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Is Transport in Porous Media Always Diffusive? A Counterexample

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For the special case of a stratified porous medium with flow parallel to the bedding it is shown that the transport of solute cannot, in general, be represented by the usual convection-diffusion equation, even for large time. The necessary conditions for the appearance of a Fickian diffusive process are discussed and compared with previous work done by Gelhar et al. (1979) and Marle et al. (1967). It is shown, however, that when the flow is not exactly parallel to the stratification, diffusive behavior is much more likely to appear. The need for further work on the mechanism of transport in porous media is then emphasized.

INTRODUCTION

The transport of solutes in porous media has generally been considered a Fickian diffusive process, i.e., a mechanism governed by the conventional convection-diffusion equation [*Scheideger,* 1954; *Bear and Bachmat,* 1967; *Fried,* 1972].

$$\operatorname{div}(\mathsf{D}\operatorname{grad} C - \mathbf{U}C) = \omega\frac{\partial C}{\partial t}$$

where

- D dispersion tensor;
- C concentration of the solution;
- $\mathbf{U}$ Darcy's velocity vector;
- ω kinematic porosity (sometimes called effective porosity).

The dispersion tensor D is generally considered to have its first principal direction parallel to the direction of the velocity $\mathbf{U}$, while the other two are orthogonal to this velocity. Dispersion, sometimes called hydrodynamic dispersion, is supposed to represent simultaneously both the molecular diffusion and the effect of the fluctuations of the true microscopic velocity around the average Darcy's velocity.

In the past, several attempts have been made to relate, theoretically at least, the magnitude of the coefficients of the dispersion tensor to a description of the heterogeneities of the permeability in the porous medium. We will refer here to the work of *Marle and Simandoux* [1966] and *Marle et al.* [1967] (hereinafter designated as Marle) and *Gelhar et al.* [1979] (hereinafter designated as Gelhar). Both authors have studied the transport in horizontal stratified media with horizontal flow and have concluded that this type of heterogeneity of the permeability (or, rather, of the velocity) leads asymptotically (i.e., at large distances or for large time) to a Fickian behavior, whose coefficients can be determined from the properties of the studied medium.

In this note we will present an unpublished result obtained by G. Matheron in 1975, which shows that for this particular type of heterogeneity, at least, one can sometimes obtain a non-Fickian type of transport, even asymptotically, i.e., which never conforms to the usual convection-diffusion equation. We will then give the conditions, which must be satisfied in order for this equation to be valid asymptotically, considering flow not only parallel to the stratification but also at an angle to it. We will then study the manner in which the asymptotic behavior is reached.

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STATEMENT OF THE PROBLEM

Consider a two-dimensional flow field in a stratified medium (see Figure 1). Let us start with the following assumptions: The direction of the velocity is always parallel to the bedding and constant for a given layer. This velocity is not modified by the presence of the solute (tracer assumption). However, the velocity is a function of the elevation z of the layer. We will use the microscopic seepage velocity $\mathbf{u} = \mathbf{U}/\omega$ (ω is kinematic porosity and $\mathbf{U}$ is Darcy's velocity) and assume that $u(z)$, the component of $\mathbf{u}$ along the x axis, is a random function representing a weakly stationary stochastic process. As $\mathbf{u}$ has only one component along the x axis, we will drop the vector notation.

In the work of Gelhar the assumptions are similar, and in addition, the random velocity $u(z)$ is taken to be generated by a random permeability field $K(z)$, a constant hydraulic gradient J, and a constant kinematic porosity ω.

In the work of Marle the flow geometry is the same, but $K(z)$ instead of being random is treated as a given function of the elevation z. The deterministic approach used by Marle can be viewed as a special case of the more general probabilistic approach: the stratified medium studied by Marle is one realization of the random function $K(z)$, which he assumes to be perfectly known. In the stochastic approach it is considered, on the contrary, that only statistical properties of the given stratified medium can be estimated (mean, variance, covariance . . .) and that these can best be utilized in considering the unknown real medium as a realization of a stochastic process, which is assumed to be weakly stationary and ergodic. For further discussion of the stochastic approach, refer to *Matheron* [1967], *Freeze* [1975], *Bakr et al.* [1978], *Gutjahr et al.* [1978], *Delhomme* [1979], and *Neuman* [1979]. Marle further assumes that the porosity ω varies with the elevation z.

Whereas Marle assumes that the medium is finite in the vertical direction only ($z \in [z_1, z_2]$), with no-flow boundary conditions for both water and solute, we will assume here, with Gelhar, that the medium is infinite both in x and z. This assumption holds even in a real porous medium as long as the tracer has not reached the boundaries.

Solute transport is assumed to be controlled by convection and dispersion with the following properties:

1. Convective transport has the local seepage velocity $u(z)$ at each elevation z of the medium.

2. Dispersive transport has a constant dispersion tensor: the dispersion coefficients D_L and D_T in the longitudinal x and transversal z directions are assumed to be constant, i.e., inde-

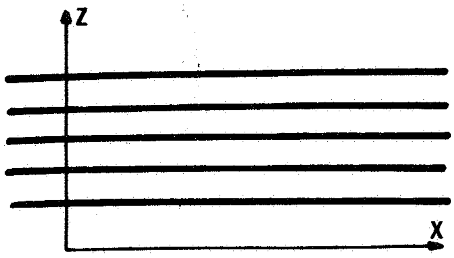

Fig. 1. Stratified medium and coordinate system.

pendent of the velocity. This transport can be viewed either as pure molecular diffusion with anisotropic (if desirable) diffusion coefficients or, preferably, as local hydrodynamic dispersion, defined on the same scale as the local velocity u, which is already an average velocity over a certain volume of porous medium. In that case, D_L and D_T should be regarded as the arithmetic averages of the dispersion coefficients of each stratum, since they would probably depend on the local velocity u. Our developments would then be approximate.

The fluctuations of the velocity $u(z)$, which is a random function of z, will then generate a differential transport at each elevation, creating what has sometimes been called 'macrodispersivity.'

We will ask the following question: Is this macrodispersivity always a Fickian process? That is, can it be represented by the convection-diffusion equation?

Conventional Model Used to Quantify Transport

With these assumptions the transport equation at the local scale can be written exactly as follows:

$$D_L \frac{\partial^2 C}{\partial x^2} + D_T \frac{\partial^2 C}{\partial z^2} - u(z)\frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \tag{1}$$

To check if this total transport equation gives rise to a global convection-diffusion equation, when the aquifer is considered as a whole, Marle computes a mean concentration by averaging C over the vertical $z \in [z_1, z_2]$ and tries to relate this average concentration C_A to the depth-averaged velocity U_A and the depth-averaged dispersion coefficient D_A. In other words, if the aquifer is taken as a single unit defined by its average properties, will transport also be diffusive in the average?

His answer is 'yes,' but only for the asymptotic displacement, i.e., when $t \to \infty$ and the displacement has occurred over large distances. Marle relates the global asymptotic longitudinal dispersion coefficient $D_A(\infty)$ to the fluctuation of the velocity (or permeability) and to the value of the local dispersion coefficients D_L and D_T. He assumes, for that matter, that D_L, D_T, and the kinematic porosity ω are also deterministic functions of the elevation z. Using the methods of moments, he finds

$$D_A(\infty) = \left[\int_{z_1}^{z_2} D_L(z)\,\omega(z)\,dz \bigg/ \int_{z_1}^{z_2} \omega(z)\,dz\right] + \left[\int_{z_1}^{z_2} \frac{\phi^2(z)}{D_T(z)\omega(z)}\,dz \bigg/ \int_{z_1}^{z_2} \omega(z)\,dz\right]$$

where

$$\phi(z) = \int_{z_1}^{z} \omega(s)\,[u(s) - \bar{u}]\,ds$$

$$u(z) = -\frac{K(z)}{\omega(z)}J$$

is the local pore velocity, J is the constant hydraulic gradient in the x direction, $K(z)$ is the permeability, and

$$\bar{u} = \left[\int_{z_1}^{z_2} \omega(z)\,u(z)\,dz \bigg/ \int_{z_1}^{z_2} \omega(z)\,dz\right]$$

is a weighted average of the velocity. This expression shows that in the long term the global longitudinal dispersion coefficient is the sum of an average of the local longitudinal dispersion coefficient plus a term representing the fluctuation of the local velocity with respect to its mean divided by the local transverse dispersion coefficient.

If we assume that D_L, D_T, and ω are independent of z, as we will do later, the above expression reduces to

$$D_A(\infty) = D_L + \left\{\int_{z_1}^{z_2}\left[\int_{z_1}^{z}[u(s) - \bar{u}]\,ds\right]^2 dz \bigg/ D_T(z_2 - z_1)\right\}$$

In his analysis, Gelhar assumes that ω is constant but that, simultaneously, $D_L(z)$, $D_T(z)$, and $K(z)$ are stationary random processes, while the longitudinal dispersivity is linked to the intrinsic permeability k by the assumption that the dispersivity α_L ($D_L = \alpha_L u$) is proportional to $\sqrt{k}$. With these assumptions, (1) becomes a stochastic partial differential equation (i.e., with stochastic parameters). Gelhar shows that the average transport process is no longer diffusive at small time, non-Fickian terms being added to the global equation. Here, the average is no longer a depth average over a finite medium but an ensemble average over all the possible realizations of the medium. The two are equivalent for a given aquifer if the solute has invaded a large portion of the aquifer in the z direction in order to reach ergodicity. For large time (or large displacement distance along x), using spectral analysis and a first-order expansion of the stochastic partial differential equation, he obtains, with our notation,

$$D_A(\infty) = \bar{D}_L + \frac{\bar{u}^2}{\bar{K}^2\bar{D}_T}\int_{-\infty}^{\infty}\frac{S_{KK}(r)}{r^2}\,dr$$

where

- $\bar{D}_L$ mean longitudinal dispersion coefficient, $E[D_L]$;
- $\bar{D}_T$ mean transversal dispersion coefficient, $E(D_T)$;
- $\bar{u}$ mean velocity u, $E(u)$;
- $\bar{K}$ mean of the hydraulic conductivity of the medium, $E(K)$;
- $S_{KK}(r)$ spectrum (or 'power density spectrum') of the permeability K, defined by the inverse Fourier transform of the covariance function of K, equal to $(1/2\pi)\int_{-\infty}^{\infty} e^{-irs}\,\mathrm{Cov}\,[K(z+s), K(z)]\,ds$;
- r integration variable, called 'the wave number' of the spectral representation.

The covariance, with the assumption of stationarity, is defined by

$$\mathrm{Cov}\,[K(z+s), K(z)] = E[K(z+s)\cdot K(z)] - \{E[K(z)]\}^2$$

It is a function of the lag s only, not of z. Gelhar first recognizes that for D_A to be finite the spectrum $S_{KK}(r)$ must approach zero near the origin so that $\int_{-\infty}^{\infty}(S_{KK}(r)/r^2)\,dr$ will be finite. This seems to be the first recognition in the literature of the fact that any arbitrary statistical distribution of permeability does not automatically give rise to diffusive transport. Gelhar further shows that in order for a higher-order term in his equation to be finite, a second constraint must be imposed on the spectrum $S_{KK}(r)$, namely, that $\int_{-\infty}^{\infty} (S_{KK}(r)/r^4)\, dr$ be finite. He then suggests one form of the spectrum satisfying this constraint:

$$S_{KK}(r) = \frac{8\sigma_K^2 l^5 r^4}{3\pi(s + l^2r^2)^3} \tag{2}$$

which is the spectrum of the following covariance:

$$\mathrm{Cov}\,[K(z + s), K(z)] = \sigma_K^2\left(1 - \frac{5}{3}\frac{s}{l} + \frac{1}{3}\frac{s^2}{l^2}\right) e^{-s/l} \qquad s \geq 0$$

where σ_K^2 is the variance of the permeability and l is a parameter called the length scale of the medium.

With such a covariance for the permeability he obtains

$$D_A(\infty) = \bar{D}_L + \frac{\bar{u}^2}{3}\frac{\sigma_K^2}{\bar{K}^2}\frac{l^2}{\bar{D}_T} \tag{3}$$

As in Marle's analysis, the asymptotical dispersion coefficient in (3) is the sum of the mean local longitudinal coefficient and a term related to the variance of the permeability divided by the mean local transverse dispersion coefficient. Gelhar also develops an expression for the dispersion coefficient at small and intermediate values of time, which is a very important achievement. This point will be discussed later.

RANDOM MOTION MODEL

G. Matheron (unpublished results, 1975) uses another representation of transport to solve the same problem: he follows the movement through the porous medium of a tracer particle located at point (x_0, z_0) at time $t = 0$ and a point (X_t, Z_t) at some later time t. Using the assumptions stated earlier (i.e., horizontal flow, horizontal velocity $u(z)$ being a weakly stationary random function, and constant dispersion coefficients), we can write

$$\begin{aligned} X_t &= x_0 + \xi_t + \int_0^t u(Z_\tau)\, d\tau \\ Z_t &= z_0 + \zeta_t \end{aligned} \tag{4}$$

where ξ_t and ζ_t represent the total dispersion process defined by a Brownian motion, i.e, Gaussian stochastic process of zero mean having a variance proportional to time:

$$\sigma_\xi^2 = 2D_L\, t$$
$$\sigma_\zeta^2 = 2D_T\, t$$

It has been shown [*Kolmogorov*, 1931] that this representation of transport is equivalent to the convection-diffusion equation (1) when the dispersion coefficients D_L and D_T are constant. The expectation of the concentration C at location (x_1, z_1) and time t_1 is equal to the probability density of the particle at (x_1, z_1, t_1).

This approach should not be confused with the classical 'random walk' model developed in diffusion theory [*Sposito et al.*, 1979]. In the latter model the scale is that of a molecule, and the movement is due to molecular collisions giving rise to true Brownian motion. In our case the scale is much larger because the seepage velocity u is a macroscopic quantity representing an average over many pores. In our model, Brownian motion represents hydrodynamic dispersion, not only molecular diffusion, as in the classical model.

The 'particle' that we will follow has no actual physical existence but is a mathematical concept. We only claim that the dispersion equation (1) and the system of equations (4) are mathematically strictly equivalent: solving (4) is the same as solving (1) except that the mathematical analysis is simpler.

In (4), X_t and Z_t are stochastic processes; solving (1) is therefore equivalent to finding the probability density functions of X_t and Z_t. We will focus on the determination of the first two moments of X_t. If transport in the stratified medium is to be Fickian, then the variance of X_t should be proportional to time:

$$\sigma_{X_t}^2 = 2\, D_A\, t \tag{5}$$

The determination of $\sigma_{X_t}^2$ is given in Appendix 1. We find that

$$\sigma_{X_t}^2 = 2D_Lt + \dot{I}(t) \tag{6}$$

where

$$\dot{I}(t) = 2\int_0^t (t - \tau)\frac{1}{2(\pi D_T\tau)^{1/2}}\int_{-\infty}^{+\infty} e^{-(s^2/4D_T\tau)}\,\mathrm{Cov}\,(s)\, ds\, d\tau$$

Cov (s) being the covariance function of the random velocity $u(z)$.

If we take the Laplace transform of $\dot{I}$, we obtain (see Appendix 2)

$$\Lambda(\dot{I}) = \frac{2}{p^{5/2}\sqrt{D_T}}\, Y[(p/D_T)^{1/2}] \tag{7}$$

where p is the Laplace variable,

$$Y(p) = \int_0^\infty e^{-ps}\,\mathrm{Cov}\,(s)\, ds$$

is the Laplace transform of the covariance function of the velocity u, and Λ denotes the Laplace transform. From these expressions one can show that an arbitrary covariance function of the velocity will not produce a Fickian transport, i.e., that $\dot{I}$ will not necessarily always be linearly proportional to time. For instance, if we choose a Gaussian covariance function, such as

$$\mathrm{Cov}\,[s] = \sigma_u^2 \exp\left(-\frac{m^2}{2}\left(\frac{s}{l}\right)^2\right)$$

where l is the length scale, m is a dimensionless constant, and σ_u^2 is the velocity variance, then we obtain

$$\dot{I} = 2\sigma_u^2\frac{l}{m}\left[\frac{1}{3D_T^2}\left(\frac{l^2}{m^2} + 2D_Tt\right)^{3/2} - \frac{1}{3D_T^2}\left(\frac{l}{m}\right)^3 - \frac{l}{mD_T}t\right]$$

Clearly, $\dot{I}$ (or $\sigma_{X_t}^2$) is a function of $t^{3/2}$ and not of t; the transport is therefore non-Fickian for all time values.

This result is puzzling because a Gaussian covariance function for the velocity (or for the permeability, if we assume a linear relationship between permeability and velocity, i.e., a constant gradient) seems a priori very acceptable. We can pursue the analysis further and, with our assumptions, give necessary and sufficient conditions for the transport to be Fickian either asymptotically or for all time values.

Asymptotic Fickian Transport

Asymptotic Fickian behavior means that for $t \to \infty$ we expect $\dot{I}(t) \to At$, where A is a constant. But this is equivalent to

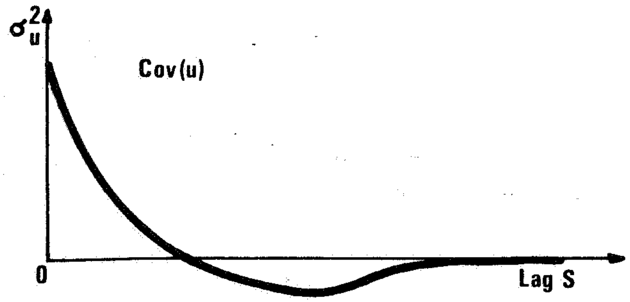

Fig. 2. Covariance function with a hole effect.

saying that in the Laplace transform domain we expect

$$\begin{aligned} \Lambda(\dot{I}) &\rightarrow \Lambda(At) \qquad \text{as } p \rightarrow 0 \\ \Lambda(\dot{I}) &\sim A/p^2 \qquad \text{as } p \rightarrow 0 \end{aligned} \tag{8}$$

Returning now to (7), we see that if $Y(0)$ is not equal to zero (e.g., $Y(0) = a$), then $\Lambda(\dot{I}) \rightarrow (2a/\sqrt{D_T})(1/p^{5/2})$ for $p \rightarrow 0$, which means that $\dot{I}$ becomes proportional to $t^{3/2}$ for large t: the asymptotic transport process is non-Fickian.

However, the condition $Y(0) = 0$ is not sufficient. Equation (7) shows indeed that $Y(p)$ must behave linearly in p near the origin for (8) to hold; if

$$Y(p) \rightarrow Bp \qquad \text{for} \quad p \rightarrow 0$$

B being a constant, then

$$\Lambda(\dot{I}) \rightarrow 2B/D_T p^2$$

$$\dot{I}(t) \rightarrow (2B/D_T)t \qquad \text{for} \quad t \rightarrow \infty$$

Recalling that

$$Y(0) = \int_0^\infty \text{Cov}\,(s)\, ds$$

these two conditions can be expressed as follows:

1. The integral of the covariance function of the velocity (or the permeability) must be zero.
2. The Laplace transform of this covariance function must behave linearly in p in the vicinity of the origin within the Laplace transform domain.

These assumptions are quite strong. In particular, condition 1 states that the covariance function must become negative, a behavior sometimes called the 'hole effect' in geostatistics, in which a positive correlation at small distances is followed by a negative correlation at larger distances, as shown in Figure 2.

Let us return for a moment to the work of Gelhar and Marle quoted earlier. Gelhar uses the following covariance for the permeability:

$$\text{Cov}_K\,(s) = \sigma_K^{\,2}\left(1 - \frac{5}{3}\frac{s}{l} + \frac{1}{3}\frac{s^2}{l^2}\right)e^{-s/l}$$

which exhibits the above hole effect.

If we assume the velocity to be proportional to the permeability with a constant gradient J and constant kinematic porosity ω, then the covariance of the velocity is simply multiplied by J^2/ω^2, or $\bar{u}^2/\bar{K}^2$, with $\bar{u} = E(u)$ and $\bar{K} = E(K)$. If we compute the Laplace transform of the covariance of this velocity, we find

$$Y(p) = \bar{u}^2 \frac{\sigma_K^{\,2}}{\bar{K}^2} \frac{p(3pl + 1)}{3l(p + 1/l)^3} \tag{9}$$

This expression satisfies our two conditions:

Condition 1

$$Y(0) = 0$$

Condition 2

$$Y(p) \sim \left(\bar{u}^2 \frac{\sigma_K^{\,2}}{\bar{K}^2}\frac{l^2}{3}\right)p \qquad \text{as} \quad p \rightarrow 0 \tag{10}$$

Furthermore, if we combine (9), (7), and (6), we find the following asymptotic expression for the longitudinal macrodispersion coefficient:

$$D_A(\infty) = D_L + \frac{\bar{u}^2}{3}\frac{\sigma_K^{\,2}}{\bar{K}^2}\frac{l^2}{D_T} \tag{11}$$

which is exactly the same as the one given by Gelhar in (3). This shows that the asymptotic transport process is governed only by the fluctuations of the velocity along the vertical and not by variations of the local dispersivity, which Gelhar considers in his first-order approximation.

It can also be shown (see Appendix 3) that our condition 2, which requires the Laplace transform of the velocity covariance function to behave linearly in p near the origin, is equivalent to the first condition obtained by Gelhar, i.e, that the integral $\int_{-\infty}^{+\infty}(S_{KK}(r)/r^2)\, dr$ of the power density spectrum of the velocity (or permeability) be finite. Furthermore, one can also show that these two conditions are satisfied if and only if the velocity (or permeability) is the derivative of a stationary random function, which is a very strong requirement.

Concerning Marle's result, it can also be shown that his assumption of a finite medium in the z direction, with no-flow boundaries, is equivalent to an assumption of an infinite periodic medium, the sequence of permeabilities of the medium being repeated by symmetry on each side of the boundaries. The covariance of the permeability (or velocity) is then periodic; one can then show that conditions 1 and 2, given earlier for the existence of asymptotic Fickian behavior, are satisfied.

Fickian Transport at All Times

If we want the transport process to remain Fickian for all values of time, integral $\dot{I}$ in (6) should always be a linear function of time t:

$$\dot{I} = At$$

A being a constant. In the Laplace transform domain this implies

$$\Lambda(\dot{I}) = A/p^2$$

Given (7), this means that

$$Y(p) = Bp$$

where B is another constant. Returning to the original domain, we find that

$$\text{Cov}\,[u] = B\frac{d}{ds}\delta(s)$$

The covariance of the velocity should be proportional to the first derivative of a Dirac function δ, thus showing no spatial correlation. But this is impossible, since the derivative of a Dirac function is not acceptable as a covariance function. With our assumption, the dispersion equation will never correctly represent the transport process for all times.

WHAT HAPPENS IF THE VELOCITY IS NOT PARALLEL TO THE STRATIFICATION?

We will now assume that the seepage velocity has two components: (1) a random horizontal component $u = u(z)$, which is a stationary stochastic process as before and (2) a constant vertical component v. In other words, the direction of flow is no longer parallel to the stratification. It is easy to show that such a situation is very likely to occur in nature, the case of zero velocity being only the exception.

Consider, for instance, an infinite section of layered porous medium, each side forming an angle with the vertical direction as shown in Figure 3. If different head values h_1 and h_2 are applied as boundary conditions on each side of the infinite section, flow will take place with velocity components in both the x and z directions. As x and z are the principal directions of the permeability tensor in the stratified medium, we can write Darcy's law as

$$U_x = -K_x(z)\frac{\partial h}{\partial x}$$

$$U_z = -K_z(z)\frac{\partial h}{\partial z}$$

where U_x and U_z represent the component of the velocity parallel and perpendicular, respectively, to the stratification. As $\partial h/\partial x$ is constant, U_x is constant inside a given stratum but varies with z. On the other hand, conservation of mass implies that U_z must be constant for all strata. This means that $\partial h/\partial z$ varies from stratum to stratum. Such a medium has therefore a constant vertical velocity component v and a random horizontal one $u(z)$. The local gradient inside the medium varies in both direction and magnitude as a function of z.

If we can assume, as before, that the local dispersion coefficients are constant, the convection-diffusion equation becomes

$$D_L\frac{\partial^2 C}{\partial x^2} + D_T\frac{\partial^2 C}{\partial z^2} - u(z)\frac{\partial C}{\partial x} - v\frac{\partial C}{\partial z} = \frac{\partial C}{\partial t} \tag{12}$$

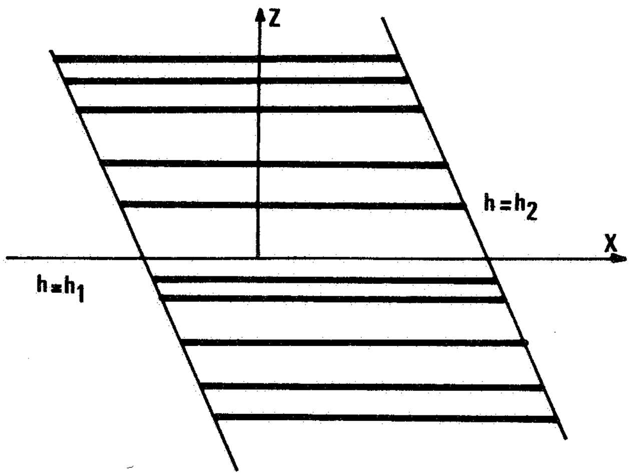

Fig. 3. Section of layered porous medium with sloping sides.

Note that in (12) we still assume that x and z are the principal directions of the dispersion tensor with principal coefficients equal to D_L and D_T. The true longitudinal and transverse directions and coefficients are different because the seepage velocity vector $\mathbf{u} = (u, v)$ is no longer parallel to either x or z. Therefore (12) is strictly correct only if D_L and D_T are equal to each other; otherwise, our result will be only approximate.

The equivalent random motion model now becomes

$$\begin{aligned} X_t &= x_0 + \xi_t + \int_0^t u(Z_\tau)\,d\tau \\ Z_t &= z_0 + \zeta_t + vt \end{aligned} \tag{13}$$

We can, as before, compute the variance of the position of the particle. From Appendix 4 we find

$$\sigma_{X_t}^2 = 2D_Lt + \dot{I}(t)$$

where

$$\dot{I}(t) = 2\int_0^t (t-\tau)\int_{-\infty}^{+\infty} g_\tau(s - v\tau)\,\mathrm{Cov}\,(s)\,ds\,d\tau \tag{14a}$$

Cov (s) is the covariance function of the horizontal velocity $u(z)$, and

$$g_\tau(y) = \frac{1}{2(\pi D_T\tau)^{1/2}}\exp\left(-\frac{y^2}{4D_T\tau}\right) \tag{14b}$$

is the Gaussian distribution function. Taking again the Laplace transform of $\dot{I}(t)$ (see Appendix 4), we find

$$\Lambda(\dot{I}) = \frac{1}{p^2(pD_T + v^2/4)^{1/2}}\left[Y\left(\frac{(pD_T + v^2/4)^{1/2} - v/2}{D_T}\right) + Y\left(\frac{(pD_T + v^2/4)^{1/2} + v/2}{D_T}\right)\right] \tag{15}$$

where $Y(p)$ is the Laplace transform of the covariance of horizontal velocity u.

Asymptotic behavior for large t will be obtained for $p \to 0$:

$$\Lambda(\dot{I}) \sim \frac{2}{vp^2}\left[Y(0) + Y\left(\frac{v}{D_T}\right)\right]$$

$$\dot{I}(t) \sim \frac{2}{v}\left[Y(0) + Y\left(\frac{v}{D_T}\right)\right]t$$

Transport will then be Fickian under the single very reasonable assumption that

$$Y(0) = \int_0^\infty \mathrm{Cov}\,(s)\,ds$$

is finite. The asymptotic macrodispersion coefficient parallel to the stratification is then given by

$$D_A(\infty) = D_L + \frac{1}{v}\left[Y(0) + Y\left(\frac{v}{D_T}\right)\right] \tag{16}$$

It is important to recognize that $D_A(\infty)$ is not the longitudinal macrodispersion coefficient because the latter is defined in the direction of the velocity vector $\mathbf{u}$ not parallel to the stratification. Thus $D_A(\infty)$ must be viewed merely as a directional dispersion coefficient defined in the direction parallel to the strata.

In the absence of local dispersion ($D_L = D_T = 0$) it is easy to

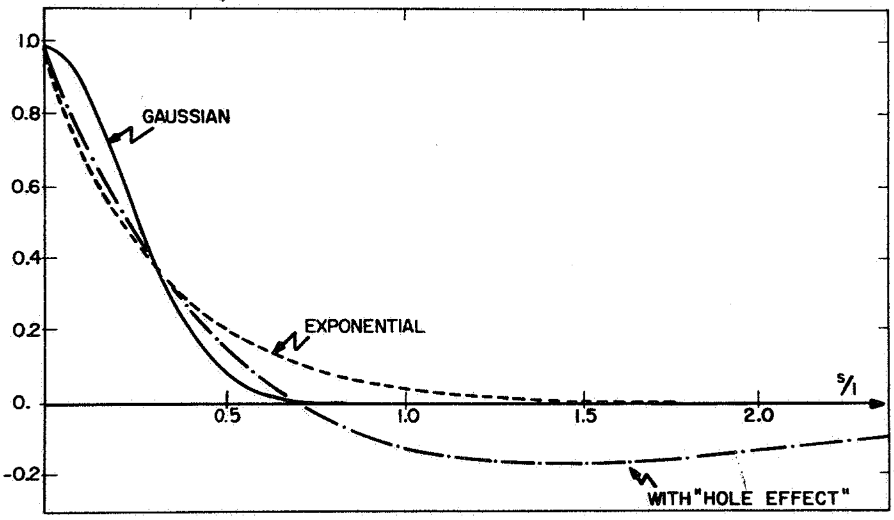

Fig. 4. Covariance functions of the velocity u as a function of the lag s/l (with hole effect (19a), exponential (20a), Gaussian (21a)).

show that the asymptotic behavior is still Fickian, and that (see Appendix 4)

$$D_{A_0}(\infty) = \frac{1}{v} Y(0) = \frac{1}{v}\int_0^{+\infty} \text{Cov}(s)\, ds$$

We can then compare the directional macrodispersion coefficients D_A and D_{A_0}:

$$D_A(\infty) = D_{A_0}(\infty) + D_L + \frac{1}{v} Y\left(\frac{v}{D_T}\right)$$

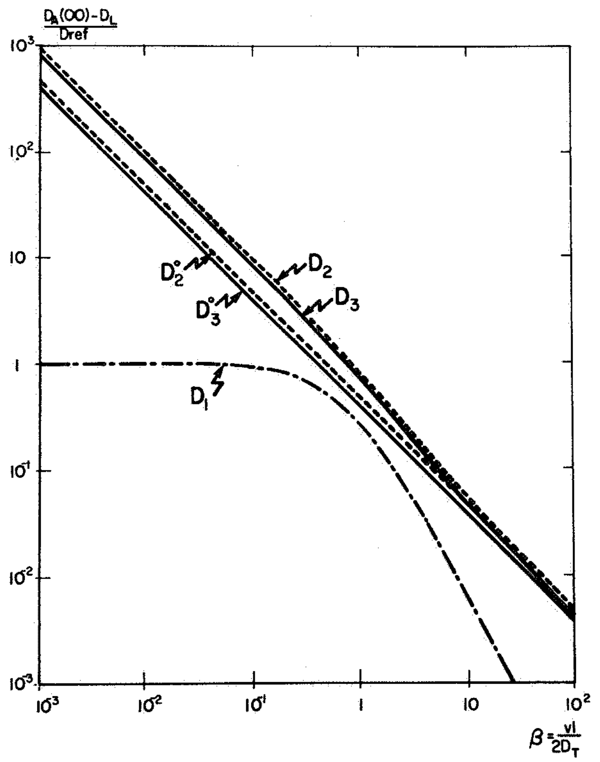

Fig. 5. Asymptotic dispersion coefficient as a function of the vertical component of velocity, where D_1 is the covariance with hole effect, D_2 is the exponential covariance, D_2^0 is the exponential covariance with no local dispersion, D_3 is the Gaussian covariance, and D_3^0 is the Gaussian covariance with no local dispersion.

As $Y(0) > Y(v/D_T)$, we can write

$$D_{A_0}(\infty) + D_L \le D_A(\infty) \le 2D_{A_0}(\infty) + D_L \qquad \forall \ D_T \tag{17}$$

$D_A(\infty)$ is of the order of $D_{A_0}(\infty)$. This result is very interesting, since it shows that the transverse local dispersion coefficient is not a very important factor in determining the value of the asymptotic directional macrodispersion coefficient parallel to the strata, contrary to the results we have obtained earlier in the one-dimensional case, where flow was strictly parallel to the stratification.

EXAMPLE

Let us return for a moment to the covariance function (2) proposed by Gelhar and used in the previous case. Its Laplace transform has been computed in (9). Using this expression in (15), we find, asymptotically,

$$D_A(\infty) = D_L + \frac{\bar{u}^2}{3} \frac{\sigma_K^2}{\bar{K}^2} \frac{l^2}{D_T}\left[\frac{1 + 3\, vl/D_T}{(1 + vl/D_T)^3}\right] \tag{18}$$

For $v = 0$ this expression is identical to (11). The influence of v is to decrease the value of $D_A(\infty)$, as the function $(1 + 3x)/(1 + x)^3$ decreases continuously with x for $x > 0$.

In order to get an impression of the influence of v we will compare this result with values given by Gelhar and compute the dispersivity:

$$A(\infty) = \frac{D_A(\infty) - D_L}{\bar{u}} = \frac{\sigma_K^2}{\bar{K}^2} \frac{l^2}{3\alpha_T}\left[\frac{1 + 3(l/\alpha_T)\mu}{(1 + (l/\alpha_T)\mu)^3}\right]$$

where $\mu = v/\bar{u}$ is the ratio of the vertical/horizontal velocity and α_T is the transverse dispersivity ($\alpha_T = D_T/\bar{u}$), although our development assumes D_T constant. We will use the same numerical values suggested by Gelhar:

$$\sigma_K^2/\bar{K}^2 = 0.25 \qquad l = 1.5 \text{ m} \qquad \alpha_T = 1 \text{ cm}$$

which gives

μ	$A(\infty)$, m
0	18.75
1/1000	17.81
1/100	6.56
1/10	0.21
1/1	2.4410^{-3}

This shows not only that the existence of a vertical velocity component makes asymptotic Fickian behavior in the direction of the layering much more likely but also that the value of the asymptotic directional macrodispersion coefficient is greatly reduced as compared to the case where $v = 0$, even if v is quite small (e.g., $E(u)/v = 1/100$).

This strong dependence of $A(\infty)$ on the flow regime suggests that it will generally be impossible to characterize a medium by constant macrodispersivities because the latter will vary with the velocity field.

The asymptotic directional macrodispersion coefficient also depends on the covariance function used to describe the velocity (or permeability) of the stratified medium. We will compare three covariance functions of the velocity and give $D_A(\infty)$ as a function of the dimensionless parameter $\beta = vl/2D_T$:

Function 1

$$\mathrm{Cov}\,(s) = \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\left(1 - \frac{5}{3}\frac{s}{l} + \frac{1}{3}\frac{s^2}{l^2}\right)e^{-s/l} \tag{19a}$$

already discussed with the hole effect, giving

$$D_A(\infty) = D_L + \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\frac{l^2}{3D_T}\left[\frac{1+6\beta}{(1+2\beta)^3}\right] \tag{19b}$$

Function 2

$$\mathrm{Cov}\,(s) = \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}e^{-n(s/l)} \tag{20a}$$

(simple exponential, l being the length scale and n a dimensionless constant) giving

$$D_A'(\infty) = D_L + \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\frac{l^2}{3D_T}\times\frac{3}{2\beta}\left[\frac{1}{n} + \frac{1}{2\beta+n}\right] \tag{20b}$$

Function 3

$$\mathrm{Cov}\,(s) = \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\exp\left(-\frac{m^2}{2}\frac{s^2}{l^2}\right) \tag{21a}$$

(Gaussian, already discussed), giving

$$D_A''(\infty) = D_L + \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\frac{l^2}{3D_T} \times\frac{3(\pi/2)^{1/2}}{2m\beta}\left[1 + \exp(2\beta^2/m^2)\,\mathrm{erfc}\left(\sqrt{2}\,\frac{\beta}{m}\right)\right] \tag{21b}$$

The three covariances, normalized by $\bar{u}^2\ (\sigma_K{}^2/\bar{K}^2)$, have been plotted in Figure 4. The resulting dispersion coefficients have been plotted in Figure 5, where $(D_A(\infty) - D_L)/D_{\mathrm{ref}}$ is taken as a function of $\beta = vl/2D_T$, and

$$D_{\mathrm{ref}} = \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\frac{l^2}{3D_T}$$

The latter is the asymptotic dispersion coefficient for the first covariance function (with hole effect) in the case of horizontal flow. From now on, D_{ref} will be used as a reference for comparison of all results.

In the exponential and Gaussian covariances we have chosen the constants n and m, so that for all three covariance functions one gets

$$\frac{\mathrm{Cov}\,(s)}{\bar{u}^2(\sigma_K{}^2/\bar{K}^2)} = \frac{1}{e} \qquad \text{for} \quad s/l = 0.31703$$

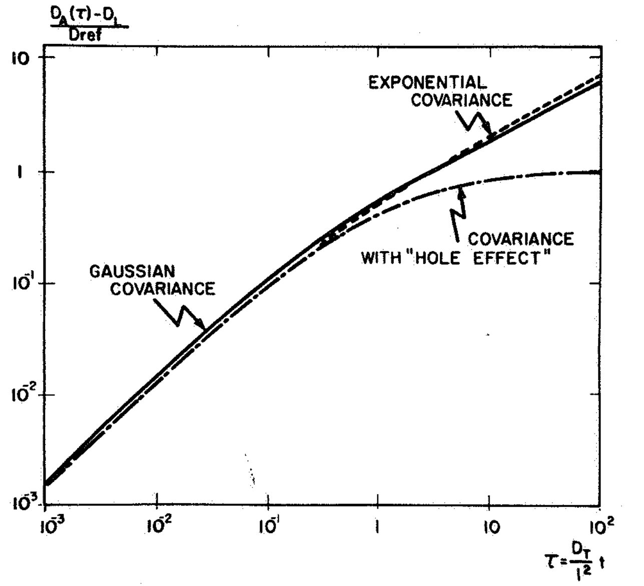

Fig. 6. Equivalent dispersion coefficient as a function of time (flow strictly parallel to stratification).

as suggested by Gelhar. This gives $n = 3.154$ and $m = 4.461$.

Figure 5 also shows the value of the asymptotic dispersion coefficient $D_{A_0}(\infty)$ for the theoretical case where $D_L = D_T = 0$:

For covariance (19a) (with hole effect)

$$D_{A_0}(\infty) = 0$$

For covariance (20a) (exponential)

$$D_{A_0}(\infty) = \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\frac{l}{nv} = D_{\mathrm{ref}}\times\frac{3}{2n\beta}$$

For covariance (21a) (Gaussian)

$$D_{A_0}(\infty) = \bar{u}^2\frac{\sigma_K{}^2}{\bar{K}^2}\frac{l}{mv}\left(\frac{\pi}{2}\right)^{1/2} = D_{\mathrm{ref}}\times\frac{3(\pi/2)^{1/2}}{2m\beta}$$

Although the three covariance functions show (Figure 4) a similar behavior at small values of s/l ($s/l < 0.5$), the corresponding asymptotic directional dispersion coefficient may be very different, especially for the covariance with the hole effect, whereas the exponential and Gaussian covariance functions give very similar results. As the accurate determination of the true covariance function of the permeability of a layered aquifer will always be difficult, especially for large s/l, this makes the a priori prediction of asymptotic macrodispersion a very difficult problem. Note also that $D_A(\infty)$ is very close to $D_{A_0}(\infty)$, showing that the local transverse dispersion D_T does not have a very important effect on the macroscale as far as dispersion parallel to the strata is concerned.

VARIATION OF THE EQUIVALENT LONGITUDINAL MACRODISPERSION COEFFICIENT WITH TIME OR SPACE: THE SCALE EFFECT

So far, we have considered only the value of the asymptotic directional macrodispersion coefficient. Our analysis gives, however, the variance of the position of a solute particle (for a pulse injection of tracer at $t = 0$) as a function of time for all t: expression (6) for zero vertical velocity, or (14) for nonzero vertical velocity. These expressions predict Fickian behavior, if any, only as $t \to \infty$.

Suppose we erroneously assumed that the dispersion equation was valid, then we could define, for any given time t_1, an 'equivalent directional macrodispersion coefficient' as

$$D_A(t_1) = \sigma_{x_t}^2(t_1)/2t_1 \tag{22}$$

This coefficient would, of course, be a function of the time t_1 (or of the distance traveled, $x_1 = E(u)t_1$). It would be the best directional coefficient to introduce into a dispersion equation as an attempt to represent at time t_1 the layered medium as an 'equivalent homogeneous medium' defined by an 'equivalent dispersion coefficient' parallel to the stratification and related to an average seepage velocity:

$$D_A(t_1)\frac{\partial^2 C}{\partial x^2} + D_T\frac{\partial^2 C}{\partial z^2} - E(u)\frac{\partial C}{\partial x} - v\frac{\partial C}{\partial z} = \frac{\partial C}{\partial t} \tag{23}$$

Note that $D_A(t_1)$ is the constant coefficient that should be used in (23) during the entire interval $t \in [0, t_1]$ in order to best reproduce the observation at time t_1 of the solute concentration in space (injected as a pulse at time $t = 0$). For a different time t_2, $D_A(t_2)$ would have the same definition. In other words, $D_A(t_1)$ is not a time-dependent coefficient in (23) but rather an equivalent constant coefficient capable of interpreting, with the aid of (23), the observation made at time t_1 in space.

Now when a real tracer experiment is performed in the field, this is what is usually done. If the tracer is injected as a pulse, the results will be interpreted at a first observation well in terms of a first dispersion coefficient D_1. At a second observation well, at a greater distance and greater time, another dispersion coefficient D_2 will usually be determined. In (23), D_1 and D_2 will be constant for $t \in [0, t_1]$ and $[0, t_2]$, respectively, t_1 and t_2 being the time at which the observations in the two wells are made (assuming that the observations are made in each well within small time intervals surrounding t_1 and t_2). Usually, $D_2 > D_1$. This fact has been reported very often in the literature [e.g., *Fried,* 1975] and is known as the 'scale effect.'

It is also possible to define a time-dependent dispersion coefficient by

$$D'(t) = \frac{1}{2}\frac{d}{dt}\sigma_{x_t}^2$$

(Gelhar). This coefficient would then vary with time in (23). We will not use this approach here.

Note also that any of those time-dependent dispersion coefficients will only make the second moment of the concentration predicted by (23) match the true one; since the process is not Fickian, higher-order moments will not be correct, i.e., the shape of the tracer plume will not be correctly represented by (23). See Gelhar's approach.

$D_A(t)$ WITHOUT A VERTICAL VELOCITY COMPONENT

Expressions (6) and (22) make it possible to determine directly $D_A(t)$, given an expression for the covariance function. To get a closed-form solution, one can either perform the two integrations in (6) or, alternatively, determine the inverse Laplace transform of (7). We have used both methods (see Appendix 5); although the integrations are quite lengthy, they present no major difficulties. This was done for the three covariance functions (19*a*), (20*a*), and (21*a*) used previously and expressed as a function of the dimensionless time $\tau = D_T t/l^2$. We obtain

For the covariance with hole effect (19*a*)

$$D_A(\tau) = D_L + \bar{u}^2\frac{\sigma_K^2}{\bar{K}^2}\frac{l^2}{3D_T}[1 + e^\tau(1 - \operatorname{erf}(\sqrt{\tau}))(4\tau - 4 + 3/\tau) - 4\sqrt{\tau}/\pi + 6/\sqrt{\pi\tau} - 3/\tau] \tag{24}$$

For the exponential covariance (20*a*)

$$D_A'(\tau) = D_L + \bar{u}^2\frac{\sigma_K^2}{\bar{K}^2}\frac{l^2}{3D_T}[4\sqrt{\tau}/n\sqrt{\pi} - 3/n^2 + 6/n^3\sqrt{\pi\tau} - 3/n^4\tau + \frac{3}{n^4\tau}e^{n^2\tau}\operatorname{erfc}(n\sqrt{\tau})] \tag{25}$$

For the Gaussian covariance (21*a*)

$$D_A''(\tau) = D_L + \bar{u}^2\frac{\sigma_K^2}{\bar{K}^2}\frac{l^2}{3D_T}\left[\frac{1}{m^4\tau}(1 + 2m^2\tau)^{3/2} - \frac{1}{m^4\tau} - \frac{3}{m^2}\right] \tag{26}$$

We know that only the first covariance will produce a finite $D_A(t)$ for $t \to \infty$, since the other two covariances do not satisfy the necessary requirements that have been established earlier. An expression similar to the one given in (24) has also been obtained by Gelhar, using a different approach.

In Figure 6 we have plotted $(D_A(\tau) - D_L)/D_{\text{ref}}$ versus τ for the three covariance functions. For $\tau < 1$, the three curves are almost identical. For $\tau > 1$, the curves for the exponential and Gaussian covariances, which are unbounded, depart from that of the covariance with the hole effect, which tends towards unity. This means that in the field, if the flow is strictly parallel to the stratification, it will be very difficult to predict $D_A(t)$ for large times from measurement made at small times. Until the asymptote is reached, one cannot say whether it actually exists unless the true covariance is completely specified.

$D_A(t)$ WHEN THERE IS A VERTICAL VELOCITY COMPONENT

Expressions (14) and (22) lead directly to $D_A(t)$. However, only the first two covariance functions lead to a closed-form solution, whereas the integration for the Gaussian covariance must be performed numerically (see Appendix 5). The results are obtained in terms of two dimensionless parameters $\tau = D_T t/l^2$ and $\beta = vl/2D_T$ used previously. We obtain

For the covariance with the hole effect (19*a*)

$$\begin{aligned} D_A(\tau) = D_L + \bar{u}^2\frac{\sigma_K^2}{\bar{K}^2}\frac{l^2}{3D_T} \times \frac{1}{2\tau}\Bigg\{\frac{1}{(2\beta - 1)^4}\Bigg[&\frac{e^{-\beta^2\tau}}{\sqrt{\pi\beta}} \\ &\cdot (2\beta - 1)\sqrt{\tau}(4\tau\beta(2\beta^2 - 3\beta + 1) \\ &- 4\beta^2 - 12\beta + 1) \\ &+ \operatorname{erfc}(\beta\sqrt{\tau})(\tau(12\beta^2 - 8\beta + 1) + 4\beta^2 - 8\beta - 3) \\ &+ e^{-(2\beta-1)\tau}\operatorname{erfc}((1 - \beta)\sqrt{\tau})(4\tau^2(\beta - 1)^2 \\ &\cdot (2\beta - 1)^2 - 2\tau(2\beta - 1)(2\beta^2 - \beta - 2) \\ &- 4\beta^2 + 8\beta + 3)\Bigg] + \frac{1}{(2\beta + 1)^4}q\Bigg\} \end{aligned} \tag{27}$$

where q is the same as the bracketed expression in (27) but with $\beta \to -\beta$.

For the exponential covariance (20a)

$$D_A(\tau) = D_L + \bar{u}^2 \frac{\sigma_K^2}{\bar{K}^2} \frac{l^2}{3D_T} \left\{ \left[\frac{3e^{-\beta^2\tau}}{4\beta^2\sqrt{\pi\tau}} \left(\frac{1}{n} - \frac{1}{2\beta - n} \right) \right.\right.$$
$$+ \frac{3 \operatorname{erf}(\beta\sqrt{\tau})}{4\beta} \left(\frac{1}{n} - \frac{1}{2\beta - n} + \frac{1}{\tau} \left(\frac{3\beta - n}{2\beta^2(2\beta - n)^2} - \frac{\beta + n}{2\beta^2 n^2} \right) \right)$$
$$+ \frac{3}{4\beta} \left(\frac{1}{n} + \frac{1}{2\beta - n} - \frac{1}{\tau} \left(\frac{3\beta - n}{2\beta^2(2\beta - n)^2} + \frac{\beta + n}{2\beta^2 n^2} \right) \right)$$
$$\left.\left. + \frac{3e^{n(n-2\beta)\tau}}{2\tau n^2(2\beta - n)^2} \operatorname{erfc}\left((n - \beta)\sqrt{\tau}\right) \right] + q' \right\} \qquad (28)$$

where q' is the same as the bracketed expression in (28) but with $\beta \to -\beta$.

For the Gaussian covariance (21a)

$$D_A(\tau) = D_L + \bar{u}^2 \frac{\sigma_K^2}{\bar{K}^2} \frac{l^2}{3D_T} \int_0^\tau \left\{ 3\left(1 - \frac{x}{\tau}\right) \right.$$
$$\left. \cdot \exp\left(- \frac{2m^2\beta^2 x^2}{2m^2 x + 1} \right) \Big/ (2m^2 + 1)^{1/2} \right\} dx \qquad (29)$$

These results are plotted in Figures 7a, 8a, and 9a in terms of $(D_A(\tau) - D_L)/D_{\text{ref}}$ versus τ and the parameter β. The same results are plotted on Figures 7b, 8b, and 9b in terms of the ratio $(D_A(\tau) - D_L)/(D_A(\infty) - D_L)$. For a given β this ratio should tend towards unity as $\tau \to \infty$. Another dimensionless time group can also be used $\tau' = (v/l)t$, as $\tau' = 2\beta\tau$.

In Figures 7a and 7b the covariance (19a) with hole effect generates a curious behavior for $D_A(\tau)$: when β is small, $D_A(\tau)$ grows toward its asymptotic value monotonically, as in the absence of vertical velocity (for $\beta = 0$, the results are identical). For $\beta \geq 1$, however, $D_A(\tau)$ first increases beyond the value of $D_A(\infty)$ and then decreases with time towards $D_A(\infty)$. In other words, the directional macrodispersion coefficient is larger at early times than its asymptotic value. This is clearly due to the negative correlation of the velocities caused by the hole effect. When a highly permeable layer is surrounded by low-permeability material, the dispersion will initially be large and then decrease, as the solute invades the aquifer in the transverse direction, until ergodicity is reached. To our knowledge, such an effect in a field experiment has never been reported in the literature. One can therefore argue that such a covariance function is not very likely to appear in nature and that if the velocity is strictly parallel to the layers, Fickian behavior will generally not hold.

In Figures 8a and 8b (exponential covariance) the growth of $D_A(\tau) \to D_A(\infty)$ is monotonous in the range of the dimensionless time $\tau > 10^{-3}$, for $\beta > 10^{-1}$; however, for small β, $D_A(\tau)$ decreases at early times and then increases toward $D_A(\infty)$. This can be related to the positive correlation structure of the velocity at small distances. One should also note that when $\beta \leq 10^{-1}$, the dimensionless time at which $D_A(\infty)$ is reached can be very large (10^2 and more), meaning that in a tracer experiment the asymptotic value of $D_A(\infty)$ will often not be obtained during the performance of the test.

In Figures 9a and 9b (Gaussian covariance) the growth of $D_A(\tau) \to D_A(\infty)$ is seen to be always monotonous.

Practical Example

Suppose that a field experiment is planned to measure the dispersion coefficient in a layered system: Can these results be used to estimate the travel distance beyond which asymptotic Fickian behavior will be reached?

Two parameters are difficult to estimate from the usual data available on aquifers: (1) the type of covariance function to be used and (2) the length scale l of this covariance. According to unpublished results obtained by J. R. MacMillan (personal communication, 1979) from the analysis of data from wells (core permeability or electric resistivity) in a sandstone-type material, an exponential covariance seems reasonable with l in the range of a few meters. We will base our assumption on these results, taking $l = 1$ m.

In addition to parameters 1 and 2 above, one also needs estimates of D_T and D_L. If only molecular diffusion is considered, $D_T \sim 10^{-9}$ m^2/s. If local transverse dispersion is considered, laboratory data on cores indicate a dispersivity α_T in the range of 1 cm. D_T can then be computed from the average horizontal velocity by $D_T = \alpha_T E(u)$. We will also test a value of 10 cm. For D_L we will assume $D_L = \alpha_L E(u)$ with $\alpha_L = 1$ m. Concerning permeability, we will assume that $E(K_h) = 10^{-3}$ m/s, $E(K_v) = 10^{-4}$ m/s, $\sigma_K^2/\bar{K}^2 = 1$, with a kinematic porosity ω of 10%.

For the flow we will consider two cases, (1) experiment with the natural velocity of the water, assuming a horizontal gradient J of 0.5%, and a vertical gradient I of 0.1% and (2) artificial increase (e.g., by pumping or injection) of the horizontal and/or vertical velocity by a factor of 10 with respect to the natural situation.

Case A, natural flow conditions. The following figures can be easily computed from the above data:

$$\bar{u} = E(u) = \frac{J\bar{K}_h}{\omega} = 5 \times 10^{-5} \text{ m/s}$$

$$v = \frac{IK_v}{\omega} = 10^{-6} \text{ m/s}$$

$$\alpha_L = 1 \text{ m}$$

thus $D_L = \alpha_L \bar{u} = 5 \times 10^{-5}$ m^2/s,

$$\alpha_T = 0.01 \text{ m}$$

thus $D_T = \alpha_T \bar{u} = 5 \times 10^{-7}$ m^2/s,

$$\beta = \frac{v}{2D_T} = 1$$

$$\tau = \frac{D_T}{l^2} t = 5 \times 10^{-7} t$$

where t is in seconds.

Using Figures 5 and 8b, we can determine the macrodispersion coefficient in the horizontal direction as a function of time (or distance of travel). We give the apparent macrodispersivities $\alpha_A = D_A(\tau)/\bar{u}$ in Table 1.

Clearly, it will be very difficult to perform an experiment long enough to reach (within 95%) the asymptotic dispersivity (600 m and 140 days), and measurements made at much shorter distances drastically underestimate the asymptotic value. It appears that only natural tracers could be used to estimate $\alpha_A(\infty)$.

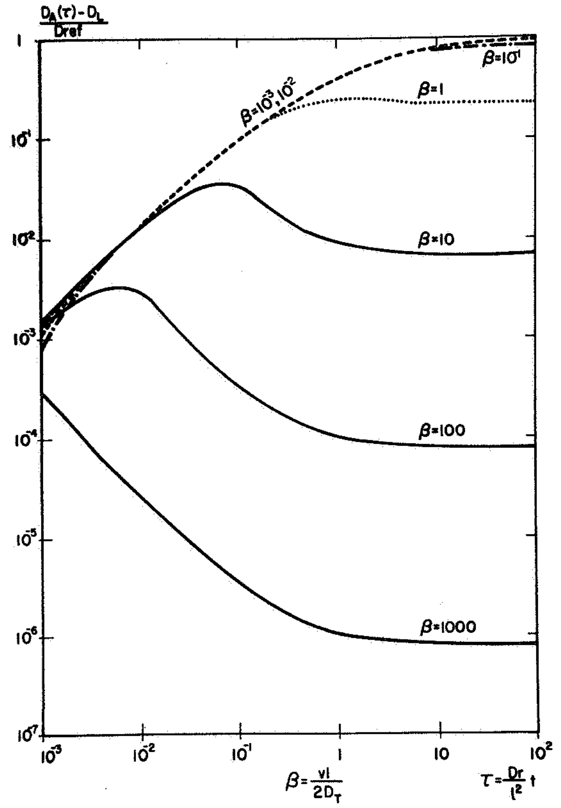

Fig. 7*a*. Equivalent dispersion coefficient as a function of time and vertical component of velocity (covariance with hole effect).

Case B, artificial flow conditions. We will test the influence of an artificial increase of the velocity in the aquifer (e.g., by pumping or injection). Would it then be easier to obtain the asymptotic macrodispersivity with the same field experiment? To explore the various possibilities, we will assume (1) that both u and v are simultaneously increased by a factor of 10, and as the local α_L and α_T are assumed constant, the local D_L and D_T will also be increased by a factor of 10, (2) that u, D_L, and D_T are increased by a factor of 10, while v remains constant, and (3) that u, D_L, and D_T remain constant while v is increased by a factor of 10. We obtain the results shown in Table 2.

It appears that increasing the horizontal velocity does not alleviate the difficulty. The experiment becomes feasible only if the vertical velocity is increased while the horizontal one remains constant. Aside from the difficulty in obtaining this situation in the field, the measured dispersivity (2.8 m) would not be representative of the true asymptotic dispersivity of the aquifer under natural flow conditions.

If we want to test the value of the transverse local dispersivity α_T, e.g., $\alpha_T = 0.1$ m instead of 0.01 m, we find for natural flow conditions (u, v, and D_L, as in case A) the same results as for case B (assumption 2): 900 days and 39 km. Since the $\alpha_T = 0.01$ m that we had chosen initially is already small, there is not much hope that this parameter may solve the problem.

Existence of Dispersivity

It is usual to assume that the coefficient of dispersion is a linear function of the velocity and to define the dispersivity α as $D = \alpha|u|$ (longitudinal or transverse). Does this relationship still hold for the directional macrodispersion coefficient?

Expressions (19*b*), (20*b*), and (21*b*) for the asymptotic values give the following (the problem to consider is whether D_T and v are linearly proportional to $\bar{u}$):

1. When $\bar{u}$ varies and v and D_T are proportional to $\bar{u}$ (i.e., constant direction of velocity), $D_A(\infty)$ is proportional to $\bar{u}$.
2. When $\bar{u}$ varies and v and D_T are constant, $D_A(\infty)$ is proportional to $\bar{u}^2$.
3. When $\bar{u}$ varies, v is constant, and D_T is proportional to $\bar{u}$, $D_A(\infty)$ has no simple relation with $\bar{u}$.

Now for the time-dependent directional macrodispersion $D_A(\infty)$, expressions (24) (25), and (26) (case without vertical velocity) show that $D_A(\tau) = \bar{a}_L\bar{u}$ if $D_T = \bar{a}_T\bar{u}$. However, as $\tau = D_Tt/l^2$, $D_A(t)$ is not proportional to $\bar{u}$. However, if instead of t we consider the average distance of displacement $\bar{x} = \bar{u}t$, then $D_A(\bar{x})$ is proportional to $\bar{u}$. In the case where the vertical velocity is not zero, (27), (28), and (29) show that the same result holds as long as both D_T and v are proportional to $\bar{u}$.

CONCLUSION

We have examined the conditions for the appearance of Fickian macrodispersion parallel to the layering of an unbounded stratified medium. It was possible to show that when the flow is strictly parallel to the stratification, Fickian behavior will, in general, not occur and that the usual convection-diffusion equation should not be used. This stems from the fact that the group of pure convection (i.e., no local dispersion, $D_L = D_T = 0$) does not cause mixing in this case. This conclusion may be particularly relevant for flow in certain types of fractured rocks, where parallel fractures may allow the propagation of dissolved species without considerable lateral mixing if the general flow direction is parallel to the fractures. If transverse local dispersion takes place (i.e., $D_T \neq 0$), Fickian behavior could eventually be reached asymptotically

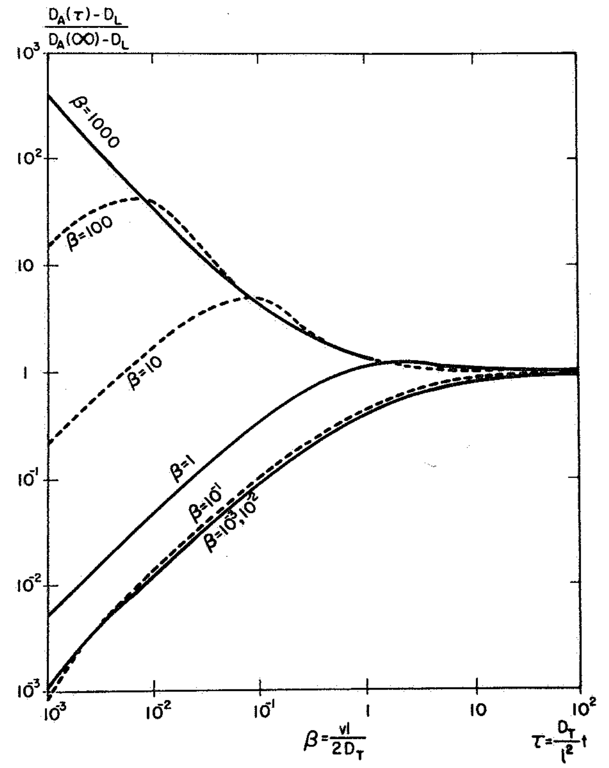

Fig. 7*b*. Same as Figure 7*a* but with equivalent dispersion coefficient as a ratio to its asymptotic value.

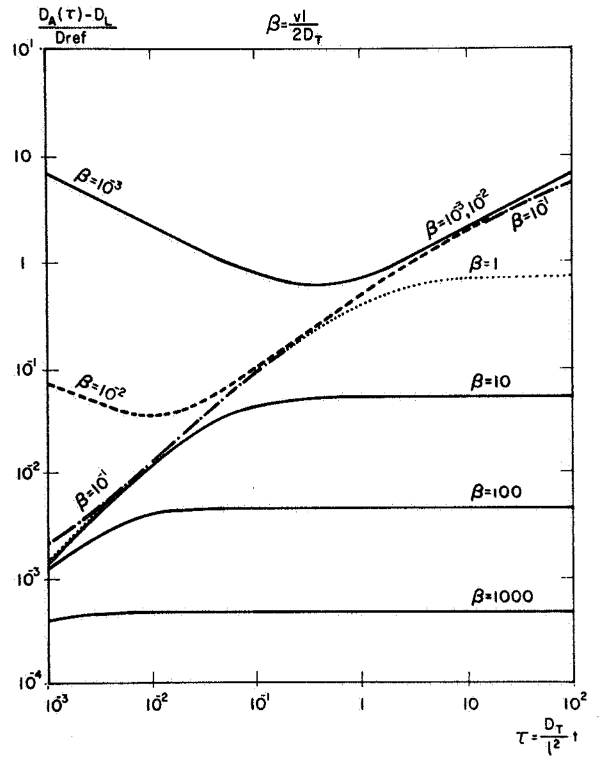

Fig. 8*a*. Equivalent dispersion coefficient as a function of time and vertical component of velocity (exponential covariance).

for large time or large displacement of the dissolved species. This, however, would only happen under the unrealistic requirement that the covariance function of the velocity (or of the permeability) exhibits a hole effect, so that its integral is zero and has a Laplace transform behaving linearly in the Laplace variable near the origin. For most reasonable covariance functions the behavior will not be Fickian, even asymptotically.

However, if the flow is not strictly parallel to the stratification (i.e., a perpendicular flow component, however small, is added), then the group of pure convection causes mixing, and Fickian behavior will take place asymptotically under the very reasonable assumption that the integral of the covariance of the parallel velocity component (or permeability) is finite. It was possible to show that the asymptotic directional macrodispersion coefficient, parallel to the layering, $D_A(\infty)$ depends more on the lateral mixing generated by the vertical velocity component than on the local transverse dispersion coefficient:

$$D_{A_0}(\infty) + D_L \leq D_A(\infty) \leq 2D_{A_0}(\infty) + D_L$$

where $D_{A_0}(\infty)$ is the hypothetical asymptotical directional macrodispersion coefficient without local dispersion ($D_L = D_T = 0$). Furthermore, the magnitude of $D_{A_0}(\infty)$ depends strongly on the magnitude of the perpendicular component of the velocity v, as

$$D_{A_0} = \frac{1}{v}\int_0^\infty \text{Cov}\,(s)\, ds$$

where Cov (s) is the covariance function of the parallel component of the velocity. This makes $D_A(\infty)$ depend very strongly on v.

The way in which this asymptotic dispersion coefficient is reached with time (or distance of travel) depends mainly on (1) the type of covariance function assumed, (2) the value of the transverse local dispersion coefficient D_T, and (3) the magnitude of the vertical velocity v.

Simple computations show that in a typical alluvial deposit the experimental determination of the asymptotic dispersion coefficient by tracer test may be meaningless unless the vertical gradient of head in the aquifer is significant (e.g., 1%) or the travel distance is very large (e.g., several hundreds of meters, which means that environmental tracers should be used). These results assume that the aquifer is of infinite thickness; however, they may be too restrictive if the aquifer is very thin. Further development is needed to include this effect of aquifer thickness in the analysis.

If D_T and v are well known, it is conceivable that a local tracer test in the field can be used to estimate the covariance function of the permeability (or rather, the parameters of such functions, assuming a given type, e.g., exponential). This may indeed be an indirect method of estimating covariances. Once this step is made, the asymptotic behavior could be predicted from nonasymptotic, local measurements. However, this idea remains highly questionable, since it assumes perfect second-order stationarity of the medium over large distances.

Indeed, even if an asymptotic Fickian behavior can be obtained as a result of the above mentioned mechanisms, it may not be applicable to real life situations: the time (or length of displacement) needed to obtain asymptotic behavior may be too large and may allow the tracer to encounter other aquifer heterogeneities (macrostructures, in the terminology of *Cherry et al.* [1979]), which can be viewed as a nonstationarity of the medium.

The proper method of modeling the movement of dissolved species in a layered aquifer remains therefore unsolved. As long as asymptotic behavior is not reached, the usual convection-diffusion equation does not hold. Making the dispersion coefficient D a function of time is only an artifact, valid approximately for a point source with a pulse injection

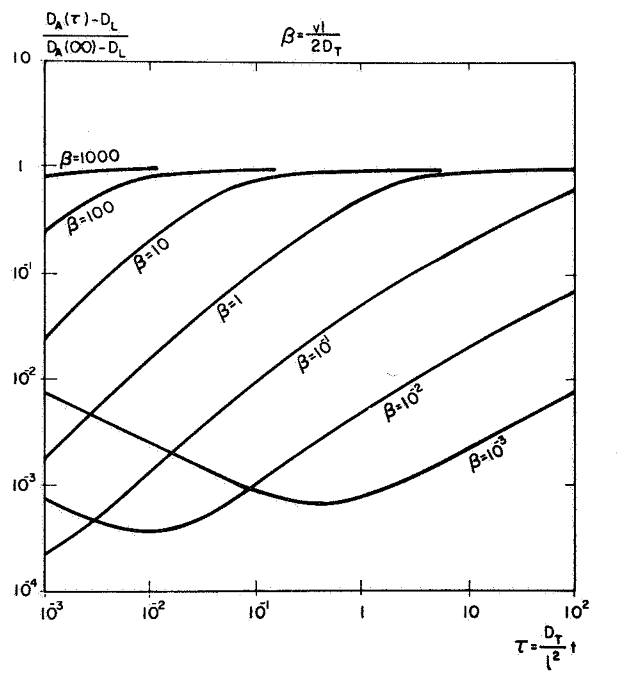

Fig. 8*b*. Same as Figure 8*a* but with equivalent dispersion coefficient as a ratio to its asymptotic value.

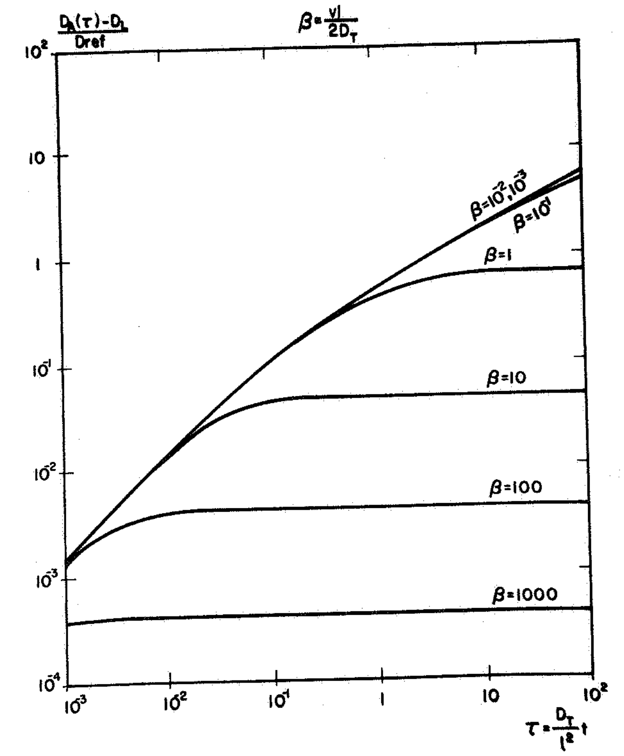

Fig. 9*a*. Equivalent dispersion coefficient as a function of time and vertical component of velocity (Gaussian covariance).

in time and giving only an approximate picture of the concentration at a given time t but not for all time between zero and t. A new simulation with another constant D is necessary for any new prediction at a different time, which makes the problem intractable. In any case, distributed sources would be very difficult to represent this way.

The present study of macrodispersion also provides an answer to the question why the usual convection-diffusion model predicts upstream migration of a solute from its injection point, when large dispersivities are used [*Simpson*, 1978; *de Marsily*, 1978]. Our study explains that this is due to the inapplicability of the dispersion equation for early time, especially if a single dispersion coefficient is used at all times.

A better mathematical formulation of the transport process in porous and fractured media, valid for all time, seems necessary. In the meantime a possible way is to include many more details in the description of the aquifer when modeling dispersion. Instead of assuming the existence of an equivalent homogenous medium, defined by an average permeability, one should try to represent in three dimensions the position and properties of each of the layers of the medium that can be identified [e.g., *Dieulin*, 1979]. This description can be seen as deterministic or even as stochastic (generated by stochastic sedimentation models). In each of these layers the appearance of asymptotic behavior will be faster (effect of the thickness), and thus the dispersion equation will be valid much earlier. Measurements of the dispersive property of the medium should be made on the same scale. Macrodispersion will then result from assembling these layers.

Although our results only apply to strictly layered media, it seems reasonable to extend these conclusions to many other media which are approximately stratified, such as lenticular alluvial deposits.

Appendix 1: Determination of the Variance of the Coordinate of the Particle in the Random Motion Model

We will assume for the sake of simplicity that at $t = 0$, $x_0 = z_0 = 0$. From (4) we have

$$X_t = \xi_t + \int_0^t u(Z_\tau)\, d\tau$$

$$Z_t = \zeta_t$$

Then

$$E[X_t] = E[\xi_t] + E\left[\int_0^t u(Z_\tau)\, d\tau\right]$$

As ξ_t is a Brownian motion with zero mean, we have

$$E[X_t] = \int_0^t E[u(Z_\tau)]\, d\tau$$

$$E[X_t] = \int_0^t E[u(z)]\, d\tau$$

$u(z)$ and Z_τ are independent

$$E[X_t] = tE[u]$$

We then have

$$\sigma_{X_t}^2 = \mathrm{Var}\,(X_t) = E[(X_t - tE[u])^2] = E[X_t^2] - t^2(E[u])^2$$

$$= E\left[\left(\xi_t + \int_0^t u(Z_\tau)\, d\tau\right)^2\right] - t^2(E[u])^2$$

$$= E[\xi_t^2] + E\left[\int_0^t u(Z_\tau)\, d\tau \int_0^t u(Z_{\tau'})\, d\tau'\right]$$

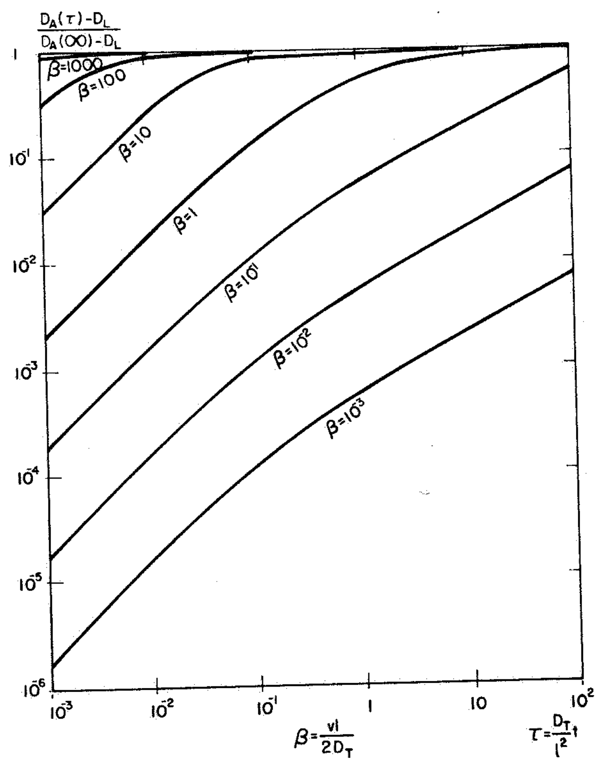

Fig. 9*b*. Same as Figure 9*a* but with equivalent dispersion coefficient as a ratio to its asymptotic value.

TABLE 1. Apparent Macrodispersivities for Natural Flow Conditions

Time of Observation, days	Average Distance of Travel, m	Apparent Macrodispersivity α_A, m
∞ (asymptotic)	∞	26.5
140*	600	25.2
14	60	10.6
1.4	6	2.1

*In order to get 95% of asymptotic dispersivity.

$$+ 2E\left[\xi_t \int_0^t u(Z_\tau)\, d\tau\right] - t^2(E[u])^2$$

$$= \mathrm{Var}\,(\xi_t) + \int_0^t \int_0^t E[u(Z_\tau)u(Z_{\tau'})]\, d\tau d\tau' - t^2(E[u])^2$$

as ξ_t and $\int_0^t u(Z_\tau)\, d\tau$ are independent (as the Brownian motions ξ_t, ζ_t, representing constant local dispersion coefficients D_L and D_T, are not functions of the velocity u). Then

$$\sigma_{x_t}^2 = 2D_L t + \int_0^t \int_0^t \{E[u(Z_\tau)u(Z_{\tau'})] - (E[u])^2\}\, d\tau' d\tau$$

This expression is symmetric with respect to τ and τ'. We can then write

$$\sigma_{x_t}^2 = 2D_L t + 2\int_0^t \int_\tau^t \{E[u(Z_\tau)u(Z_{\tau'})] - (E[u])^2\}\, d\tau' d\tau$$

The expected value E is taken over all possible values of the random variables $u(z)$, Z_τ, and $Z_{\tau'}$. Letting $p(u, Z_\tau, Z_{\tau'})$ be the joint probability distribution function of these three variables, we can write the expectation of any function F as

$$E[F] = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \int_0^{\infty} p(u, Z_\tau, Z_{\tau'})\, F(u, Z_\tau, Z_{\tau'})\, du\, dZ_{\tau'}\, dZ_\tau$$

However, as u and Z_τ or $Z_{\tau'}$ are independent and as Z_τ and $Z_{\tau'}$ represent the same Brownian motion process with $\tau' \geq \tau$, we can write

$$p(u, Z_\tau, Z_{\tau'}) = p(u) \times p(Z_\tau) \times p(Z_{\tau'}|Z_\tau)$$

Thus the first integral over u of the expectation for Z_τ and $Z_{\tau'}$ kept constant, is

$$\dot{I}_1 = \int_0^{\infty} p(u)\, [u(Z_\tau)u(Z_{\tau'}) - (E[u])^2]\, du$$

$$= \int_0^{\infty} p(u)u(Z_\tau)u(Z_{\tau'})\, du - (E[u])^2$$

which is by definition the covariance function of the velocity u, taken at the lag distance $Z_{\tau'} - Z_\tau$; $I_1 = \mathrm{Cov}\,(Z_{\tau'} - Z_\tau)$. We can now express the probability density function (pdf) of the Brownian motions Z_τ and $Z_{\tau'}|Z_\tau$. The first is normal with zero mean and variance $2D_T\tau$. The second is also normal with mean Z_τ and variance $2D_T(\tau' - \tau)$:

$$p(Z_\tau) = \frac{1}{2(\pi D_T \tau)^{1/2}} \exp\left(-\frac{Z_\tau^2}{4D_T\tau}\right)$$

$$p(Z_{\tau'}|Z_\tau) = \frac{1}{2(\pi D_T(\tau' - \tau))^{1/2}} \exp\left(-\frac{(Z_{\tau'} - Z_\tau)^2}{4D_T(\tau' - \tau)}\right)$$

We can then compute the second integral over $Z_{\tau'}$, for a given Z_τ:

$$I_2 = \frac{1}{2(\pi D_T(\tau' - \tau))^{1/2}} \cdot \int_{-\infty}^{+\infty} \exp\left(-\frac{(Z_{\tau'} - Z_\tau)^2}{4D_T(\tau' - \tau)}\right) \mathrm{Cov}\,(Z_{\tau'} - Z_\tau)\, dZ_{\tau'}$$

We define the variable $s = Z_{\tau'} - Z_\tau$ (as the velocity u is assumed stationary) and get

$$I_2 = \frac{1}{2(\pi D_T(\tau' - \tau))^{1/2}} \int_{-\infty}^{+\infty} \exp\left(-\frac{s^2}{4D_T(\tau' - \tau)}\right) \mathrm{Cov}\,(s)\, ds$$

Then the third integral over Z_τ reduces to 1, since $\dot{I}_2$ is not a function of Z_τ:

$$\dot{I}_3 = \frac{1}{2(\pi D_T \tau)^{1/2}} \int_{-\infty}^{+\infty} \exp\left(-\frac{Z_\tau^2}{4D_T\tau}\right) \dot{I}_2\, dZ_\tau = \dot{I}_2$$

The integral I_2, which is a function of $(\tau' - \tau)$, must be integrated twice over time. We change a variable, integrate by part, and change a variable again:

$$\dot{I}_4 = \int_0^t \int_\tau^t I_2\,(\tau' - \tau)\, d\tau'\, d\tau$$

$$\dot{I}_4 = \int_0^t \int_0^{t-\tau} \dot{I}_2(y)\, dy\, d\tau \qquad \text{with} \qquad y = \tau' - \tau$$

$$I_4 = \left[\tau \int_0^{t-\tau} \dot{I}_2(y)\, dy\right]_0^t - \int_0^t \tau \frac{\partial}{\partial\tau}\left[\int_0^{t-\tau} \dot{I}_2(y)\, dy\right] d\tau$$

$$= 0 + \int_0^t \tau\, \dot{I}_2\,(t - \tau)\, d\tau = \int_0^t (t - \tau)\, \dot{I}_2(\tau)\, d\tau$$

with $\tau = t - \tau$

As $\sigma_{x_t}^2 = 2D_L t + 2\dot{I}_4$, we finally obtain

$$\sigma_{x_t}^2 = 2D_L t + 2\int_0^t (t - \tau) \frac{1}{2(\pi D_T \tau)^{1/2}} \cdot \int_{-\infty}^{+\infty} \exp\left(-\frac{s^2}{4D_T\tau}\right) \mathrm{Cov}\,(s)\, ds d\tau$$

TABLE 2. Apparent Macrodispersivities for Artificial Flow Conditions

	Assumption 1*	Assumption 2†	Assumption 3‡
Asymptotic dispersivity, m	26.5	317	2.8
Time of observation needed to obtain 95% of asymptotic dispersivity, days	14	900	9
Average travel distance, m	600	39×10^3	40

*Where $10u = 5 \times 10^{-4}$ m/s, $10v = 10^{-5}$ m/s, $10D_L = 5 \times 10^{-4}$ m²/s, and $10\, D_T = 5 \times 10^{-6}$ m²/s.
†Where $10u = 5 \times 10^{-4}$ m/s, $v = 10^{-6}$ m/s, $10D_L = 5 \times 10^{-4}$ m²/s, and $10D_T = 5 \times 10^{-6}$ m²/s.
‡Where $u = 5 \times 10^{-5}$ m/s, $10v = 10^{-5}$ m/s, $D_L = 5 \times 10^{-5}$ m²/s, and $D_T = 5 \times 10^{-5}$ m²/s.

APPENDIX 2: DETERMINATION OF THE LAPLACE TRANSFORM OF THE INTEGRAL $\dot{I}$

$$\dot{I} = 2\int_0^t (t-\tau)\frac{1}{2(\pi D_T \tau)^{1/2}}\int_{-\infty}^{\infty} e^{-(s^2/4D_T\tau)}\,\text{Cov}(s)\,ds\,d\tau$$

The Laplace transform of $\dot{I}(t)$ is defined by

$$\Lambda(\dot{I}) = \int_0^{\infty} e^{-pt}\,\dot{I}(t)\,dt$$

The integral $\dot{I}(t)$ is the convolution product of the two functions

$$F_1(t) = 2\frac{t}{\sqrt{D_T}}$$

$$F_2(t) = \int_0^{\infty}\frac{e^{-(s^2/4D_T t)}}{\sqrt{\pi}\sqrt{t}}\,\text{Cov}(s)\,ds$$

since the integral from $-\infty$ to $+\infty$ is twice the integral from zero to ∞ (symmetric function). In the transform domain the convolution reduces to a simple product; $\Lambda(\dot{I}) = \Lambda(F_1)\,\Lambda(F_2)$

$$\Lambda(F_1) = \frac{2}{\sqrt{D_T}}\cdot\frac{1}{p^2}$$

To determine the transform of F_2, we use the following theorem [*Carslaw and Jaeger,* 1963]. If

$$\Lambda[K(t,s)] = \phi(p)\,e^{-s\psi(p)} \qquad \Lambda[y(t)] = Y(p) \qquad \text{for } p > 0$$

then

$$\Lambda\left[\int_0^{\infty} K(t,s)\,y(s)\,ds\right] = \phi(p)\,Y[\psi(p)]$$

Here we have

$$K(t,s) = \frac{e^{-(s^2/4D_T t)}}{\sqrt{\pi}\sqrt{t}} \qquad y(s) = \text{Cov}(s)$$

Let us denote $Y(p) = \Lambda(\text{Cov}(s)) = \int_0^{\infty} e^{-ps}\,\text{Cov}(s)\,ds$

$$\Lambda[K(t,s)] = \frac{e^{-s(p/D_T)^{1/2}}}{\sqrt{p}}$$

that is,

$$\phi(p) = 1/\sqrt{p} \qquad \psi(p) = (p/D_T)^{1/2}$$

Then

$$\Lambda(F_2) = \frac{1}{\sqrt{p}}\cdot Y[(p/D_T)^{1/2}]$$

We finally obtain

$$\Lambda(\dot{I}) = \Lambda(F_1)\,\Lambda(F_2) = \frac{2}{p^{5/2}\sqrt{D_T}}\,Y[(p/D_T)^{1/2}]$$

$Y(p)$ being the Laplace transform of the covariance function of the velocity u.

APPENDIX 3: EQUIVALENCE OF THE CONDITIONS ON THE LAPLACE OR FOURIER TRANSFORM OF THE COVARIANCE OF THE VELOCITY

Our condition is that the integral $I(t)$ in (6) behaves linearly in t for large t, i.e.,

$$\frac{\dot{I}(t)}{t} \to A \qquad \text{for } t \to \infty$$

From (6), if we define

$$f(\tau) = \frac{1}{2(\pi D_T\tau)^{1/2}}\int_{-\infty}^{\infty} e^{-(s^2/4D_T\tau)}\,\text{Cov}(s)\,ds$$

then

$$\frac{\dot{I}(t)}{t} = 2\int_0^t \left|1 - \frac{\tau}{t}\right| f(\tau)\,d\tau$$

as $f(\tau) \geq 0$, we can then write

$$\int_0^{t/2} f(\tau)\,d\tau \leq \frac{\dot{I}(t)}{t} \leq 2\int_0^t f(\tau)\,d\tau$$

For $t \to \infty$, our condition requires that $\int_0^{\infty} f(\tau)\,d\tau < \infty$ and then the constant A is given by

$$A = 2\int_0^{\infty} f(\tau)\,d\tau$$

However, we can show that $f(\tau)$ can be expressed as a function of the density power spectrum of the velocity, which is defined by

$$S_{uu}(r) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-irs}\,\text{Cov}(s)\,ds$$

which is equivalent to

$$\text{Cov}(s) = \int_{-\infty}^{\infty} e^{irs}\,S_{uu}(r)\,dr$$

We can then rewrite $f(\tau)$ in terms of $S_{uu}(r)$ instead of Cov (s)

$$f(\tau) = \frac{1}{2(\pi D_T\tau)^{1/2}}\int_{-\infty}^{+\infty} e^{-(s^2/4D_T\tau)}\int_{-\infty}^{+\infty} e^{irs}\,S_{uu}(r)\,dr\,ds$$

$$= \frac{1}{2(\pi D_T\tau)^{1/2}}\int_{-\infty}^{+\infty} S_{uu}(r)\int_{-\infty}^{+\infty}\exp\left|irs - \frac{s^2}{4D_T\tau}\right| ds\,dr$$

$$= \int_{-\infty}^{+\infty} S_{uu}(r)\,e^{-D_T\tau r^2}\,\frac{1}{\sqrt{\pi}}$$

$$\cdot\int_{-\infty}^{+\infty}\exp\left(-\left(\frac{s - 2iD_T\tau r}{2\sqrt{D_T\tau}}\right)^2\right)\frac{ds}{2\sqrt{D_T\tau}}\,dr$$

when changing the variable of integration from s to

$$v = \frac{s - 2iD_T\tau r}{2\sqrt{D_T\tau}}$$

one obtains the error function. Then

$$f(\tau) = \int_{-\infty}^{+\infty} S_{uu}(r)\,e^{-D_T\tau r^2}\,dr$$

We then have

$$\int_0^t f(\tau)\,d\tau = \int_{-\infty}^{+\infty} S_{uu}(r)\int_0^t e^{-D_T\tau r^2}\,d\tau\,dr$$

$$= \frac{1}{D_T}\int_{-\infty}^{+\infty}\frac{S_{uu}(r)}{r^2}(1 - e^{-D_T t r^2})\,dr$$

For $t \to \infty$ we find

$$\int_0^\infty f(\tau)\, d\tau = \frac{1}{D_T}\int_{-\infty}^{+\infty} \frac{S_{uu}(r)}{r^2}\, dr$$

that is to say that the limit A of $I(t)/t$ for $t \to \infty$ is

$$A = \frac{2}{D_T}\int_{-\infty}^{+\infty} \frac{S_{uu}(r)}{r^2}\, dr$$

or for the asymptotic dispersion coefficient,

$$D_A = D_L + \frac{1}{D_T}\int_{-\infty}^{+\infty} \frac{S_{uu}(r)}{r^2}\, dr$$

which is exactly the result obtained by Gelhar if the velocity is taken to be proportional to the permeability with a constant gradient J, i.e., the power spectrum of the velocity is that of the permeability multiplied by $\bar{u}^2/\bar{K}^2$.

The condition imposed by Gelhar that this integral be finite is thus strictly equivalent to our condition on the Laplace transform of the covariance of the velocity.

APPENDIX 4: VARIANCE OF THE PARTICLE COORDINATE, RANDOM MOTION MODEL WITH TWO COMPONENTS ON THE VELOCITY

Let us assume that in (12), $x_0 = Z_0 = 0$. Then

$$X_t = \xi_t + \int_0^t u(Z_\tau)\, d\tau$$

$$Z_t = \zeta_t + vt$$

We now have

$$E[Z_t] = vt$$

$$\sigma_{Z_t}{}^2 = 2D_T t$$

The Brownian motion process Z_t no longer has a zero mean but rather a mean vt with the same variance $2D_T t$. The computation of the variance of X_t is identical to Appendix 1, except for the introduction of the mean vt in the pdf of Z_τ. One obtains

$$\sigma_{X_t}{}^2 = 2D_L t + 2\int_0^t (t-\tau)\frac{1}{2(\pi D_T \tau)^{1/2}} \cdot \int_{-\infty}^{+\infty} \exp\left(-\frac{(s-v\tau)^2}{4D_T\tau}\right) \mathrm{Cov}\,(s)\, ds\, d\tau$$

In the case where $D_L = D_T = 0$ one immediately obtains

$$\sigma_{X_t}{}^2 = \int_0^t \int_0^t \mathrm{Cov}\,[Z_{\tau'} - Z_\tau]\, d\tau' d\tau$$

By symmetry, changing of variable as in Appendix 1, and substitution of Z_τ by $v\tau$ one finds

$$\sigma_{X_t}{}^2 = \frac{2}{v^2}\int_0^{vt} (vt - s)\, \mathrm{Cov}\,(s)\, ds$$

for $t \to \infty$

$$\sigma_{X\infty}{}^2 \to 2t \times \frac{1}{v}\int_0^\infty \mathrm{Cov}\,(s)\, ds$$

The computation of the Laplace transform of the integral, in the general case, is very similar to that presented in Appendix 2. Following the same method, we obtain

$$\dot{I}(t) = \int_0^t F_1(t-\tau)\, F_2(\tau)\, d\tau$$

with

$$F_1(t) = 2\frac{t}{\sqrt{D_T}}$$

$$F_2(t) = \int_{-\infty}^{\infty} \frac{e^{-(v^2t/4D_T)} \cdot e^{vs/2D_T} \cdot e^{-(s^2/4D_T t)}}{2\sqrt{\pi t}}\, \mathrm{Cov}\,(s)\, ds$$

We have again

$$\Lambda(\dot{I}) = \Lambda(F_1)\, \Lambda(F_2)$$

$$\Lambda(F_1) = \frac{2}{\sqrt{D_T p^2}}$$

We can write $F_2(t)$ as

$$F_2(t) = e^{-(v^2t/4D_T)}\left[\int_0^\infty \frac{e^{s^2/4D_T}\, e^{vs/2D_T}}{2\sqrt{\pi t}}\, \mathrm{Cov}\,(s)\, ds + \int_0^\infty \frac{e^{-(s^2/4D_T t)} e^{-(vs/2D_T)}}{2\sqrt{\pi t}}\, \mathrm{Cov}\,(s)\, ds\right]$$

Let $F_3(t)$ be the sum of the two integrals in brackets:

$$F_2(t) = e^{-(v^2t/4D_T)}\, F_3(t)$$

Multiplication by e^{at} is a translation from p to $p - a$ in the Laplace domain:

$$\Lambda(F_2)(p) = \Lambda(F_3)\left(p + \frac{v^2}{4D_T}\right)$$

We will use the same theorem as in Appendix 2 to compute the Laplace transform of each integral. With the same notations we have

$$K(t, s) = \frac{e^{-(s^2/4D_T t)}}{\sqrt{\pi t}} \qquad \Lambda[K] = \frac{e^{-s(p/D_T)^{1/2}}}{\sqrt{p}}$$

$$y(s) = \tfrac{1}{2}\, e^{\pm(vs/2D_T)}\, \mathrm{Cov}\,(s) \qquad \Lambda(y) = \tfrac{1}{2}\, Y\left(p \pm \frac{v}{2D_T}\right)$$

where we note again

$$Y(p) = \Lambda(\mathrm{Cov}\,(s)) = \int_0^\infty e^{-ps}\, \mathrm{Cov}\,(s)\, ds$$

then

$$\Lambda(F_3)(p) = \frac{1}{\sqrt{p}} \cdot \frac{1}{2}\left[Y\left(\left(\frac{p}{D_T}\right)^{1/2} - \frac{v}{2D_T}\right) + Y\left(\left(\frac{p}{D_T}\right)^{1/2} + \frac{v}{2D_T}\right)\right]$$

and, finally,

$$\Lambda(\dot{I}) = \frac{1}{p^2(pD_T + v^2/4)^{1/2}}\left[Y\left(\frac{(pD_T + v^2/4)^{1/2} - v/2}{D_T}\right) + Y\left(\frac{(pD_T + v^2/4)^{1/2} + v/2}{D_T}\right)\right]$$

APPENDIX 5: DETERMINATION OF THE EQUIVALENT DISPERSION COEFFICIENT AS A FUNCTION OF TIME

Without Vertical Component of the Velocity

From (5) and (7) we have

$$D_A(\tau) = D_L + \frac{1}{2t}\dot{I}(t)$$

Laplace transform of $\dot{I}$

$$\Lambda(\dot{I}) = \frac{2}{p^{5/2}\sqrt{D_T}}\,Y\left[\left(\frac{p}{D_T}\right)^{1/2}\right]$$

Covariance (19) (with the hole effect).

$$Y(p) = \bar{u}^2\,\frac{\sigma_K^2}{\bar{K}^2}\,\frac{p(3pl+1)}{3l(p+1/l)^3}$$

This value of Y was substituted in $\Lambda(\dot{I})$, and the inverse Laplace transform computed. One obtains expressions such as

$$\frac{A}{p\sqrt{p}((p+b)^{1/2})^3} \qquad \frac{C}{p^2((p+b)^{1/2})^3}$$

the $\sqrt{p}$ can be eliminated by the relations [*Carslaw and Jaeger*, 1963]

$$\Lambda^{-1}\left(\frac{1}{\sqrt{p}}\,\bar{x}(\sqrt{p})\right) = \int_0^\infty \frac{1}{\sqrt{\pi t}}\,e^{-u^2/4t}\,x(u)\,du$$

or

$$\Lambda^{-1}(\bar{x}(\sqrt{p})) = \int_0^\infty \frac{u}{2\sqrt{\pi t^3}}\,e^{-u^2/4t}\,x(u)\,du$$

and then expressions in $(1/p^n)\bar{x}(p)$ are integrated by

$$\Lambda^{-1}\left(\frac{1}{p^n}\,\bar{x}(p)\right) = \int_0^t \int_0^{\lambda_{n-1}} \cdots \int_0^{\lambda_1} x(\lambda_0)\,d\lambda_0 \cdots d\lambda_{n-1}$$

the computation is straightforward.

Covariance (20) (exponential).

$$Y(p) = \bar{u}^2\,\frac{\sigma_K^2}{\bar{K}^2}\,\frac{1}{p+n/l}$$

again the inverse Laplace transform was used after substitution; the relation $(1/\sqrt{p})\,\bar{C}(\sqrt{p})$ was used to eliminate $\sqrt{p}$. Then a simple expression as $[1/(p^4(p+b))]$ was obtained and integrated.

Covariance (21) (Gaussian).

$$Y(p) = \bar{u}^2\,\frac{\sigma_K^2}{\bar{K}^2}\,\frac{l}{m}\left(\frac{\pi}{2}\right)^{1/2}\exp\left(\frac{l^2p^2}{2m^2}\right)\operatorname{erfc}\left(\frac{lp}{\sqrt{2}m}\right)$$

after substitution, the inverse Laplace transform was computed using the relation [*Erdélyi et al.*, 1954, p. 267]:

$$\Lambda^{-1}\left(\frac{1}{\sqrt{p}}\,e^{ap}\operatorname{erfc}(\sqrt{ap})\right) = \frac{1}{\sqrt{\pi}}\,\sqrt{t+a}$$

followed by two integrations in time.

With a Vertical Component of the Velocity

Covariance (19) (with the hole effect). Given the expression for $\Lambda(\dot{I})$ in (15) and that for $Y(p)$ in (9), it is possible to write it, after substitution and rearranging, as

$$\Lambda(\dot{I}) = \frac{J^2\sigma_K^2}{u(u-x)^2}\left[\frac{u^2+Cu+d}{(u-y)^3} + \frac{u^2+C'u+d'}{(u-y')^3}\right]$$

where

$$u = \sqrt{p} + u^2/4D_T$$

$$x = v^2/4D_T$$

$$C = \frac{\sqrt{D_T}}{l^3} - \frac{v}{\sqrt{D_T}}$$

$$d = \frac{v}{2\sqrt{D_T}}\left(\frac{v}{2\sqrt{D_T}} - \frac{\sqrt{D_T}}{3l}\right)$$

$$y = \frac{v}{2\sqrt{D_T}} - \frac{\sqrt{D_T}}{l}$$

with C', d', and y' changing v in $(-v)$.

The translation $p + u^2/4D_T$ is a multiplication by $\exp(-(v^2/4D_T)t)$ in the time domain; furthermore, $\sqrt{p}$ can be eliminated by the transformation $(1/\sqrt{p})\bar{x}(\sqrt{p})$. Then, we can write that $\bar{x}(p) = J^2\sigma_K^2[(p^2+ap+b)/(p^2-x)^2(p-y)^3)]$ plus similar expression, $v \to (-v)$. This is a rational function of polynomials in p, $(Q(p)/P(p))$, with $P(p) = (p-a_1')^{m_1} \cdots (p-a_n)^{m_n}$, $Q(p)$ is a polynomial of degree inferior to $m_1 + \cdots + m_n - 1$, where $a_i \neq a_k$ and $i \neq k$. The inverse Laplace transform of such functions is [*Erdélyi et al.*, 1954]

$$\Lambda^{-1}\left(\frac{P}{Q}\right) = \sum_{k=1}^{n}\sum_{l=1}^{m}\frac{\phi_{kl}(a_k)}{(m_k-l)!(l-1)!}\,t^{m_k-l}\,e^{a_kt}$$

with

$$\phi_{kl}(x) = \frac{d^{l-1}}{dx^{l-1}}\left|\frac{Q(x)}{P_k(x)}\right|$$

$$P_k(x) = \frac{P(x)}{(x-a_k)^{m_k}}$$

The inverse Laplace transform of $\Lambda(\dot{I})$ can be determined from these relations without major difficulty.

Covariance (20) (exponential). Use exactly the same procedure.

Covariance (21) (Gaussian). Could not easily obtain an inverse Laplace transform; $D_A(\tau)$ was computed directly from (15) by a simple integration over s, while the integration over τ in (29) was done numerically.

NOTATIONS

- A, a constant.
- $A(\infty)$ asymptotic longitudinal macro-dispersivity.
- B constant.
- C concentration.
- $\mathrm{Cov}(F)$ covariance function of random function F.
- d diffusion coefficient in porous media.
- $\mathbf{D}$ dispersion tensor.
- $D_A(\tau)$ equivalent longitudinal macrodispersion coefficient of a homogeneous medium.
- $D_{A_0}(\tau)$ equivalent longitudinal macrodispersion coefficient of a homogeneous medium for pure convection ($D_L = D_T = 0$).
- D_L, D_T local longitudinal and transversal dispersion coefficient.
- D_{ref} reference for dispersion coefficient, equal to $\bar{u}^2(\sigma_K^2/\bar{K}^2)(l^2/3d_T)$.

erf (x) error function, equal to $(2/\sqrt{\pi}) \int_0^x e^{-x^2} dx$.
erfc (x) complementary error function, equal to $1 - \text{erf}(x)$.
$E(\ )$ expected value.
F any function.
$g_\tau(u)$ Gaussian distribution function.
h hydraulic head.
$\dot{I}$ integral.
J hydraulic gradient in the porous medium.
k intrinsic permeability of the porous medium.
K hydraulic conductivity of the porous medium.
$\bar{K}$ $E(K)$.
l length scale of the porous medium, in covariance functions.
m constant, Gaussian covariance of K; in example, $m = 4.461$.
n constant, exponential covariance of K; in example, $n = 3.154$.
p Laplace variable.
$p(Z)$ probability distribution function of Z.
$p(Z|Z')$ conditional probability distribution function of Z, given Z'.
r wave number, in spectrum $S_{KK}(r)$.
s lag, in covariance Cov(s).
$S_{KK}(r)$ spectrum of permeability K.
t time.
$\mathbf{u}$ local microscopic velocity vector.
u idem, horizontal component along x.
$\bar{u}$ expected value of u, equal to $E(u)$.
$\mathbf{U}$ Darcy's velocity vector.
U Darcy's velocity vector, horizontal component along x.
v vertical component of $\mathbf{u}$, along z.
Var(F) variance of random function F.
x horizontal axis.
X_t horizontal component of the position of the particle at time t.
$Y(p)$ Laplace transform of the covariance function of the velocity u.
z vertical axis.
Z_t ordinate of particle at time t.
α_L, α_T local longitudinal and transversal dispersivity.
β dimensionless parameter, equal to $vl/2D_T$.
δ Dirac function.
Δt time step.
$\Lambda(F)$ Laplace transform of function F.
μ ratio vertical/horizontal velocity, equal to $v/\bar{u}$.
ω kinematic porosity (or effective).
σ_F^2 variance of random function F.
τ dimensionless time, equal to $(D_T/l^2)t$, or integration variable.
ξ_t Brownian motion along x axis.
ζ_t Brownian motion along z axis.
$\sum$ summation sign.

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