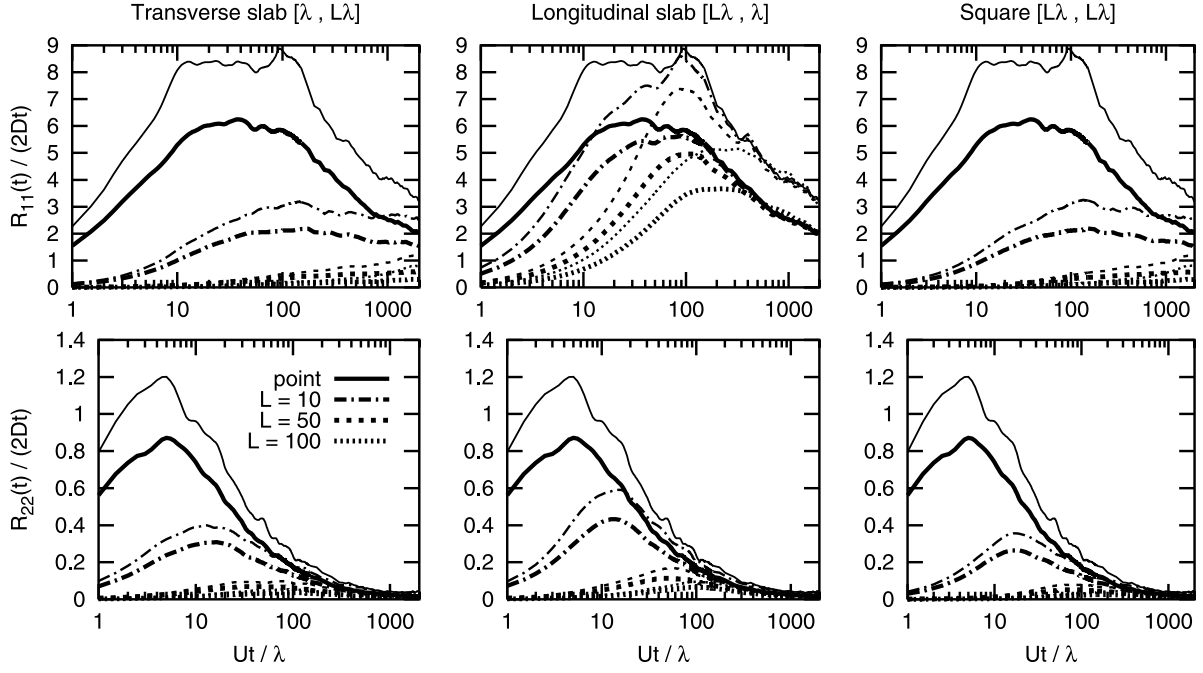


Figure 1. Variance of the plume center of mass R_{II} and corresponding standard deviation $SD(r_{II})$ (thin lines) for different shapes and extensions of the initial plume.

[11] A similar ergodic argument suggests that the second term of (2) behaves like X_{II} for large initial plumes. But even if (10) can approximate the expectation, it might not be accurate for the actual dispersion. Indeed, (3) shows that for large sources the memory terms can be important. So, at best, the actual dispersion can be approximated as

$$s_{II} \approx S_{II}(0) + X_{II} + m_{II}. \quad (11)$$

[12] The relevance of the stochastic description for the dispersion observable in a real case; that is, the ergodicity in a broad sense, can be assessed quantitatively by an “ergodicity range” defined as root mean square deviation of observable quantities from stochastic model predictions [Suciu et al., 2006a]. The numerical experiment presented in the following shows that for large initial plumes the ergodicity range of $s_{II} - S_{II}(0)$ with respect to X_{II} can be estimated, according to (11), by standard deviations of memory terms m_{II} .

3. Memory Effects and Ergodicity

[13] We considered an isotropic two-dimensional aquifer system, characterized by log-hydraulic conductivity with small variance equal to 0.1 and exponentially decaying isotropic correlation with correlation length $\lambda = 1$ m. First-order approximations for incompressible Darcy velocity fields were generated numerically. The task was achieved with the Kraichnan algorithm by using 6400 periodic modes, which guarantees accurate simulations of transport in Gaussian fields over thousands of heterogeneity scales [Eberhard et al., 2007]. For fixed mean flow velocity $U = 1$ m/d and isotropic local dispersion with constant coefficient $D = 0.01$ m²/d, the Péclet number got a typical value $Pe = U\lambda/D = 100$. In every velocity realization, 10^{10}

particles, that were initially uniformly distributed in rectangular domains $L_1\lambda \times L_2\lambda$ or released from the origin of the computational grid, were tracked simultaneously with the GRW algorithm (for details on the implementation of the numerical method see Suciu et al. [2006a, Appendix A]). For all cases investigated here we simulated 1024 realizations, which rendered the statistical oscillations of the estimated means and standard deviations of the dispersion terms (4), (5), and (6) smaller than half the local dispersion Dt uniformly in time (see the preprint by Suciu et al., 2008).

[14] Figure 1 shows that the dispersion of the center of mass R_{II} decays monotonically with the increasing dimensions of the source, irrespective of its shape and orientation. The standard deviations $SD[r_{II}]$ were found to be of the same order of magnitude. They are significantly larger than R_{II} only for small sources. The results for $L \geq 50$ provide a numerical support for the ergodic argument which suggests that for large plumes $R_{II} \approx 0$. Hence, if relation (9) holds, then the expectation of the second moment of the actual concentration field can be approximated with relation (10) by the sum between the deterministic initial second moment $S_{II}(0)$ and the one-particle dispersion X_{II} .

[15] Numerical results for the second moment of the ensemble averaged concentration Σ_{II} (not presented here) show that for slab sources perpendicular to l -axis X_{II} can be accurately estimated by $\Sigma_{II} - S_{II}(0)$, in keeping with theoretical predictions for transport in statistically homogeneous velocity fields (compare (7) and (9)). For sources with large extensions on l -axis we found differences between the two quantities (of the order of a few local dispersions $2Dt$ for $L = 100$) which indicate non-vanishing mean memory terms M_{II} . Both situations described above (for transverse slab sources only) and the “ergodic” dispersion coefficients $X_{II}/(2Dt)$ (derived from point source simulations) were previously presented by Suciu et al. [2006a, Figures 11 and 13]. The non-zero values of M_{II} can be explained by small

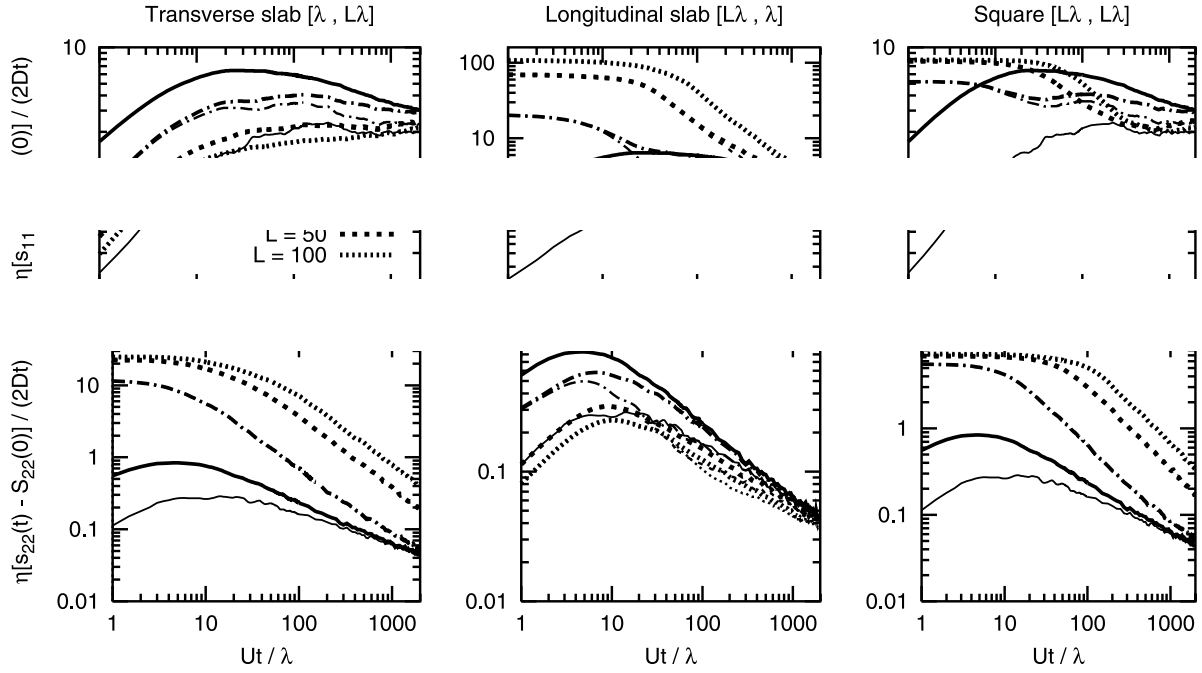


Figure 2. Ergodicity range η with respect to the memory-free dispersion and standard deviation $SD(s_{||})$ of the actual dispersion (thin lines) for different shapes and extensions of the initial plume.

variations with the initial position of the simulated mean Lagrangian velocity [Suciu and Vamoş, 2007, Figure 2].

[16] Similar GRW simulations for smoother velocity fields and larger ensembles of realizations led to values M_{22} smaller than Dt [Suciu et al., 2007b]. Preliminary test simulations show that increasing the number of periodic modes in the Kraichnan routine from 64 to 6400, for transverse sources with $L = 100$ and a fixed number of 256 realizations, causes the increase of M_{22} from values smaller than $2Dt$ to about $5Dt$ [Suciu et al., 2008]. Since larger number of modes yield closer approximations for the random velocity fields with exponential correlations, considered in this numerical setup, the occurrence of non-zero mean memory terms can be associated with the lack of smoothness of the samples of such random fields [Yaglom, 1987]. Simulations based on the mixed finite element method for both water flow [Radu et al., 2004] and solute transport [Radu et al., 2008] will be compared in a forthcoming paper with the present simulations, which are based on first-order approximations of the velocity field. A conclusion is yet premature and further work is needed to clarify whether the mean memory terms reflect irregularities of the velocity model or whether they are finite size effects inherent in numerical simulations which always reproduce the nominal values of the velocity statistics with some finite precision.

[17] The mean memory terms are expected to be quantitatively important for transport in inhomogeneous velocity fields, when relation (9) is no longer verified and has to be replaced by (7) and (8) [Suciu and Vamoş, 2007]. Here we investigate memory effects on dispersion in single realizations of a homogeneous random velocity field. Nevertheless, because single realization quantities such as $s_{||}$ and $m_{||}$ are random variables, we cannot give up using ensemble averages. Like in statistical inference problems [Yaglom,

1987], we need estimations by distances in the mean square sense (ergodicity ranges), computed by averaging over statistical ensembles. Since in this case the mean memory terms are 1 or 2 orders of magnitude smaller than the large standard deviations of the actual dispersion (see Figure 2), they can be neglected. On the other hand, their standard deviation will be shown to be relevant for the quantitative evaluation of the memory effects presented in the following.

[18] Memory effects on single-realization dispersion are demonstrated by the strong influence of the shape and dimension of the source on the ergodicity range η of $s_{||} - S_{||}(0)$ with respect to the memory-free dispersion $X_{||}$ presented in Figure 2. For large slab sources perpendicular to l -axis η decreases with L , whereas for large extensions of the source on the l -axis η strongly increases to values that at early times are 1 or 2 orders of magnitude larger than for a point source. The strongest increase is found in case of longitudinal dispersion for longitudinal slabs.

[19] The ergodicity range $\eta[s_{||} - S_{||}(0)] = \langle (s_{||} - S_{||}(0) - X_{||})^2 \rangle_V^{1/2}$ was computed from the deviation of the mean $S_{||} - S_{||}(0) - X_{||} = \eta_1$ and the standard deviation $SD(s_{||}) = \eta_2$ via $\eta = [\eta_1^2 + \eta_2^2]^{1/2}$ [Suciu et al., 2006a]. The longitudinal one-particle dispersion X_{11} was estimated by $\Sigma_{11} - S_{11}(0)$ in case of transverse slab $\lambda \times 100\lambda$ and the transverse one, X_{22} , by $\Sigma_{22} - S_{22}(0)$ in case of longitudinal slab $100\lambda \times \lambda$. Given the smallness of η in these cases, we approximate $s_{||} - S_{||}(0) \approx X_{||}$. From (3) and the Cauchy-Schwartz inequality, $m_{||}^2(t) \leq 4S_{||}(0)\langle X_{||}^2 \rangle_{X_0}$, we also find that the memory terms for slabs perpendicular to l -axis can be neglected as compared with those for slabs oriented along l . In these conditions, the second term of (2) estimates $X_{||}$ and the approximation (11) can be adopted. Since $SD(s_{||})$ and $\eta[s_{||} - S_{||}(0)]$ practically coincide for $L \geq 10$, regardless the shape and the orientation of the source, it follows that $\eta_1 \approx 0$; that is, the actual dispersion is an (almost) unbiased estima-

tor of X_{ll} . Then the ergodicity range is given by $\eta_2 = SD(s_{ll})$, which according to (11) is just the standard deviation $SD(m_{ll})$ of the memory terms.

4. Discussion and Conclusions

[20] Persistent influences of the shape and dimension of the source on the second moments of the plume reported in this paper are certainly related to memory terms consisting of correlations between initial positions and displacements of solute molecules. Such correlations naturally occur as components of the dispersion whenever the dependence of the trajectory on the initial position is considered explicitly, as previously shown by *Sposito and Dagan* [1994]. For a given direction, the memory terms increase with the increase of the source dimension in the same direction and are responsible for nonergodic behavior of the actual dispersion with respect to the memory-free one-particle dispersion. The uncertainty caused by the randomness of the center of mass, instead, is reduced by increasing the dimension of the source and is less sensitive to its shape and orientation.

[21] The analysis of the GRW simulations reveals that for a typical situation of contaminant transport in aquifers the memory terms can be tens to hundreds of times larger than the one-particle dispersion. Their observed decay after some hundreds of heterogeneity scales indicates that the displacements averaged over local dispersion, $\langle \tilde{X}_l \rangle_D$, gradually decorrelates from the initial position and (3) goes to zero. This behavior is somehow expected for non-vanishing local dispersion and ergodic velocity fields. The situation could be different for advective transport in Darcy velocity fields. *Sposito* [2001] has shown that the trajectories of deterministic Darcy flows are generally confined on invariant subsets of the flow domain. It is therefore possible that solute molecules, when driven by Darcy flows and in absence of local dispersion, never lose the memory of initial position and memory terms persist indefinitely, as suggested by numerical investigations of *Fiori and Jancović* [2005].

[22] Another extreme situation is when the relative displacement $\tilde{X}_l$ is independent of initial position X_{0l} and the memory term m_{ll} defined by (3) vanishes. This happens, for instance, when $\tilde{X}_l$ is the superposition of a diffusion process and a uniform movement with constant velocity, as in case of confined stratified flows through a single fracture in geological media [see, e.g., *Dentz and Carrera*, 2007]. Then, the transverse dimension of the source governs the interplay between the local dispersion and the coherent cross-section velocity profile. The significant relation for the longitudinal dispersion is obtained from (1) by adding and subtracting $\langle \langle \tilde{X}_1 \rangle_D^2 \rangle_{X_0}$, where $\langle \tilde{X}_1 \rangle_D$ is the center of mass of a solute plume originating from a point source,

$$s_{11} = S_{11}(0) + \langle \langle [\tilde{X}_1 - \langle \tilde{X}_1 \rangle_D]^2 \rangle_D \rangle_{X_0} + \langle \langle [\tilde{X}_1]_D - \langle \tilde{X}_1 \rangle_{DX_0} \rangle_D^2 \rangle_{X_0}. \quad (12)$$

The last term in (12) is a spatial variance of the point-source center of mass which carries the memory of initial conditions: when it becomes negligible, the reduced dispersion $s_{11} - S_{11}(0)$ behaves as a superposition of point-source dispersions, $\langle \langle [\tilde{X}_1 - \langle \tilde{X}_1 \rangle_D]^2 \rangle_D \rangle_{X_0}$.

[23] Memory terms of form (3) always vanish for point sources, even if the relative displacements depend on initial positions. But if the only available information is the concentration distribution at a post-injection stage, which has to be taken as an initial condition, memory terms cannot be disregarded until the dispersion has reached a linear time behavior [*Sposito and Dagan*, 1994]. Explicit expressions of memory terms (3), derived from trajectory equations, are then given by time integrals of correlations of the Lagrangian velocity at pre- and post-injection times, which account for the non-linear behavior of dispersion in pre-asymptotic regime (see the preprint by *Suciu et al.*, 2008).

[24] The dependence of the memory effects on the source extension and anisotropy can be relevant for the calibration of the model, predictions, and procedures for identification of the source of contamination from available measurement data. For example, it is known that for small sources the best fit of measured second moments and theoretical memory-free dispersion in velocity fields with finite correlation range, X_{ll} , can underestimate the variance and the correlation length of the hydraulic conductivity [*Suciu et al.*, 2006b]. As shown in Figure 2, erroneous estimations will also be obtained for sources with large extension on the l direction. Instead, for large narrow sources perpendicular to l , the ergodic behavior of the actual dispersion with respect to X_{ll} can be used in practice to improve the parameter identification from field experiments.

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