Stochastic Analysis of Nonstationary Subsurface Solute Transport
1.   Unconditional Moments

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This paper applies stochastic methods to the analysis and prediction of solute transport in heterogeneous saturated porous media. Partial differential equations for three unconditional ensemble moments (the concentration mean, concentration covariance, and velocity concentration cross covariance) are derived by applying perturbation techniques to the governing transport equation for a conservative solute. Concentration uncertainty is assumed to be the result of unmodeled small-scale fluctuations in a steady state velocity field. The moment expressions, which describe how each moment evolves over time and space, resemble the classic deterministic advection-dispersion equation and can be solved using similar methods. A solution procedure based on a Galerkin finite element algorithm is illustrated with a hypothetical two-dimensional example. For this example the required steady state velocity statistics are obtained from an infinite domain spectral solution of the stochastic groundwater flow equation. The perturbation solution is shown to reproduce the statistics obtained from a Monte Carlo simulation quite well for a natural log conductivity standard deviation of 0.5 and moderately well for a natural log conductivity standard deviation of 1.0. The computational effort required for a perturbation solution is significantly less than that required for a Monte Carlo solution of acceptable accuracy. Sensitivity analyses conducted with the perturbation approach provide qualitative confirmation of a number of results obtained by other investigators for more restrictive special cases.

INTRODUCTION

It is now widely recognized that the dispersion of solutes moving in the subsurface environment is due, in large part, to natural heterogeneity. Important sources of heterogeneity include spatial variations in soil properties such as hydraulic conductivity and spatial and temporal variations in hydrologic inputs such as recharge. Small-scale velocity fluctuations induced by these heterogeneities enhance spreading and contribute to the irregularity and "randomness" which is frequently observed in field scale tracer plumes. The connection between velocity variability and solute transport has been investigated both theoretically (see, for example, the review papers by *Gelhar* [1986, 1987], and *Dagan* [1987]) and in natural gradient tracer tests [*Freyberg*, 1986; *Sudicky*, 1986; *Garabedian*, 1987]. Much of this work has focused on evaluations of spatially invariant macrodispersivity coefficients, largely because of the close connection between macrodispersion and dilution. Less attention has been devoted to the impact of natural heterogeneity and related data limitations on the uncertainty of concentration predictions.

It is often convenient to treat heterogeneous soil and hydrologic variables as if they were samples drawn at random from a population (or ensemble) of physically plausible functions. This approach, which is now commonly used in groundwater hydrology, allows both spatial variability and prediction uncertainty to be analyzed with the tools of probability theory. Probabilistic methods are most useful if the ensemble description incorporates everything known about solute transport at a particular site. This can be accomplished in two ways. First, the ensemble statistics (e.g., the ensemble mean and covariance) can be derived from physically based models which rely on concepts such as mass conservation. Second, these statistics

can be conditioned on actual measurements of solute concentration and other related hydrologic variables. The two approaches are complementary and should be combined whenever possible. The ultimate objective is to make the ensemble definition specific, so that it admits only those replicates which are realistic and consistent with observations.

Since a properly constructed ensemble description provides a concise summary of available information about a particular site, it may be used to make predictions about the transport of subsurface solutes. Bayesian estimation theory establishes a systematic framework for deriving predictions with desirable properties [*Schweppe*, 1973]. This theory may be used, for example, to show that the ensemble mean is the minimum variance unbiased estimate of the actual solute concentration for any concentration probability distribution with finite moments [*Jazwinski*, 1978]. The ensemble variance is a useful measure of the uncertainty associated with this estimate, particularly if the concentration is normally distributed. In this paper, we are interested primarily in the derivation of unconditional ensemble moments; that is, moments which do not depend on concentration observations. Such moments may be used to predict the impact of contamination incidents which have not yet actually occurred. If, at some point, concentration measurements become available, the unconditional moments may be conditioned (or updated) so that the predictions can take advantage of this additional information [*McLaughlin and Graham*, 1986; *Graham*, 1988].

A number of investigators have attempted to derive the unconditional statistics of hydrologic variables directly from the local equations which describe subsurface flow and solute transport. This approach is particularly useful if the heterogeneous inputs to these equations are stationary (or, more strictly, ergodic) over the region of interest, so that their ensemble moments can be estimated from sample spatial statistics. The most common example of this concept is the derivation of the ensemble statistics of a possibly nonstationary hydraulic head field from the estimated ensemble statistics of a stationary hydraulic conductivity field. Some representative

examples include studies by *Freeze* [1975], *Sagar* [1978], *Smith and Freeze* [1979a, b], *Bakr et al.* [1978], *Gutjahr and Gelhar* [1981], *Dettinger and Wilson* [1981], *Dagan* [1982a, b], *Townley and Wilson* [1985], and *McLaughlin and Wood* [1988a, b]. The application of derived head statistics to Bayesian estimation is discussed by *Dagan* [1982b], *Hoeksema and Kitanidis* [1985], and *Gutjahr and Wilson* [1985].

The basic concepts used in the above studies of groundwater flow extend naturally to solute transport. In this case, the ensemble concentration moments may be derived from velocity statistics which may, in turn, be derived from the ensemble moments of the hydraulic conductivity field. Much of the research in stochastic solute transport has emphasized evaluations of concentration statistics and related macrodispersivity coefficients for the special case of one-dimensional flow through perfectly layered random media [*Gelhar et al.*, 1979; *Matheron and de Marsily*, 1980; *Gelhar et al.*, 1981; *Black and Freyberg*, 1987]. Our interest here is in more general multidimensional problems similar to the one investigated by *Smith and Schwartz* [1980, 1981]. They used Monte Carlo techniques to investigate solute transport in a finite two-dimensional domain with a spatially correlated hydraulic conductivity field. The synthetically generated concentration distributions obtained in this study were very sensitive to the structure of the underlying hydraulic conductivity fields. Moreover, a constant Fickian macrodispersivity coefficient could not be identified for any individual realization or for the ensemble as a whole.

Gelhar and Axness [1983] used spectral perturbation techniques to evaluate spatially invariant steady state ensemble macrodispersivity coefficients in an infinite three-dimensional domain. These anisotropic coefficients, which are derived from ensemble macrodispersive fluxes, depend on the local dispersivities, the mean flow direction, and the statistics of the log hydraulic conductivity field. The Gelhar and Axness approach is based on an assumption that the concentration field is locally stationary in the sense that the mean concentration gradient is approximately constant over many concentration correlation scales. *Sudicky* [1986] found that ensemble macrodispersivities evaluated from expressions in Gelhar and Axness are similar to those obtained from a spatial moment analysis of concentration data collected at the Borden site.

Dagan [1982b, 1984, 1986, 1987] used Lagrangian perturbation techniques to analyze stochastic solute transport in two- and three-dimensional domains. He derived closed-form expressions for the time-dependent ensemble displacement statistics of a solute particle moving in a random steady state velocity field with a uniform mean hydraulic gradient. Spatially invariant ensemble macrodispersivity coefficients inferred from Dagan's displacement covariance compare favorably with those obtained from an analysis of the spatial moments of the tracer plume at the Borden site [*Sudicky*, 1986]. The leading terms of Dagan's asymptotic longitudinal macrodispersivity expression match the result derived by *Gelhar and Axness* [1983].

Dagan's approach also provides expressions for the ensemble moments of a random concentration field. The ensemble concentration mean is the solution to an advection-dispersion equation which uses macrodispersivity coefficients obtained from the time-dependent displacement covariance expressions given by *Dagan* [1984]. Although Dagan derived an exact equation for the ensemble concentration variance, he presents no solution to this equation for the general case. His

qualitative analysis indicates that the time-varying concentration variance should depend on the statistics of the hydraulic conductivity field, the pore scale dispersivity, and the volumes over which the source and observed concentrations are averaged [*Dagan*, 1984].

Vomvoris [1986] and *Vomvoris and Gelhar* [1986] used infinite domain spectral techniques to derive closed form expressions for the ensemble covariance of a locally stationary three-dimensional steady state concentration field. Their analysis indicates that the concentration variance depends on the statistics of the hydraulic conductivity field, the mean concentration gradient, and the local dispersivity. It also shows that the concentration covariance is highly anisotropic with very large spatial persistence in the direction of mean flow. Although Vomvoris' closed-form expressions provide useful insight into the mechanisms which influence concentration uncertainty, they depend on stationarity assumptions which can be restrictive in some applications. For example, the spatial correlations inferred from these expressions are valid only when applied to points which are closely spaced in comparison to the scale of a solute plume. This requirement may not be met in estimation applications where solute observation points are scattered over an extended region.

Neuman et al. [1987] used a semigroup approach to derive spatially invariant ensemble macrodispersivity coefficients from a general description of a three-dimensional subsurface velocity field. In the special case of a steady state, uniform density flow field, these coefficients are the same as the macrodispersivities obtained from the leading terms of *Dagan*'s [1984] asymptotic expressions and from the *Gelhar and Axness* [1983] analysis. The approach adopted by *Neuman et al.* [1987] assumes that the ensemble concentration mean tends toward Fickian behavior at some scale but does not provide any indication of the times or travel distances at which this assumption is valid.

This paper describes a new approach to the derivation of solute concentration statistics which differs in a number of ways from the studies cited above. Our emphasis is on spatially variable ensemble moments rather than on spatially invariant spatial moments and effective parameters, primarily because we are ultimately interested in Bayesian estimation and monitoring design. We use first-order Eulerian methods to derive a set of coupled partial differential equations for the transient nonstationary first and second unconditional moments of concentration. The basic approach is similar to the perturbation technique used in the groundwater flow investigations described by *Sagar* [1978], *Dettinger and Wilson* [1981], *Townley and Wilson* [1985], and *McLaughlin and Wood* [1988a, b]. In the transport application we assume that the underlying source of concentration uncertainty is small-scale variability in a steady state velocity field. This variability may, in turn, be related to heterogeneities in soil properties and hydrologic inputs.

Our concentration moment equations constitute a generalized transport model which explicitly accounts for the effects of hydrogeologic variability on both large-scale dispersion and prediction uncertainty. The macrodispersive flux used in the mean equation is obtained from a set of closure equations in a manner similar to that described by *Cushman* [1983], *Gutjahr et al.* [1985], and *Tompson and Gray* [1986a, b]. This flux does not generally have a Fickian form and may vary over both time and space. The concentration covariance equation, which provides the information needed to quantify the uncer-

tainty of the mean prediction, is also dependent on the macrodispersive flux. All of these equations can be solved numerically over a finite domain, both near and far from sources, for a variety of boundary conditions and random velocity characteristics. Our approach provides results similar to those obtained from a Monte Carlo analysis, but with less computational effort. Moreover, the moment equations can reveal qualitative information about the evolution of uncertainty even before detailed numerical solutions are computed.

The perturbation procedure used to derive the concentration moment equations is described in the next section. This is followed by a brief discussion of a finite element algorithm which may be used to obtain approximate numerical solutions for two-dimensional problems. We then present results from a relatively simple example which reveals a number of interesting points about near-source ensemble concentration statistics.

DERIVATION OF THE MOMENT EQUATIONS

The governing equation for the transport of a conservative solute in a random saturated porous medium with constant porosity is [Bear, 1979]

$$\frac{\partial c}{\partial t} + \frac{\partial}{\partial x_i}(v_i c) - \frac{\partial}{\partial x_i}\left[D_{ij}\frac{\partial c}{\partial x_j}\right] = 0 \qquad x \in D \qquad (1a)$$

$$c = c_b \qquad x \in \partial D \qquad (1b)$$

$$c = c_0 \qquad x \in D \qquad t = t_0 \qquad (1c)$$

where the concentration $c(x, t)$ is assumed to be a nonstationary random function of the location vector x (with coordinates x_i) and time t. Equation (1) is defined over a finite two or three-dimensional domain D with boundary ∂D. If D is two-dimensional the vector indices are equal to 1 or 2, the velocity and concentration are assumed to be vertically homogeneous and the flow depth is assumed constant; if D is three-dimensional the vector indices are equal to 1, 2, or 3. Summation over repeated indices is implied. The pore velocity $v_i(x)$ is assumed to be a nonstationary time-invariant random function which is defined over an infinite spatial domain. The initial and boundary conditions $c_0(x)$ and $c_b(x, t)$ appearing in (1b) and (1c) could be random functions but are assumed, for simplicity, to be known constants. Our use of Dirichlet conditions presumes that the boundary is located beyond the range of the solute plume in a region where concentrations are fixed either at zero or at some low background level.

The local (pore scale) dispersion tensor D_{ij} is assumed to be a deterministic variable which is related to the mean velocity as follows:

$$D_{ij} = \alpha_T \bar{v}\delta_{ij} + (\alpha_L - \alpha_T)\frac{\bar{v}_i \bar{v}_j}{\bar{v}} \qquad (2)$$

where α_T and α_L are the transverse and longitudinal local dispersivities (assumed constant over the domain D); $\bar{v}$ is the magnitude of the mean velocity vector; and molecular diffusion is neglected [Bear, 1979; Gelhar and Axness, 1983]. Note that overbars are used to represent mean values (mathematical expectations) and that location and time arguments (x and t) are omitted unless required for clarification. The use of constant α_L and α_T in (2) reflects the implicit assumption that spatial variations in the processes responsible for local dispersion are small compared to spatial variations in the local velocity field. This is a common assumption [Gelhar and Axness,

1983; Dagan, 1987] which is supported by an analysis presented by Vomvoris [1986].

If the random velocity and concentration variables appearing in (1) are each expanded into the sum of a spatially variable mean and a small perturbation around this mean, the following expression results:

$$\frac{\partial(\bar{c} + \delta c)}{\partial t} + \frac{\partial}{\partial x_i}\left[(\bar{v}_i + \delta v_i)(\bar{c} + \delta c)\right]$$

$$- \frac{\partial}{\partial x_i}\left[D_{ij}\frac{\partial(\bar{c} + \delta c)}{\partial x_j}\right] = 0 \qquad x \in D \qquad (3a)$$

$$\bar{c} + \delta c = c_b \qquad x \in \partial D \qquad (3b)$$

$$\bar{c} + \delta c = c_0 \qquad x \in D \qquad t = t_0 \qquad (3c)$$

where the quantities prefixed by δ are zero-mean perturbations. The ensemble mean concentration equation is obtained by expanding all products in (3) and then taking the expectation of the resulting expression:

$$\frac{\partial\bar{c}}{\partial t} + \frac{\partial}{\partial x_i}(\bar{v}_i\bar{c}) - \frac{\partial}{\partial x_i}\left[D_{ij}\frac{\partial\bar{c}}{\partial x_j}\right] + \frac{\partial}{\partial x_i}[J_{Di}] = 0 \qquad (4a)$$

$$x \in D$$

$$\bar{c} = c_b \qquad x \in \partial D \qquad (4b)$$

$$\bar{c} = c_0 \qquad x \in D \qquad t = t_0 \qquad (4c)$$

Here $J_{Di}(x, t)$ is the cross covariance between velocity and concentration perturbations at location x:

$$J_{Di}(x, t) = P_{v_i c}(x, x, t) \qquad (5)$$

and $P_{v_i c}(x', x, t)$ is the general cross covariance between velocity and concentration perturbations at two independent locations x' and x, respectively:

$$P_{v_i c}(x', x, t) = \overline{\delta v_i(x')\delta c(x, t)} \qquad (6)$$

The vector J_{Di} is an ensemble macrodispersive flux which accounts for spreading caused by velocity fluctuations too small to be included in the mean advective term but too large to be included in the pore scale dispersion term.

We assume here that the time-invariant mean $\bar{v}_i(x)$ and covariance $P_{v_i v_j}(x', x)$ of the random velocity field are known. The velocity covariance definition is similar to (6):

$$P_{v_i v_j}(x', x) = \overline{\delta v_i(x')\delta v_j(x)} \qquad (7)$$

If the fluid density is assumed to be constant, the mass conservation principle implies that the divergence of the random velocity must be zero at any x:

$$\partial v_i(x)/\partial x_i = 0 \qquad (8)$$

If we take the expectation of this equation, it becomes apparent that the divergences of the velocity mean and perturbation must also be zero at any x:

$$\partial\bar{v}_i(x)/\partial x_i = \partial\delta v_i(x)/\partial x_i = 0 \qquad (9)$$

These conditions must be satisfied by the specified velocity statistics. In particular, the second equality in (9) implies that the velocity covariance must satisfy the following conditions:

$$\frac{\partial P_{v_i v_j}(x', x)}{\partial x_i'} = \frac{\partial P_{v_i v_j}(x', x)}{\partial x_j} = \frac{\partial^2 P_{v_i v_j}(x', x)}{\partial x_i' \, \partial x_j} = 0 \qquad (10)$$

where, as before, the location vectors x and x' are assumed to

be independent. The velocity statistics used in this paper are derived from a perturbation analysis of the steady state stochastic groundwater flow equation [*Gelhar*, 1986; *McLaughlin and Wood*, 1986*b*]. These statistics satisfy the mass conservation requirements stated in (9) and (10).

The mean solute transport equation has the same form as the traditional advection-dispersion equation with one additional term, the divergence of the macrodispersive flux J_{D_i}. Equation (5) indicates that J_{D_i} can be obtained from the more general cross covariance $P_{v_ic}(x', x, t)$, which is also needed to find the concentration covariance (see equation (14) below). A first-order approximation for P_{v_ic} can be derived by subtracting (4) from (3) and multiplying each term of the resulting equation by the velocity perturbation $\delta v_i(x')$. Since this perturbation depends on the coordinate x' (which is assumed to be independent of x) it can be brought inside derivatives which are taken with respect to the spatial variable x. If differences between products of small perturbations and their means are neglected to obtain closure, the resulting (approximate) cross covariance equation is

$$\frac{\partial}{\partial t} P_{v_ic}(x', x, t) + \frac{\partial}{\partial x_j} [\bar{v}_j(x)P_{v_ic}(x', x, t)]$$

$$- \frac{\partial}{\partial x_j} \left[D_{jk} \frac{\partial}{\partial x_k} P_{v_ic}(x', x, t) \right]$$

$$+ \frac{\partial}{\partial x_j} [\bar{c}(x, t)P_{v_iv_j}(x', x)] \simeq 0 \qquad x, x', \in D \qquad (11a)$$

$$P_{v_ic}(x', x, t) = 0 \qquad x \in \partial D \qquad x' \in D \qquad (11b)$$

$$P_{v_ic}(x', x, t) = 0 \qquad x, x' \in D \qquad t = t_0 \qquad (11c)$$

Here it is understood that each part of (11) applies for $i = 1$ to M, where M is the spatial dimension of the domain D. Note that the auxiliary conditions in this equation are both homogeneous because the coefficients c_b and c_0 are assumed to be known perfectly.

Each component of the linearized cross-covariance expression has the form of a traditional M-dimensional transport equation with a forcing term which is the divergence of the vector $\bar{c}(x, t)P_{v_iv_j}(x', x)$ (with i fixed). This vector, which introduces velocity uncertainty into the concentration moment equations, plays a role similar to the macrodispersive flux in the mean equation. It is important to note that (11) is an approximation which is strictly valid only in the limit as the higher-order moments of the velocity and concentration perturbations become arbitrarily small. The adequacy of the small perturbation approximation is explored for a particular example later in this paper. An extensive discussion of this topic is also provided in the work by *Ababou* [1988].

A first-order equation for the concentration covariance may be derived in much the same way as (11). This covariance is defined as follows:

$$P_{cc}(x', x, t) = \overline{\delta c(x', t)\delta c(x, t)} \qquad (12)$$

If (12) is differentiated with respect to time and the expectation and differentiation operators are interchanged, the result is

$$\frac{\partial}{\partial t} P_{cc}(x', x, t)$$

$$= \overline{\delta c(x', t)\left[\frac{\partial}{\partial t} \delta c(x, t)\right]} + \overline{\left[\frac{\partial}{\partial t} \delta c(x', t)\right]\delta c(x, t)} \qquad (13)$$

The two temporal derivatives appearing in right-hand side of this expression are obtained by subtracting (4) from (3), with the spatial arguments set equal to x and x', respectively. As before, perturbations which depend on the x' argument can be taken inside x derivatives and perturbations which depend on x can be taken inside x' derivatives. If differences between products of small perturbations and their means are neglected to obtain closure, the resulting (approximate) concentration covariance equation is

$$\frac{\partial}{\partial t} P_{cc}(x', x, t) + \frac{\partial}{\partial x_i} [\bar{v}_i(x)P_{cc}(x', x, t)]$$

$$+ \frac{\partial}{\partial x_i'} [\bar{v}_i(x')P_{cc}(x, x', t)] - \frac{\partial}{\partial x_i}\left[D_{ij}\frac{\partial}{\partial x_j} P_{cc}(x', x, t) \right]$$

$$- \frac{\partial}{\partial x_i'}\left[D_{ij}\frac{\partial}{\partial x_j'} P_{cc}(x, x', t) \right] + \frac{\partial}{\partial x_i} [\bar{c}(x, t)P_{cv_i}(x', x, t)]$$

$$+ \frac{\partial}{\partial x_i'} [\bar{c}(x', t)P_{cv_i}(x, x', t)] \simeq 0 \quad x, x' \in D \qquad (14a)$$

$$P_{cc}(x', x, t) = 0 \qquad x \in \partial D \qquad x' \in D \qquad (14b)$$

$$P_{cc}(x, x', t) = 0 \qquad x' \in \partial D \qquad x \in D \qquad (14c)$$

$$P_{cc}(x', x, t) = 0 \qquad x, x' \in D \qquad t = t_0 \qquad (14d)$$

As before, this first-order approximation is strictly valid only in the limit as the higher-order velocity and concentration moments become arbitrarily small. The concentration covariance expression has the form of a $2M$-dimensional transport equation with a forcing term which is the divergence of the $2M$-dimensional vector obtained by combining the M-dimensional vectors $\bar{c}(x, t)P_{cv_i}(x', x, t)$ and $\bar{c}(x', t)P_{cv_i}(x, x', t)$. Here again, there is a close structural analogy with the original mean equation.

Equations (4), (11), and (14) form a system of $2 + M$ coupled partial differential equations which describe the propagation of the mean concentration, the velocity-concentration cross covariance, and the concentration covariance through a random velocity field with known mean $\bar{v}_i(x)$ and covariance $P_{v_iv_j}(x', x)$. These equations can be solved in a variety of ways, using transform methods, Green's function techniques, or numerical solution algorithms [*McLaughlin and Wood*, 1988*b*]. The first two approaches require simplifying assumptions which may not be appropriate in certain applications. The numerical approach is considerably more general but is computationally demanding, especially for large three-dimensional domains which extend over many velocity correlation scales. A numerical solution is used here, primarily because we wish to examine nonstationary near-source effects which are difficult to address with the other methods.

NUMERICAL SOLUTION OF THE MOMENT EQUATIONS

A number of different numerical algorithms could be used to solve the advective-dispersive moment equations derived in the previous section. The results presented in our example problem were computed with a modified Galerkin finite element algorithm. The coupled system of partial differential equations obtained from the moment analysis may be simplified if the macrodispersive term appearing in each equation is evaluated explicitly, using moments computed on the last time step. Since the resulting decoupled equations all have the same basic form, they can all be solved with the same two-dimensional finite element transport algorithm.

The mean concentration equation depends on only one spatial vector x and can be discretized in the same way as a deterministic transport equation. The variables in the macrodispersive flux and concentration covariance equations must, however, be treated differently since they depend on two spatial vectors x and x'. The covariance functions appearing in these equations may be approximated by a quadratic form containing two interpolation functions, one for each spatial vector [*Courant and Hilbert*, 1953]. These covariance approximations have the general form

$$P_{uv}(x, x', t) \simeq \sum_{m=1}^{N} \sum_{n=1}^{N} [P_{uv}]_{mn}\phi_m(x)\phi_n(x') \qquad (15)$$

where $\phi_m(x)$ is the interpolation function associated with node m, evaluated at location x, N is the number of non-Dirichlet nodes in the domain, and $[P_{uv}]_{mn}$ is the generic covariance between the discretized variables u_m (evaluated at node m) and v_n (evaluated at node n).

If appropriate versions of (15) are substituted into each of the moment equations, the Galerkin finite element discretization procedure can be carried out in the usual way [*McLaughlin and Wood*, 1988b; *Graham*, 1988]. First, a weighted residual error is constructed by multiplying each discretized equation by the weighting function $\phi_m(x)$, where $m = 1, \cdots, N$ [*Pinder and Gray*, 1977]. This weighted residual is then integrated over the x coordinate, with the x' coordinate treated as a parameter. Next, the resulting equation in x' is multiplied by the weighting function $\phi_n(x')$, where $n = 1, \cdots, N$, to give a second weighted residual error. This residual is, in turn, integrated over x'. This two-step procedure yields N systems of ordinary differential equations ($n = 1, \cdots, N$), each containing N equations ($m = 1, \cdots, N$) in N unknowns (the elements of $[P_{uv}]_{mn}$), for each of the original covariance equations. The resulting discretized moment equations can be decomposed into a mathematically equivalent "square root" form which reduces computer storage and run times considerably [see *McLaughlin and Wood*, 1988a, Appendix B; *Graham*, 1988]. The desired covariance matrices $[P_{v_ic}]_{mn}$ and $[P_{cc}]_{mn}$ are constructed directly from the solutions to the decomposed covariance equations.

EXAMPLE PROBLEM

This section describes a simple two-dimensional example which illustrates the solution of the concentration moment equations for an evolving plume. In this example, a conservative solute is assumed to be released from a localized continuous line source into a confined aquifer (see Figure 1). The source location and the initial release time are both assumed to be known and the initial concentration is assumed to be zero everywhere except at the source, which is treated as a boundary. The source concentration is assumed to be fixed at the known solubility of the solute. The remaining boundaries are assumed to be located far enough from the solute source so that homogeneous Dirichlet boundary conditions apply on all sides of the domain. The velocity field is assumed to be at steady state with a spatially uniform mean which is aligned with the x_1 direction.

In order to apply the concentration moment equations derived earlier to this problem we must specify the mean and covariance of the steady state velocity field. Since we assume that velocity fluctuations are caused by small-scale variations in the log hydraulic conductivity, it is reasonable to obtain the velocity statistics from a stochastic analysis of the ground-
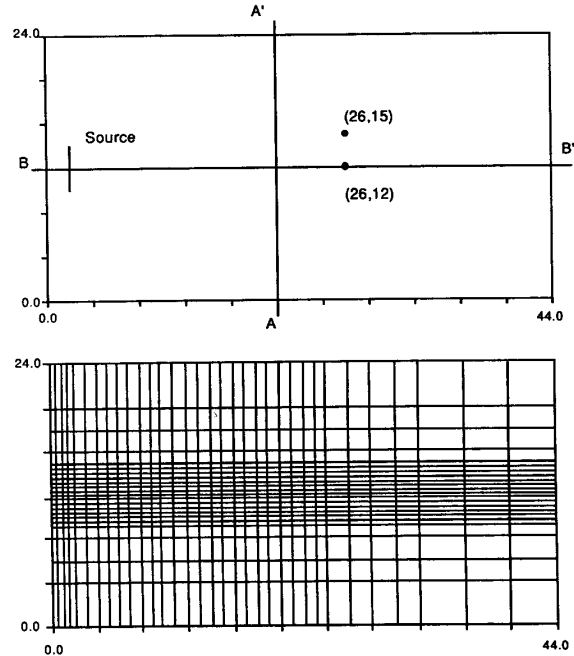

Fig. 1. Computational domain for the example problem.

water flow equation. Here we assume that the velocity and log hydraulic conductivity fields are both stationary over the domain of interest and derive the velocity mean and covariance using the steady state infinite domain spectral approach described by *Gelhar* [1986]. This approach is attractive because it provides closed-form expressions for both moments. The mean velocity vector for two-dimensional flow is [*Gelhar*, 1986]

$$\bar{v}_i = K_G J_i/n \qquad (16)$$

where J_i $\partial \bar{h}/\partial x_i$ is the mean head gradient vector, n is the porosity, and K_G is the geometric mean of the hydraulic conductivity $K(x)$, defined as

$$K_G = \exp \overline{[\ln [K(x)]]} \qquad (17)$$

The velocity spectral density function for two-dimensional flow is [*Gelhar and Axness*, 1983]

$$S_{v_iv_j}(k_1, k_2) = \bar{v}_m\bar{v}_l\left[\delta_{im} - \frac{k_ik_m}{k^2}\right]\left[\delta_{jl} - \frac{k_jk_l}{k^2}\right]S_{ff}(k_1, k_2) \qquad (18)$$

where k is the magnitude of the spectral wave number vector k_l, the function δ_{jl} is one if j is equal to l and zero otherwise, and all repeated indices are understood to be summed from 1 to 2. The function $S_{ff}(k_1, k_2)$ is the log hydraulic conductivity spectral density. The derivation of (18) is based on linearized perturbation expansions of both the groundwater flow equation and Darcy's law and, like any stochastic perturbation analysis, is strictly valid only when the higher-order moments of perturbed variables are sufficiently small. A detailed simulation study presented by *Ababou* [1988] suggests that the *Gelhar and Axness* [1983] results are reasonably accurate for the range of log hydraulic conductivity standard deviations (0.5–1.0) used in our example. Also, it should be noted that the velocity perturbation expressions used in the Gelhar and

TABLE 1. Inputs for the Sample Problem

Parameter	Case					
	1(Nominal)	2	3	4	5	6
$\bar{v}$	0.1 m/day	⋯	⋯	⋯	⋯	⋯
σ_f	0.5	1.0	⋯	⋯	⋯	⋯
λ	2.0 m	⋯			1.0	4.0
α_L	0.25 m	⋯	0.125	0.500	⋯	⋯
α_T	0.10 m	⋯	0.050	0.200	⋯	⋯
Domain size	44 by 25 m	⋯	⋯	⋯	⋯	⋯
Simulation period	225 days	⋯	⋯	⋯	⋯	⋯

Axness analysis satisfy the mass conservation conditions stated in (9) and (10).

The desired two-dimensional velocity covariance function is obtained by taking the inverse Fourier transform of (18). For this example, the log hydraulic conductivity spectrum is assumed to be the two-dimensional isotropic hydraulic conductivity function used by *Mizell* [1982]:

$$S_{ff}(k_1, k_2) = \frac{2\sigma_f{}^2\left(\dfrac{\pi}{4\lambda}\right)^2 k^2}{\pi\left[k^2 + \left(\dfrac{\pi}{4\lambda}\right)^2\right]^3} \tag{19}$$

where $\sigma_f{}^2$ and λ are the variance and integral scale of the log hydraulic conductivity field, respectively. The elements of the resulting velocity covariance matrix are derived in the appendix. The expressions provided in the appendix indicate that the velocity covariance functions are anisotropic even though the log hydraulic conductivity statistics used to generate them are isotropic. This is because the mean head gradient establishes a preferential direction that influences the way random velocity perturbations at different points are correlated. This anisotropy of the velocity covariance propagates through the concentration moment equations into the concentration covariance, which is also anisotropic.

Table 1 summarizes the values used for the example problem input parameters and identifies a nominal case as well as several additional cases used to investigate sensitivity. In each sensitivity case one or two inputs are changed while the others are held fixed at their nominal values. The values of the changed inputs are noted in the appropriate columns. The mean velocity and the nominal statistics of the log hydraulic conductivity field are roughly based on values determined by *Sudicky* [1986] for the experimental tracer test site in Borden, Ontario. The nominal pore scale dispersivities are in the upper range of those determined by *Klotz et al.* [1980].

Figure 2 shows the nominal (case 1 in Table 1) concentration moments obtained by solving the perturbation moment equations with the Galerkin finite element algorithm. The plots on the left show all four moments 75 days after the start of solute release while those on the right show the same moments after 150 days. It is apparent that the area of maximum concentration prediction uncertainty and maximum longitudinal macroscopic flux propagates downstream with the leading edge of the mean concentration plume. The area of maximum transverse macroscopic flux tends to remain close to the source, where the transverse mean concentration gradients are the greatest. The concentration uncertainty is highest in areas where the mean concentration gradients are large and the peak value decreases with time as the plume disperses throughout the simulation domain.

It is instructive to compare the moments obtained from our perturbation approach with sample estimates derived from a Monte Carlo simulation. The Monte Carlo sample moments presented here were computed from a set of 500 statistically independent numerically generated replicates. This sample size was based on a standard error analysis which specified that the 95% confidence intervals for any sample statistic should not be larger than 10% of the estimated value along the portion of the plume axis estending downstream of the source. It should be noted that classical standard error estimates, which rely on normality assumptions [*Kendall and Stuart*, 1977], provide only rough approximations in this application, since the concentration probability density function can be skewed, especially near the boundaries of the simulation domain. These standard error estimates also neglect the important effects of numerical errors introduced in the Monte Carlo transport simulations.

Each Monte Carlo replicate consists of a two-dimensional time-dependent concentration field obtained from a conventional finite element solution of (1), the local transport equation. Velocity fields for our example were obtained from a multivariate multidimensional turning bands algorithm similar to those described in the work by *Shinozuka and Jan* [1972] and *Mantoglou* [1987]. This algorithm generates correlated (and mass conservative) v_1 and v_2 velocity components which reproduce the ensemble statistics given in (16) and (A4).

The finite element grid used in the Monte Carlo simulations must be able to resolve abrupt fluctuations in the random velocity and concentration fields. By contrast, the grid used to solve the perturbation moment equations only needs to resolve relatively smooth changes in the velocity and concentration moment functions. Also, the Monte Carlo solution algorithm must be able to solve a local transport equation which does not have macrodispersive terms and which includes, for this example, advection in two directions rather than one. The uniform node spacing used in the Monte Carlo grid (0.5 m) is based on a criterion of approximately 4 nodes/correlation length suggested by *Ababou* [1988] and on Peclet number constraints. The variable node spacing used in the perturbation solution can be significantly larger, particularly in areas far from the source (see Figure 1). As a result, the perturbation technique requires only one fifth the number of nodes (782 versus 4361) and only one quarter the computation time of the Monte Carlo simulation for the same time step. A time step of 5 days was used for the sample problem.

Figure 3 compares the nominal perturbation and Monte Carlo moments at 225 days. The perturbation method gives a slightly higher concentration standard deviation along the centerline of the mean plume. Although the problem formulation forces the true ensemble moments to be symmetric about the axis of the mean plume, the Monte Carlo sample statistics exhibit minor asymmetries. These asymmetries, which are due to sampling errors inherent in the method, give some indication of the confidence that can be placed in the Monte Carlo results.

Figure 4 compares the perturbation and Monte Carlo moments at 225 days for a log hydraulic conductivity standard deviation of 1.0 (case 2 in Table 1). In this case the discrepancy observed along the centerline of the standard deviation plume is larger, with the perturbation method giving a more pronounced peak and a somewhat higher maximum value. Both solution methods indicate that the mean plume is more dispersed than for the nominal case, due to increases in the mac-
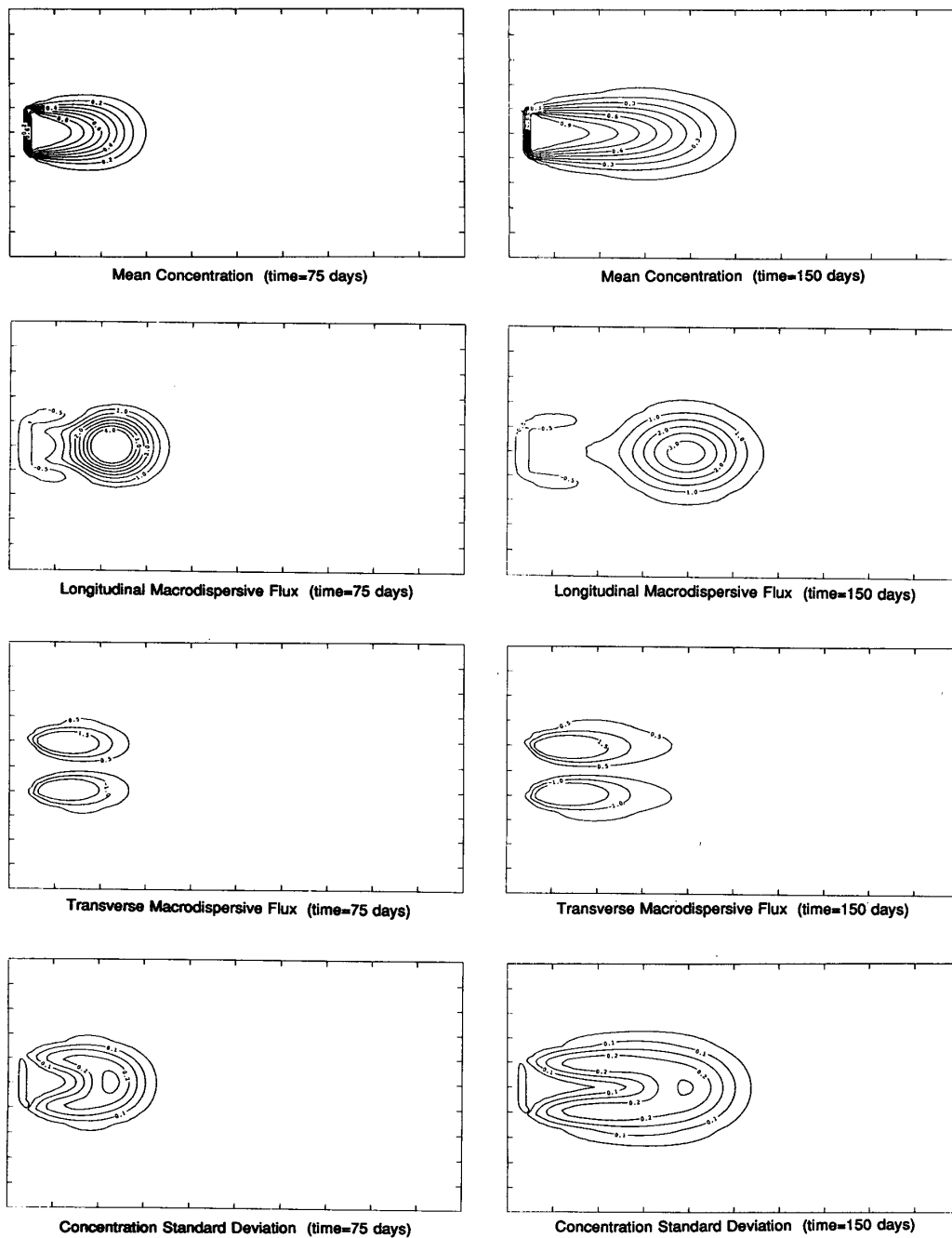
Fig. 2.   Concentration moments for nominal case at 75 and 150 days (perturbation method).

rodispersive flux terms caused by increased variability of the random velocity field. As expected, the concentration standard deviations are higher for this case than for the nominal case and the uncertainty is spread over a larger portion of the domain.

It should be noted that differences between the two sets of moment results compared in Figures 3 and 4 reflect errors contributed by each solution method [*Graham and McLaughlin*, 1988]. As input variability increases, the Monte Carlo method becomes less accurate because of the effects of statistical sampling and numerical error. The Monte Carlo simulations are particularly vulnerable to numerical dispersion, since they must contend with highly variable velocity fields and small dispersivities. The perturbation method also becomes less accurate as input variability increases, since the small perturbation assumption begins to break down. The relative contributions of these various error sources are difficult to quantify and it is not clear, at this point, which method is most

Mean Concentration (Perturbation)

Mean Concentration (Monte Carlo)

Longitudinal Macrodispersive Flux (Perturbation)

Longitudinal Macrodispersive Flux (Monte Carlo)

Transverse Macrodispersive Flux (Perturbation)

Transverse Macrodispersive Flux (Monte Carlo)

Concentration Standard Deviation (Perturbation)

Concentration Standard Deviation (Monte Carlo)

Fig. 3.   Comparison of perturbation and Monte Carlo concentration moments at 225 days ($\sigma_f = 0.5$).

accurate. Nevertheless, it is encouraging to see that the two solution approaches predict most of the same structural features and are qualitatively similar in many respects.

Figures 5 and 6 summarize selected sensitivity analyses of the perturbation moment solutions. In each case the nominal (case 1) result is indicated with a solid line and the results for other sets of input parameters are displayed with dashed lines. The transverse and longitudinal cross sections plotted in Figure 5 show the effect of local dispersivity on the ensemble

mean and standard deviation (the locations of these cross sections are indicated in Figure 1). The longitudinal and transverse local dispersivities are both scaled up and down, relative to the nominal values, by a factor of two (cases 3 and 4 in Table 1).

Changes in local dispersivity have a similar effect on the ensemble mean and standard deviation. In both cases, lower values produce steeper, less dispersed moment distributions. The maximum concentration standard deviation (or uncer-

Mean Concentration (Perturbation)

Mean Concentration (Monte Carlo)

Longitudinal Macrodispersive Flux (Perturbation)

Longitudinal Macrodispersive Flux (Monte Carlo)

Transverse Macrodispersive Flux (Perturbation)

Transverse Macrodispersive Flux (Monte Carlo)

Concentration Standard Deviation (Perturbation)

Concentration Standard Deviation (Monte Carlo)

Fig. 4. Comparison of perturbation and Monte Carlo concentration moments at 225 days ($\sigma_f = 1.0$).

tainty) increases when the local dispersivity is decreased. This result is due to the fact that large local dispersivity values tend to smooth out random plume irregularities, whereas small local dispersivities tend to preserve steep concentration gradients. Note, however, that the areal coverage of the uncertainty distribution decreases with decreasing local dispersivity, reflecting decreased spreading of the random solute plume.

Figure 6 shows the effect of the log hydraulic conductivity correlation scale on the ensemble mean and standard deviation along the cross sections considered above. The correlation scale is varied by a factor of two above and below the nominal value (cases 5 and 6 in Table 1). Higher values produce a more dispersed mean plume as well as higher concentration standard deviations throughout the domain. A larger correlation scale indicates that hydraulic conductivity anomalies (deviations from the mean value) are likely to persist over greater distances and therefore are likely to have a greater effect on the shape of the resulting concentration
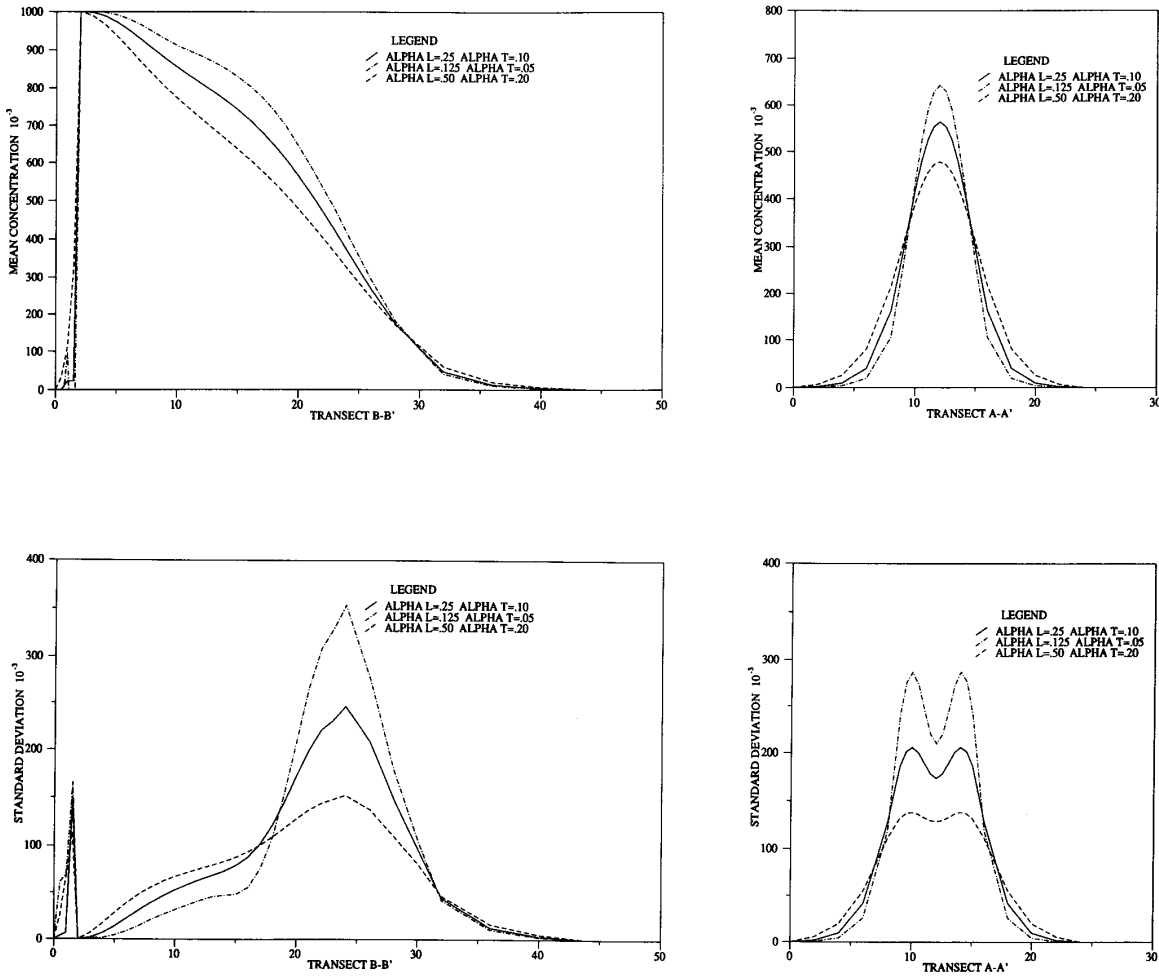
Fig. 5. Sensitivity of concentration mean and standard deviation at 225 days to changes in local dispersivity.

plume. This effect increases variability between plumes across the ensemble, leading both to a more dispersed mean plume and to greater concentration uncertainty.

The correlations between concentrations at different points in a plume indicate how far the information from discrete field observations can be extrapolated over space. For this reason, correlation (or covariance) functions play an important role in spatial estimation and monitoring design (see, for example, *Journel and Huijbreghts* [1978] and *Delhomme* [1979]). Each of the two contour plots in Figure 7 show the normalized covariance (or correlation) between the concentration at a particular location and every other point in the simulation domain. The locations of the two reference points (located at coordinates (26, 12) and (26, 15)) are indicated on Figure 2 with filled circles. The directional dependence (anisotropy) of the concentration correlation is apparent in each of the plots. The nonstationarity of the correlation function is less obvious but may be inferred by comparing the two figures. A careful inspection reveals that the correlation between points with the same separation (magnitude and direction) can be different in different parts of the simulation domain. Some of this non-stationarity may be due to boundary effects, which are more

noticeable in the normalized correlation functions than in the nonnormalized covariances.

Figure 7 indicates that random deviations from the ensemble mean are correlated for longer distances along the streamlines of the mean velocity field than across. Concentration perturbations are positively correlated between points on the same side of the mean plume and slightly negatively correlated between points on opposite sides. That is, point measurements convey considerable information about concentrations downstream of the measurement point but relatively little about concentrations in the transverse direction.

These results, which are application dependent, have clear implications for field sampling. In particular, they support the rather intuitive strategy of sampling along a series of transects running perpendicular to the longitudinal axis of a solute plume. Samples on each transect should be closely spaced relative to the distance between transects and the spacing in each direction should be on the order of one concentration correlation scale. Since the correlation scale and the direction of the plume axis may vary over space (and possibly time) in a real field application, the sampling network should be devel-
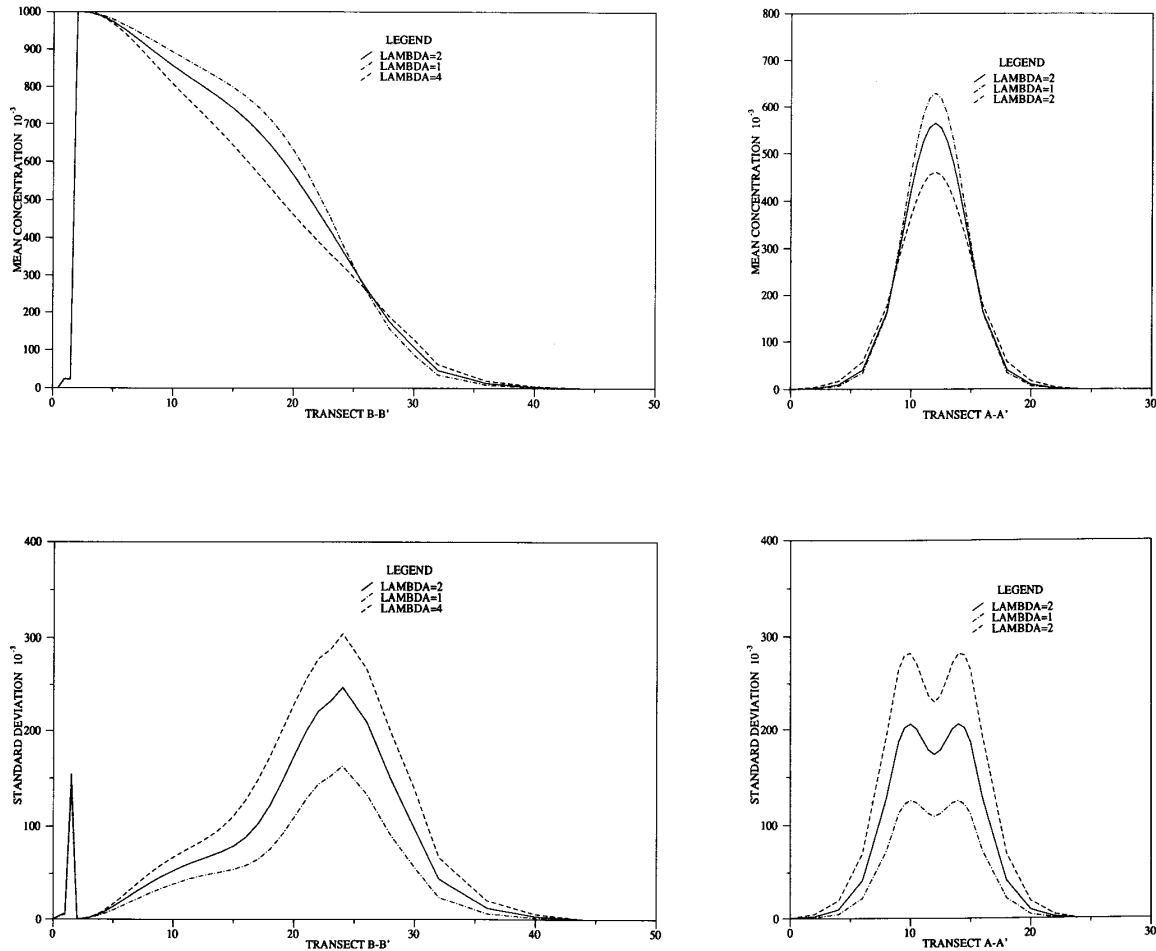
Fig. 6. Sensitivity of concentration mean and standard deviation at 225 days to changes in log conductivity correlation scale.

oped sequentially, with sampling resources allocated over an extended period. This allows monitoring decisions to take advantage of the information gained from measurements collected in earlier rounds of sampling. The conditional moments obtained from a perturbation analysis similar to the one described here may be used to compare the performance of alternative sampling networks and to guide the design of a sequential sampling strategy [McLaughlin and Graham, 1986].

DISCUSSION

The stochastic mass transport model described in this paper provides a convenient method for predicting the movement of a solute plume in heterogeneous aquifers where hydrogeological properties are unknown or highly uncertain. The model computes the unconditional ensemble mean, macrodispersive flux, and covariance of the concentration at any time or location. Each of these moments provides useful information about plume behavior. When measurements of concentration and related variables are unavailable, the unconditional mean is the best (minimum variance unbiased) estimate of the true concentration in any given replicate. The ensemble macrodispersive flux determines how the mean

plume spreads over time and space. Since this flux is derived it does not need to be specified a priori or obtained through a calibration exercise. Macrodispersion is not assumed to be Fickian, although the magnitude of the macrodispersive flux does depend in a complex way on the mean concentration gradient. The unconditional ensemble variance (or the coefficient of variation) may be used to determine the likely range of variability of the true concentration about the ensemble mean. Finally, the ensemble covariance may be used to guide sampling design decisions and to derive conditional concentration estimates [Graham, 1988].

The numerically based approach we use to compute ensemble moments is computationally intensive but considerably more general than analytical approaches which do not apply in near-source regions and cannot deal with forcing functions or boundaries. It is also less computationally demanding than Monte Carlo simulation, at least for problems of the size examined in this paper. Although the straightforward numerical solution algorithm used in this paper is feasible for two-dimensional problems, its computational demands make it impractical for three-dimensional problems of realistic size. In such cases, the moment equations will probably need to be

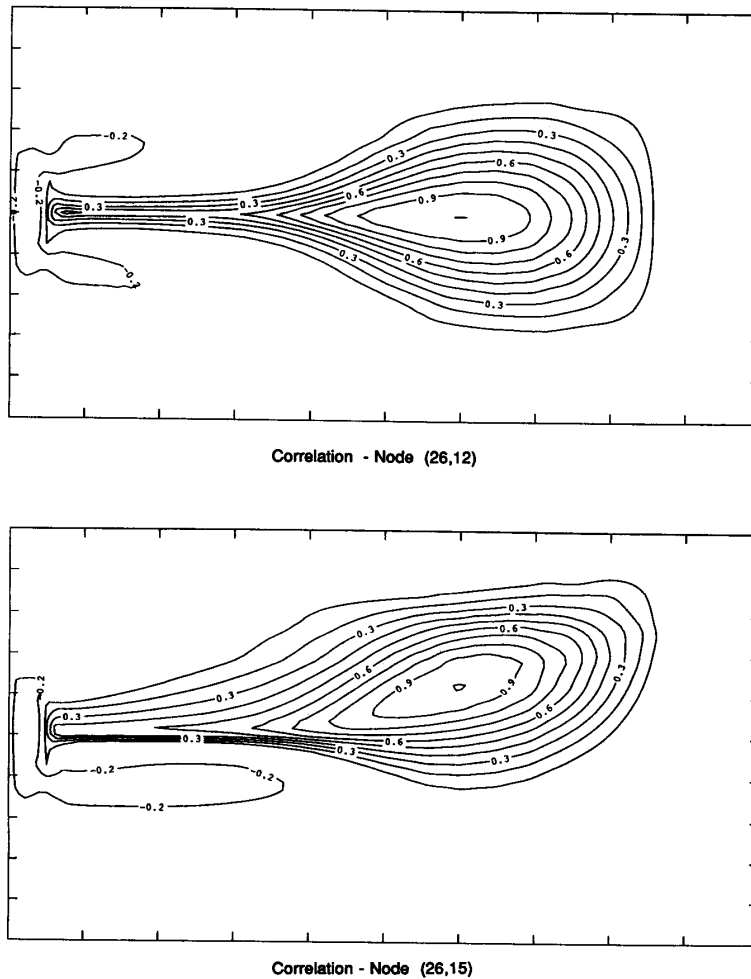Correlation - Node (26,12)

Correlation - Node (26,15)

Fig. 7. Nominal concentration correlations at 225 days, relative to locations (26, 12) and (26, 15).

solved with more sophisticated numerical algorithms which use decomposition techniques and/or take advantage of parallel processing.

Our approach, like many others, relies on small perturbation assumptions which may not be valid in geological formations with log hydraulic conductivity variances much larger than one. The limitations of the small perturbation assumption may be circumvented, at least to some extent, in situations where a larger-scale velocity trend (i.e. a nonstationary ensemble mean) can be identified [McLaughlin and Wood, 1988a]. In such cases, the variance about the ensemble mean is reduced and the small perturbation assumption becomes more acceptable.

The concentration moments obtained for the sample problem show the same general behavior as similar results developed by others. In particular, the ensemble longitudinal and transverse macrodispersive fluxes increase with increasing hydraulic conductivity variance and correlation scale, thus causing increased spreading of the mean plume. Similar behavior is reported for the steady state asymptotic macrodispersivity tensor derived by Gelhar and Axness [1983] and the transient macrodispersivity tensor derived by Dagan [1987]. Also, the

ensemble concentration variance increases with increasing log hydraulic conductivity variance and correlation scale over the entire domain. These general trends are consistent with results reported by Dagan [1984] and Vomvoris [1986]. Finally, random concentration fluctuations appear to be correlated for longer distances along the mean flow direction than across, as found by Dagan [1984], Vomvoris [1986], and Vomvoris and Gelhar [1986].

As mentioned above, the ensemble macrodispersive flux derived in our analysis is a vector which need not behave in a Fickian manner. When applied to the ensemble mean, the Fickian assumption states that the macrodispersive flux vector is related to the mean concentration gradient (at all times and locations) as follows:

$$J_{Di} = -\bar{v}A_{ij}(\partial \bar{c}/\partial x_j) \tag{20}$$

where A_{ij} is a time and space-invariant macrodispersivity tensor. The validity of this assumption may be checked by evaluating spatially integrated time-dependent macrodispersion coefficients derived from the spatial moments of the concentration distribution [Aris, 1956; Graham, 1988]. If these coefficients approach a constant value, the mac-
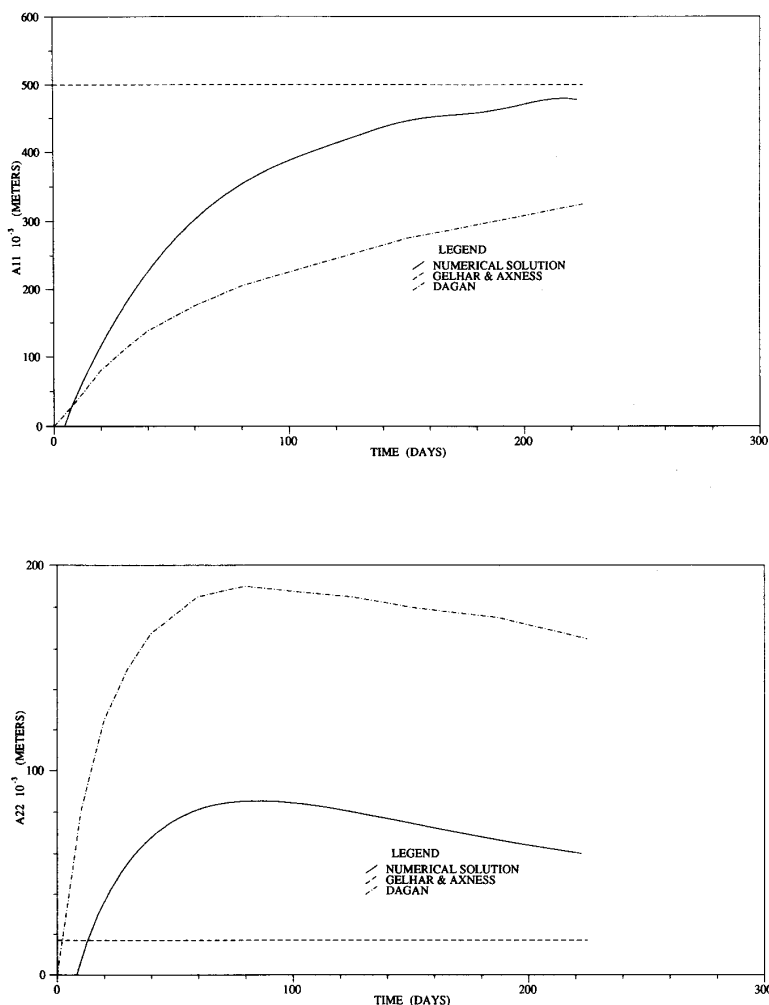
Fig. 8. Evolution of macrodispersivity coefficients derived from a spatial moment analysis of the ensemble mean concentration $(\sigma_f = 0.5)$.

rodispersion process can be treated as asymptotically Fickian at an appropriately large scale.

Figure 8 shows the evolution of the longitudinal (A_{11}) and transverse (A_{22}) macrodispersivities obtained from a spatial moment analysis of our nominal ensemble mean concentration plume, with the temporal and spatial characteristics of the continuous source taken into account and the effects of local dispersivity removed. Also plotted are the macrodispersivity coefficients obtained from the theories of Gelhar and Axness [1983] and Dagan [1984]. The longitudinal macrodispersivity obtained from our moment equations increases momotonically with time and approaches the Gelhar and Axness limit more quickly than the curve obtained from Dagan's analysis. Our transverse macrodispersivity shows behavior similar to Dagan's, rising past the Gelhar and Axness limit to peak at about 80 days (equivalent to a mean displacement of 4λ). It should be noted that the Gelhar and Axness macrodispersivity derivation assumes $\alpha_L/\lambda \ll 1$, while the Dagan derivation assumes $\alpha_L/\lambda = \alpha_T/\lambda = 0$. The macrodispersivities obtained for our nominal case are, by contrast,

based on an α_L/λ value of 0.125. The differences between the numerical and Dagan macrodispersivity curves plotted in Figure 8 are probably due, at least in part, to the relatively large α_L/λ ratio used in our example.

Figure 8 suggests that the ensemble mean plume approaches Fickian behavior (i.e., the macrodispersivities approach constant values) as time progresses. It should be recalled, however, that the ensemble macrodispersive flux accounts for differences among the trajectories of random concentration replicates over the ensemble, rather than the macroscopic spreading of a single plume [Sposito et al., 1986]. The first effect (ensemble uncertainty) influences our ability to predict where a plume is likely to move, while the second effect (spatial dispersion) influences the degree of dilution experienced in any given contamination incident. Figures 9 and 10 illustrate the difference between these effects for two typical Monte Carlo replicates. The two concentration distributions plotted in Figure 9 are taken from the case 1 (nominal, $\sigma_f = 0.5$) ensemble while those plotted in Figure 10 are taken from the case 2 $(\sigma_f = 1.0)$ ensemble. The 0.1 ensemble mean con-
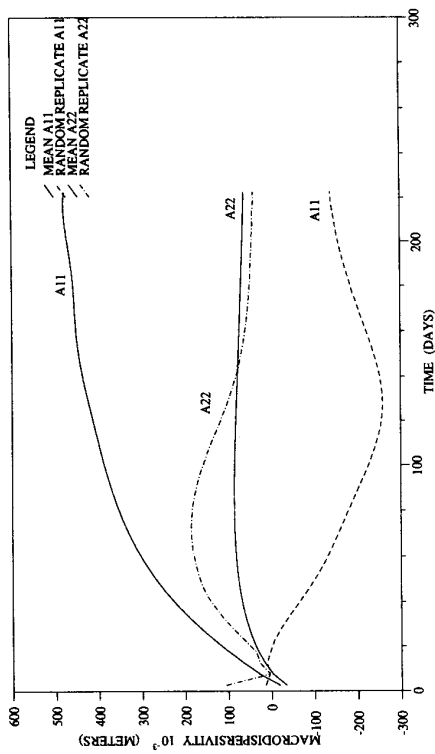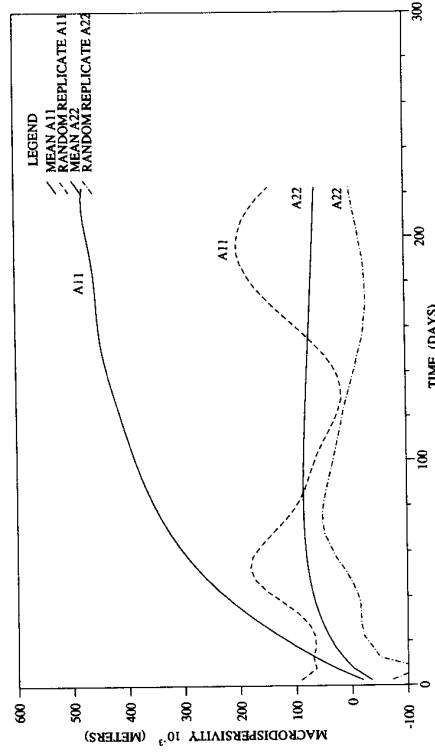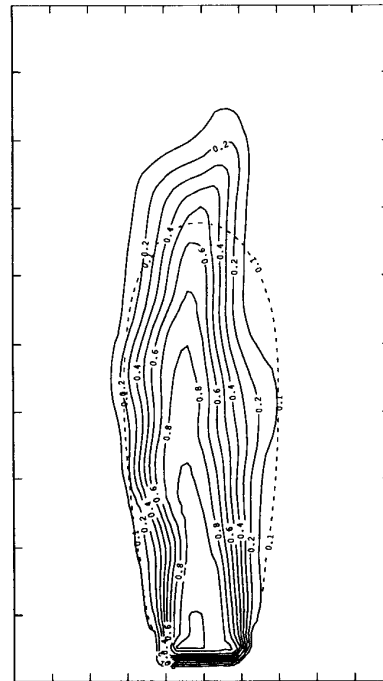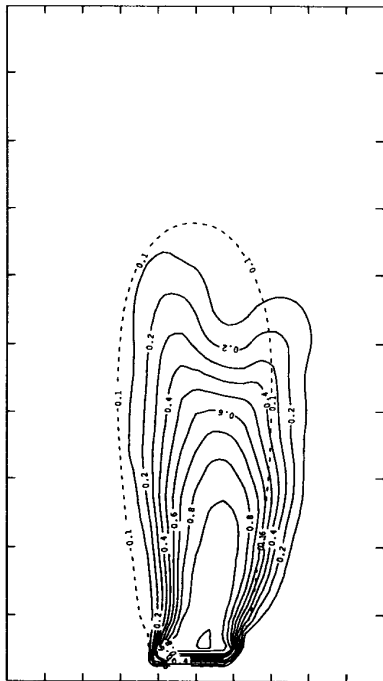
Fig. 9.   Typical random concentration replicates at 225 days ($\sigma_f = 0.5$; dashed lines indicate ensemble mean concentration contours) and associated single-replicate macrodispersivities.
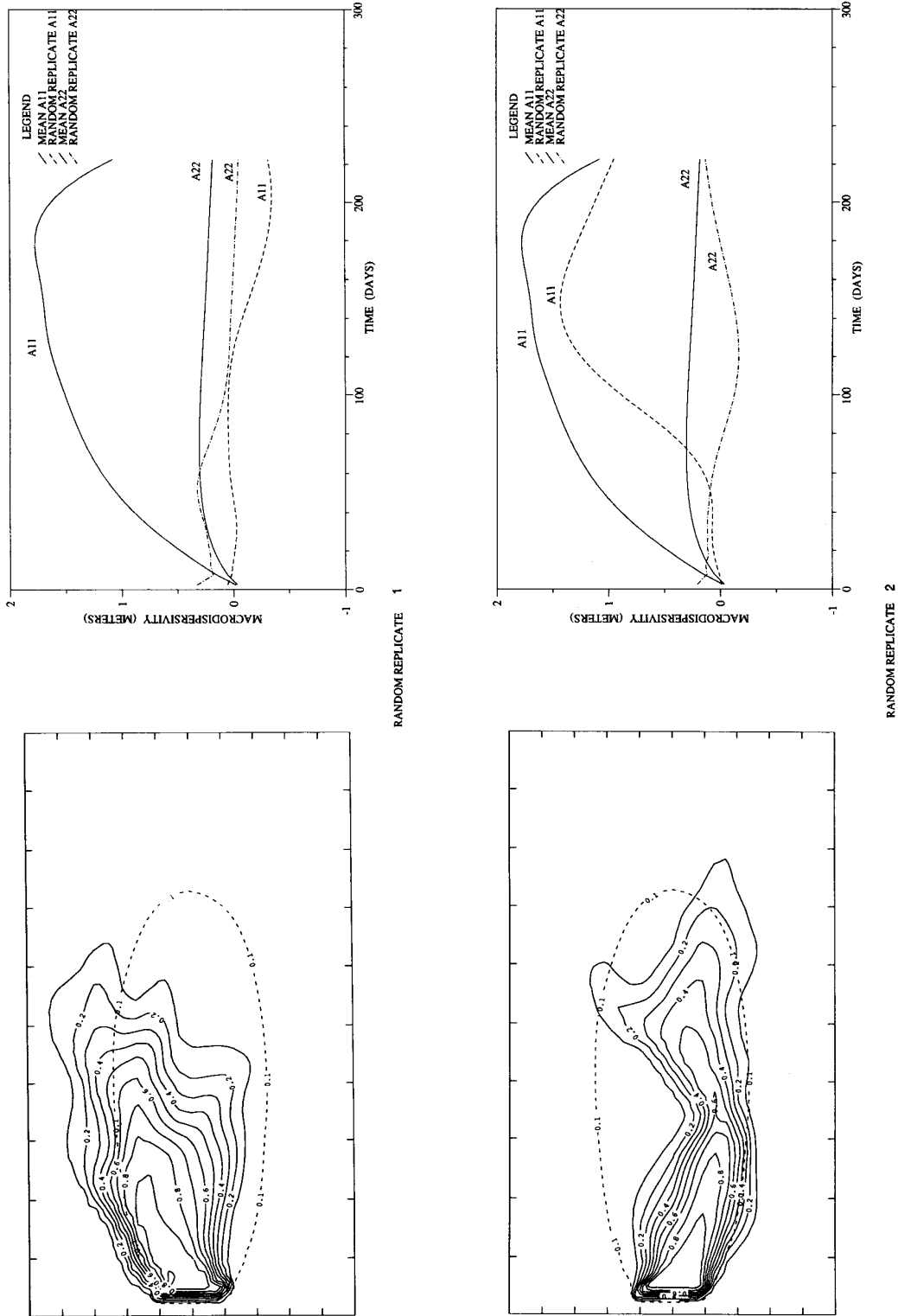
Fig. 10. Typical random concentration replicates at 225 days ($\sigma_f = 1.0$; dashed lines indicate ensemble mean concentration contours) and associated single-replicate macrodispersivities.

tour from the perturbation solution is superimposed on each of the replicate contour plots. Macrodispersivities computed from a spatial moment analysis of each replicate are plotted, together with our ensemble mean macrodispersivities, next to the concentration distributions. Note that the ensemble mean A_{11} value is affected by the boundary of the computational grid at times greater than 170 days in the $\sigma_f = 1.0$ case.

Figures 9 and 10 indicate that individual solute plumes belonging to the same ensemble can differ significantly, depending on the replicate selected. In particular, macrodispersivities computed from spatial moment analyses of individual replicates either do not approach asymptotic values or approach asymptotic values which differ from the ensemble macrodispersivities. The negative dispersivities obtained for some of the random replicates provide additional evidence that the uniform velocity Fickian assumption does not hold at the time and space scales considered here. These results suggest that dilution computations or remediation strategies based on unconditional ensemble mean statistics may not be appropriate for particular site-specific applications, especially in near-source regions.

It is useful to recall that the ensemble description should (ideally) incorporate everything known about solute transport at a particular site. If relatively little site-specific information is available, the ensemble description will be naive and general. It is not likely that the statistics of such an ensemble will provide a good description of an individual plume, especially if the velocity field is very heterogeneous. If, on the other hand, field measurements are abundant, the ensemble description can provide a more specific and informed picture of local conditions. This may be accomplished by conditioning the ensemble or, more strictly, the ensemble statistics, on available measurements of concentration, head, log hydraulic conductivity, and other relevant variables [Delhomme, 1979; Dagan, 1984]. Conditional velocity and concentration statistics may be derived, subject to certain assumptions, directly from unconditional means and covariances [Schweppe, 1973; McLaughlin and Graham, 1986; Graham, 1988]. These conditional statistics depend, as might be expected, on the values and locations of the conditioning measurements. We plan to discuss the implications of conditioning in more detail in future papers.

APPENDIX A: DERIVATION OF THE VELOCITY COVARIANCE

The stationary covariance of a random velocity field can be derived by taking the inverse Fourier transform of the velocity power spectral density function. The general expression for the two-dimensional velocity covariance between two points in space is

$$P_{v_i v_j}(\xi_1, \xi_2)$$
$$= \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \exp\left[i(k_1\xi_1 + k_2\xi_2)\right]S_{v_i v_j}(k_1, k_2)\, dk_1\, dk_2 \quad (A1)$$

where ξ_l is the separation vector extending from the first point to the second, k_l is the spectral wave number vector, i is the imaginary constant $\sqrt{-1}$, and $S_{v_i v_j}$ is the velocity spectral density. Equation (14) relates the velocity spectral density to the spectral density of the log hydraulic conductivity function. If the isotropic log hydraulic conductivity spectrum of (19) is substituted into (18) and the resulting velocity spectral density is substituted into (A1), the velocity covariance expression becomes

$$P_{v_i v_j}(\xi_1, \xi_2) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\pi\bar{v}^2\sigma_f^2}{8\lambda^2}\frac{k^2}{[k^2 + (\pi/4\lambda)^2]^3}$$
$$\cdot \left[\delta_{1i} - \frac{k_1 k_i}{|k|^2}\right]\left[\delta_{j1} - \frac{k_j k_1}{|k|^2}\right]\exp\left[i(k_1\xi_1 + k_2\xi_2)\right]\, dk_1\, dk_2 \quad (A2)$$

All variables appearing here are defined in the text accompanying (16) through (19). Note that the unsubscripted variables J and k are understood to be the magnitudes of the vectors J_l and k_l. Also, the ξ_1 axis is assumed to be aligned with the local mean flow direction so that the ξ_2 component of the hydraulic gradient vector J_l is zero.

Equation (A2) may be evaluated analytically, using contour integration in the complex plane [Erdelyi, 1954; Abramowitz and Stegun, 1970]. The resulting components of the symmetric 2 by two-dimensional velocity covariance matrix are

$$P_{v_1 v_1}(\xi_1, \xi_2) = \bar{v}^2\sigma_f^2/4[A(\xi_1, \xi_2, \lambda) + B(\xi_1, \xi_2, \lambda)K_0(\alpha\xi) + C(\xi_1, \xi_2, \lambda)K_1(\alpha\xi)] \quad (A3a)$$

$$P_{v_2 v_2}(\xi_1, \xi_2) = \bar{v}^2\sigma_f^2/4[D(\xi_1, \xi_2, \lambda) + E(\xi_1, \xi_2, \lambda)K_0(\alpha\xi) + F(\xi_1, \xi_2, \lambda)K_1(\alpha\xi)] \quad (A3b)$$

$$P_{v_1 v_2}(\xi_1, \xi_2) = \bar{v}^2\sigma_f^2/4[G(\xi_1, \xi_2, \lambda) + H(\xi_1, \xi_2, \lambda)K_0(\alpha\xi) + I(\xi_1, \xi_2, \lambda)K_1(\alpha\xi)] \quad (A3c)$$

where K_n is the modified Bessel function of order n, $\alpha = \pi/(4\lambda)$, and the unsubscripted variable ξ is understood to be the magnitude of the vector ξ_l. The algebraic functions $A(\xi_1, \xi_2, \lambda)$ through $I(\xi_1, \xi_2, \lambda)$ are defined as follows:

$$A(\xi_1, \xi_2, \lambda) = \frac{16}{\alpha^4}\left[\frac{6}{\xi^4} - \frac{48\xi_1^2}{\xi^6} + \frac{48\xi_1^4}{\xi^8}\right] \quad (A4)$$

$$B(\xi_1, \xi_2, \lambda) = -6 + 2\xi_1^2\alpha^2 - 2\xi_2^2\alpha^2 - 2\frac{\xi_1^4}{\xi^2}\alpha^2 + 48\frac{\xi_1^2}{\xi^2}$$
$$- 32\frac{\xi_1^4}{\xi^4} - \frac{48}{\alpha^2\xi^2} + \frac{384\xi_1^4}{\alpha^2\xi^2} - \frac{16\xi_1^4}{\xi^4} \quad (A5)$$

$$C(\xi_1, \xi_2, \lambda) = 12\alpha\frac{\xi_1^2}{\xi} - 12\alpha\frac{\xi_1^4}{\xi^3} - \frac{24}{\alpha\xi} + \frac{192\xi_1^2}{\alpha\xi^3}$$
$$- \frac{192\xi_1^4}{\alpha\xi^5} - \frac{96}{\alpha^3\xi^3} + \frac{768\xi_1^2}{\alpha^3\xi^5} - \frac{768\xi_1^4}{\alpha^3\xi^7} \quad (A6)$$

$$D(\xi_1, \xi_2, \lambda) = \frac{16}{\alpha^4}\left[-\frac{6}{\xi^4} + \frac{48\xi_1^2\xi_2^2}{\xi^8}\right] \quad (A7)$$

$$E(\xi_1, \xi_2, \lambda) = 6 - 2\alpha^2\frac{\xi_1^2\xi_2^2}{\xi^2} - 48\frac{\xi_1^2\xi_2^2}{\xi^4} + \frac{48}{\alpha^2\xi^2} - 384\frac{\xi_1^2\xi_2^2}{\alpha^2\xi^6}$$
$$\quad (A8)$$

$$F(\xi_1, \xi_2, \lambda) = 2\alpha\xi - 12\alpha\frac{\xi_1^2\xi_2^2}{\xi^3} + \frac{24}{\alpha\xi} - 192\frac{\xi_1^2\xi_2^2}{\alpha\xi^5}$$
$$+ \frac{96}{\alpha^3\xi^3} - \frac{768\xi_1^2\xi_2^2}{\alpha^3\xi^7} \quad (A9)$$

$$G(\xi_1, \xi_2, \lambda) = \frac{16}{\alpha^4}\left[-\frac{24\xi_1\xi_2}{\xi^5} + \frac{48\xi_1^3\xi_2}{\xi^8}\right] \quad (A10)$$

$$H(\xi_1, \xi_2, \lambda) = 2\alpha^2\xi_1\xi_2 - 2\alpha^2\frac{\xi_1^3\xi_2}{\xi^2} - 24\frac{\xi_1\xi_2}{\xi^2} - 48\frac{\xi_1^3\xi_2}{\xi^4}$$
$$+ \frac{192\xi_1\xi_2}{\alpha^2\xi^4} - \frac{384\xi_1^3\xi_2}{\alpha^2\xi^6} \quad (A11)$$

$$I(\xi_1, \xi_2, \lambda) = \frac{6\alpha\xi_1\xi_2}{\xi} - \frac{12\alpha\xi_1{}^3\xi_2}{\xi^3} + 96\frac{\xi_1\xi_2}{\alpha\xi^3} + \frac{384\xi_1\xi_2}{\alpha^3\xi^5} - \frac{768\xi_1{}^3\xi_2}{\alpha^3\xi^7}$$

(A12)

The zero-lag velocity variances and cross covariance can be evaluated either by taking the limits of (A3a) through (A3c) as ξ approaches zero or by evaluating (A2) directly with ξ equal to zero. The resulting expressions are

$$P_{v_1v_1}(0, 0) = \tfrac{3}{8}\bar{v}^2\sigma_f{}^2$$ (A13a)

$$P_{v_2v_2}(0, 0) = \tfrac{1}{8}\bar{v}^2\sigma_f{}^2$$ (A13b)

$$P_{v_1v_2}(0, 0) = 0$$ (A13c)

NOTATION

A_{ij} macrodispersivity tensor.
$c(x, t)$ solute concentration at location x and time t.
c_0 initial solute concentration (solubility).
D spatial domain of interest.
D_{ij} pore scale dispersion tensor.
∂D boundary of D.
i imaginary constant $\sqrt{-1}$.
J_i hydraulic head gradient vector.
J_{Di} macrodispersive flux vector.
k_i Fourier wave number vector.
K_G hydraulic conductivity geometric mean.
n porosity.
N number of non-Dirichlet nodes in the computational grid used to discretize the distributed moment equations.
$P_{uv}(x, x', t)$ cross covariance between the generic variables $u(x, t)$ and $v(x', t)$.
$[P_{uv}]_{ij}$ spatially discretized approximation to the cross-covariance function P_{uv}.
$S_{uv}(k)$ power spectral density between the generic time-invariant variables $u(x)$ and $v(x')$.
t time.
t_0 initial time.
$\bar{u}, \bar{u}_i$ mean of the scalar or vector function u.
$v_i(x)$ velocity vector at location x.
x_i, x_i' spatial location vectors.
α_L, α_T longitudinal and transverse pore scale dispersivities.
δ_{ij} function equal to 1 if $i = j$ and equal to 0 otherwise.
$\delta(x)$ Dirac delta function.
δu perturbation of the function u about its mean.
$\phi_i(x)$ finite element interpolation functions associated with node i of a computational grid.
λ log hydraulic conductivity correlation scale.
$\sigma_f{}^2$ log hydraulic conductivity variance.
ξ_i separation vector between two locations.

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