Comment on "Nonstationary flow and nonergodic transport in random porous media" by G. Darvini and P. Salandin

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[1] *Darvini and Salandin* [2006] addressed the extremely important problem of the implications of the statistical inhomogeneity of the velocity field for the solute transport in saturated aquifers. The authors considered the velocity inhomogeneity due to the limited size of the computational domain which occurs in numerical simulations for assumed statistically homogeneous hydraulic conductivity. In this comment we question the correctness of the estimation for the expected second moment of the solute plume in the paper of *Darvini and Salandin* [2006]. Even though the explicit formula used to compute this quantity was not provided, there are indications that relevant terms accounting for the inhomogeneity of the random velocity field were ignored. Clarifying this issue is essential for an evaluation of the results. We also show that the validity of the basic relation for the expected value of the second spatial moment of the plume [*Darvini and Salandin*, 2006, equation (19)] extends beyond the frame of their first-order approach for purely advective transport. Finally we argue that the approach of *Darvini and Salandin* [2006] is well suited and can be used to produce a detailed explanation for the nonergodic behavior of the second moments caused by the statistical inhomogeneity of the velocity field.

[2] With a finite element method using Taylor series expansions, a realization of the velocity field was computed as sum of a deterministic component (ensemble mean) and a fluctuating one, $\mathbf{v}(\mathbf{x}) = \mathbf{v}_0(\mathbf{x}) + \mathbf{v}'(\mathbf{x})$ [*Darvini and Salandin*, 2006, equation [10]]. Because of the influence of the boundaries for small computational domains, the ensemble mean velocity $\mathbf{v}_0 = \langle \mathbf{v} \rangle$ is space dependent, thus the random field is not statistically homogeneous. The local dispersion is neglected and trajectories of the solute particles, starting from $\mathbf{x} = \mathbf{a}$ at time $t = t_0$, are approximated by a first iteration of the equation of motion

$$X_i(t;\mathbf{a},t_0) = X_{0i}(t;\mathbf{a},t_0) + \int_{t_0}^{t} v_i'(\mathbf{X}_0(t';\mathbf{a},t_0))dt', \qquad (1)$$

where $i = 1, 2, 3$ and

$$X_{0i}(t;\mathbf{a},t_0) = a_i + \int_{t_0}^{t} v_{0i}(\mathbf{X}_0(t';\mathbf{a},t_0))dt' \qquad (2)$$

is the deterministic trajectory of the mean velocity. This consistent first-order approximation in velocity fluctuations yields robust estimations [*Suciu et al.*, 2006b] for dispersion coefficients defined as time integrals of Lagrangian velocity covariance [see, e.g., *Salandin and Fiorotto*, 1998]. The equations (1) and (2), written for our convenience in a more explicit form, correspond exactly to the Lagrangian approach of *Darvini and Salandin* [2006, section 3].

[3] To simplify the writing, in the following we note by

$$\langle f(\mathbf{a})\rangle_{\mathbf{a}} = \frac{1}{V_0}\int_{V_0} f(\mathbf{a})d\mathbf{a},$$

the average of a function f over the initial plume of constant concentration in the volume V_0. So, the coordinate of the center of mass is $R_i = \langle X_i\rangle_{\mathbf{a}}$ and the diagonal components of the second moment can be written as $S_{ii} = \langle [X_i - R_i]^2\rangle_{\mathbf{a}}$. As pointed out by *Darvini and Salandin* [2006, p. 4], in statistically inhomogeneous velocity fields, both R_i and S_{ii} depend on the size, the shape, and the location of the initial plume. The expectation of the second moment has the following equivalent forms:

$$\begin{aligned}\langle S_{ii}\rangle &= \langle\langle [X_i - R_i]^2\rangle_{\mathbf{a}}\rangle = \langle\langle X_i^2\rangle_{\mathbf{a}}\rangle - \langle R_i^2\rangle \\ &= \langle\langle X_i^2\rangle_{\mathbf{a}}\rangle - \langle R_i\rangle^2 - R_{ii},\end{aligned}$$

where $R_{ii} = \langle R_i^2\rangle - \langle R_i\rangle^2$ is the variance of the center of mass. Assuming that the trajectory is continuous as a function of the initial position, by virtue of Fubini's theorem the ensemble average permutes with the integral with respect to $\mathbf{a}$ and one obtains

$$\langle S_{ii}\rangle = \langle X_{ii}\rangle_{\mathbf{a}} - R_{ii} + \langle\langle X_i\rangle^2\rangle_{\mathbf{a}} - \langle R_i\rangle^2, \qquad (3)$$

where $X_{ii} = \langle X_i^2\rangle - \langle X_i\rangle^2$ is the moment computed by ensemble averaging for a fixed initial position $\mathbf{a}$; that is, it is the one-particle displacements variance [*Dagan*, 1990]. The relation (3), written here for diagonal components of the expected second moment, is just the equation (19) presented by *Darvini and Salandin* [2006] in the frame of their first-order approximation. However, this relation is not based on the approximate form of the equations (1)–(2) and is valid under the only assumption that the averages over initial position and velocity realizations permute. As already

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shown by *Suciu et al.* [2006a, p. 9] a relation of the same form also holds in the general case which considers local dispersion.

[4] By using the displacement $\widetilde{X}_i = X_i - a_i$ relative to the initial position, the last two terms of (3) give $S_{ii}(0) + M_{ii} + Q_{ii}$, where $S_{ii}(0) = \langle a_i^2 \rangle_{\mathbf{a}} - \langle a_i \rangle_{\mathbf{a}}^2$ is the moment of the initial plume and

$$M_{ii} = 2\langle [a_i - \langle a_i \rangle_{\mathbf{a}}] \langle \widetilde{X}_i \rangle \rangle_{\mathbf{a}} \quad (4)$$

$$Q_{ii} = \langle \langle \widetilde{X}_i \rangle^2 \rangle_{\mathbf{a}} - \langle \langle \widetilde{X}_i \rangle \rangle_{\mathbf{a}}^2. \quad (5)$$

With (4) and (5), (3) takes the equivalent form

$$\langle S_{ii} \rangle = S_{ii}(0) + \langle X_{ii} \rangle_{\mathbf{a}} - R_{ii} + M_{ii} + Q_{ii}. \quad (6)$$

The term M_{ii} is the mean correlation between initial positions and particle displacements. *Sposito and Dagan* [1994, p. 587] show that such correlations must be incorporated into the prediction of both actual and expected second moment of the plume. Because M_{ii} carries the information about the initial position of the particles, we call it "memory term". The term Q_{ii} occurring in (6) is a spatial correlation of the expected relative displacement for different initial positions and, as shown in the following, it is a key tool which relates the time behavior of the expected second moment to the velocity inhomogeneity. We also note that $\langle X_{ii} \rangle_{\mathbf{a}} + Q_{ii} = \langle \langle \widetilde{X}_i^2 \rangle_{\mathbf{a}} \rangle - \langle \langle \widetilde{X}_i \rangle_{\mathbf{a}} \rangle^2 = \widetilde{X}_{ii}$, which gives another expression, equivalent with (3) and (6), for the expectation of the second moment:

$$\langle S_{ii} \rangle = S_{ii}(0) + \widetilde{X}_{ii} - R_{ii} + M_{ii}. \quad (7)$$

The assumption of "Lagrangian stationarity" renders the quantities $\langle \widetilde{X}_i \rangle$ and X_{ii} independent of the initial position $\mathbf{a}$. Consequently, the terms (4) and (5) vanish and (6) becomes the well known relation of *Dagan* [1990, equation (11)],

$$\langle S_{ii} \rangle = S_{ii}(0) + X_{ii} - R_{ii}. \quad (8)$$

Following *Dagan* [1990], *Darvini and Salandin* [2006, p. 5] denote by "ergodicity" the relevance of the one particle variance X_{ii} for the expected second moment of the plume (8), which, in this case, is ensured by a negligible small variance R_{ii} of the center of mass.

[5] Before proceeding with the inhomogeneous case, let us shortly discuss the relation (7) from the perspective of the two-dimensional simulations of *Suciu et al.* [2006a]. In that paper, accurate simulations of advection-dispersion in statistically homogeneous random velocity fields, for log-hydraulic conductivity of variance 0.1 and correlation length $\lambda = 1$ m, were presented in detail. With a constant mean velocity $U = 1$ m/d and an isotropic local dispersion coefficient $D = 0.01$ m^2/d, the Péclet number was fixed at $U\lambda/D = 100$, a reasonably high value which is representative for real aquifer systems. The accuracy of the transport simulations was ensured by tracking 10^{10} particles in a given velocity realization with the "global random walk" algorithm [*Vamoş et al.*, 2003]. By using 256 velocity realizations and 6400 periodic modes in the Kraichnan routine the resulting statistical ensemble allows reliable simulations of the self-averaging transport process over thousands of dimensionless times Ut/λ [*Eberhard et al.*, 2007].

[6] In the case of nonvanishing local dispersion the relations presented above hold true with the only difference that the averages over initial positions and velocity realizations are preceded by the average over the realizations of the local dispersion process. It was found that for point and slab sources oriented across the i axis, for which the memory term (4) vanishes or is negligible, the moment $\widetilde{X}_{ii}$ is independent of initial conditions and (7) reduces to (8). On the contrary, for sources oriented along the i axis the relation (8) is no longer verified. Assuming that $\widetilde{X}_{ii}$ is practically the same as that for point sources in all cases, we estimate the transverse memory terms for transverse sources from relation (7). The result presented in Figure 1 shows significant memory terms, which increase with the source dimension. The same increase with the source dimensions of the single realization memory terms (i.e., defined by (4) without ensemble averaging) was found by *Fiori and Jancović* [2005, Figure 5] by simulations of the purely advective transport.

[7] Since the mean displacement $\langle \widetilde{X}_i \rangle$ in (4) is a time integral of the ensemble mean of the velocity field sampled on trajectories, $\langle v_i(\mathbf{X}(t;\ \mathbf{a},\ t_0)) \rangle$, nonvanishing ensemble average memory terms occur only if $\langle v_i \rangle$ is not constant as a function of $\mathbf{a}$. Figure 2 shows that, for the simulations presented in Figure 1, the mean $\langle v_2 \rangle$ varies with the initial position $\mathbf{a}$ of the simulated advection-dispersion processes. Significant differences occur at less than 100 dimensionless times. The multiplication by $[a_2 - \langle a_2 \rangle_{\mathbf{a}}]$ in (4) explains the mean memory terms shown in Figure 1 and their increase with the source dimension L. The average of $\langle v_2(\mathbf{X}(t;\mathbf{a}, t_0)) \rangle$ over $\mathbf{a}$ (solid line in Figure 2) is also nonvanishing in the preasymptotic regime and differs from the numerically estimated Eulerian mean velocity $\langle v_2(\mathbf{x}) \rangle$, which belongs to the range $[-0.00072, -0.00031]$ [*Suciu et al.*, 2006a, Table B1]. A comparison of the curves presented in Figure 2 indicates that the term Q_{22}, which depends on the spatial variance of the ensemble mean velocity on trajectories, is about two orders of magnitude smaller than the local dispersion. Therefore the term $\widetilde{X}_{22}$ in (7) is practically independent of the initial conditions. This example shows that, even for accurate simulations of transport in statistically homogeneous fields, the Lagrangian stationarity cannot be assumed and the mean memory terms M_{ii} are nonvanishing for asymmetric initial plumes. Since the ensemble average statistics of the velocity simulated by the finite element method varies from point to point, even in the core region nonaffected by boundaries [*Bellin et al.*, 1992, p. 2217], one expects that in the paper commented here the simulated second moments $\langle S_{ii} \rangle$ also contain significant mean memory terms M_{ii}.

[8] The ergodicity issue was found to be more subtle than one can expect from analyzes on the basis of relation (8) only. Though R_{ii} can be neglected for large plumes [*Suciu et al.*, 2006a, Figure 12] the one-particle moment $X_{ii} \simeq \widetilde{X}_{ii}$ becomes relevant for the ensemble average of the actual moment $\langle S_{ii} \rangle - S_{ii}(0)$ only when the memory term M_{ii} also becomes negligible (see Figure 1). Moreover, the standard deviation of S_{ii} increases with the extension L of the source on the i axis [*Suciu et al.*, 2006a, Figure 8] and it can be shown that for large sources (longitudinal and transverse

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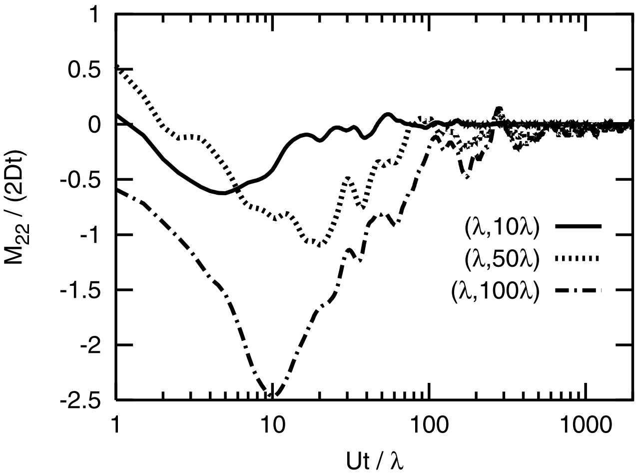

Figure 1. Transverse memory terms for slab sources (λ, $L\lambda$) oriented across the mean flow, computed from data published by *Suciu et al.* [2006a, Figure 13].

slabs and squares) it is well estimated by the standard deviation of M_{ii} (Suciu et al., Memory effects induced by dependence on initial conditions and ergodicity of transport in heterogeneous media, submitted to *Water Resources Research*, 2007). Summarizing the facts, we see that the ergodicity of the actual moment $S_{ii} - S_{ii}(0)$ with respect to the one-particle moment X_{ii} is quantified by both the bias of its ensemble average, which according to (7) is $M_{ii} - R_{ii}$, and by its standard deviation [see *Suciu et al.*, 2006a, definition (5)]. It is noteworthy to point out that such ergodic properties, often formulated in the hydrological literature, differ from the ergodicity of a given random function. The latter denotes the convergence toward the ensemble mean of an unbiased estimator defined by time or space averages of a single realization of the random function.

[9] The expectation of the second moment in the inhomogeneous case is given explicitly by (6). The terms $\langle X_{ii}\rangle_{\mathbf{a}}$ and R_{ii} can be expressed in the above first-order approximation with the aid of the one- and two-particle Eulerian velocity covariances evaluated at points on the trajectory $\mathbf{X}_0(t;\ \mathbf{a},\ t_0)$ of the mean velocity [*Darvini and Salandin*, 2006, equations (22) and (23)]. Using (1) and (2), the last two terms of (6) are obtained from (4) and (5) as

$$M_{ii}(t,t_0) = 2\int_{t_0}^{t} \left\langle \left[a_i - \langle a_i\rangle_{\mathbf{a}}\right] v_{0i}(\mathbf{X}_0(t';\mathbf{a},t_0))\right\rangle_{\mathbf{a}} dt' \qquad (9)$$

$$Q_{ii}(t,t_0) = \int_{t_0}^{t}\int_{t_0}^{t} \left[\langle v_{0i}(\mathbf{X}_0(t';\mathbf{a},t_0))v_{0i}(\mathbf{X}_0(t'';\mathbf{a},t_0))\rangle_{\mathbf{a}} - \langle v_{0i}(\mathbf{X}_0(t';\mathbf{a},t_0))\rangle_{\mathbf{a}}\langle v_{0i}(\mathbf{X}_0(t'';\mathbf{a},t_0))\rangle_{\mathbf{a}}\right] dt'dt''. \qquad (10)$$

Thus the first-order approximation (1)–(2) describes contributions to the expected second moment (6) due to the spatial inhomogeneity of the mean Eulerian velocity field, sampled on the zeroth-order trajectories, by the spatial correlation of the mean velocity (10) and by its correlation with the initial positions of the solute particles (9).

[10] As the authors stated in the abstract, the objective of the paper was "to describe nonergodic transport of inert solutes by spatial moments in a domain of finite size." In other words, according to the sense of ergodcity in their paper, the aim was to characterize the deviation, due to velocity inhomogeneity in a domain of finite size, of the expected second moment from the one particle variance X_{ii}. However, the authors do not explain how they obtained the results on the expected second moment presented in their Figures 10–12. The only indication is that "plume statistics are computed by numerical quadrature on the same grid adopted in the FE solution by relationships described in section 3" [*Darvini and Salandin*, 2006, p. 5]. These are (3) and (8) discussed here and explicit forms for X_{ii} and R_{ii}.

[11] The moments $\langle S_{ii}\rangle - S_{ii}(0)$ were found to be in excellent agreement with Monte Carlo simulations [*Darvini and Salandin*, 2006, Figure 11] developed following the approach previously used by *Salandin and Fiorotto* [1998]. The method described in the latter paper consisted in simulating the advective displacement of 40 particles, equally spaced by a heterogeneity scale, in 500 Monte Carlo runs. The statistics was evaluated "by averaging the single realization results on all the Monte Carlo runs" [*Salandin and Fiorotto*, 1998, pp. 953–954]. Finally, dispersion coefficients "were computed by the integration of the Lagrangian velocity covariance" [*Salandin and Fiorotto*, 1998, p. 958]. That means, first evaluating the velocity covariance on the particle trajectory $\mathbf{X}_i(t;\ \mathbf{a},\ t_0)$ in a given velocity realization (Monte Carlo run) by an average $\langle v_i v_i\rangle_{\mathbf{a}} - \langle v_i\rangle_{\mathbf{a}}\langle v_i\rangle_{\mathbf{a}}$ over all particles identified by their initial position (and possibly by a time average along particles trajectories, which for the sake of simplicity we do not consider here), then averaging over velocity realizations and integrating in time to obtain the moments

$$Y_{ii}(t,t_0) = \int_{t_0}^{t}\int_{t_0}^{t} \left[\langle\langle v_i(\mathbf{X}(t';\mathbf{a},t_0))v_i(\mathbf{X}(t'';\mathbf{a},t_0))\rangle_{\mathbf{a}}\rangle - \langle\langle v_i(\mathbf{X}(t';\mathbf{a},t_0))\rangle_{\mathbf{a}}\langle v_i(\mathbf{X}(t'';\mathbf{a},t_0))\rangle_{\mathbf{a}}\rangle\right] dt'dt''. \qquad (11)$$

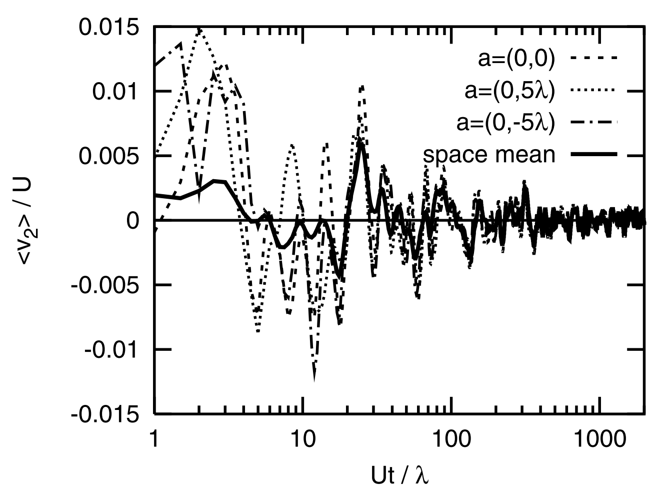

Figure 2. Ensemble average of the transverse velocity on the trajectories of the advection-dispersion processes starting from different initial positions **a**; the solid line represents the space average of $\langle v_2\rangle$ with respect to **a** for a transverse slab source with dimensions (λ, 10λ).

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Since the particle trajectory is given by

$$X_i(t;\mathbf{a},t_0) = a_i + \int_{t_0}^{t} v_i(\mathbf{X}(t';\mathbf{a},t_0))dt',$$

from (11) and (4) it follows that

$$\begin{aligned} Y_{ii} &= \langle\langle X_i^2\rangle_{\mathbf{a}}\rangle - \langle R_i^2\rangle - \left[\langle a_i^2\rangle_{\mathbf{a}} - \langle a_i\rangle_{\mathbf{a}}^2\right] - M_{ii} \\ &= \langle S_{ii}\rangle - S_{ii}(0) - M_{ii}. \end{aligned} \quad (12)$$

[12] The agreement of the first-order estimations with the Monte Carlo simulations suggests that the quantity presented in Figures 10–12 of *Darvini and Salandin* [2006] might be an estimation of Y_{ii}. With the first-order approximation (1)–(2), this estimation is obtained from (11) by replacing the argument $\mathbf{X}$ of v_i by $\mathbf{X}_0$. However, as follows from the definition of the uncertainty of the second moment with respect to the one-particle displacements covariance [*Darvini and Salandin*, 2006, p. 12], one can also suppose that the authors estimated $\langle X_{ii}\rangle_{\mathbf{a}} - R_{ii}$. This can be achieved by replacing in (11) v_i by v_i' and $\mathbf{X}$ by $\mathbf{X}_0$, as in relations (22) and (23) of *Darvini and Salandin* [2006]. From (12) and (6) we have $Y_{ii} = \langle X_{ii}\rangle_{\mathbf{a}} - R_{ii} + Q_{ii}$. Thus the estimation of the second moment by $\langle X_{ii}\rangle_{\mathbf{a}} - R_{ii}$ disregards not only the memory term M_{ii} (as the numerical approach based on (12) does) but also the correlation of the mean velocity Q_{ii} (accounted for by the numerical simulations). This choice is justified if a highly accurate numerical solution for the zeroth-order potential head [*Darvini and Salandin*, 2006, equation (6)] was obtained or if no numerical solution was computed at all and a constant head gradient, as resulting from boundary conditions, was imposed. This leads to a constant zeroth-order velocity and to the cancellation of both M_{ii} and Q_{ii} in (9)–(10). In this case, the comparison with the simulation results [*Darvini and Salandin*, 2006, Figure 11] indicates that the variance of the mean Lagrangian velocity near the left boundary, corresponding to the finite element solution of the exact flow equations in the Monte Carlo simulations, does not produce significant terms Q_{ii}.

[13] If, instead of following exactly the same approach as *Salandin and Fiorotto* [1998], the expected second moment was computed from the mean square displacement of the simulated particles trajectories, then the Monte Carlo results correspond to the exact relation (6). Hence, even if Q_{22} can be negligible, significant memory terms M_{22}, as in our example presented in Figure 1, can occur for the transverse line source placed near the left boundary. Figure 11 of *Darvini and Salandin* [2006] shows that this was not the case. To help the interested reader to understand their results, the authors should explain how the expected second moment was computed in both the first-order stochastic finite element approach and Monte Carlo simulations. In addition, we suggest that in a reply to this comment the authors of the paper commented here should present some results on the path line analysis of the mean Lagrangian velocity field. This can be achieved by evaluations of the correlation terms (9)–(10), which are readily obtainable by their approach, or, as in our example from Figure 2, by presenting the dependence of the numerically derived mean Lagrangian velocity on initial positions of the particles. If one proves that such dependencies can be neglected, the effect of spatial inhomogeneity of the random velocity field caused by the finite size of the domain, considered by *Darvini and Salandin* [2006], can be completely described in terms of one- and two-particle velocity covariances.

[14] For physically relevant statistical inhomogeneity of the velocity field, as that caused by a trend in the mean hydraulic conductivity, the bias of the expected second moment $\langle S_{ii}\rangle - S_{ii}(0)$ with respect to the space average of the one-particle variance $\langle X_{ii}\rangle_{\mathbf{a}}$ is given, according to (6), by $M_{ii} + Q_{ii} - R_{ii}$. Since in the first-order approximation M_{ii} (9) and Q_{ii} (10) quantify the effect of the space variation of the mean Eulerian velocity, these terms are particularly relevant for the inhomogeneous case. To complete the assessment of ergodicity with respect to $\langle X_{ii}\rangle_{\mathbf{a}}$, in addition to the bias of the expectation, the standard deviation of S_{ii} must be computed as well [*Suciu et al.*, 2006a]. Both tasks can be achieved with the first-order stochastic finite element method of *Darvini and Salandin* [2006]. By solving a deterministic problem for given mean and correlation functions of the hydraulic conductivity, such an approach avoids cumbersome repeated Monte Carlo computations.

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