Hydrologic Theory of Dispersion in Heterogeneous Aquifers

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Abstract: A general methodology to develop dispersion models in three-dimensional heterogeneous aquifers under nonstationary conditions is presented. Under this procedure, fundamental hydrologic processes influencing chemical dispersion in porous media, such as recharge rate, ground-water flow boundary conditions, regional hydraulic gradients and their transient behavior, as well as nonstationary statistical properties of the flow and dispersion parameters (if known), may be included in the analysis. The method of decomposition, which is a general analytic technique not requiring many of the restricting assumptions of the current small perturbation methods, is used as a basic tool to solve the resulting stochastic partial differential equations. A special application that reproduces the enhanced dispersion of the Borden aquifer tests is presented. The results suggest that the longitudinal and transverse field dispersion coefficients do not exhibit asymptotic values, even in the absence of recharge, and rather grow as linear functions of time and the corresponding longitudinal and transverse velocity variances. If the regional recharge rate is important, the rate of growth of the dispersion coefficients is expected to increase, and the corresponding mean concentration plume would be nonsymmetric with respect to its center of mass. Finally, a comparison between perturbation and decomposition solutions to ground-water equations, for the case of large conductivity variance, is presented.

INTRODUCTION

Fundamental hydrologic processes affecting contaminant migration in soils (the unsaturated zone) include the precipitation regime in the area of interest and its interaction with soil surface characteristics such as vegetation type and density, soil type, properties, and their spatial distribution, evapotranspiration regime and its controlling variables (water vapor pressure gradient distribution, solar radiation intensity, latitude, etc.). The infiltration rate reflects the complex interaction between these intervening variables and should be an integral part in a dispersion model. The infiltration rate is responsible for the vertical displacement of the center of mass of a contaminant plume, and therefore affects advection terms in the resulting transport differential equations. The infiltration rate also determines the magnitude of the dispersion coefficients if a Fickian approximation is adopted.

In the saturated zone, the infiltration rate (and ultimately the recharge rate) interacts with the aquifer hydraulic conductivity spatial distribution and controls the hydraulic heads, the hydraulic gradients, and the aquifer flow velocities. Since the flow velocity determines the magnitude of the dispersion coefficients and the advective terms in the differential equation, the recharge rate should be part of a contaminant dispersion model in the region. In addition, the precipitation regime, and the resulting watershed hydrologic response, also controls the levels in streams and lakes, which are a fundamental component of the boundary conditions in an underlying ground-water flow model. Therefore a ground-water boundary value problem built for predicting contaminant migration in an aquifer should include, at least in steady form, a set of boundary conditions and a specification of the recharge rate based on a hydrologic analysis of the region.

The incorporation of temporal and spatial variations in recharge and boundary conditions, in addition to a statistical representation of aquifer heterogeneity in the hydraulic conductivity, significantly increases the complexity of the dispersion equations and the level of difficulty in their solution. This is one reason for the proliferation of dispersion models that neglect the regional hydrology. The inability to obtain exact solutions of the differential equations has resulted in a multitude of dispersion schemes that manipulate the field conditions to fit limited mathematical methods, such as small perturbation procedures [i.e., Bakr et al. (1978), Dagan (1984), Gelhar and Axness (1983), Neuman et al. (1987), Rubin (1991)]. For a summary and a critical review of the existing stochastic theories of transport the reader is referred to Cushman (1987, 1983) and Sposito and Jury (1986).

Previous works by the writer have studied the effect of recharge and hydrologic physical variables on the dispersion parameters in deterministic one-dimensional aquifers (Serrano, 1992a), and the relative importance of recharge versus aquifer heterogeneity, as expressed by a stationary random field (Serrano and Adomian, in press, 1996; Serrano 1995a, 1993). The purpose of this paper, besides motivating a return to the fundamental physical hydrology as a basis to study field dispersion, is to present a general methodology based on new refinements in decomposition theory to study three-dimensional domains subject to nonstationary random fields in the aquifer velocity distribution. Applications to special cases of dispersion in two-dimensional aquifers with Dupuit assumptions are illustrated. Emphasis is placed on a corresponding boundary value problem as a basis of analysis: the solution of the underlying regional ground-water flow problem (and the consideration of the regional hydrology). These form the basis to define the specific form of the dispersion parameters, the form of the dispersion equation, and its subsequent solution. The method of decomposition, which has become a general analytic technique for dynamic systems, is used as a solution procedure for the differential equations, since it does not require the usual restrictive assumptions of the commonly used methods, and it allows the construction of a series (much like the Fourier series) that converge fast to the exact solution (Serrano and Adomian, in press, 1995; Adomian 1994, 1991, 1986). Finally, a comparison is made between the perturbation and decomposition solutions of ground-water equations subject to large variances in the hydraulic conductivity to illustrate the conceptual difficulties of perturbation methods.

DISPERSION IN A THREE-DIMENSIONAL AQUIFER UNDER NONSTATIONARITY

Neglecting compressive storage, the transient regional ground-water flow problem in an unconfined heterogeneous aquifer is described by

$$\nabla \cdot (K \nabla \phi) = 0, \quad \phi \in \Gamma \tag{1}$$

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J. Hydrol. Eng., 1996, 1(4): 144-151

where $\phi(x, y, z, t)$ = hydraulic head (m) within a three-dimensional domain; $\Gamma \subset \Re^3$; $K(x, y, z)$ = (random) hydraulic conductivity field; t = time coordinate; and (x, y, z) = spatial coordinates, with z the vertical dimension. Eq. (1) is subject to

$$Q\phi = J, \quad \phi \in \partial\Gamma \tag{2a}$$

$$(K \cdot \nabla\phi + I) \cdot \nabla(\phi - z) = n \frac{\partial \phi}{\partial t}, \quad \text{on} \quad z = h \tag{2b}$$

$$\phi = h, \quad \text{on} \quad z = h \tag{2c}$$

where Q = a boundary operator; $J(t)$ = a known function; I = aquifer recharge from rainfall (m/month); n = effective porosity; and $h(x, y)$ = elevation of the free surface (m). Eq. (2*a*) represents the presence of Dirichlet, Neumann (or a combination of the two) boundary conditions; (2*b*) satisfies continuity at the free surface; and (2*c*) requires that the hydraulic head at the free surface must equal its elevation head.

After solving (1) and (2) one obtains the transient distribution of the hydraulic heads and subsequently the statistical properties of the x, y, z components of the velocity field, u_x, u_y, and u_z, respectively. In general the velocity components are functions of x, y, z, t, ω, where ω is the probability variable. The properties of the velocity field constitute vital information for the definition of terms containing dispersion coefficients and advection terms in the corresponding contaminant transport model. In a dispersion model built on the basis of the underlying hydrologic problem, the coefficients of the dispersion differential equation could be ultimately expressed in terms of the recharge rate, the boundary conditions, and their regime. An additional advantage of a hydrologically based dispersion model is the possibility of expressing spatial or scale variability in the dispersion parameters as functions of physically measurable hydrologic variables. Serrano and Adomian (in press, 1996) and Serrano (1993, 1992) found that by introducing recharge from rainfall in the analysis, the resulting dispersion parameters were scale-dependent, and functionally related to recharge as well as the statistical properties of the transmissivity field.

Consider, for example, the three-dimensional solute continuity equation in a large domain, after neglecting the local scale hydrodynamic dispersion

$$\frac{\partial C}{\partial t} + \frac{\partial}{\partial x}(u_x C) + \frac{\partial}{\partial y}(u_y C) + \frac{\partial}{\partial z}(u_z C) = 0,$$

$$C(x, y, z, t) \in \Gamma, \quad 0 < t \tag{3a}$$

$$C = 0, \quad C \in \partial\Gamma \tag{3b}$$

$$C(x, y, z, 0) = f(x, y, z) \tag{3c}$$

where C = solute concentration (mg/L); f = known initial contaminant distribution; and u_x, u_y, u_z = three-dimensional nonstationary transient velocity components whose statistical properties are functions of the known statistical properties of the hydraulic conductivity, the aquifer boundary conditions, and the recharge rate.

As usual in the decomposition method, the solution of (3) could be attempted in one of several ways. One could rewrite (3) as

$$L_t C + \frac{\partial}{\partial x}(u_x C) + \frac{\partial}{\partial y}(u_y C) + \frac{\partial}{\partial z}(u_z C) = 0 \tag{4}$$

where $L_t = (\partial/\partial t)$, and its inverse, L_t^{-1} is the definite integral from 0 to t. Operating with the inverse, the solution to (4) is

$$C = f - L_t^{-1}\frac{\partial}{\partial x}(u_x C) - L_t^{-1}\frac{\partial}{\partial y}(u_y C) - L_t^{-1}\frac{\partial}{\partial z}(u_z C) \tag{5}$$

C on the right-hand side of (5) may be expressed as the series

$$C = C_0 + C_1 + C_2 + \cdots \tag{6}$$

or after taking expectations

$$\bar{C} = \bar{C}_0 + \bar{C}_1 + \bar{C}_2 + \cdots \tag{7}$$

where C, $\bar{C}_0$, $\bar{C}_1$, $\bar{C}_2$, ..., = $E\{C\}$, $E\{C_0\}$, $E\{C_1\}$, $E\{C_2\}$, ..., respectively; and $E\{\ \}$ = expectation operator. The first term in the decomposition series is

$$C_0 = \bar{C}_0 = f \tag{8}$$

Thus the first term in the series is the initial condition. Following decomposition theory (Adomian 1994; Serrano and Adomian, in press, 1996), each term in the series is obtained from (5) by operating on the previous term. The second term in the series is

$$C_1 = -L_t^{-1}\frac{\partial}{\partial x}(u_x C_0) - L_t^{-1}\frac{\partial}{\partial y}(u_y C_0) - L_t^{-1}\frac{\partial}{\partial z}(u_z C_0) \tag{9a}$$

$$C_1 = -L_t^{-1}\frac{\partial u_x}{\partial x} f - L_t^{-1} u_x \frac{\partial f}{\partial x} - L_t^{-1}\frac{\partial u_y}{\partial y} f - L_t^{-1} u_y \frac{\partial f}{\partial y} - L_t^{-1}\frac{\partial u_z}{\partial z} f - L_t^{-1} u_z \frac{\partial f}{\partial z} \tag{9b}$$

$$\bar{C}_1 = -L_t^{-1}\frac{\partial \bar{u}_x}{\partial x} f - L_t^{-1} \bar{u}_x \frac{\partial f}{\partial x} - L_t^{-1}\frac{\partial \bar{u}_y}{\partial y} f - L_t^{-1} \bar{u}_y \frac{\partial f}{\partial y} - L_t^{-1}\frac{\partial \bar{u}_z}{\partial z} f - L_t^{-1} \bar{u}_z \frac{\partial f}{\partial z} \tag{9c}$$

Thus (9*b*) is based on the previously computed (9*a*) and on the known first moment of the velocity. In general any term could be computed as

$$C_i = -L_t^{-1}\frac{\partial}{\partial x}(u_x C_{i-1}) - L_t^{-1}\frac{\partial}{\partial y}(u_y C_{i-1}) - L_t^{-1}\frac{\partial}{\partial z}(u_z C_{i-1}), \quad i \geq 1 \tag{10}$$

that is, the ith-order term is based on the $(i - 1)$ order term, and the expected ith-order term is based on the ith order, or lower order, moment of the random input terms, but not higher order (i.e., hierarchy methods). This feature allows the modeler to compute decomposition terms up to the corresponding known order of the random terms in the differential equation without having to neglect unknown higher-order moments. If the field in question is Gaussian, then an infinite set of terms (an exact solution) could, in principle, be found. Without information on the particular probability law of the velocity field, which is the usual case, we may still approximate a solution to the transport differential equation if information on the lower order moments of the random functions is available. For instance, if information up to the second-order moment in the input quantities is available, the expected mean term in the foregoing series is, from (10)

$$\bar{C}_2 = L_t^{-1}L_t^{-1}\left(2\frac{\partial \overline{u_x u_x}}{\partial x}\frac{\partial f}{\partial x} + \frac{\partial^2 \overline{u_x u_x}}{\partial x^2} f + \overline{u_x u_x}\frac{\partial f}{\partial x} + \frac{\partial^2 \overline{u_x u_y}}{\partial x \partial y} f \right.$$
$$+ \frac{\partial \overline{u_x u_y}}{\partial y}\frac{\partial f}{\partial x} + \frac{\partial \overline{u_x u_y}}{\partial x}\frac{\partial f}{\partial y} + \overline{u_x u_y}\frac{\partial^2 f}{\partial x \partial y} + \frac{\partial^2 \overline{u_x u_z}}{\partial x \partial z} f + \frac{\partial \overline{u_x u_z}}{\partial z}\frac{\partial f}{\partial x}$$
$$+ \frac{\partial \overline{u_x u_z}}{\partial x}\frac{\partial f}{\partial z} + \overline{u_x u_z}\frac{\partial^2 f}{\partial x \partial z} + \frac{\partial^2 \overline{u_x u_y}}{\partial x \partial y} f + \frac{\partial \overline{u_x u_y}}{\partial x}\frac{\partial f}{\partial y} + \frac{\partial \overline{u_x u_y}}{\partial y}\frac{\partial f}{\partial x}$$
$$+ \overline{u_x u_y}\frac{\partial^2 f}{\partial x \partial y} + \frac{\partial^2 \overline{u_y u_y}}{\partial y^2} f + 2\frac{\partial \overline{u_y u_y}}{\partial y}\frac{\partial f}{\partial y} + \overline{u_y u_y}\frac{\partial^2 f}{\partial y^2} + \frac{\partial^2 \overline{u_y u_z}}{\partial y \partial z} f$$
$$+ \frac{\partial \overline{u_y u_z}}{\partial z}\frac{\partial f}{\partial y} + \frac{\partial \overline{u_y u_z}}{\partial y}\frac{\partial f}{\partial z} + \overline{u_y u_z}\frac{\partial^2 f}{\partial y \partial z} + \frac{\partial^2 \overline{u_x u_z}}{\partial x \partial z} f + \frac{\partial \overline{u_x u_z}}{\partial x}\frac{\partial f}{\partial z}$$

J. Hydrol. Eng., 1996, 1(4): 144-151

$$+\frac{\partial \overline{u_x u_z}}{\partial z}\frac{\partial f}{\partial x}+\overline{u_x u_z}\frac{\partial^2 f}{\partial x\partial z}+\frac{\partial^2 \overline{u_y u_z}}{\partial y\partial z}f+\frac{\partial \overline{u_y u_z}}{\partial y}\frac{\partial f}{\partial z}+\frac{\partial \overline{u_y u_z}}{\partial z}\frac{\partial f}{\partial y}$$

$$+\overline{u_y u_z}\frac{\partial^2 f}{\partial y\partial z}+\frac{\partial^2 \overline{u_z u_z}}{\partial z^2}f+2\frac{\partial \overline{u_z u_z}}{\partial z}\frac{\partial f}{\partial z}+\overline{u_z u_z}\frac{\partial^2 f}{\partial z^2}\Bigg) \quad (11)$$

where $\overline{u_i u_j}$ = (cross) correlation function $E(u_i u_j)$. Eq. (11) indicates that the expected third term in the decomposition series requires up to the second-order moment of the velocity field, but not higher than two. All the terms in (11) are computable and analytic if the velocity correlation functions (i.e., if the transmissivity correlation) and the initial conditions are analytic.

An additional advantage of the decomposition series is its fast convergence rate. Usually two or three terms are sufficient to obtain an accurate solution. Serrano and Adomian (in press, 1996) present several comparisons between decomposition and exact solutions of various scale-dependent dispersion equations. Serrano and Unny (1987) showed several comparisons between different decomposition and exact solutions of transient ground-water flow equations. A convergence theorem with proof for the advective dispersive differential equation is developed by Serrano (1992b). Other decomposition schemes for the one-dimensional advective dispersive equation are illustrated in Serrano (1988). For a rigorous mathematical treatment of the convergence problem of decomposition series the reader is referred to Gabet (1994, 1993, 1992), Abbaoui and Cherruault (1994), Cherruault (1989), and Cherruault et al. (1992).

From (6), and truncating at a level consistent with the desired level of accuracy, or the available statistics information on the input parameters, it is possible to obtain a solution from which sample functions, the mean, and the correlation function of the concentration field may be calculated. This information should provide the bases for a ground-water pollution forecasting model, or for an investigation into the specific form of the dispersion parameters. In the preceding treatment a logarithmic transformation to reduce the variance of the parameters was not required. Similarly, the application of decomposition to ground-water equations that contain the hydraulic conductivity as a physical parameter do not require a logarithmic transformation either. No special assumptions on the probability law of the input processes or on the maximum size of the parameter variance (i.e., the subjective assumption required by small perturbation methods) are needed. Obviously the simulation time step is restricted by the magnitude of the input variance, which is a minor modeling problem (Serrano 1992b). Restricting the computational time step of a solution is a better alternative than having to neglect terms in the differential equation to make the solution possible.

APPLICATIONS TO SPECIAL CASES

Due to the frequent occurrence in nature, and in the hydrologic literature, of unconfined aquifers where Dupuit assumptions are reasonable, an exploration of contaminant dispersion in two-dimensional, plan-view aquifers is interesting. For nearly horizontal flow in mildly sloping aquifers, the hydraulic head along the vertical is similar to that at the free surface, that is $\phi \approx h$. In a recent work (Serrano 1995b) that presented analytical solutions to the nonlinear Boussinesq flow eqaution and to the exact steady version of (1), subject to the nonlinear boundary condition in (2), it was found that the Dupuit assumptions are reasonable when the regional gradients are mild (even though the local gradients may be large) and the hydraulic conductivities are large. This is the common Dupuit assumption, which obviates the vertical coordinate, neglects the vertical component of flow, eliminates the need for the free-surface boundary conditions in (2), and results in the Boussinesq flow equation

$$\nabla_2\cdot(Kh\nabla_2 h) = -I, \quad h \in \Gamma_1 \subset \mathcal{R}^2 \quad (12a)$$

$$Q_1 h = J_1, \quad h \in \partial\Gamma_1 \quad (12b)$$

where ∇_2 = a horizontal divergence operator; Γ_1 = two-dimensional areal domain corresponding to Γ; Q_1 = boundary operator; and J_1 = a known function. Considering a steady condition, and defining the term $T = Kh$ as a vertically averaged hydraulic conductivity, or transmissivity, (12) reduces to

$$\frac{\partial}{\partial x}\left(T\frac{\partial h}{\partial x}\right)+\frac{\partial}{\partial y}\left(T\frac{\partial h}{\partial y}\right) = -I, \quad 0 \le x \le l_x, \quad 0 \le y \le l_y \quad (13a)$$

$$h(0, y) = H_1, \quad h(l_x, y) = H_2, \quad h(x, y) = g(x) \quad (13b)$$

$$\frac{\partial h(x, 0)}{\partial y} = \frac{\partial h(x, l_y)}{\partial y} = 0 \quad (13c)$$

where l_x = x-dimension of the aquifer; l_y = y-dimension of the aquifer; and the rest of the terms are as defined before. The restriction $h(x, y) = g(x)$ implies an aquifer with a regional gradient in the x-direction and negligible in the y-direction commonly studied in contaminant hydrogeology.

Aquifer heterogeneity will be characterized as a stationary random field with known second-order statistical properties. Clearly the foregoing mathematical procedure is not restricted to stationarity. We consider this approach because it has received considerable attention in the last two decades. However, in this study the statistical properties of the "raw" transmissivity, rather than its logarithm, will be considered, and no particular probability law will be assumed. Thus the transmissivity field is modeled as

$$T(x, y, \omega) = \bar{T} + T'(x, y, \omega) \quad (14a)$$

$$E[T'(x, y)] = 0, \quad E[T'(x_1, y_1)T'(x_2, y_2)] = R_T(d) = \sigma_T^2 e^{-\rho d^2} \quad (14b)$$

where $\bar{T}$ = mean transmissivity (m^2/month); T' = a stationary random field representing the spatial variability in the transmissivity (m^2/month); $E(\,)$ = expectation operator; R_T = two-point correlation function; σ_T^2 = variance parameter (m^2/month)^2; ρ = correlation decay parameter (m^{-2}); and $d^2 = d_x^2 + d_y^2 = (x_1 - x_2)^2 + (y_1 - y_2)^2$ = square of the distance (m^2) between the points with coordinates (x_1, y_1) and (x_2, y_2).

Using the method of decomposition, Serrano (1995b) solved (13) and derived an expression for the longitudinal component of the velocity field, $u_x = \bar{u}_x + u_x'$, where

$$\bar{u}_x = \left(\frac{C_v^2 - 1}{h_0}\right)(\bar{T}A - Ix) = (C_v^2 - 1)\,\bar{T}\left(\frac{h_0'}{h_0}\right) \quad (15)$$

$C_v = \sigma_T/\bar{T}$ = coefficient of variability of the transmissivity

$$h_0 = Ix^2/2\bar{T} + Ax + H_1, \quad A = [(H_2 - H_1)/(l_x) + (Il_x/2\bar{T})] \quad (16a)$$

$$h_0' = \partial h_0/\partial x \quad \text{and} \quad u_x' = \frac{1}{h_0}\left(T'\frac{\partial h_0}{\partial x} + \int \frac{\partial T'}{\partial x}\frac{\partial h_0}{\partial x}\,dx\right) \quad (16b,c)$$

The process u_x' has the following mean and correlation functions, respectively

$$E\{u_x'\} = 0; \quad R_{u_x}(d_x, d_y) = \frac{\sigma_T^2 e^{-\rho d}}{h_0(x_1)h_0(x_2)}\{h_0'(x_1)h_0'(x_2)$$

$$+ h_0'(x_2)[2A - h_0'(x_2)] + h_0'^2\} \quad (17a,b)$$

If $x_1 = x_2 = x$ we obtain the variance of the longitudinal velocity as

J. Hydrol. Eng., 1996, 1(4): 144-151

$$R_{u_x}(0, 0) = \overline{u_x' u_x'} = \frac{\sigma_T^2 h_0'}{h_0^2} (h_0' - 2A) \quad (18)$$

It is clear that the longitudinal velocity and its statistical properties are functions of distance, aquifer heterogeneity, and recharge rate. Therefore, an aquifer dispersion coefficient functionally defined in terms of the longitudinal velocity should naturally reflect a (scale) dependency on distance. A similar result was obtained by Serrano (1992) for one-dimensional homogeneous aquifers.

Similarly, the transverse component of the velocity is

$$u_y = -\left\{\frac{[2A - h_0'(x)] \dfrac{\partial T'}{\partial x}}{h_0(x)}\right\} \left(1 + \frac{T'}{\bar{T}}\right) \quad (19)$$

The mean, correlation function and variance are (respectively) given by

$$E\{u_y\} = \bar{u}_y = 0 \quad (20a)$$

$$R_{u_y}(d_x, d_y) = \frac{2\rho\sigma_T^2 e^{-\rho d^2}(2\rho d_x^2 - 1)h_0'(x_1)h_0'(x_2)}{h_0(x_1)h_0(x_2)} \quad (20b)$$

$$R_{u_y}(0, 0) = \overline{u_y' u_y'} = 2\rho\sigma_T^2 \left[\frac{h_0'(x)}{h_0(x)}\right]^2 \quad (20c)$$

Eqs. (19) and (20) indicate that the transverse velocity does not depend on y. The correlation function depends on x distance. The variance is a constant function of y. There is a marked dependence on x, aquifer heterogeneity, and recharge.

To study the solute dispersion problem in this aquifer, let us imagine that a contaminant is released at a location $x = b_x$, $y = b_y$. From (3), the solute continuity equation is then

$$\frac{\partial C}{\partial t} + \frac{\partial}{\partial x'}(u_x C) + \frac{\partial}{\partial y'}(u_y C) = 0 \quad (21a)$$

$$C(x', y', 0) = f(x', y') \quad (21b)$$

where C = integrated over the vertical concentration; and $x' = x + b_x$, $y = y' + b_y$.

It is now accepted that at least two distinct mechanisms of dispersion are present (Cushman 1987): one primarily operating at the small scale where the dispersion is controlled by the variability in the pore size and the pore velocity at this scale; and one operating at the large scale, where the dispersion is controlled by the aquifer heterogeneity at this scale. Thus the large-scale pore velocity in the x-direction could be expressed as $u_x(x) = \bar{u}_x + u_{px}' + u_x'$, where u_{px}' represents the random component of the small-scale pore velocity, and u_x' represents the random component of the large-scale pore velocity. With the x-coordinate coinciding with the (mean) regional ground-water pore velocity, the y (transverse) component of the pore velocity is defined as $u_y = u_{py}' + u_y'$, where u_{py}' represents the random component in the y-direction of the pore velocity, and u_y' represents the random component of the large-scale pore velocity in the y-direction. Thus (21) becomes

$$\frac{\partial C}{\partial t} + \frac{\partial(\bar{u}_x C)}{\partial x'} + \frac{\partial(u_{px}' C)}{\partial x'} + \frac{\partial(u_x' C)}{\partial x'} + \frac{\partial(u_{py}' C)}{\partial y'} + \frac{\partial(u_y' C)}{\partial y'} = 0 \quad (22)$$

Adopting the Fickian approximation at the small scale, $u_{px}'C \simeq -D_x(\partial C/\partial x)$ and $u_{py}'C \simeq -D_y(\partial C/\partial y)$, where D_x and D_y are the small-scale dispersion coefficients (m^2/month) in x and y, respectively, defined as the product of a small (laboratory) scale dispersivity times the mean longitudinal pore velocity. Using the concept of separation of variables for a plume size much smaller than $(l_x \times l_y)$, the concentration distribution of a transport equation may be written as (Barry and Sposito 1989)

$$C(x', y', t) = C_i X(x', t) \cdot Y(y', t) \quad (23)$$

where $X(x', t)$ satisfies

$$\frac{\partial X}{\partial t} - D_x \frac{\partial^2 X}{\partial x'^2} + \bar{u}_x \frac{\partial X}{\partial x'} + \frac{\partial u_x'}{\partial x'} X + u_x' \frac{\partial X}{\partial x'} = 0 \quad (24a)$$

$$X(x', 0) = f_x(x') \quad (24b)$$

$Y(y', t)$ satisfies

$$\frac{\partial Y}{\partial t} - D_y \frac{\partial^2 Y}{\partial y'^2} + u_y' \frac{\partial Y}{\partial y'} = 0 \quad (25a)$$

$$Y(y', 0) = f_y(y') \quad (25b)$$

and $f = f_x \cdot f_y$. Using a decomposition scheme similar to the one outlined earlier, one finds that the mean x-component of the concentration is given by the series [from (24)]

$$\bar{X}(x', t) = \bar{X}_0 + \bar{X}_1 + \bar{X}_2 + \cdots, \quad \bar{X}_0 = f_x \quad (26a)$$

$$\bar{X}_1 = t(D_x f_x'' - \bar{u}_x f_x') \quad (26b)$$

$$\bar{X}_2 = \frac{t^2}{2}\left(D_x^2 f_x^{iv} - D_x \frac{\partial^2 \bar{u}_x}{\partial x'^2} f_x - 2D_x \frac{\partial \bar{u}_x}{\partial x'} f_x'' - 2D_x \bar{u}_x f_x''' + \bar{u}_x \frac{\partial \bar{u}_x}{\partial x'} f_x' + \bar{u}_x^2 f_x'' + 2\frac{\partial^2 \overline{u_x' u_x'}}{\partial x'^2} f_x + 3\frac{\partial \overline{u_x' u_x'}}{\partial x'} f_x' + \overline{u_x' u_x'} f_x''\right)$$

$$\vdots \quad (26c)$$

where $f_x^i = (\partial^i f_x / \partial x'^i)$, and the velocity correlation terms are easily derived from (16)–(18).

Similarly, from (25) the mean y-component of the concentration is given by the series

$$\bar{Y}(y', t) = \bar{Y}_0 = \bar{Y}_1 + \bar{Y}_2 + \cdots, \quad \bar{Y}_0 = f_y \quad (27a)$$

$$\bar{Y}_1 = D_y t f_y''; \quad \bar{Y}_2 = \frac{t^2}{2}(D_y^2 f_y^{iv} + \overline{u_y' u_y'} f_y'') \quad (27b,c)$$

$$\vdots$$

where $f_y^i = (\partial^i f_y / \partial y'^i)$, and the velocity correlation terms are easily derived from (19) and (20).

Because of the exponential form of the transmissivity correlation assumed, the velocity cross-correlation terms are negligible (this is easily verified analytically or numerically) and $\bar{C}(x', y', t) \approx \bar{X}(x', t) \cdot \bar{Y}(y', t)$. Thus (26) and (27) constitutes a simplified model whereby the mean concentration distribution may be forecasted, knowing the mean monthly recharge, the regional ground-water flow boundary conditions, and the statistical properties of the transmissivity.

The foregoing formulation is used to investigate the functional form of an equivalent field dispersion coefficient. As an illustration let us estimate the plume longitudinal second moment, $\phi_x = \int_{-\infty}^{\infty} (x' - \bar{u}_x t)^2 \bar{X}(x', t)\, dx'$. Without loss of generality, and as an example, let us assume an initial condition of the form

$$f_x(x') = \frac{C_i}{\sqrt{2\pi\sigma_i^2}} e^{-x'^2/2\sigma_i^2} \quad (28)$$

which is a Gaussian distribution with initial plume variance σ_i^2. As $\sigma_i^2 \to 0$, $f_x(x') \to C_i\delta(x)$, a Dirac delta function of strength C_i, theoretically a point spill. To make the results comparable to the existing effective dispersion coefficients, we consider the case where the recharge rate is zero. Thus from (26)

J. Hydrol. Eng., 1996, 1(4): 144-151

$$\phi_x = \sigma_i^2 + 2D_x t + \frac{\sigma_T^2 A^2 t^2}{n^2 h_0^2} \quad (29)$$

Similarly, the transverse plume second moment is calculated from $\phi_y = \int_{-\infty}^{\infty} y'^2 \bar{Y}(y', t)\, dy'$, assuming $f_y(y') = [e^{-(y'^2/2\sigma_i^2)}/(\sqrt{2\pi\sigma_i^2})]$, and from (27) as

$$\phi_y = \sigma_i^2 + D_y t + \frac{2\rho\sigma_T^2 A^2 t^2}{n^2 h_0^2} \quad (30)$$

Eqs. (29) and (30) imply that an equivalent convection dispersion [or a variable dispersion equation (VDE)] model for the mean concentration would satisfy

$$\frac{\partial \bar{C}}{\partial t} - \bar{D}_x(t)\frac{\partial^2 \bar{C}}{\partial x'^2} - \bar{D}_y(t)\frac{\partial^2 \bar{C}}{\partial y'^2} = 0 \quad (31)$$

The solution to (31) is (Barry and Sposito 1989)

$$\bar{C}(x', y', t) = C_i \frac{e^{-[(x'-\bar{u}_x t)^2/(2\phi_x)]}}{\sqrt{2\pi\phi_x}} \cdot \frac{e^{-(y'^2/2\phi_y)}}{\sqrt{2\pi\phi_y}} \quad (32)$$

Now since the plume moments are the integrals of the effective dispersion coefficients, $\bar{D}_x$, $\bar{D}_y$, then from (29) and (30)

$$\bar{D}_x(t) = D_x + \sigma_{u_x}^2 t = D_x + \frac{\sigma_T^2 A^2}{n^2 h_0^2} t \quad (33a)$$

$$\bar{D}_y(t) = D_y + \sigma_{u_y}^2 t = D_y + \frac{2\rho\sigma_T^2 A^2}{n^2 h_0^2} t \quad (33b)$$

where $\sigma_{u_x}^2 = \overline{u_x' u_x'}$, and $\sigma_{u_y}^2 = \overline{u_y' u_y'}$. Thus we obtain effective longitudinal and transverse dispersion coefficients that are functions of time as well as the corresponding longitudinal and transverse velocity variances. Again (33) also indicates that the effective dispersion coefficients are ultimately functions of the aquifer boundary conditions, regional hydraulic gradient, aquifer mean porosity, transmissivity variance, and correlation decay parameter. The foregoing results were obtained from (26) and (27) after we set $I = 0$. In general the effective dispersion coefficients are also functions of the recharge rate, and the resulting mean concentration distribution is nonsymmetric with respect to its center of mass.

The simplified result of (33) emphasizes a difference with respect to the existing theories, namely, that the effective dispersion coefficients are not asymptotic functions of time [i.e., Dagan (1984), (1986)], and rather grow linearly with a slope equal to the velocity variance. A possible explanation for this discrepancy lies in the differences in the mathematical procedure used. The Dagan model assumes a Gaussian probability distribution for $\log(K)$, whereas the present work does not assume a particular probability law and considers the statistical properties of the actual transmissivity; also, the Dagan model uses the small perturbation method of solution of the differential equations, whereas the present work uses the method of decomposition and does not impose a restriction on the size of the random terms. Serrano (1992a, 1993) also found that in the presence of recharge the dispersion coefficient does not appear to have an asymptotic value either. The nonexistence of an asymptotic value in the field dispersion coefficient was also observed by Paredes and Elorza (1992) using a dispersion model determined by nonstationary random walk techniques, and the concept of fractal geometry.

FIELD VERIFICATION AND COMPARISON BETWEEN PERTURBATION AND DECOMPOSITION

Preliminary verification of scale-dependent models of dispersion that use other versions of the method of decomposition was presented in Serrano (1995a). In that work, an initial attempt to model dispersion in a one-dimensional flow field led to results similar to (33). Comparisons with the classical convection dispersion equation and with the field experiments at the Borden aquifer indicated that the decomposition-based models reproduced the basic features of field-measured contaminant plumes: peak magnitude, peak time, and contaminant spread. In this section we first illustrate an application of (31)–(33) to the Borden site experiments.

The results of the Borden site experiment have been extensively documented in the literature (Mackay et al. 1986). We focus our attention on the implementation of the two-dimensional Dagan model to vertically averaged bromide and chloride concentrations at the Borden site reported by Barry et al. (1988). The parameter values for the aquifer are: $\bar{u}_x = 2.73$ m/month, $\sigma_Y^2 = 0.24$, $l_y = 2.8$ m, $D_x = 0.03$ m^2/month, $D_y = 0.009$ m^2/month, $h_0 = 6.0$ m, $n = 0.33$, $h_0' = 0.0056$; the mean transmissivity is (Serrano 1993) $\bar{T} = 1{,}112.4$ m^2/month, and $\sigma_T = 579.34$ m^2/month as calculated from the log-conductivity variance; the bromide initial concentration is 324 mg/L, and the initial volume is 12.0 m^3.

The log-conductivity correlation length for the Borden aquifer was estimated as $L_y = 2.8$ m. It is logical to assume that the correlation length of the raw hydraulic conductivity is substantially greater than that of the log conductivity. Furthermore, (14) adopts a transmissivity correlation that decays exponentially with the square of the distance. Since such value is not available to estimate its inverse, the parameter ρ of the transmissivity correlation function, a small value consistent with a mildly heterogeneous aquifer, was adopted. The hydraulic conductivity variance of the Borden aquifer was estimated based on small-scale point samples. Small-scale measurements of the hydraulic conductivity offer better resolution, but comparatively greater variances, than large-scale measurements (Cushman 1987). Presumably, large-scale measurements of the hydraulic conductivity, such as those obtained from pumping tests, should produce smaller variances in the transmissivity and consequently smaller, more reasonable, values of the dispersion coefficient. The small-scale value was used in the present simulations, keeping in mind that (33) is sensitive to the magnitude of large-scale transmissivity variance, and that large-scale values should be used in the equations.

After adjusting the values of H_1 and H_2 to reflect a regional gradient consistent with the Borden aquifer, a calculation of the dispersion coefficients for the Borden aquifer was done. Fig. 1 illustrates the longitudinal dispersion coefficients according to (33). Again the main differences between perturbation and decomposition models are illustrated: the decomposition dispersion coefficients increase over time and do not exhibit an asymptotic value, in contrast to the perturbation models. In this application, (33) produces significantly higher values of the dispersion coefficient.

Fig. 2 shows the mean longitudinal bromide concentration distribution nine months after injection as simulated by (31)–(33). While a comparison of the mean concentration is difficult to assess with respect to a single realization measured in the field (Barry et al. 1988), the results approximately reproduce the main features of the plume: peak magnitude, peak time, and enhanced spatial contaminant spread.

To this writer's knowledge, there has not been a comparison of a ground-water small perturbation solution with respect to its corresponding exact solution. There have been accounts of the limitations of small perturbation solutions to simulate hydrologic systems [i.e., Chen et al. (1994), Cushman (1983)]. Decomposition solutions, on the other hand, have been successfully tested with respect to exact analytical solutions [for ground-water equations see Serrano and Adomian (in press, 1996), Serrano (1992b), Serrano and Unny (1987)]. A comparison between decomposition and perturbation solutions of ground-water equations should be made under identical con-

J. Hydrol. Eng., 1996, 1(4): 144-151

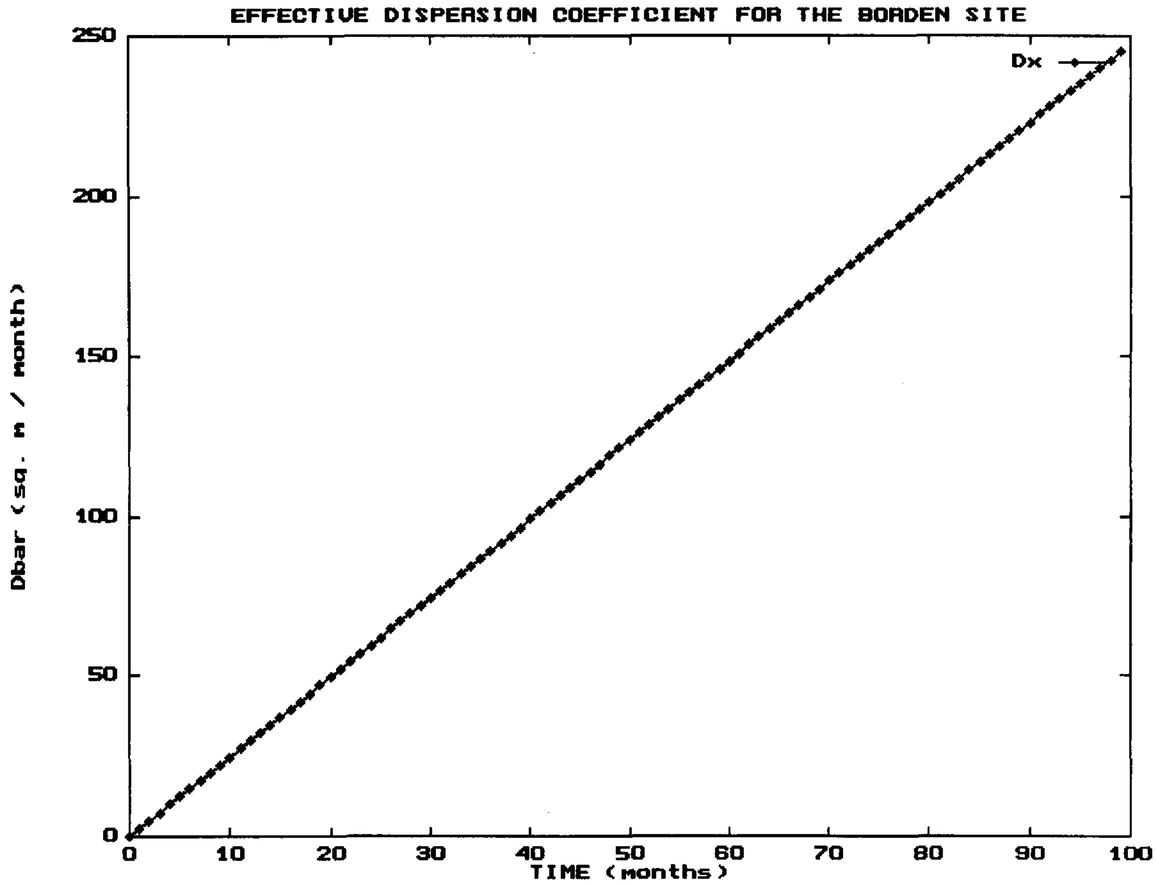

FIG. 1. Field Dispersion Coefficient for Borden Site

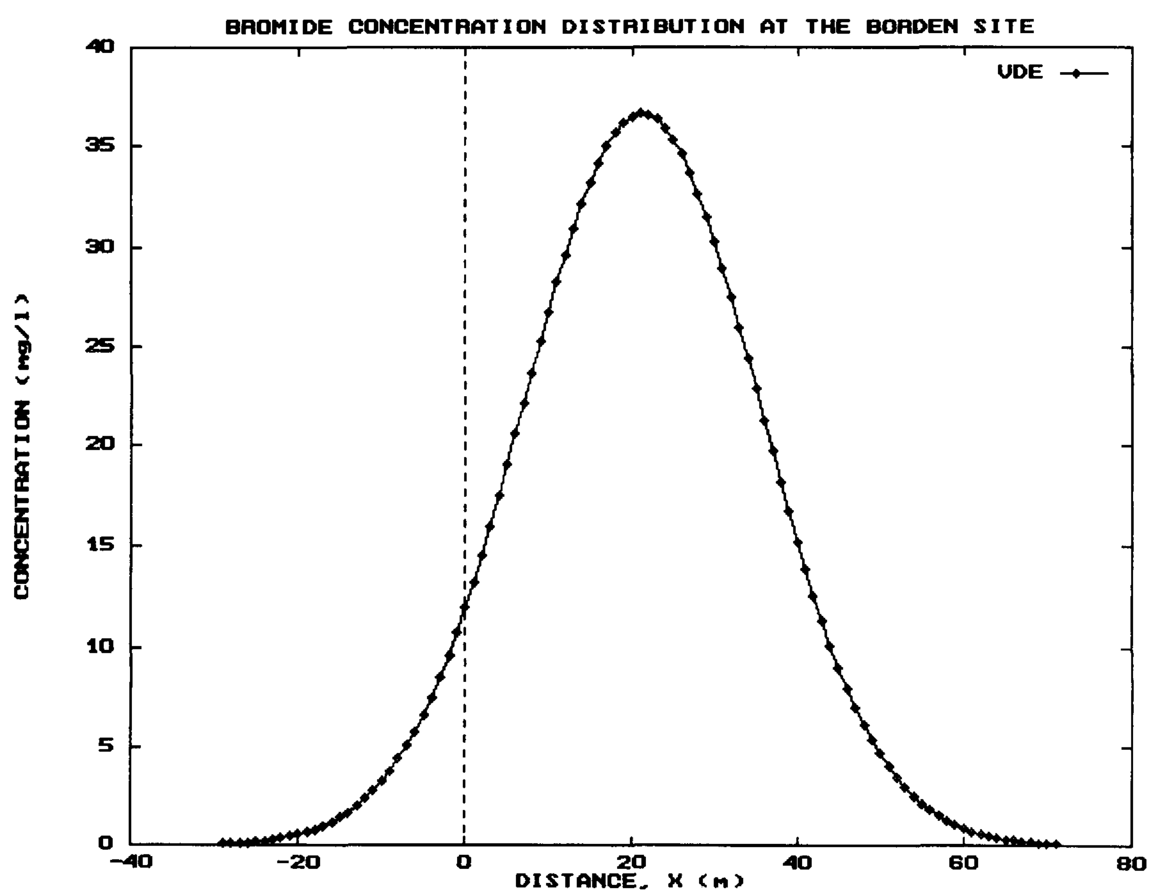

FIG. 2. Longitudinal Bromide Breakthrough Mean Concentration Distribution for Borden Aquifer 648 days after Injection

J. Hydrol. Eng., 1996, 1(4): 144-151

ditions. For example, an objective comparison should observe the performance of both methods as applied to the same equation subject to logarithmically transformed conductivity. To illustrate the conceptual difficulties associated with the foundations of the small perturbation theory of ground-water flow, we consider its classical representation under the light of the method of decomposition and show that it may constitute an improperly posed boundary value problem. The starting classical model is (Bakr et al. 1978)

$$\frac{d}{dx}\left[K(x)\frac{dh}{dx}\right] = 0, \quad 0 \leq x \leq l_x \tag{34a}$$

$$h(0) = H_1, \quad h(l_x) = H_2 \tag{34b}$$

where h = hydraulic head (m); x = horizontal distance (m); l_x = aquifer length (m); and $K(x)$ = hydraulic conductivity (m/month). We have added a set of boundary conditions. Define $L_x = (\partial^2/\partial x^2)$, and write (34) as

$$L_x h = -\frac{dLnK(x)}{dx}\frac{dh}{dx} \tag{35}$$

or

$$h = -L_x^{-1}\frac{dLnK(x)}{dx}\frac{dh}{dx} \tag{36}$$

Use of the decomposition method will result in the series $h = h_0 + h_1 + h_2 + \cdots$, where

$$h_0 = H_1 + ax, \quad a = \frac{H_2 - H_1}{l_x} \tag{37a}$$

$$h_1 = -L_x^{-1}\frac{dLnK(x)}{dx}\frac{dh_0}{dx} = -a\int LnK(x)\,dx \tag{37b}$$

$$h_2 = -L_x^{-1}\frac{dLnK(x)}{dx}\frac{dh_1}{dx} = aL_x^{-1}\frac{dLnK(x)}{dx}LnK(x) \tag{37c}$$

$$\vdots$$

Following the classical small perturbation formulation, $LnK(x) = K_l + K'(x)$, where K_l is a constant; and the process $K'(x)$ has the properties $[K'(x)] = 0$, $[K'(x_1)K'(x_2)] = \sigma_y^2 e^{-\rho(x_1-x_2)^2}$, where σ_y^2 is the log hydraulic conductivity variance parameter, and ρ is the correlation decay parameter. Taking expectations on (37), we obtain the series for the mean hydraulic head

$$\langle h_0\rangle = H_1 + ax; \quad \langle h_1\rangle = -aK_l x; \quad \langle h_2\rangle = a\sigma_y^2 x \tag{38a-c}$$

$$\vdots$$

As an example, consider an aquifer with a large mean hydraulic conductivity of 200.0 m/month and a large variability in the log hydraulic conductivity $\sigma_y^2 = 3.0$, which is well outside the range of validity of small perturbation solutions. For a log-normal distribution this translates into $K_l = 3.8$. Set $H_1 = 10.0$ m, $H_2 = 11.0$ m, and $l_x = 1{,}000.0$ m. Thus the maximum value for the first term in (38) $\max[|\langle h_0\rangle|] = 11.0$ m, $\max[|\langle h_1\rangle|] = 3.8$ m, $\max[|\langle h_2\rangle|] = 3.0$ m, etc. Clearly the decomposition solution (38) exhibits uniform convergence, even in the case of very large variances. This result has been obtained without neglecting elements of the differential equation judged to be "small." The series may be truncated after a convergent series gives a solution at the desired resolution. In this example an infinite set of terms may be included because of the log-normality assumption. While this result is mathematically correct, on close examination one discovers that $\sigma_y^2 = 3.0$ corresponds to a coefficient of variability $C_v = 1{,}908.55\%$ and a standard deviation in the field hydraulic conductivity of $\sigma_k = 3{,}817.1$ m/month. This contradicts any observed hydraulic conductivity record and implies the existence of physically nonrealizable negative conductivities. While recent modifications of the small perturbation conception of ground water (Neuman and Orr 1993) claim accuracy for $\sigma_y^2 = 7.0$, clearly the underlying conductivity field is fictitious. This leads to the conclusion that coefficients of variability of the order of 50% in the field hydraulic conductivity are indeed large and that values beyond 100% represent academic examples.

On the other hand, one may attempt to solve (34) by properly constructing a decomposition series without the logarithmic transformation. It is easy to show that on taking expectations, all the terms in the series vanish except the first, h_0 in (38), thus giving the correct physical result: in the absence of recharge, the mean steady hydraulic head is independent of the hydraulic conductivity and its variance, and must equal the deterministic solution (a straight line between the two boundaries). Therefore, while the logarithmic transformation conveniently adjusts the variances for small perturbation solutions, it probably yields an incorrect hydrologic model (Cushman 1983; Unlu et al. 1989).

SUMMARY AND CONCLUSIONS

A general methodology to develop dispersion models in three-dimensional heterogeneous aquifers under nonstationary conditions was presented. Under this procedure, fundamental hydrologic processes influencing chemical dispersion in porous media, such as recharge rate, ground-water flow boundary conditions, regional hydraulic gradients and their transient behavior, as well as nonstationary statistical properties of the flow and dispersion parameters (if known), may be included in the analysis. The method of decomposition, which is a general analytic technique not requiring many of the restricting assumptions of the current methods, is used as a basic tool to solve the resulting stochastic partial differential equations.

Inclusion of the regional hydrologic processes in dispersion analysis offers an understanding of the physical factors affecting dispersion phenomena, and results in scale-dependent dispersion parameters that are functionally, and explicitly, related to the regional hydrology. In addition, the inclusion of the natural boundary conditions in deterministic, stochastic, steady, or transient form, results in well-posed boundary value problems, and more meaningful mathematical models than those that neglect them.

The dispersion problem in the extensively studied two-dimensional, plan-view aquifer with Dupuit assumptions was reformulated using the decomposition method as a special application. Natural boundary conditions and recharge were added to the problem, and the "raw" transmissivity field, rather than $\log(K)$ was represented as a random field without any particular assumption on its probability law or the size of the variances. A ground-water pollution forecasting model was developed using the solution of the differential equations. Finally, the effective longitudinal and transverse dispersion coefficients were derived and used to reproduce the bromide concentration distribution of the Borden aquifer test. The results reproduced the enhanced longitudinal dispersion reported in the literature. The results also suggest that the longitudinal and transverse field dispersion coefficients do not exhibit asymptotic values, even in the absence of recharge. In this work the dispersion coefficients grow linearly as functions of time and the corresponding longitudinal and transverse velocity variances. If the regional recharge rate is important, the rate of growth of the dispersion coefficients is expected to increase, and the corresponding mean concentration plume would be nonsymmetric with respect to its center of mass.

A comparison between a perturbation solution of ground-water equations with respect to decomposition solutions indicated that perturbation models with logarithmically trans-

J. Hydrol. Eng., 1996, 1(4): 144-151

formed conductivities may generate improperly posed boundary value problems.

Future research should be devoted to investigation into the possible analytical forms of nonstationary transmissivity fields occurring in nature and the derivation of corresponding forecasting models and effective dispersion coefficients using a methodology such as the one presented here. An important item should be the consideration of transient (seasonal) effects of the recharge rate, the boundary conditions, the phreatic surface, and the hydraulic gradients. It is hoped that the regional hydrology will be included in future, more realistic, dispersion analyses.

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J. Hydrol. Eng., 1996, 1(4): 144-151