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Dispersion of a passive solute in non-ergodic transport by steady velocity fields in heterogeneous formations

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An inert solute is convected by a steady random velocity field, which is associated with flow through a heterogeneous porous formation. The log conductivity and the velocity are stationary random space functions. The log conductivity Y is assumed to be normal, with an isotropic two-point correlation of variance σ_Y^2 and of finite integral scale I. The solute cloud is of a finite input zone of lengthscale l. The transport is characterized with the aid of the spatial moments of the solute body. The effective dispersion coefficient is defined as half of the rate of change with time of the second spatial moment with respect to the centroid. Under the ergodic hypothesis, which is bound to be satisfied for $l/I \gg 1$, the centroid moves with the mean velocity U and the longitudinal dispersion coefficient $\mathscr{D}_L$ tends to its constant, Fickian, limit. Under a Lagrangian first-order analysis in σ_Y^2 it has been found that $\mathscr{D}_L = \sigma_Y^2 UI$.

This study addresses the computation of the effective longitudinal dispersion coefficient for a finite input zone, for which ergodic conditions may not be satisfied. In this case the centroid trajectory and the second spatial moments are random variables. In line with a previous work (Dagan 1990) the effective dispersion coefficient D_L is defined as half the rate of change of the expected value of the second spatial moment for large transport time. The aim of the study is to derive D_L and its dependence upon l/I and in particular to determine the conditions under which it tends to the ergodic limit $\mathscr{D}_L$. The computation is carried out separately for a thin body aligned with the mean flow and one transverse to it. In the first case it is found that D_L is equal to zero, i.e. the streamlined body does not disperse in the mean. This result is explained by the correlation between the trajectories of the leading and trailing edges, respectively, once the latter reaches the position of the first. The relatively modest increase of the mean second spatial moment is effectively computed. In the case of a thin body initially transverse to the mean flow, D_L may reach the ergodic limit $\mathscr{D}_L$ for a ratio l/I of the order 10^2. For smaller values, D_L is found to be bounded from above, and its maximum depends on l but not on I. The uncertainty caused by the randomness of the velocity field is manifested in the trajectory of the centroid rather than in the effective dispersion.

1. Introduction

The mechanism of spreading of a solute carried by a fluid in natural porous formations is dominated by the large-scale heterogeneity of the formation properties. Field studies have shown that the hydraulic conductivity (permeability) K varies in an erratic manner in space and its scale of variation, related to the geological non-uniform setting, is much larger than the pore scale. Hence, at this scale the fluid motion can be averaged over the pore scale and the continuous velocity field satisfies Darcy's law and the mass conservation equation. Solute particles, large compared to the pore scale but small at the heterogeneity scale, are convected by the Darcian velocity field and diffuse due to the effect of pore-scale structure. The effect of the latter mechanism, coined 'hydrodynamic dispersion' in the literature, is generally small compared to that of large-scale K variations and can be neglected altogether. There are exceptional cases, like perfectly stratified formations and flow parallel to the bedding (see Matheron & de Marsily 1980 or for generalizations, Bouchaud *et al.* 1990) in which pore-scale dispersion plays an important role, but they are not of our concern here. Recent field experiments (e.g. Sudicky 1986; Garabedian 1987) confirm this picture: the spatial distribution of the solute concentration C is irregular, it precludes a deterministic description and the scale of spatial variations is quite large. The plumes can at best be characterized by some global measures, the most common being the spatial moments of the cloud (mass, centroid, second moment). These moments are functions of time and the second one portrays the spreading of the solute around the cloud centroid. The velocity fields caused by natural gradients are generally steady or slowly varying in time and both field experiments and theory indicate that the longitudinal spreading is much stronger than the transverse. Effective (or macro) dispersion coefficients and associated dispersivities can be defined as half the rate of change of spatial second moments of the plume.

The main role of the theory is to predict transport for a given heterogeneous structure and for given boundary and initial conditions for pressure head and concentration. The investigation of transport here is focused on relatively simple conditions: K is modelled as a random stationary (homogeneous) space function, the flow domain is regarded as unbounded (i.e. the plume is far from boundaries), the flow is driven by a constant average pressure-head gradient, the solute is a passive scalar, the initial concentration of a finite cloud is constant and only longitudinal dispersion is considered. These conditions are satisfied approximately in many field cases and our purpose is anyway to discuss some basic issues of transport using only simple computations. Still, even under these conditions, prediction of the flow field and of concentration is a formidable problem. Its solution has been achieved either by numerical methods or by approximate analytical ones. We shall rely here on the latter, numerical results serving merely as illustrative simulative experiments.

We have employed in the past the Lagrangian approach (Dagan 1984, 1987), which is well suited to depict the solute-body spatial moments, in order to solve the transport problem. The results of the analysis compared favourably with some field experiments (e.g. Freyberg 1986). The theory, ether Lagrangian or Eulerian, and its application to analysis of field experiments, was underlain by the *ergodic hypothesis*. In simple terms it is assumed that the ensemble means of the spatial moments of the trajectories of one particle in various realizations of the random heterogeneous structure are equal to those of the trajectories of the many particles making up the finite solute body in any particular realization. The ergodic hypothesis for the cloud spatial moments is bound to be satisfied if the lengthscale characterizing the size of the solute body is much larger than the heterogeneity scale I. With neglect of the effect of pore-scale dispersion, the lengthscale l of the initial cloud and its ratio with the heterogeneity scale I is the parameters playing the major role. The ergodic hypothesis for spatial moments was apparently satisfied for the field experiments mentioned above. Indeed, the heterogeneity scales (of the order of centimetres, vertically, and of metres, horizontally), related mainly to the local sedimentary features of the formations, were small compared to l. The question has been raised recently (e.g. Philip 1986 and Neuman 1990) about the impact of larger heterogeneity scales upon transport. Such scales are encountered for instance, in regional flows (Dagan 1986, 1989). Indeed, natural formations are generally shallow and for transport distances that are large compared to the thickness, new heterogeneity scales, of the vertically averaged conductivity, appear. At these scales transport may be regarded as two-dimensional in the horizontal plane, with transmissivity heterogeneity scales found to be of the order of hundreds to thousands of metres (Delhomme 1979; Hoeksema & Kitanidis 1984). The salient question is what is the impact of such large scales upon the spatial moments of a solute cloud of size which is not large compared to I, i.e. for non-ergodic transport? The aim of the present study is to investigate this issue along the lines of Dagan (1990). The plan of the paper is as follows: for completeness, we define in the next Section the transport problem in mathematical terms and review briefly previous results of the Lagrangian analysis under ergodic conditions; in §3 we recall briefly the definition of effective longitudinal dispersion coefficient under non-ergodic conditions (Dagan 1990) and relate it to the velocity and conductivity fields; the main original contribution is in §4, in which we derive the expressions of the effective dispersion coefficient in terms of the conductivity statistical parameters and the initial solute-body size, for two-dimensional velocity fields. As a by-product we arrive at the limiting conditions for which ergodicity may be obeyed. The main result is that under non-ergodic conditions the controlling lengthscale of the solute-body dispersion is its initial size l, rather than the heterogeneity scale I.

Although the results are derived for transport in heterogeneous porous formations, it is believed that they are of interest to other convective transport phenomena by steady velocity fluid fields.

2. Mathematical statement of the problem; ergodic transport by the Lagrangian approach

For the sake of completeness we recall here briefly the mathematical statement of the flow and transport problem (for details see for instance Dagan 1989). Flow of an incompressible fluid takes place in a domain Ω in the horizontal x_1, x_2-plane. The steady velocity field $\boldsymbol{V}(\boldsymbol{x})$ satisfies Darcy's law

$$\boldsymbol{V} = -\frac{K}{n}\nabla H, \tag{1}$$

where $K(\boldsymbol{x})$ is the conductivity (vertically averaged), n is the effective porosity and $H(\boldsymbol{x})$ is the head. In a heterogeneous formation n is generally spatially variable, but to a much smaller extent than the conductivity and it is, therefore, assumed to be constant. The velocity also obeys the continuity equation

$$\nabla \cdot \boldsymbol{V} = 0. \tag{2}$$

On the boundary $\partial\Omega$ of the domain, $H = -\boldsymbol{J}\cdot\boldsymbol{x}$, where $\boldsymbol{J}$ is a constant vector. The conductivity is a random space function and in line with empirical findings (Delhomme 1979; Hoeksema & Kitanidis 1984) we assume that it is lognormal and stationary. Thus, $Y = \ln K$ is completely characterized by its mean $\langle Y\rangle$ and by the two-point covariance $C_Y(\boldsymbol{r}) = \langle[Y(\boldsymbol{x}+\boldsymbol{r})-\langle Y\rangle][Y(\boldsymbol{x})-\langle Y\rangle]\rangle = \sigma_Y^2\rho_Y(\boldsymbol{r})$, where σ_Y^2 is the variance and ρ_Y is the auto-correlation. C_Y is assumed to be isotropic, i.e. a function of $r = |\boldsymbol{r}|$, and of finite integral scale $I = \int_0^\infty \rho_Y(r_1, 0)\,\mathrm{d}r_1$. The domain Ω is of a dimension much larger than I, such that ergodic arguments about Y are bound to hold. The velocity is a random space function due to the dependence on K, equation (1). Its constant mean $\boldsymbol{U} = \langle \boldsymbol{V} \rangle$ is given by $\boldsymbol{U} = (K_{ef}/n)\boldsymbol{J}$ and the fluctuation $\boldsymbol{u} = \boldsymbol{V} - \boldsymbol{U}$ is characterized by the two-point covariance $u_{ij}(\boldsymbol{r}) = \langle u_i(\boldsymbol{x}+\boldsymbol{r})\, u_j(\boldsymbol{x})\rangle$ $(i,j = 1, 2)$, as well as by higher-order moments. Deriving the relationship between K_{ef} and u_{ij} on one hand and the statistical parameters of Y on the other, is one of the central problems of flow through heterogeneous formations, to be briefly discussed below.

In the Lagrangian approach transport is described in terms of the motion of indivisible solute particles which are convected by the fluid. The trajectory is related to the velocity field by the kinematic relationship

$$\frac{\mathrm{d}\boldsymbol{X}}{\mathrm{d}t} = \boldsymbol{V}(\boldsymbol{X}); \quad \boldsymbol{X} = \boldsymbol{a} \quad \text{for} \quad t = 0, \tag{3}$$

where $\boldsymbol{x} = \boldsymbol{X}(t, \boldsymbol{a})$ is the equation of the trajectory of a particle which at $t = 0$ is at $\boldsymbol{x} = \boldsymbol{a}$. We have neglected the effect of pore-scale or local heterogeneity, characterized by scales much smaller than I, and manifested in a diffusive displacement that supplements $\boldsymbol{X}$. As already mentioned in §1, the effect of such a term is negligible for the type of heterogeneous structure investigated here and for the transport times of interest. The aim of the Lagrangian theory is to determine the statistical moments of $\boldsymbol{X}$, i.e. $\langle \boldsymbol{X} \rangle, X_{ij}(t, 0) = \langle X_i'(t, \boldsymbol{a}) X_j'(t, \boldsymbol{a})\rangle$, etc. in terms of the statistical moments of $\boldsymbol{V}$, which in turn depend on those of Y. The solute concentration is related to $\boldsymbol{X}$ through the relationship $C(\boldsymbol{x}, t) = (m/n)\,\delta(\boldsymbol{x} - \boldsymbol{X})$, where m is the solute mass. From this definition it is seen that $\langle C \rangle = (m/n) f(\boldsymbol{x}, t)$, where f is the p.d.f. (probability density function) of X. We concentrate here, however, on characterizing C of a finite solute body by its spatial moments. The initial condition is $C(\boldsymbol{x}, 0) = C_0 =$ const. in an initial area A_0, whose lengthscale is l. The spatial moments are defined as follows:

$$\left.\begin{aligned} M &= \int nC\,\mathrm{d}\boldsymbol{x} = nC_0 A_0; \quad \boldsymbol{R} = \frac{1}{M}\int nC\boldsymbol{x}\,\mathrm{d}\boldsymbol{x} = \frac{1}{A_0}\int_{A_0} \boldsymbol{X}(t, \boldsymbol{a})\,\mathrm{d}\boldsymbol{a}, \\ S_{ij} &= \frac{1}{M}\int n(x_i - R_i)(x_j - R_j)\,C\,\mathrm{d}x = \frac{1}{A_0}\int_{A_0} [(X_i(t, \boldsymbol{a}) - R_i]\,[X_j(t, \boldsymbol{a}) - R_j]\,\mathrm{d}\boldsymbol{a}, \end{aligned}\right\} \tag{4}$$

where M is the total mass of the conservative solute, $\boldsymbol{R}$ is the coordinate of the centroid of the cloud and S_{ij} are second spatial moments, proportional to the moments of inertia of the cloud.

Under ergodic conditions for the spatial moments, supposed to prevail if $l \gg I$, one has $\boldsymbol{R} \approx \langle \boldsymbol{R} \rangle = \bar{\boldsymbol{a}} + \boldsymbol{U}t$, where $\boldsymbol{x} = \bar{\boldsymbol{a}}$ is the centroid of A_0. By the same token, $S_{ij}(t) \approx \langle S_{ij}(t) \rangle = S_{ij}(0) + X_{ij}(t, 0)$, where the last relationship stems from ensemble averaging (4) and replacing $\boldsymbol{X}$ by $\boldsymbol{a} + \boldsymbol{U}t + \boldsymbol{X}'$ and $\boldsymbol{R}$ by $\bar{\boldsymbol{a}} + \boldsymbol{U}t$, respectively. Hence, it is seen that the one-particle-trajectory statistical moments characterize the solute-body spatial moments. The results are simplified considerably for a transport distance $L = Ut$ large compared to I. By invoking arguments relying on the central-limit theorem one may assume that $f(\boldsymbol{x}, t)$ becomes Gaussian and completely defined by $\langle \boldsymbol{X} \rangle$ and X_{ij}. Furthermore, $X_{ij}(t, 0) \rightarrow 2\mathscr{D}_{ij}t$, where $\mathscr{D}_{ij}$ is the tensor of constant effective dispersion coefficients, while $\mathscr{D}_{ij}/U$ are effective dispersivities. At the large-t limit and for ergodic transport, the spatial moments are completely characterized by U and $\mathscr{D}_{ij}$. The restricted scope of the theory, which is all that is pursued here, is to derive these entities in terms of the statistical parameters of the velocity field and of Y. Even this limited objective is a formidable one and it can be generally attained only by numerical methods. In contrast, simple results of an analytical nature can be obtained by a first-order approximation in σ_Y^2. Recent numerical simulations (for instance Valocchi 1990 and Quinodoz & Valocchi 1991; see figure 1) shows that the first-order approximation of X_{ij} may be quite accurate for $\sigma_Y^2 = 0.5$ and even larger. Again, for the sake of completeness, we review briefly the main results of the first-order analysis.

With $K = \exp(\langle Y \rangle + Y') = K_G \exp(Y') = K_G(1+Y'+\dots)$, where $K_G = \exp(\langle Y \rangle)$ is the conductivity geometric mean, and with $H = -\boldsymbol{J}\cdot\boldsymbol{x}+h$, the expansion of Darcy's law (1) yields at first-order in Y' and h

$$\boldsymbol{V} = \frac{K_G}{n}(\boldsymbol{J}-\boldsymbol{J}Y'-\nabla h), \quad \text{i.e.} \quad \boldsymbol{U} = \frac{K_G}{n}\boldsymbol{J}, \quad \boldsymbol{u} = \frac{K_G}{n}(\boldsymbol{J}Y'-\nabla h), \tag{5}$$

where quadratic terms in Y' and h have been neglected. Elimination of $\boldsymbol{u}$ from (5) and (2) yields the simplified equation for the head fluctuation

$$\nabla^2 h = \boldsymbol{J}\cdot\nabla Y' \quad (x \in \Omega); \qquad h = 0 \quad (x \in \partial\Omega). \tag{6}$$

Without loss of generality, the mean flow is assumed to be in the x_1 direction, i.e. $\boldsymbol{U}(U,0)$. Then, the longitudinal velocity covariance $u_{11} = O(\sigma_Y^2)$, resulting from (5), is given by

$$u_{11}(\boldsymbol{r}) = U^2 C_Y(\boldsymbol{r}) + \frac{2U^2}{J}\frac{\partial C_{YH}(\boldsymbol{r})}{\partial r_1} - \frac{U^2}{J^2}\frac{\partial^2 C_H(\boldsymbol{r})}{\partial r_1^2}. \tag{7}$$

In (7), $C_{YH}(\boldsymbol{r}) = \langle Y'(\boldsymbol{x}+\boldsymbol{r})\,h(\boldsymbol{x})\rangle$ and $C_H(\boldsymbol{r}) = \langle h(\boldsymbol{x}+\boldsymbol{r})\,h(\boldsymbol{x})\rangle$ are logconductivity and head covariances, respectively, and they can be evaluated with the aid of (6). Indeed, multiplying (6) by $Y'(\boldsymbol{x}+\boldsymbol{r})$ and by $h(\boldsymbol{x}+\boldsymbol{r})$, leads to differential equations for C_{YH} and C_H, respectively. Explicit expressions for C_{YH} and C_H for an exponential C_Y are given in Dagan (1989), while those for u_{ij} were derived by Rubin (1990). The following general relationships are of interest here:

$$C_{YH} = \partial C_H/\partial r_1 = 0 \quad \text{for} \quad r_1 = 0; \qquad C_{YH} = 0, \quad \nabla C_H = 0 \quad \text{for} \quad r \to \infty.$$

Under the same linearized approximation, the trajectory $\boldsymbol{X}$, solution of (3), and the covariance X_{11}, are given by

$$\left.\begin{aligned} \boldsymbol{X}(t,\boldsymbol{a}) &= \boldsymbol{a} + \boldsymbol{U}t + \int_0^t \boldsymbol{u}(\boldsymbol{a}+\boldsymbol{U}t')\,\mathrm{d}t', \\ X_{11}(t,0) &= \int_0^t\int_0^t u_{11}(Ut', Ut'')\,\mathrm{d}t'\,\mathrm{d}t'' = 2\int_0^t (t-t')\,u_{11}(Ut',0)\,\mathrm{d}t', \end{aligned}\right\} \tag{8}$$

the approximation being that the actual trajectory in the argument of $\boldsymbol{u}$ (3), is replaced by its mean. This approximation is consistent with the linearization of the flow equations (5), both implying neglection of terms $O(\sigma_Y^4)$ in various covariances.

For large $tU/I, X_{11}$ tends to the 'Fickian' limit

$$X_{11}(t,0) \to 2t\int_0^\infty u_{11}(Ut',0)\,\mathrm{d}t' = \frac{2t}{U}\int_0^\infty u_{11}(r_1,0)\,\mathrm{d}r_1. \tag{9}$$

Finally, from (7) and (9) and with the aforementioned properties of C_{YH} and C_H, we arrive at the result

$$\mathscr{D}_L = \mathscr{D}_{11} = \frac{1}{2}\frac{\mathrm{d}X_{11}}{\mathrm{d}t} = \frac{1}{U}\int_0^\infty u_{11}(r_1,0)\,\mathrm{d}r_1 = U\int_0^\infty C_Y(r_1,0)\,\mathrm{d}r_1 \to \sigma_Y^2 UI \quad \text{for} \quad tU/I \gg 1,$$

i.e.

$$\alpha_L = \frac{\mathscr{D}_{11}}{U} = \sigma_Y^2 I. \tag{10}$$

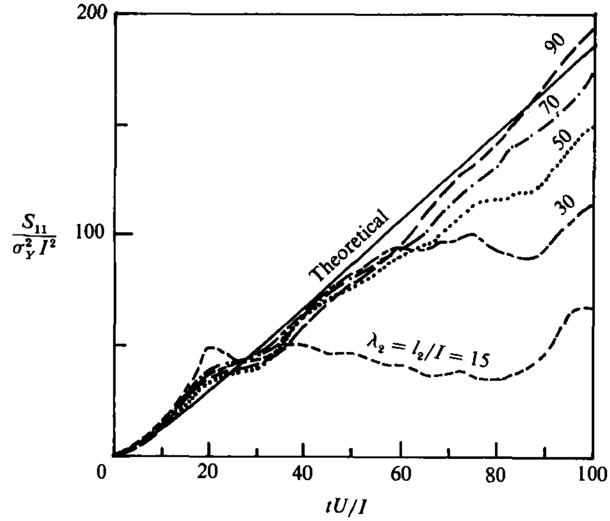

FIGURE 1. The dependence of the second longitudinal spatial moment S_{11} upon travel time for solute bodies of different initial transverse size l_2. Based on a numerical simulation of a single realization of the logconductivity and velocity fields (from Valocchi 1990, and Quinodoz & Valocchi 1991).

This result for the longitudinal dispersivity α_L has been obtained using the Lagrangian approach by Dagan (1982*a*, 1984, 1987) and with the aid of the Eulerian one by Gelhar & Axness (1983) and Neuman, Winter & Newman (1987), for different flow configurations and with the inclusion of the diffusive term.

These derivations summarize the previously obtained results of relevance to the present study. It is emphasized that $\mathscr{D}_L$ represents half the rate of change of the second spatial moment S_{11}, (4), only under ergodic conditions, i.e. for $l/I \gg 1$. The tendency of S_{11} to the ergodic limit is illustrated by the results of a numerical simulation (Valocchi 1990; Quinodoz & Valocchi 1991) shown in figure 1. The authors have conducted a very detailed study: the conductivity was generated on a dense grid, with an exponential $\rho_Y = \exp(-r/I)$ and with $\sigma_Y^2 = 0.5$; the solute input zone was a line of dimension l_2 normal to the mean flow direction; the velocity field was determined by solving numerically (1), (2) for boundary conditions of average uniform gradient, and transport has been simulated by tracking a large number of particles. The results pertain to a *single* realization in which the ratio $\lambda_2 = l_2/I$ has been varied systematically. Figure 1 depicts the dependence of S_{11} of (4), the second spatial moment in the longitudinal direction of the particles cloud, upon time. The theoretical curve (Dagan 1982*a*) has the slope of its linear portion equal to $2\mathscr{D}_{11}$ of (10). For the largest $\lambda_2 = 90$, the agreement of (10) with the numerical simulations is seen to be quite good.

3. Definition of effective dispersion coefficient for non-ergodic transport

In the case in which l/I is not large enough to warrant ergodicity, the spatial moments (4) of a plume in each realization may differ from their ensemble mean. This point is illustrated in figure 1 in which S_{11} is represented as function of time and for input zones A_0 of $l_1 = 0$ and l_2 of diminishing magnitude, for the same realization of Y and $\boldsymbol{u}$, It is seen that S_{11} may differ considerably from the result of (10), though good agreement was obtained for the largest λ_2. The salient question is how to characterize transport and its uncertainty under such circumstances. Following Dagan (1989, 1990) we now regard $\boldsymbol{R}$ and S_{ij} as random variables, represented by their statistical moments. Under the conditions enumerated above, the first two moments of the centroid longitudinal trajectory are given by

$$\langle R_1\rangle = \bar{a}_1+Ut;\quad R_{11} = \langle (R_1-\langle R\rangle)^2\rangle = \frac{1}{A_0^2}\int_{A_0}\int_{A_0} X_{11}(t,\boldsymbol{b})\,\mathrm{d}\boldsymbol{a}'\,\mathrm{d}\boldsymbol{a}''\quad (\boldsymbol{b}=\boldsymbol{a}'-\boldsymbol{a}''), \tag{11}$$

where $X_{11}(t,\boldsymbol{a}',\boldsymbol{a}'') = \langle X'(t,\boldsymbol{a}')\,X'(t,\boldsymbol{a}'')\rangle$ is the covariance of the trajectories of two particles originating from two different points in A_0. By the stationarity of the velocity field it is a function of $\boldsymbol{b} = \boldsymbol{a}'-\boldsymbol{a}''$ (see the next Section).

In a similar manner the expected value of S_{11} from (4) is given by

$$\langle S_{11}(t)\rangle = S_{11}(0)+X_{11}(t,0)-R_{11}(t,l). \tag{12}$$

This simple and fundamental relationship, obtained by Kitanidis (1988) and by Dagan (1989, 1990) by different methods, reads that $X_{11}(t,0)$, the trajectory variance with respect to the mean centroid, is equal to the sum of $\langle S_{11}\rangle$, the variance with respect to the realization centroid, and of R_{11}, the variance of the centroid trajectory. The 'actual dispersion coefficient' $\bar{D}_{11}$ is defined by $\frac{1}{2}\mathrm{d}S_{11}/\mathrm{d}t$ and it is a random variable as well. The natural definition of the effective dispersion coefficient D_{11} is the expected value of $\bar{D}_{11}$, i.e. by (12)

$$D_{11}(t,l) = \frac{1}{2}\frac{\mathrm{d}\langle S_{11}\rangle}{\mathrm{d}t} = \frac{\mathrm{d}X_{11}(t,0)}{\mathrm{d}t}-\frac{\mathrm{d}R_{11}(t,l)}{\mathrm{d}t} = \mathscr{D}_{11}-\frac{\mathrm{d}R_{11}}{\mathrm{d}t}. \tag{13}$$

Under the ergodic conditions mentioned above $D_{11}\to\mathscr{D}_{11}$. The tendency to ergodicity may be assessed with the aid of the ratio var $(S_{11})/\langle S_{11}\rangle^2$. Its computation in terms of X_{ij} becomes cumbersome, even if $\boldsymbol{X}$ is assumed to be Gaussian (Dagan 1990). We shall pursue here only the computation of R_{11} and D_{11} and examine the tendency of the latter to $\mathscr{D}_{11}$ in (10) as a measure of approaching ergodic conditions.

After these preparatory steps we are in a position to define in precise terms the aim of the present study: we seek to derive expressions for R_{11} and D_{11} for large transport time $Ut/I \gg 1$ in terms of the statistical parameters of $Y(\langle Y\rangle,\sigma_Y^2,I)$ and of the lengthscale l of the input zone A_0. This is achieved by using the first-order approximation in σ_Y^2, which has served in the past to derive $\mathscr{D}_{11}$, see (10).

4. Computation of the effective longitudinal dispersion coefficient for non-ergodic transport

We proceed now with the computation of $D_{\mathrm{L}} = D_{11}(\infty,l)$, the asymptotic value of the longitudinal dispersion coefficient for a finite input zone of lengthscale l. Since $\mathscr{D}_{11}$ has already been evaluated (see (13)), the crux of the matter is to calculate $\mathrm{d}R_{11}/\mathrm{d}t$ in (13). In turn, the latter is related to $X_{11}(t,\boldsymbol{b})$, the two-particle-trajectories covariance. From the first-order approximation (8) of $\boldsymbol{X}'$, we get for the latter the general relationship

$$X_{ij}(t,\boldsymbol{b}) = \int_0^t\int_0^t u_{ij}[U(t'-t'')+b_1,b_2]\,\mathrm{d}t'\,\mathrm{d}t'',$$

i.e.
$$\frac{\mathrm{d}X_{ij}}{\mathrm{d}t} = \int_0^t \{u_{ij}[U(t-t')+b_1,b_2]+u_{ij}[U(t'-t)+b_1,b_2]\}\,\mathrm{d}t'\quad (i,j=1,2). \tag{14}$$

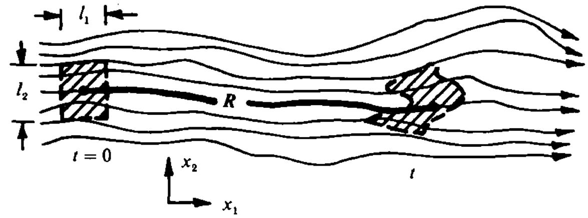

FIGURE 2. Sketch of motion of a solute body in steady flow.

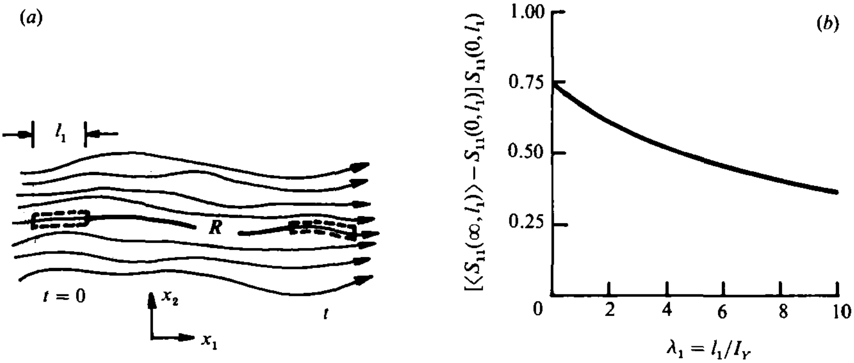

FIGURE 3. (*a*) Sketch of the motion of a thin solute body aligned with the mean flow, and (*b*) the dependence of the second spatial moment $S_{11}(\infty, l_1)$ upon the ratio $\lambda_1 = l_1/I$ (equations (21), (22)).

Next, we select the input zone A_0 to be a rectangle defined by $0 < x_1 < l_1, 0 < x_2 < l_2$, as shown in figure 2. Substituting (14) into (11) gives the following expression of the effective dispersion tensor in (13):

$$D_{ij}(t, l_1, l_2) = \frac{4}{l_1^2 l_2^2} \int_0^{l_1} \int_0^{l_2} \int_0^t (l_1 - b_1)(l_2 - b_2)\,[u_{ij}(Ut', 0) - \tfrac{1}{2}u_{ij}(Ut' + b_1, b_2) - \tfrac{1}{2}u_{ij}(Ut' - b_1, b_2)]\,\mathrm{d}t'\,\mathrm{d}b_2\,\mathrm{d}b_1. \tag{15}$$

In turn u_{ij} is related to the logconductivity by (7) and substitution in (15) yields for $D_\mathrm{L} = D_{11}$, after integration over t and for $t \to \infty$ and $i = j = 1$,

$$D_\mathrm{L} = \frac{4U\sigma_Y^2}{l_1^2 l_2^2} \int_0^{l_1} \int_0^{l_2} \int_0^\infty (l_1 - b_1)(l_2 - b_2)\,[\rho_Y(r, 0) - \tfrac{1}{2}\rho_Y(r + b_1, b_2) - \tfrac{1}{2}\rho_Y(r - b_1, b_2)]\}\,\mathrm{d}r\,\mathrm{d}b_2\,\mathrm{d}b_1. \tag{16}$$

It is emphasized that the terms stemming from the derivatives of C_{YH} and C_H in (7) do not appear in (16) because the integration is up to $t \to \infty$. Indeed, since C_{YH} and $\partial C_H/\partial r_1$ vanish for $r_1 = 0$ and $r_1 \to \infty$ and are antisymmetric in r_1, they drop out from (16). For simplicity, we examine now separately the impact of the longitudinal dimension l_1 and of the transverse one l_2 upon D_L, since they have a profoundly different influence.

4.1. *Thin body aligned with the mean flow direction* ($l_2 = 0$)

Such a body is depicted in figure 3(*a*). For $l_2 = 0$, D_L in (16) becomes

$$D_\mathrm{L} = \frac{2U\sigma_Y^2}{l_1^2} \int_0^{l_1} (l_1 - b_1) \int_0^\infty [\rho_Y(r, 0) - \tfrac{1}{2}\rho_Y(r + b_1, 0) - \tfrac{1}{2}\rho_Y(r - b_1, 0)]\,\mathrm{d}r\,\mathrm{d}b_1. \tag{17}$$

The integral over r in (17) is identically zero due to the symmetry of the autocorrelation ρ_Y and we arrive at the unexpected result that the longitudinal dispersion coefficient $D_{\mathrm{L}} = 0$ for any finite l_1. In contrast, we obtain from (13) that the centroid variance is given by $R_{11} \rightarrow 2U\sigma_Y^2 It$ for $t \rightarrow \infty$, i.e. the velocity randomness manifests in the uncertainty of R_1. It is of interest to evaluate the variance of $\bar{U}_1 = \mathrm{d}R_1/\mathrm{d}t$, the velocity of the solute-body centroid. From (4) it is easy to ascertain that it is equal to

$$\bar{U}_{11} = \frac{2}{l_1^2}\int_0^{l_1} (l_1 - b_1)\, u_{11}(b_1, 0)\, \mathrm{d}b_1. \tag{18}$$

The variance can be evaluated for any given ρ_Y by substituting u_{11} from (7) into (18). For large $\lambda_1 = l_1/I$ the result is

$$\bar{U}_{11} \approx \frac{2\sigma_Y^2 U^2}{l_1^2}\int_0^{l_1} (l_1 - b_1)\, \rho_Y(b_1, 0)\, \mathrm{d}b_1 \approx \frac{2\sigma_Y^2 U^2}{\lambda_1} \quad \text{with} \quad \lambda_1 = l/I. \tag{19}$$

It is seen therefore that the ratio of $\bar{U}_{11}$ to $\langle \bar{U}_1 \rangle^2 = U^2$ tends to zero like $2\sigma_Y^2/\lambda_1$ for $\lambda_1 \gg 1$ and the exchange between the space average of the Lagrangian velocity and the Eulerian ensemble mean is then permissible. This is in agreement with a general result of Lumley (1962), which was proved, however, for a solute body of a unbounded spatial extent.

The vanishing of D_{L} does not imply that $\langle S_{11} \rangle$ does not change with time, sine D_{L} is the limit of $\frac{1}{2}\mathrm{d}S_{11}/\mathrm{d}t$ for $t \rightarrow \infty$. This can be shown by computing $\langle S_{11}(\infty, l_1) \rangle$ from (4), which is given in terms of X_{11} (14) as follows:

$$\langle S_{11}(t, l_1) \rangle - S_{11}(0, l_1) = \frac{2}{l_1^2}\int_0^{l_1} (l_1 - b_1)\left[X_{11}(t, 0) - \tfrac{1}{2}X_{11}(t, b_1, 0) - \tfrac{1}{2}X_{11}(t, -b_1, 0)\right] \mathrm{d}b_1. \tag{20}$$

For $t \rightarrow \infty$ (20) reduces to the simple formula

$$\langle S_{11}(\infty, l_1) \rangle - S_{11}(0, l_1) = \frac{2}{l_1^2}\int_0^{l_1} (l_1 - b_1)\, X_{11}(b_1/U, 0)\, \mathrm{d}b_1, \tag{21}$$

where $X_{11}(b_1/U, 0)$ is the one-particle-trajectory covariance (8) in which the argument t is replaced by b_1/U. X_{11} has been evaluated in a closed form for the exponential $\rho_Y = \exp(-r/I)$ (Dagan 1984, equation 4.5 and figure 1*a*) and is reproduced here:

$$X_{11} = 2\beta_1 - 3\ln\beta_1 + 1.5 - 3E + 3\left[\mathrm{Ei}(-\beta_1) + \frac{(1+\beta_1)\exp(-\beta_1) - 1}{\beta_1^2}\right] \quad \text{with} \quad \beta_1 = b_1/I, \tag{22}$$

where E is Euler constant and Ei is the exponential integral. $\langle S_{11}(\infty, l) \rangle$ of (21) has been computed numerically for X_{11} and the result is represented in a dimensionless form in figure 3(*b*). For the assumed shape of the initial solute body, $S_{11}(0, l) = \frac{1}{12}l_1^2$. Figure 3(*b*) shows that the increase of the second-order spatial moment in terms of its initial value is quite modest. At any rate the effective dispersion coefficient has to grow from zero at $t = 0$ to a maximal value and to drop again to zero, as proved above.

The result concerning D_{L} can be interpreted by considering flow and transport under general conditions. Indeed, we refer now to a curvilinear thin initial solute

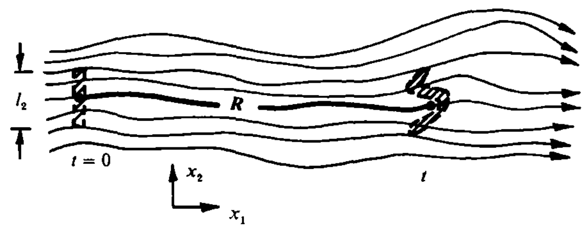

FIGURE 4. Sketch of the motion of a thin solute body transverse to the mean flow.

body lying on a streamline of the steady velocity field. Let $X(t, a)$ be now the intrinsic coordinate of a particle at t which was at the curvilinear coordinate $s = a$ at $t = 0$. With l the initial length of the body, we have $l = X(0, l) - X(0, 0)$. The curvilinear length L at any time is therefore given by $L = X(t, l) - X(t, 0)$, since the body preserves its continuity. The variance of the elongation $L - l$ is expressed by var $(L - l) = \langle [X'(t, l) - X'(t, 0)]^2 \rangle$, where X' is the fluctuation of X, which is assumed to be a stationary random function of t. The displacement X can be related to the Lagrangian velocity of the particle along the streamline by $\mathrm{d}X'(t, a)/\mathrm{d}t = v(t, a)$. Now, owing to the steadiness of the Eulerian velocity the important relationship $X(t, l) = X(t - T, 0)$, is satisfied for $t > T$. Here T is the time required for the trailing edge of the solute body to reach the initial location of the leading edge. This simple relationship shows that the motion of the two ends of the thin body become correlated for $t > T$. By using this relationship and assuming that $v(t, 0)$, is stationary and of covariance $C_v(t)$, we get for the elongation

$$\operatorname{var}(L - l) = 2 \int_0^T (T - t')\, C_v(t')\, \mathrm{d}t' \quad (t > T), \tag{23}$$

which is fixed and does not depend on the time t. Hence, the elongation of the thin, streamline-aligned, solute body grows in the mean for a while and then does not increase anymore, in agreement with the result found by the first-order analysis. Generally speaking, a diffusion process occurs when the various parts of the solute body move independently in a statistical sense, but this does not happen for a body of *finite* length, since the motion of each particle becomes identical to the one preceding it after a fixed time lag, equal to T for the end points. The argument does not hold, of course, for the theoretical but unrealistic case of an infinite solute body, for which the ergodic argument applies.

Summarizing this paragraph, we have found that for steady velocity fields and for a thin solute body aligned with a streamline, i.e. in the mean flow direction under the first-order approximation, the motion of the body centroid is subjected to uncertainty. In contrast, the expected value of the second spatial moment increases from its initial value to an asymptotic, fixed one, which is attained in practice after a travel time $t > l_1/U$. Since S_{11} is a random variable, its complete characterization is achieved by computing its statistical higher-order moments, but this task is not undertaken here.

4.2. *Thin body initially normal to the mean flow direction* ($l_1 = 0$, *figure* 4)

The general relationship (16) reduces now to

$$D_{\mathrm{L}} = \sigma_Y^2\, U \left[\int_0^\infty \rho_Y(r_1, 0)\, \mathrm{d}r_1 - \frac{2}{l_2^2} \int_0^\infty \int_0^{l_2} (l_2 - b_2)\, \rho_Y(r, b_2)\, \mathrm{d}r\, \mathrm{d}b_2 \right]. \tag{24}$$

It is easy to estimate the limit values of D_L for $\lambda_2 \to 0$ and $\lambda_2 \to \infty$, respectively, with $\lambda_2 = l_2/I$. Indeed, in the first case we get from (24)

$$D_L \to \sigma_Y^2 U\left[I - \lim_{l_2 \to 0} \frac{2}{l_2^2} \int_0^\infty \int_0^{l_2} (l_2 - b_2)\rho_Y(r, 0)\,dr\,db_2\right] = 0, \tag{25}$$

where neglected terms are $O(l_2^2)$. This result is to be expected since at the limit $\lambda_2 \to 0$ the solute body degenerates into an indivisible particle.

At the other limit, $\lambda_2 \to \infty$, and for an integrable ρ_Y, we get from (24)

$$D_L \to \sigma_Y^2 U\left[I - \frac{2}{l_2} \int_0^\infty \int_0^\infty \rho_Y(r, b_2)\,dr\,db_2\right] = \sigma_Y^2 UI + O(I/\lambda_2) \tag{26}$$

and D_L tends to the asymptotic limit $\mathscr{D}_L$, (10). Hence, unlike the elongated solute body, the transverse one tends to disperse according to the ergodic limit for a sufficiently large solute body. This is understandable in the light of the discussion of the preceding paragraph: particles making up the solute body which move along remote streamlines no longer have correlated trajectories.

If we wish to follow the variation of D_L for a fixed initial solute body, of fixed l_2, but for media of different I, it is appropriate to make D_L dimensionless with respect to $\sigma_Y^2 Ul_2$. The dimensionless dispersion coefficient becomes a function of λ_2 solely and it is seen from (25) and (26) that $D_L/\sigma_Y^2 Ul_2$ tends to zero for both limits $\lambda_2 \to 0$ and $\lambda_2 \to \infty$. Since D_L is positive it follows that it must have a maximum. To grasp this result in quantitative terms we have considered two particular examples of logconductivity autocorrelation: the exponential $\rho_Y = \exp(-r/I)$ and the Gaussian $\rho_Y = \exp(-\pi r^2/4I^2)$. The first one pertains to a medium made up from blocks of constant K, the slope of ρ_Y at the origin being related to the specific interface area between blocks. The Gaussian ρ_Y depicts a medium of continuous Y, with a sharp drop of the correlation with distance.

The result of integration for the exponential ρ_Y in (26) is as follows:

$$D_L/(\sigma_Y^2 Ul_2) = (1/\lambda_2)\{1 - \pi[K_1(\lambda_2)L_0(\lambda_2) + L_1(\lambda_2)K_0(\lambda_2)] - 2K_2(\lambda_2) + 4/\lambda_2^2\}, \tag{27}$$

where K_i and L_i are modified Bessel and Struve functions, respectively (Abramowitz & Stegun 1965). Asymptotic results are as follows:

$$\frac{D_L}{\sigma_Y^2 Ul_2} \to -\frac{1}{12}\lambda_2 \ln \lambda_2 \quad (\lambda_2 \to 0); \qquad \frac{D_L}{\sigma_Y^2 Ul_2} \to \frac{1}{\lambda_2}\left(1 - \frac{\pi}{\lambda_2}\right) \quad (\lambda_2 \to \infty). \tag{28}$$

Similarly, for the Gaussian ρ_Y the integration in (26) yields

$$\left.\begin{aligned} &\frac{D_L}{\sigma_Y^2 Ul_2} = \frac{1}{\lambda_2}\left\{1 - \frac{2}{\lambda_2}\operatorname{erf}\left(\frac{\pi^{\frac{1}{2}}\lambda_2}{2}\right) + \frac{4}{\pi\lambda_2^2}\left[1 - \exp\left(-\frac{\pi\lambda_2^2}{4}\right)\right]\right\}, \\ &\frac{D_L}{\sigma_Y^2 Ul_2} \to \frac{\pi}{24}\lambda_2 \quad (\lambda_2 \to 0); \qquad \frac{D_L}{\sigma_Y^2 Ul_2} \to \frac{1}{\lambda_2}\left(1 - \frac{\pi^{\frac{1}{2}}}{\lambda_2}\right) \quad (\lambda_2 \to \infty). \end{aligned}\right\} \tag{29}$$

The dimensionless dispersion coefficient is represented as function of $1/\lambda_2$ for both cases (27) and (29) in figure 5 and the overall behaviour is similar. The different asymptotic results for $\lambda_2 \to 0$ can be attributed to the different structures of C_Y near $r = 0$. The striking result, however, is that $D_L/(\sigma_Y^2 Ul_2)$ reaches a maximum of around 0.15 for $\lambda_2 \approx 2$. Hence, the effective dispersion coefficient has an upper bound which depends only on l_2, no matter how large I is. This result contradicts the intuitive one

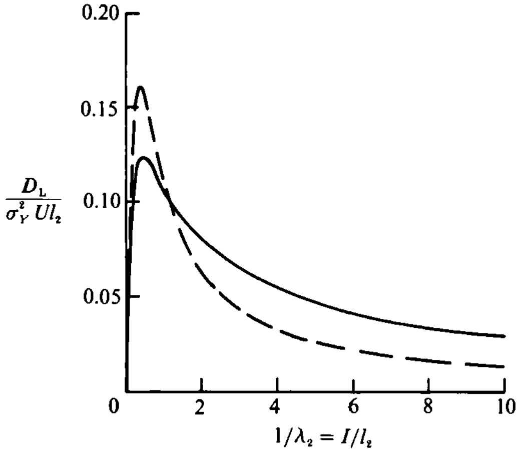

FIGURE 5. The dependence of the effective dispersion coefficient D_L on the ratio $1/\lambda_2$ between the logconductivity integral scale I and the solute body initial size l_2 (full line (27) and dashed line (29)).

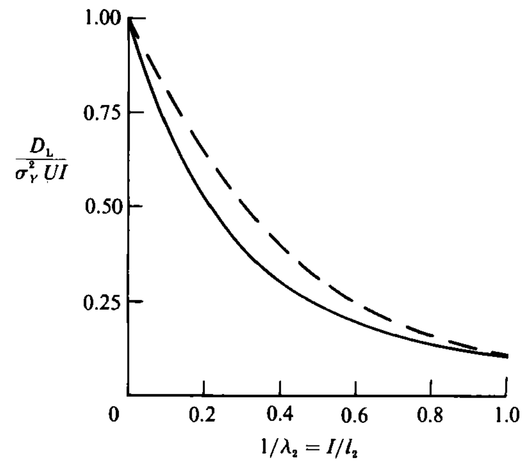

FIGURE 6. Same as figure 5, but with a different dimensionless representation of D_L.

based on inspection of the ergodic limit (10), which is underlain however by the requirement $\lambda_2 \gg 1$. This point is illustrated further in figure 6, in which we have rendered D_L dimensionless with respect to I, corresponding to a given formation and a variable l_2. The ergodic limit $D_L/(\sigma_Y^2 UI) = 1$ of (10) is attained for $1/\lambda_2 \rightarrow 0$. Adopting, for instance $D_L/(\sigma_Y^2 UI) = 0.99$ as a criterion, we get from (28) and (29) $\lambda_2 \approx 300$ and $\lambda_2 \approx 180$, respectively. Again, this point is somewhat illustrated by figure 1, which is limited however to a single realization, whereas the above results are for expected values. An assessment of the uncertainty of S_{11} may be achieved by calculating its variance, but this task is not followed here.

Summarizing this paragraph, we have found that for a solute body lying across the mean flow, the longitudinal asymptotic dispersion coefficient may reach its ergodic limit if $\lambda_2 = l_2/I$ is sufficiently large. However, for smaller values of λ_2, D_L decreases and is controlled by the body size rather than I. since the sum $\mathrm{d}\langle S_{11}\rangle/\mathrm{d}t + \mathrm{d}R_{11}/\mathrm{d}t$ is constant (13), this means that the effect of velocity randomness manifests in the centroid trajectory to compensate for the reduction of D_L. At the limit $\lambda_2 \rightarrow 0$ the body degenerates into an indivisible particle which obviously does not disperse at all.

5. Summary and conclusions

The motion of a solute body by convective transport in a steady two-dimensional velocity field was investigated under a few simplifying conditions: stationary and isotropic logconductivity of finite integral scale and travel distance large compared to I. The dispersion coefficient has been defined as half of the rate of change of the expected value of the solute-body second spatial moment around its centroid. The main objective was to determine the dependence of D_{L} upon the parameters characterizing statistically the heterogeneous structure and the flow, and upon the size of the solute input zone. It was expected that the ergodic limit of D_{L} will be reached for $l \gg I$.

The first main finding is that the longitudinal extent l_1, in the mean flow direction does not influence D_{L}, which tends to zero. Thus, ergodic conditions are not reached in such a case. This result could be explained *a posteriori* by realizing that the trajectories of the different particles making up the cloud are correlated, since they lie on the same streamline of the steady flow.

The second main finding is D_{L} may tend to the ergodic limit $\sigma_Y^2\, UI$ for an input zone lying in the transverse direction. However, if l_2 is not larger than I by two orders of magnitude, D_{L} is smaller than the above limit and is controlled by l_2 rather than by I. This result suggests that for the case of a 'point source', i.e. of a cloud or plume of fixed l_2, and for a formation of large heterogeneity correlation scale, the solute body will disperse modestly around its centroid. However, the centroid itself is subjected to a random motion of increasing uncertainty.

The results of this study are underlain by the assumption of negligible transverse diffusive effects. This assumption may be justified by the smallness of the transverse pore-scale dispersivity or of the transverse macrodispersivity associated with local heterogeneity. Still, for a sufficiently large time for which the diffusive spreading mechanism across streamlines would ensure mixing over the velocity correlation scale, the transport will become again Fickian. Such a large limit may be, however, beyond the range of interest in application. This and many other issues of interest related to the present study deserve further investigations. A few examples are: the dependence of the spatial moments on travel time, the derivation of various statistical moments of the spatial moments, the impact of large logconductivity variance, reduction of uncertainty of spatial moments by conditioning on measured values and transport in formations of unbounded heterogeneity correlation scale.

REFERENCES

ABRAMOWITZ, M. & STEGUN, I. A. 1965 *Handbook of Mathematical Functions*. Dover.

BOUCHAUD, J.-P., GEORGES, A., KOPLIK, J., PROVATA, A. & REDNER, S. 1990 Superdiffusion in random velocity fields. *Phys. Rev. Lett.* **64**, 2503–2506.

DAGAN, G. 1982*a* Stochastic modelling of groundwater flow by unconditional and conditional probabilities 2. The solute transport. *Water Resour. Res.* **18**, 835–848.

DAGAN, G. 1982*b* Analysis of flow through heterogeneous random aquifer 2. Unsteady flow in confined formations. *Water Resour. Res.* **18**, 1571–1585.

DAGAN, G. 1984 Solute transport in heterogeneous porous formations. *J. Fluid Mech.* **145**, 151–177.

DAGAN, G. 1986 Statistical theory of groundwater flow and transport: pore to laboratory, laboratory to formation and formation to regional scale. *Water Resour. Res.* **22**, 120S–125S.

DAGAN, G. 1987 Theory of solute transport by groundwater. *Ann. Rev. Fluid Mech.* **19**, 183–215.

DAGAN, G. 1989 *Flow and Transport in Porous Formations*. Springer.

DAGAN, G. 1990 Transport in heterogeneous formations: spatial moments, ergodicity and effective dispersion. *Water Resour. Res.* **26**, 1281–1290.

DELHOMME, J. P. 1979 Spatial variability and uncertainty in groundwater flow parameters: a geostatistical approach. *Water Resour. Res.* **15**, 269–280.

FREYBERG, D. L. 1986 A natural experiment on solute transport in a sand aquifer. 2. Spatial moments and the advection and dispersion of nonreactive tracers. *Water Resour. Res.* **22**, 2031–2046.

GARABEDIAN, S. P. 1987 Large-scale dispersive transport in aquifers: field experiments and reactive transport theory. Ph.D. dissertation. M.I.T.

GELHAR, L. J. & AXNESS, C. L. 1983 Three-dimensional stochastic analysis of macrodispersion in aquifers. *Water Resour. Res.* **19**, 161–180.

HOEKSEMA, R. J. & KITANIDIS, P. K. 1984 An application of the geostatistical approach to the inverse problem in two dimensional groundwater modelling. *Water Resour. Res.* **20**, 1009–1020.

KITANIDIS, P. K. 1988 Prediction by the method of moments of transport in a heterogeneous formation. *J. Hydrol.* **102**, 453–473.

LUMLEY, J. L. 1962 The mathematical nature of the problem of relating Lagrangian and Eulerian statistical functions in turbulence. In *Mecanique de la Turbulence* (ed. A. Favre), pp. 17–26, Paris: CNRS.

MATHERON, G. & MARSILY, G. DE 1980 Is transport in porous media always diffusive? A counterexample. *Water Resour. Res.* **16**, 901–917.

NEUMAN, S. P. 1990 Universal scaling of hydraulic conductivities and dispersivities in geologic media. *Water Resour. Res.* **26**, 1749–1758.

NEUMAN, S. P., WINTER, C. L. & NEWMAN, C. M. 1987 Stochastic theory of field-scale Fickian dispersion in anisotropic porous media. *Water Resour. Res.* **23**, 453–466.

PHILIP, J. R. 1986 Issues in flow and transport in heterogenous porous media. *Transport in Porous Media* **1**, 319–338.

QUINODOZ, H. A. M. & VALOCCHI, A. J. 1991 Macrodispersion in heterogeneous aquifers: numerical experiments. In *Proc. Conf. on Transport and Mass Exchange Processes in Sand and Gravel Aquifers: Field and Modeling Studies, Ottawa, Oct. 1990.*

RUBIN, Y. 1990 Stochastic modelling of macrodispersion in heterogeneous porous media. *Water Resour. Res.* **26**, 133–142.

SUDICKY, E. A. 1986 A natural-gradient experiment on solute transport in a sand aquifer: spatial variability of hydraulic conductivity and its role in the dispersion process. *Water Resour. Res.* **22**, 2069–2082.

VALOCCHI, A. J. 1990 Numerical simulation of the transport of absorbing solutes in heterogeneous aquifers. In *Computational Methods in Subsurface Hydrology* (ed. G. Gambolati, A. Rinaldo, C. A. Brebbia, W. C. Gray & G. F. Pinder), pp. 373–382. Computational Mechanics Publications and Springer.