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Multiple meanings of ergodicity in real life problems

Nicolae Suciu[*], **Călin Vamoş**[†] , and **Harry Vereecken** [‡]

Ergodic properties such as convergence of time averages, for dynamical systems and stochastic processes, convergence of space averages, for random fields, and self-averaging are often assumed in stochastic modeling of transport in heterogeneous media. However, no general theory has been provided so far which ensures the reliability of the stochastic model from ergodic assumptions. We propose instead an operational concept, "ergodicity in the large sense", which assesses the validity of the model by mean square deviations of actual observables from theoretical predictions. This approach is mainly useful for elaborated models such as the macrodispersion (up-scaled) process or the memory-free dispersion, which do not consist of predefined random functions. After a short review on ergodicity issue, we investigate numerically the ergodicity in the large sense with respect to a memory-free dispersion derived from an approximate solution of the Itô equation. We find that the ergodicity strongly depends on initial conditions and the transport shows significant memory-effects for large anisotropic supports of the initial concentration.

1. Introduction

The concept of "ergodicity" originates in statistical physics and is chiefly used in the mathematical theory of dynamical systems to denote the convergence of the time average of an observable to its space average with respect to an invariant measure associated to the dynamical system [*Cornfeld et al.*, 1982]. A dynamical system approach can be used to describe transport in heterogeneous media when

[*]Institute of Applied Mathematics, Friedrich-Alexander University Erlangen-Nürnberg, Germany, e-mail: suciu@am.uni-erlangen.de
[†]"T. Popoviciu" Institute of Numerical Analysis, Romanian Academy, e-mail: cvamos@ictp.acad.ro
[‡]Research Center Jülich, ICG-IV: Institute of Agrosphere, Germany, e-mail: h.vereecken@fz-juelich.de

diffusive mechanisms are neglected and if the velocity field has suitable smoothness and boundedness properties [*Sposito*, 1997]. A theoretical result, for a large class of laminar flows, is that nonergodicity of the flow implies a t^2 time behavior of the dispersion (defined by the mean square displacement of the solute particles) [*Mezić and Wiggins*, 1994]. It follows that for such laminar flows there is no normal diffusive behavior (i.e. linear dispersion). When a local diffusion-like process is considered, the situation changes and diffusive large time behavior of the dispersion can take place. For advection dominated transport, the nonergodicity of the advective flow implies a Pe^2 dependence of the effective dispersion on Péclet number [*Mezić and Wiggins*, 1994]. For instance, steady groundwater flows governed by Darcy law were shown to be in general nonergodic, a property which renders questionable the "ergodicity conditions" under which Lagrangian approaches to purely advective transport are developed [*Sposito*, 1997, 2001]. The ergodicity is forbidden in this case by the existence of invariant sub-sets smaller than the flow domain, on which the trajectories of the solute particles are confined (a property which is common to flows proportional to the gradient of a scalar function, structurally equivalent to Hamiltonian flows). However, ergodicity can be expected for unsteady Darcy flows [Sposito, 2006].

Another often mentioned meaning of "ergodicity" is the convergence of the space average of an observable, defined for a realization of a random space function, to its ensemble average. The existence of a finite integral (or correlation) range implies the ergodicity of the spatial mean velocity (and in the case of Gaussian fields also that of the spatial velocity correlations) [*Chilès and Delfiner*, 1999]. A practical criterion for ergodic estimations through space averages is to ensure that the problem spatial dimensions are much larger than the integral range. It is reasonable to assume that the ergodicity of the velocity implies an "ergodic behavior of the transport process". In this larger sense, ergodicity means that observables of interest in single realizations behave closely to their ensemble average and converge for large times to predictions provided by theoretical models (e.g. an up-scaled Gaussian diffusion called "macrodispersion" process [*Dagan*, 1984]). Even though assuming the ergodicity of the random velocity field could be a starting point for theoretical investigations, the large-sense ergodicity of the transport is not straightforward and it seems that no general proofs of this issue have been provided so far [*Sposito et al.*, 1986; *Kabala and Sposito*, 1994]. Some advances have been achieved by numerical simulations which indicate that transport in velocity fields with finite correlation range is asymptotically ergodic in the large sense [*Suciu et al.,* 2006a]. However, for real life problems the ergodicity timescale is often too large to permit contamination risk assessments based only on the asymptotic ergodicity of the transport.

For the pre-asymptotic regime of transport it is even more intricate to assess the reliability of the stochastic modeling. Since the ensemble average predictor of the dispersive flux is non-local in space-time and non-Fickian, the macrodispersion concept makes no sense for small and intermediate times. Even if localized forms can be derived under some restrictive conditions, the localized dispersion coefficients still depend on space and time [*Morales-Casique et al.*, 2006]. Moreover, since the velocity field is highly variable and actually known only in a mean (statistical) sense, the transport equations cannot be in general solved to provide a complete description of the transport for given velocity realizations. Some information about the behavior of the transport is supplied by the second spatial central moments of the plume (or by their rate of increase with time, which defines dispersion coefficients). Such dispersion quantities can be derived by different approximation techniques, without solving the transport equations and regardless the Fickian or non-Fickian behavior of the process [*Suciu et al.*, 2006b]. Our objective is to show that the ergodicity issue for pre-asymptotic regime can be formulated with respect to "memory-free" quantities provided by stochastic models and to apply this approach for contaminant transport in groundwater.

The paper is organized as follows. In the next section several notions which are generically referred to as ergodic properties are discussed from the point of view of their relation with the reliability of the stochastic modeling. Section 3 introduces a memory-free dispersion which can be used to assess the ergodicity of the transport in the pre-asymptotic regime. The case of transport in saturated aquifers is investigated numerically in Section 4. Some conclusions are drawn in Section 5.

2. Ergodicity concepts

A dynamical system $S_t : M \longmapsto M$ in a measure space $(M, \mathcal{A}, \mu)$, where M is a state space endowed with a σ-algebra $\mathcal{A}$ and a measure μ, is measure preserving if $\mu(S_t^{-1}(A)) = \mu(A)$ for all $A \in \mathcal{A}$. Ergodic theorems state **the convergence of the time average** of an observable f:

$$\lim_{T \longrightarrow \infty} \frac{1}{T} \int_0^T f(S_t(x)) \mathrm{d}t = \frac{1}{\mu(M)} \int f(x) \mu(\mathrm{d}x) = f^* = \text{const.}$$

For ergodic systems the only invariant sets, i.e. $S_t(A) = A$, are sets of null measure, $\mu(A) = 0$, or sets with the measure equal to that of the state space, $\mu(A) = \mu(M)$. An equivalent formulation uses the evolution operator U in $L^p(M, \mu)$, $p \geq 1$, defined by $(U_t f)(x) = f(S_t(x))$. Birkhoff's theorem states the convergence in L^1 and von Neumann's theorem the convergence in L^2. When ergodicity holds, the fix points f of the evolution operator, $U_t f = f$, are constant functions [*Sinai*, 1976]. The physically relevant meaning of ergodicity is that the orbit of the ergodic dynamical system passes through almost all possible states. In particular, written for the indicator function of the set A, $f(x) = \chi_A(x)$,

$$\lim_{T \longrightarrow \infty} \frac{1}{T} \int_0^T \chi_A(S_t(x)) \mathrm{d}t = \frac{1}{\mu(M)} \int \chi_A(x) \mu(\mathrm{d}x) = \frac{\mu(A)}{\mu(M)},$$

the ergodicity allows us to define probability densities $p(x)$, $\int_A p(x)dx = P(A) = \mu(A)/\mu(M)$, and to arrive at a statistical description of the physical system. Intuitively, that means to associate a "stochastic model" (ensemble average) to a measurement modeled by a time average [*Cornfeld et. al.*, 1982].

The extension to stochastic processes says that **time and ensemble averages are interchangeable** [*Kloeden and Platen*, 1995]. A strong ergodicity condition is given by the mean-square convergence of the time average to the average with respect to the probability measure $p(x)dx$ of the process X_t [*Gardiner*, 1985],

$$m.s. \lim_{T\longrightarrow\infty} \frac{1}{T}\int_0^T f(X_t(x))\mathrm{d}t = \int f(x)p(x)\mathrm{d}x = \langle f(x)\rangle \,.$$

Ergodic theorems for dynamical systems are retrieved from the above property for "degenerate Markov processes" [*Suciu*, 2001].

As a property of random space functions $F(x)$ (random fields), ergodicity states that **space and ensemble averages are interchangeable**,

$$\lim_{\mathcal{V}(\Omega)\longrightarrow\infty} \frac{1}{\mathcal{V}(\Omega)}\int_\Omega F(x)\mathrm{d}x = \langle F(x)\rangle \,,$$

where $\mathcal{V}(\Omega)$ is the volume of the domain $\Omega \subset \mathbb{R}^3$. The L^2-convergence is ensured by Slutsky's condition of finite integral of the correlation function $\langle F(x)F(x+y)\rangle$, which, in turn, is ensured by the existence of a finite correlation range $\int\langle F(x)F(x+y)\rangle dy/\langle F(x)^2\rangle$ of the random function [*Chilès and Delfiner*, 1999]. In this case, by virtue of ergodicity one associates an ensemble average to a measurement described by a space average.

A strong property of transport in random environments, formulated for stochastic processes (or sequences), is the "**self-averaging**". That means the convergence of the un-averaged function of the trajectory of the process $f(X_t(x))$ to its ensemble average with respect to the probability measure of the process [*Bouchaud and Georges*, 1990],

$$\lim_{t\longrightarrow\infty} f(X_t(x)) = \int f(x)p(x)\mathrm{d}x = \langle f(x)\rangle \,.$$

A sufficient condition for self-averaging is a vanishing variance $\langle[f-\langle f\rangle]^2\rangle$ of the observable f in the large time limit.

The ergodic properties described above have in common the following practical meaning. They express the reliability of the probability measures inferred from experiments [*Gardiner*, 1985; *Chilès and Delfiner*, 1999] for measurements and observations. The latter are modeled by random variables (dynamical systems included as particular cases) or by time/space averages of random variables. Therefore ergodicity concepts prove their utility in stochastic modeling of transport processes. Let us consider the purely advective transport described by the advection equation for the concentration field $c(\mathbf{x},t)$,

$$\partial_t c + \mathbf{V}(\mathbf{x})\nabla c = 0. \tag{1}$$

This equation is also the Liouville equation for the dynamical system $\mathbf{S}_t(\mathbf{x}) = \mathbf{X}(t;\mathbf{x},0)$, where $\mathrm{d}\mathbf{X}(t)/\mathrm{d}t = \mathbf{V}(\mathbf{X}(t))$. *Mezić and Wiggins* [1994] have shown that if the velocity is periodic or quasiperiodic in the l-direction, $l = 1$, 2,or 3, then the dispersion in that direction, $\langle[X_l(t)-\langle X_l(t)\rangle]^2\rangle$, may increase linearly in time (Fickian behavior) only if the dynamical system $\mathbf{S}_t$ is ergodic. It was also proved that the nonergodicity of $\mathbf{S}_t$ is a necessary and sufficient condition for a t^2 behavior of the dispersion. A situation where the ergodicity is vitiated is the existence of invariant sets of the dynamical system that are subsets of the state space. This is the case when the velocity is the gradient of a scalar function (Darcy law, Hamiltonian systems) and the trajectories $\mathbf{X}(t)$ are confined on "Lamb surfaces" [*Sposito*, 1997, 2001]. Then the dynamical system does not explore the entire phase space and ergodicity fails, i.e.

$$\lim_{T\longrightarrow\infty}\frac{1}{T}\int_0^T \mathbf{S}_t(\mathbf{x})\mathrm{d}t \neq \langle\mathbf{x}\rangle \,.$$

Nevertheless, a stochastic approach to advective transport leads to large time Fickian behavior under the requirements that the velocity $\mathbf{V}(\mathbf{x})$ is an homogeneous random field with non-vanishing mean $\langle\mathbf{V}(\mathbf{x})\rangle = \mathbf{U}$ which has small fluctuations with suitable strong-mixing properties [*Kesten and Papanicoulaou*, 1979]. In these conditions there exists an up-scaling, $\langle c\rangle \rightarrow c^*$, of the mean concentration to a Gaussian process described by an advection-diffusion equation with constant coefficients $\mathbf{U}$ and $\mathbf{D}$,

$$\partial_t c^* + \mathbf{U}\nabla c^* = \mathbf{D}\nabla^2 c^*. \tag{2}$$

The question arises, and it is a central one in stochastic modeling, how much is the model (2) relevant for the actual realization of the transport described by (1). Is the transport "ergodic" in the sense that c or some "coarse-grained" concentration tends to c^*? The versions of the ergodicity discussed above (convergence of time or space averages and self-averaging) do not cover this eventuality because they only refer to convergence to the ensemble average of a predefined random function. Here we have to deal with relations between actual observables, c, ensemble averages, $\langle c\rangle$, and up-scaled quantities, c^* (and the same for other observables as dispersion or dispersion coefficients). This situation requires the definition of a new ergodic property, which we call hereafter "ergodicity in the large sense".

Let $A(t)$ be an observable, $A^*(t)$ the theoretical prediction and $\Delta_A = \langle[A - A^*]^2\rangle^{1/2}$ the root mean square deviation from theory.

DEFINITION 1: **The observable A is ergodic within the range η, $\eta > 0$, if $\Delta_A \leq \eta$.**

When A^* is an asymptotic limit and A is known at finite times it is convenient to use the relation $\Delta_A^2 = \sigma_A^2 + \Delta_{\langle A\rangle}^2$, where $\sigma_A = \langle[A - \langle A\rangle]^2\rangle^{1/2}$ is the standard deviation of A and $\Delta_{\langle A\rangle} = |\langle A\rangle - A^*|$ the deviation of the mean. With this, one obtains the following equivalent definition.

DEFINITION 2: **The observable A is ergodic within the range $\eta = (\eta_1^2 + \eta_2^2)^{1/2}$ if**

$$\text{(e1) } \Delta_{\langle A\rangle} \leq \eta_1, \text{ (e2) } \sigma_A \leq \eta_2. \tag{3}$$

This definition was proposed by *Suciu et al.* [2006a] and was used in investigations on advective-dispersive transport in saturated aquifers. It was shown in the cited paper that previously used notions of "ergodicity" can be obtained as particular cases of (3):

- The strong requirement for the reliability of the up-scaled model (2) formulated by *Sposito et al.* [1986] corresponds to the case where A is the concentration and (e1, e2) hold for $t \longrightarrow \infty$ and arbitrary small and positive η_1 and η_2.

- When the observable A is a space averaged concentration and (e1, e2) hold at finite times for large averaging domains, then A converges in the mean square limit to c^* and the space and ensemble averages are interchangeable. *Dagan* [1984] used (e2) under the assumption that the ensemble averaged concentration $\langle c \rangle$ is already close to the Gaussian concentration c^* at finite times.
- The self-averaging property of the effective coefficients considered in [*Clincy and Kinzelbach*, 2001; *Eberhard*, 2004] is given by the condition (e2) alone, for $t \longrightarrow \infty$.
- The condition (e1) applied to ensemble averaged dispersion (or dispersion coefficients) is often referred to as "ergodicity condition" [*Fiori*, 1998; *Naff et al.*, 1998].
- *Kabala and Sposito* [1994] defined an "operational ergodicity" which seeks conditions that lead to acceptably small deviations of the experimentally observable concentrations from the predictions of the stochastic model. This corresponds to (e1, e2), for finite times and ranges η.

3. Memory-free dispersion

We consider the advection-dispersion equation for the concentration field $c(\mathbf{x}, t)$,

$$\partial_t c + \mathbf{V}(\mathbf{x})\nabla c = D\nabla^2 c. \tag{4}$$

The constant D describes a "local dispersion" which is produced by molecular diffusion and small scale hydrodynamic mixing. The velocity $\mathbf{V}(\mathbf{x})$ is a realization of a statistically homogeneous random space function with constant mean $\langle \mathbf{V}(\mathbf{x}) \rangle = \mathbf{U}$. For instance, this model corresponds to turbulent diffusion in the atmosphere or to non-reactive transport in natural porous media with constant or slowly variable porosity. An equivalent description of the transport problem formulated for the partial derivative equation (4) is given by the solutions of the Itô equation [*Gardiner*, 1985]. The trajectories starting at $t = 0$ from $\mathbf{X}_0$ of the advection-dispersion process with probability densities obeying (4) are solutions of the integral Itô equation

$$X_l(t) = X_{0l} + \int_0^t V_l(\mathbf{X}(t'))\mathrm{d}t' + \int_0^t \mathrm{d}W_l(t'). \tag{5}$$

The Wiener process $W_l(t) = \int_0^t \mathrm{d}W_l(t')$ is defined by the Itô stochastic integral which has the properties [*Kloeden and Platen*, 1995, pp. 85, 190]

$$\langle W_l \rangle_w = 0, \ \langle W_l \rangle_w^2 = 2Dt, \ \left\langle \int_0^t \int_0^t V_l(\mathbf{X}(t'))\mathrm{d}t' \mathrm{d}W_l(t'') \right\rangle_w = 0, \tag{6}$$

where $\langle \cdots \rangle_w$ is the average with respect to the probability measure of the Wiener process. The joint n-times probabilities are related with the trajectories of the advection-dispersion process by the definition [*van Kampen*, 1981]

$$p(\mathbf{x}_1, t_1; \mathbf{x}_2, t_2; \cdots ; \mathbf{x}_n, t_n) =$$

$$\langle\delta[(\mathbf{x}_1-\mathbf{X}(t_1)]\delta[(\mathbf{x}_2-\mathbf{X}(t_2)]\cdots\delta[(\mathbf{x}_n-\mathbf{X}(t_n)]\rangle_{wX_0},$$

where the subscripts wX_0 denote the average over trajectories and initial positions. In particular, the normalized concentration is defined by $c(\mathbf{x},t)=p(\mathbf{x},t)$ and is a solution of (4).

From (7) it follows that the dispersion $s_{ll}(t)=\langle[X_l(t)-\langle X_l(t)\rangle_{wX_0}]^2\rangle_{wX_0}$ coincides with the second moment of the concentration with respect to the center of mass of the solute plume,

$$s_{ll}(t)=\int_{\mathbb{R}^3}[\mathbf{x}-\bar{\mathbf{x}}(t)]^2\,c(\mathbf{x},t)\mathrm{d}\mathbf{x},\text{ where }\bar{\mathbf{x}}(t)=\int_{\mathbb{R}^3}\mathbf{x}c(\mathbf{x},t)\mathrm{d}\mathbf{x}.$$

Hereafter we make no distinction between dispersion and second moment.

Assuming the necessary smoothness and boundedness conditions for the realizations of $\mathbf{V}$ which ensure the existence for the solutions of the Itô equation [*Gardiner*, 1985, section 4.3.1] and using (5) and (6) one obtains the dispersion

$$\begin{aligned}s_{ll}(t)\quad&=\left\langle[X_{0l}-\langle X_{0l}\rangle_{X_0}]^2\right\rangle_{X_0}+2Dt+\int_0^t\int_0^t\langle V_l(\mathbf{X}(t'))V_l(\mathbf{X}(t''))\rangle_{wX_0}\,\mathrm{d}t'\mathrm{d}t''\\&+2\int_0^t\left\langle[X_{0l}-\langle X_{0l}\rangle_{X_0}]V_l(\mathbf{X}(t'))\right\rangle_{wX_0}\mathrm{d}t'\\&-\int_0^t\int_0^t\langle V_l(\mathbf{X}(t'))\rangle_{wX_0}\langle V_l(\mathbf{X}(t''))\rangle_{wX_0}\,\mathrm{d}t'\mathrm{d}t''.\end{aligned}$$

The relation (8) expresses the dispersion as a sum between the initial dispersion $S(0)=\langle[X_{0l}-\langle X_{0l}\rangle_{X_0}]^2\rangle_{X_0}$, the local dispersion $2Dt$, and contributions due to velocity correlations, to spatial correlations between initial positions and velocity on trajectories and to fluctuations of the mean velocity on trajectories.

The substitution into (8) of $V_l(\mathbf{x})=u_l(\mathbf{x})+U_l$, where U_l is the constant mean velocity, yields a relation of the same form, with V_l replaced by the velocity fluctuation u_l. By collecting the second and third term of this relations one obtains the dispersion $x_{ll}(t)$ of the trajectories (5) with respect to the trajectory of the mean velocity $X^{(0)}(t)=X_0+U_lt$,

$$\begin{aligned}x_{ll}(t)\quad&=\left\langle[X_l(t)-X^{(0)}(t)]^2\right\rangle_{wX_0}\\&=2Dt+\int_0^t\int_0^t\langle u_l(\mathbf{X}(t'))u_l(\mathbf{X}(t''))\rangle_{wX_0}\,\mathrm{d}t'\mathrm{d}t''.\end{aligned}\qquad(7)$$

This relation will be used in the following to derive a memory-free quantity, depending on the physical parameters of the system but not on the initial conditions.

Using (7) and the projection of the Eulerian velocity fluctuations $u_l(\mathbf{x})$ on the trajectories of the advection-dispersion process, $u_l(\mathbf{X}(t))=\int u_l(\mathbf{x})\delta[\mathbf{x}-\mathbf{X}(t)]\mathrm{d}\mathbf{x}$, the integrand of the double integral in (9) becomes

$$\begin{aligned}
&\langle u_l(\mathbf{X}(t';\omega);\omega)u_l(\mathbf{X}(t'';\omega);\omega)\rangle_{wX_0}\\
&=\int\int u_l(\mathbf{x}';\omega)u_l(\mathbf{x}'';\omega)\langle\delta[\mathbf{x}'-\mathbf{X}(t';\omega)]\delta[\mathbf{x}''-\mathbf{X}(t'';\omega)]\rangle_{wX_0}\mathrm{d}\mathbf{x}'\mathrm{d}\mathbf{x}''\\
&=\int\int u_l(\mathbf{x}';\omega)u_l(\mathbf{x}'';\omega)p(\mathbf{x}',t';\mathbf{x}'',t'';\omega)\mathrm{d}\mathbf{x}'\mathrm{d}\mathbf{x}''\\
&=\int c(\mathbf{x}_0)\mathrm{d}\mathbf{x}_0\int\int u_l(\mathbf{x}';\omega)u_l(\mathbf{x}'';\omega)p(\mathbf{x}',t';\mathbf{x}'',t''|\mathbf{x}_0;\omega)\mathrm{d}\mathbf{x}'\mathrm{d}\mathbf{x}'',
\end{aligned}$$

The argument ω is used in (10) to show the dependence on the given realization of the velocity field. The last equality in (10) follows from the consistency condition which gives the two-times joint probability density $p(\mathbf{x}',t';\mathbf{x}'',t'';\omega)$ as an integral of the conditional probability density $p(\mathbf{x}',t';\mathbf{x}'',t''|\mathbf{x}_0;\omega)$ with respect to the initial normalized concentration distribution $c(\mathbf{x}_0)\mathrm{d}\mathbf{x}_0$ [*Gardiner*, 1985]. Similar expressions can be derived as well for the integrals in the last two terms of (8).

Due to the highly non-linear dependence of probability densities p on velocity fluctuations $u_l(\mathbf{X}(t;\omega);\omega)$, the ensemble average of (10) can be computed exactly only in some particular cases, as for instance the problem of transport in perfectly stratified flows [see e. g. *Clincy and Kinzelbach*, 2001]. In principle, to deal with such a nonlinearity one iterates indefinitely the transport equation around an unperturbed solution independent of velocity realization [*Bouchaud and Georges*, 1990]. However, since iterations of order larger than one become very involved, the mostly used are first-order approximations in velocity fluctuations equivalent to the approximation of the solution $\mathbf{X}(t;\omega)$ by the first iteration of the Itô equation (5) [*Suciu et al.*, 2006b],

$$X_l(t;\omega)\approx X_l^{(1)}(t;\omega)=X_{0l}+\int_0^t V_l(\mathbf{X}^{(0)}(t');\omega)\mathrm{d}t'+\int_0^t \mathrm{d}W_l(t'), \tag{8}$$

where the unperturbed solution $\mathbf{X}^{(0)}$ is either the mean trajectory $\mathbf{X}_0+\mathbf{U}t$ (as in relation (9) above) or the trajectory of the diffusion process of coefficient D and constant drift $\mathbf{U}$. From (11) it follows that the argument of u_l in (9) has to be replaced by $\mathbf{X}^{(0)}$, which is independent of the realization ω of the velocity field. Consequently, the conditional probability density in (10) also becomes independent of ω. Here we consider only the unperturbed solution given by the trajectory of the ensemble mean advective velocity. This yields consistent expansions of the Itô equation (5) for the advection-dominated transport problem considered in the simulations presented in the next section, where the velocity fluctuations are of the order of $Pe^{-1/2}$ [*Suciu et al.*, 2006b]. Since in this case $\mathbf{X}^{(0)}$ no longer depends of the realizations of the Wiener process, the conditional probability p in (10) degenerates to a Dirac delta function, $p(\mathbf{x}',t';\mathbf{x}'',t''|\mathbf{x}_0)=\delta[\mathbf{x}'-(\mathbf{x}_0+\mathbf{U}t')]\delta[\mathbf{x}''-(\mathbf{x}_0+\mathbf{U}t'')]$. With these, the ensemble average $X_{ll}=\langle x_{ll}\rangle$ of (9) yields

$$X_{ll}(t)=2Dt+\int_0^t\int_0^t \mathrm{d}t'\mathrm{d}t''\int u_{ll}(\mathbf{x}_0+\mathbf{U}t',\mathbf{x}_0+\mathbf{U}t'')c(\mathbf{x}_0)\mathrm{d}\mathbf{x}_0, \tag{9}$$

where

$$u_{ll}(\mathbf{x}', \mathbf{x}'') = \int\int u_l' u_l'' p_u(\mathbf{u}', \mathbf{x}'; \mathbf{u}'', \mathbf{x}'')\mathrm{d}\mathbf{u}'\mathrm{d}\mathbf{u}''$$

is the Eulerian velocity correlation and $p_u(\mathbf{u}', \mathbf{x}'; \mathbf{u}'', \mathbf{x}'')$ is the joint two-point probability density of the velocity fluctuations. Since the velocity field is statistically homogeneous, the density p_u is invariant to translations and the correlation u_{ll} in (12) depends only on $\mathbf{U}(t' - t'')$ and does not depend on $\mathbf{x}_0$. It follows that the first-order approximation of the dispersion with respect to the mean trajectory is independent of initial conditions and is identical to X_{ll} computed for a singular initial concentration distribution localized at the origin of the coordinate system, $c(\mathbf{x}_0) = \delta(\mathbf{x}_0)$. Therefore, hereafter X_{ll} will be referred to as "memory-free" dispersion.

The result (12) is identical with the well known expression of *Dagan* [1984], which was found to be a very robust approximation. For instance, it was shown that (12) is practically identical with the approximation derived from the iteration (11) with respect to the solution of diffusion in the mean velocity field $\mathbf{U}$ [*Suciu et al.*, 2006b, Figure 2]. Nevertheless, it should be emphasized that $X_{ll}(t)$ derived by first-order approximations is different from the dispersion with respect to the ensemble averaged center of mass. The latter is obtained from (8) if the velocity fluctuations are defined with respect to the ensemble average of the velocity on trajectories, $u_l(\mathbf{X}(t), t) = V_l(\mathbf{X}(t)) - \langle\langle V_l(\mathbf{X}(t))\rangle_{wX_0}\rangle$, and not by the simple projection on the trajectories of the Eulerian velocity fluctuations with respect to the constant mean U_l. This approach has been followed in (N. Suciu et al., manuscript submitted to Water Resources Research, 2006) to derive numerically the memory-free component of the dispersion and to quantify the "memory effects" produced by the term of (8) accounting for correlations between initial positions and velocity on trajectories. It was found that the approximation (12) is close, uniformly in time, to the numerically derived memory-free dispersion in a range of the order of the local dispersion $2Dt$. Moreover, for the order of magnitude relation between velocity fluctuations and Pe considered here, the large time limit of $X_{ll}(t) - 2Dt$ corresponds to the up-scaled dispersion predicted by *Kesten and Papanicoulaou* [1979], which is in excellent agreement with numerical simulations [*Suciu et al.*, 2006a]. These arguments recommend the approximate memory-free moment (12) as an useful reference for investigations on ergodicity of the pre-asymptotic regime transport.

4. Numerical results

We consider isotropic two-dimensional diffusion ($D_1 = D_2 = D = 0.01$ m^2/day) in a groundwater flow modeled by a random velocity field $\mathbf{V}$ with ensemble mean $\mathbf{U} = (U, 0)$, $U = 1$ m/day. The velocity field is generated, with the Kraichnan routine, as a superposition of 6400 random sin modes which approximates a Gaussian field. This Gaussian field is an approximation of a time stationary Darcy velocity field in saturated groundwater formations, for log-hydraulic conductivity with variance of 0.1, exponentially correlated, and with finite isotropic correlation length $\lambda = 1$ m. The transport over 2 000 days in given realizations of the velocity field, for point instantaneous injection conditions and uniform distributions in rectangles $(L_1\lambda, L_2\lambda)$, is simulated by simultaneously tracking $N = 10^{10}$ computational particles with the "global random walk" (GRW) algorithm [*Vamoş et al.*, 2003]. Details on the implementation of the numerical method can be found in [*Suciu et al.*, 2006a]. In that paper ergodicity with respect to the up-scaled "macrodispersion" process of *Dagan* [1984] was investigated for transverse slab sources $(\lambda, L\lambda)$. The main result was a numerical evidence of asymptotic ergodicity.

We present here, within a more suggestive graphical representation, the ergodicity range for the cross-section space averaged concentration evaluated at the plume center of mass $\langle x_1 \rangle$,

$$C(\langle x_1 \rangle, t) = \frac{1}{B} \int_0^B c(\langle x_1 \rangle, x_2, t) \mathrm{d}x_2,$$

where B is the transverse dimension of the grid and $c(x_1, x_2, t)$ is the GRW simulated concentration. We evaluate the ergodicity range, according to Definition 2, by $\eta = \left(\eta_1^2 + \eta_2^2\right)^{1/2}$, where $\eta_1 = |\langle C \rangle - C^*|$ is the deviation of the average of C over the ensemble of 256 velocity realizations from the corresponding up-scaled value C^* and $\eta_2 = \sigma_c$ is the standard deviation of C^* (numerical data from [*Suciu et al.*, 2006a]). The results for point source and three different transverse sources $(\lambda, L\lambda)$, presented in Figure 1, indicate a general decreasing trend of the ergodicity range η, thus an asymptotically ergodic behavior with respect to the up-scaled process. It is also noticeable that though the increase of transverse dimension L of the source causes a decrease of η at early time, the ergodicity range reaches a plateau value which persists over large dimensionless times. Since the simulated velocity field approximates a stationary Darcy flow, the associated dynamical system is very likely nonergodic, as indicated by theoretical results [*Sposito*, 1997, 2001]. To round off the issue of ergodicity in the large sense of the advective transport described by equation (1), formulated in Section 2, we also simulated for a transverse source $(\lambda, 100\lambda)$ the case $Pe = \infty$ by dropping the local dispersive step in our GRW algorithm. (The simulations were interrupted after about $700\, Ut/\lambda$, when numerical trapping phenomena occurred.) In spite of the existence of the macrodispersive behavior described by equation (2), the results presented in Figure 2 clearly prove the nonergodicity in the large sense of the cross-section space averaged concentration with respect to the up-scaled concentration C^*.

As shown by Figure 1, the timescale of asymptotic ergodicity could be impractical for predictions at finite times. We investigated therefore the ergodicity with respect to the memory free dispersion (12) predicted by the approximate stochastic model. To do that, we evaluated numerically the Eulerian correlation and the dispersion $X_{ll}(t)$ (12). The correlation was computed by averages over 512 realizations of the numerical velocity field. Further, an "ergodic" dispesion $X_{ll}^{erg}(t)$ was defined by a mean square fitting of $X_{ll}(t)$. The results, normalized by the local dispersion $2Dt$, are given in Figure 3.

Using the ergodic dispersion from Figure 3 as a reference, the ergodicity range was computed according to Definition 1 from actual dispersions s_{ll} given by 1024

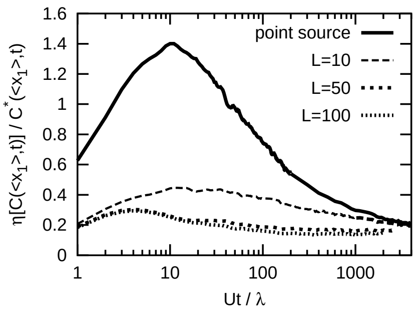

Fig. 1. Concentration ergodicity ranges for different initial conditions.

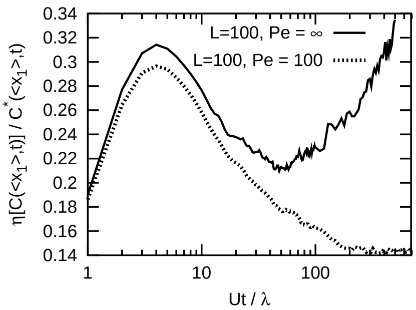

Fig. 2. Concentration ergodicity ranges for $Pe = \infty$ and $Pe = 100$.

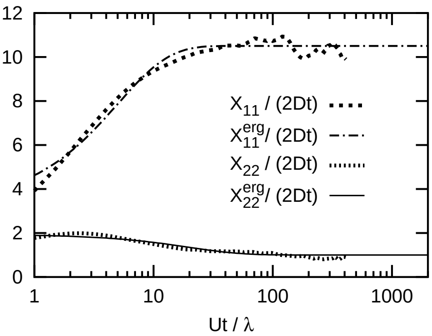

Fig. 3. Ergodic dispersion.

GRW simulations, for point sources, longitudinal and transverse slabs, and square sources, by the formula

$$\eta\left[s_{ll}(t) - S_{ll}(0)\right] = \left\langle \left[s_{ll}(t) - S_{ll}(0) - X_{ll}^{erg}(t)\right]^2 \right\rangle^{1/2} .$$

The results presented in Figure 4 indicate an ergodic behavior at early times of the actual dispersion in the l-direction with respect to the memory-free dispersion for large slab sources perpendicular to l. But for large extensions of the source in the l-direction s_{ll} shows a memory effect indicated by large ergodicity ranges η.

5. Conclusions

The "ergodicity in the large sense" is an operational concept which quantifies the departure of observables in actual realizations of the physical system from predictions provided by stochastic modeling of transport in heterogeneous media. We illustrated it in this paper by investigations on asymptotic ergodicity (with respect

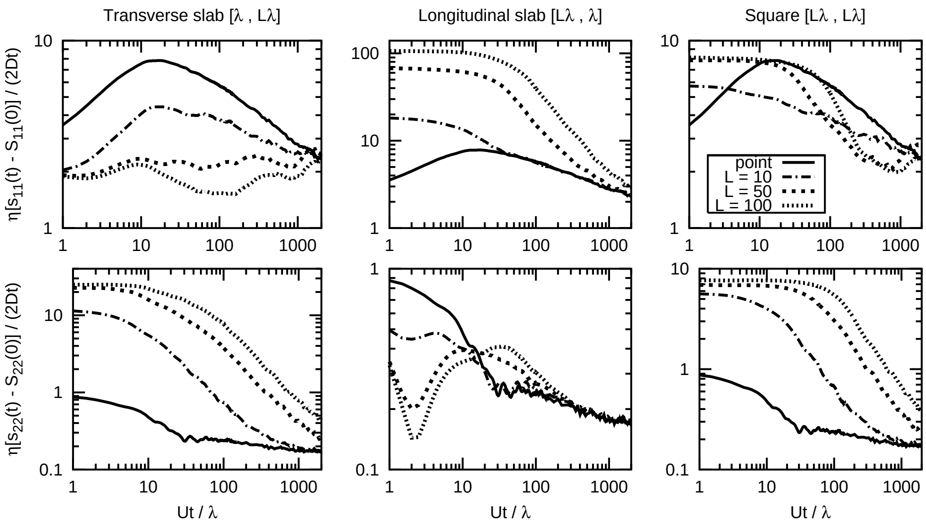

Fig. 4. Ergodicity ranges of dispersion for different initial conditions.

to the up-scaled macrodispersive limit behavior) and on ergodicity with respect to memory-free dispersion. The latter is characteristic to a given environment and depends only on local dispersion coefficient and velocity correlation function. Since it can be evaluated at finite times by analytical or numerical approaches, the memory-free dispersion is a reference for the actual dispersion which can be used to assess the ergodicity of the pre-asymptotic regime.

Though no rigorous mathematical results are available, large sense ergodicity can be related to the notions of ergodicity as formulated for dynamical systems, and random functions. For instance, the numerical results of Figure 2 indicate that advective transport by nonergodic dynamical systems is also nonergodic in the large sense. This happens although the associated stochastic model admits a diffusive up-scaling and in spite of the ergodicity of the homogeneous space random function (with finite correlation length) which describes the velocity field. The situation changes completely if the stochastic model considers a diffusion-like mechanism. In this case the transport behaves asymptotically ergodic with respect to the up-scaled process (Figure 1) and, at finite times, an ergodic behavior with respect to the memory-free dispersion can also be expected (Figure 4).

The reliability of theoretical memory-free dispersion for transport originating from large initial plumes seems to be the mostly used sense of ergodicity in applications for contaminant hydrology. The ergodicity ranges presented in Figure 4 clearly indicate that the memory-free dispersion provides useful predictions for the actual dispersion in a given direction only for anisotropic sources with large dimensions on the perpendicular direction. But the dispersion in the direction of the anisotropy can be strongly nonergodic and affected by persistent memory effects. This relation between the ergodicity range and the anisotropy of the initial condition can be used to identify the source of contamination from comparisons of measured and theoretical dispersions.

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