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Stochastic Hydrology and Hydraulics

Development of stochastic partial differential equations for subsurface hydrology

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Abstract: The use of stochastic models in subsurface hydrology is growing at a logistic pace. To tie together a number of different stochastic methodologies for deriving subsurface transport equations, we have put together a brief review of some of the more common techniques. Our attention is confined to a few select methodologies so that we might delve in detail into assumptions required by the various approaches and their strengths and weaknesses. The methods reviewed include: Martingale, stochastic-convective, stochastic-relativist, spectral-integral, perturbative, statistical-mechanical, and generalized hydrodynamics. Within this list, we also have included a few stochastic methodologies which have been used solely to develop expressions for the dispersion tensor.

Key words: Martingale, stochastic-convective, stochastic-relativist, spectral-integral, perturbative, statistical-mechanical.

1 Introduction

When I was asked to review stochastic differential equations for *Stochastic Hydrology and Hydraulics*, I was somewhat at a loss as to where I should start and what material I should cover. As everyone is aware, stochastic hydrology has become extremely broad-based over the last decade and appears to be growing logistically (Dagan 1986). At first thought, being somewhat of an applied mathematician, I considered reviewing various methodologies for solving the stochastic transport equations of subsurface hydrology. I have decided against this approach for a number of reasons, not the least of which is that to do it "right", a large number of functional analytic results must be presented in an intuitive fashion so that the reader unfamiliar with the field can comprehend the methodology – this would make the document extremely long. My second thought was to present a general survey of the literature on the use of stochastic differential equations in subsurface transport. While such a survey may be interesting, it would be quite dry for those researchers working in the field, such as the readers of this journal. After much more deliberation, I have decided to review a few "select" methodologies for deriving subsurface stochastic transport equations. Actually, I will do a little more than review existing methodologies, for I am also going to take the liberty of briefly introducing a methodology that has been used with great success in other fields, but has yet to be introduced to hydrologists – that is, generalized hydrodynamics.

I apologize in advance to those authors whose methodologies I do not review; I

have tried to select the techniques most familiar to me and the methodologies that I perceive as having the widest influence and applicability. Thus, this document is not a full review of the field, but rather a review of select papers which should be of interest to those working on the foundations of stochastic subsurface transport.

The remainder of this review is broken into the following parts. Section 2 reviews the origins of stochasticity in subsurface transport. Section 3 presents several select methodologies for developing stochastic equations when natural scales of motion are in evidence. Section 4 presents a methodology for developing stochastic transport equations when no known natural scales exist. In Section 5 we present the method of generalized hydrodynamics. Section 6 reviews in detail two methodologies for developing asymptotic transport coefficients of relevance to the stochastic convective-dispersion equation. The paper is concluded with a brief summary in Section 7.

2 Origins of randomness

To be able to appreciate the need for stochastic differential equations in the field of porous media transport, an understanding of the origins of randomness in porous formations is required. To this end, an appreciation is necessary for the role scale of observation plays in all transport processes. The easiest way to see the fundamental role that spatial scale of observation plays in a heterogeneous environment is to imagine oneself looking at the world thorough a camera with an infinitely fast shutter speed and a zoom lens which is capable of resolving scales on the order of miles down to scales on the order of angstroms. This imaginary camera viewpoint is not physically unrealistic in the sense that every measurement process resolves around the taking of a snapshot (measurement) of some phenomenon on a scale which corresponds to the scale of resolution of the instrument.

Let us take, as an example, a picture of a planar cross-section of the surface of the earth. Our initial picture of this cross-section centered at the point $\mathbf{x}$ might have a minimal resolution of meters (e.g., a landsat image). On this scale of resolution we would be able to see geologic heterogeneities larger than a square meter ("field-scale" heterogeneities), but we would not see heterogeneities on a scale smaller than this. Suppose next that we increase our resolution by two orders of magnitude (say a modern U-2 type photo) so that we can observe heterogeneities on scales larger than a square centimeter. Our picture of the world, centered at $\mathbf{x}$, has now completely changed. What had previously appeared to be homogeneous (deterministic in a certain sense) now appears to be heterogeneous (random in the same sense), i.e., spatial details on scales smaller than a square meter, yet larger than a square centimeter, which had previously appeared uniform, now appear to be quite non-uniform owing to the granular structure of the soil. By continuing to increase the resolution, we successively see detail on scales of square millimeters, square microns, and etc. until finally we are on a scale of resolution of say square angstroms. On the square millimeter scale we can pick out details such as worm holes and small cracks and fractures that had previously been unnoticeable. On the square micron scale we can actually see grains and pores, and on the angstrom scale we view individual atoms and molecules. With each increasing level of resolution we see randomness manifest on a new scale which had not previously appeared. From this discussion it appears that randomness is a function of the scale of observation.

The reader should note that there is an upper bound on the amount of information an instrument of a given technology can process in a finite period of time. Assuming the camera processes the same amount of information on each scale, we necessarily must have the maximum window size (field of view) decreasing as the resolution increases. It is this fact that leads to an additional source of randomness

in heterogeneous media. That source is parameter uncertainty which is due to the finite window size, i.e., the field is generally large enough to preclude us from taking enough pictures to cover the whole field. Larger windows generally are associated with less parameter uncertainty but poor resolution, while smaller windows have higher resolution but more parameter uncertainty. Thus both the resolution of the instrument and the instrument window size affect stochasticity.

What about temporal resolution as it relates to randomness ? The spatial evolution of any dynamic variable is governed by the Liouville equation (McQuarrie 1976, p. 120). Hence, spatial and temporal scales of motion are intimately connected. When we take a snapshot with a high spatial resolution, whether or not that snapshot will be blurred or sharp, depends on the dynamic evolution of the process being measured. In general, if the dynamic variable is evolving at a fixed rate, then for a camera to obtain a sharp, high-resolution image of the dynamic variable, it must have a shutter speed much faster than this rate of evolution. We may perform a Gedanken experiment in time similar to our earlier spatial experiment. In this case, we now allow our shutter speed to vary. We maintain an infinite spatial resolution and an infinite spatial window. For very slow shutter speeds (say on the order of centuries) we might resolve glacial movements, but nothing of shorter duration. For shutter speeds on the order of years, we might see houses come and go, but one would not observe the actual building of the house. If we continue to increase the shutter speeds (temporal resolution), we eventually can view fluid motion, atomic motion, subatomic motion, and etc. On each one of these temporal scales randomness is manifested in a different fashion. And as in the spatial case, we also have parameter uncertainty varying with the temporal window.

In summary, a process (dynamic variable) may appear deterministic (uniform or smooth) on one scale, but chaotic or random on a smaller or larger scale. Thus stochasticity is a function of the scale of observation, i.e., it is a function of the scale of the instrumentation used in any given experiment -- by scale of instrumentation I mean as an upper bound the window size and as a lower bound the resolution of the instrument. It should be noted that in some experiments the window size and the scale of resolution are identical. In conclusion, any rational theory of random media must specify a priori the spatial and temporal resolution of the experimental apparatus and the associated window size. To the author's knowledge, only four investigators have attempted to build stochastic theories with this in mind, Clifton and Neuman (1982), Neuman and Depner (1987), Dagan (1986) and Cushman (1984, 1986).

3 Natural scales

It is common to assume porous media have a natural hierarchy of distinct spatial and temporal scales, e.g., atomic, hydrodynamic, pore, REV, lab, field, etc. On each of these scales, the transport equations take basically the same form; only the interpretation of the parameters differs (Cushman 1984). If there are natural scales of motion, according to the discussion in the last section, it is important that instruments be designed with the aim of measuring motions on these scales. In Fig. 1. we have presented a hypothetical plot of a dispersion curve for a porous medium. The function $\omega_{max}(k)$ is the frequency at which, for fixed k, the power spectrum of the current correlation function is a maximum. Regions where the plot is linear are natural scales and the slopes of the linear segments of the plot are the adiabatic sound speeds on these scales. Regions over which the slope of the curve is nonlinear are known as preasymptotic regions and one would expect transport parameters to be wave number and frequency dependent in these regions. This point will be discussed in more detail in a later section.

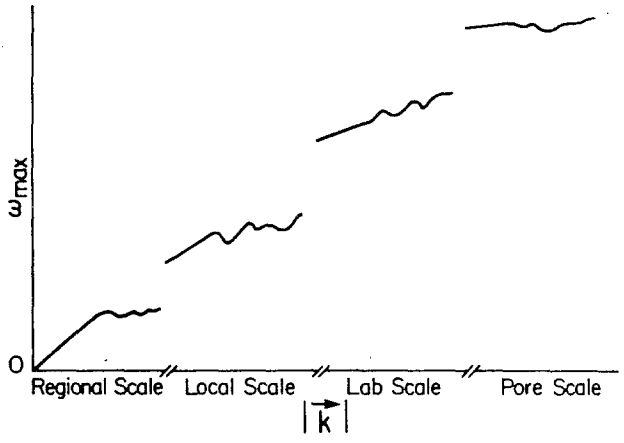

Figure 1. The dispersion curve for the power spectra of the current correlation function, ω_{max} is the frequency at which the power spectra is a maximum. Slopes of the linear regions of the plot represent the adiabatic sound speed at that scale

We will illustrate a few methods for developing stochastic transport equations when a natural hierarchy of scales is assumed. Bhattacharya and co-workers (Bhattacharya 1985; Bhattacharya and Gupta 1979, 1983, 1984; Bhattacharya et al. 1976; Gupta and Bhattacharya 1983; and Gupta et al. 1981) have a particularly notable approach to the multiscale problem. Bhattacharya's method is what I would classify as a Martingale method (For recent easy to read references on Brownian motion and Martingale's see e.g., Durret 1984; Williams 1979; Harrison 1985; the classical references on Martingale's are Doob 1953; and McKean 1969. The two most practical engineering type books I've seen on the subject are Schuss 1980; and Karlin and Taylor 1981, Vol. II). Bhattacharya and co-workers basically assume there are at least four natural scales of motion; the kinetic, microscopic, Darcy, and field scale. The three lower scales are defined as:

Kinetic. The time scale is defined by the time over which a solute molecule undergoes a large number of collisions with the surrounding liquid molecules, but which is not larger than its average collision time with the solid phase. The spatial scale (not explicitly specified by Bhattacharya) would be larger than the molecule mean free path, yet smaller than the average pore width.

Micro. "To specify the microscopic scale, fix a point within a porous medium. Consider volumes around this point gradually increasing in size starting from a small volume, say, lying entirely within the liquid phase. The average liquid velocities over these volumes will initially exhibit extreme variations because of the irregularity in the porous medium geometry at the pore level. However, as the volume is further increased, these extreme variations are averaged out and the average velocity stabilizes. The smallest volume over which this stability takes place is a microscopic (velocity) REV. The mean time in which a solute molecule traverses a microscopic REV specifies a microscopic unit of time."

Darcy. "... a spatial scale larger than the microscopic at which the equations governing solute transport become translation invariant. For example, at this scale the volume average of the microscopic liquid velocity does not vary with spatial coordinates. The smallest volume beyond the microscopic scale at which this translation invariance manifests itself is a Darcy scale REV. The mean time that a

solute molecule takes to traverse such an REV will be taken to be a Darcy scale time unit. It is assumed that a large number of microscopic REV's make up a single Darcy REV."

I am not quite sure what Bhattacharya means by this last definition. I assume he means the dispersion tensor and average convective velocity are independent of **x**. If so, this is a non-operational definition in that it can never be checked in the field.

Bhattacharya and co-workers begin the development of their equations by assuming that on the kinetic scale the velocity of a "large", nonreactive, solute molecule, $\mathbf{V}(s)$, is stationary and ergodic on some finite interval $s \varepsilon [0, t_o]$ where t_o is a microscopic time unit. For times much greater than the average molecular collision time with a pore wall, the displacement $X(t_o) - x_o$ of the molecule from its initial position x_o satisfies a central limit theorem (CLT). The displacement,

$$\int_o^{t_o} \mathbf{V}(s)\, ds,$$

is Brownian with mean drift velocity $\bar{\mathbf{U}}(\mathbf{x}_0)$ and variance

$$\sigma^2(\mathbf{x}_o)\mathbf{I},$$

where **I** is the identity matrix,

$$\bar{\mathbf{U}}(\mathbf{x}_o) = f(\mathbf{x}_o)\mathbf{U}(\mathbf{x}_o), \tag{1}$$

$$(1/2)\sigma^2(\mathbf{x}_o) = g(\mathbf{x}_o)D_o, \tag{2}$$

D_o is the coefficient of binary diffusion in bulk liquid, f and g lie between 0 and 1, and **U** is the microscopic liquid velocity. The CLT shows that the displacement is Gaussian. The displacement

$$\int_{t_o}^{2t_o} \mathbf{V}(s)\, ds$$

is again Gaussian and hence the differential

$$d\mathbf{X}(t) = \mathbf{X}(t + dt) - \mathbf{X}(t) \tag{3}$$

satisfies an Itô equation

$$d\mathbf{X}(t) = \bar{\mathbf{U}}(\mathbf{X}(t))\, dt + \sigma(\mathbf{X}(t))\, d\mathbf{B}(t) \tag{4}$$

where $\mathbf{B}(t)$ is a standard 3-D Brownian motion. Assuming the molecular motions are independent (low concentration), the microscopic solute concentration satisfies

$$\frac{\partial C}{\partial t} = \sum_{i=1}^{3} \frac{\partial^2}{\partial x_i^2}(D_o'(\mathbf{x}_o)C) - \sum_{i=1}^{3} \frac{\partial}{\partial x_i}(\bar{U}(\mathbf{x})C) \tag{5}$$

where $D_0'(\mathbf{x}) = 1/2\,\sigma^2(\mathbf{x}) = g(\mathbf{x})D_o$. (6)

Bhattacharya next proceeds to assume the porous medium is homogeneous on the Darcy scale, convection is absent, and that

$$\lim_{t \to \infty} 1/t \int_o^t \sigma^2(\mathbf{X}(s))\, ds \equiv \bar{\sigma}^2 = constant \tag{7}$$

or equivalently

$$\lim_{t \to \infty} 1/t \int_o^t g(\mathbf{X}(s))\, ds = \bar{g} \tag{8}$$

with $0 \leq \bar{g} \leq 1$ and thus

$$(1/2)\bar{\sigma}^2 = \bar{g}D_o. \tag{9}$$

This line of reasoning provides the pore diffusion coefficient on the micro scale as $\bar{g}D_o$ which is less than the bulk liquid phase diffusion coefficient, D_o, of the same bulk fluid in equilibrium with the pore fluid. It also shows that pore diffusion manifests itself in going from the kinetic to the microscale.

The transition from the microscale to the Darcy scale is more complex, however, mathematically the development follows the same line of reasoning as transition from the kinetic scale to the microscale. A CLT produces an Itô equation on the Darcy scale and a corresponding dispersion-convection equation. It is shown that mechanical dispersion is manifest in going from the micro to the Darcy scale. The major assumptions that are needed to go from the micro to the Darcy scale include: (i) a Darcy-scale velocity REV exists and is constant throughout the flow domain, (ii) a CLT (not rigorously proven) holds from the micro to the Darcy scale, (iii) the solute convection velocity on the micro scale is a fraction of the liquid velocity, (iv) flow at the micro scale is laminar, and (v) the analysis presented for a homogeneous medium applies to a heterogeneous medium.

It is instructive to examine the advantages and disadvantages of using the Martingale methodology for developing porous media transport equations. The main advantages of the Martingale approach are: (i) the physical assumptions necessary to make the method mathematically rigorous can be explicitly displayed, (ii) it provides explicit forms for the dispersion coefficient in terms of molecular quantities, (iii) it highlights the important role scale plays in solute transport, and (iv) it is consistent with the classical kinetic theory of fluids. There are a number of disadvantages and drawbacks to the method. Among them are: (i) the method suffers from the major drawbacks the volume averaging approach suffers from (e.g., existence and invariance of REV's; see Baveye and Sposito 1984; Cushman 1984), (ii) the method has been shown to be mathematically valid only for large, nonreacting molecules in dilute solution, (iii) the CLT used to go from the micro scale to the Darcy scale has only been proven for very simple nonphysical media (e.g., periodic media), (iv) the method will not work for materials which have a high clay content (In regard to this point, it has been shown (Schoen et al. 1987a) that simple fluids in small pores do not behave as their bulk counterparts, e.g., diffusion coefficients parallel to the pore wall are higher in the midpore region than near the surface of the pore wall and even more importantly, higher than in the bulk fluid. This observation holds out to at least 30 molecular diameter pore wall separations for simple fluids. Thus the classical Itô type equations will break down in small pores; see also Mulla et al. (1984) for comments on this.), v) there is no effective way to develop macroscopic boundary conditions using this approach, and vi) the theory is nonoperational in the sense that the measurement process, stressed in Section 2, is not accounted for in autocorrelation functions.

Another interesting approach for developing transport equations using stochastic pde's is the statistical mechanical approach. Sposito and Chu (Sposito 1978a, b; Sposito and Chu 1981) following earlier classical works by Kirkwood (1967) developed the mass, momentum, and energy equations of a fluid in a porous medium at the pore scale. They then transform these equations to the "micro" scale by volume averaging techniques (Bear 1972). We will briefly outline the Kirkwood approach to developing statistical mechanical transport equations in the next few paragraphs.

The starting point in the statistical mechanical approach is the Liouville equation as presented in the special form (Kirkwood 1967)

$$\frac{\partial}{\partial t}E(A) = E\{\sum_{k=1}^{N}[\frac{\mathbf{P}_k}{m_k}\cdot\nabla_{\mathbf{R}_k}A - \nabla_{\mathbf{R}_k}U\cdot\nabla_{\mathbf{P}_k}A]\} \tag{10}$$

where A is a dynamic variable, $\mathbf{P}_k$ is the center of mass momentum of the kth molecule, m_k is the mass of the kth molecule and $\mathbf{R}_k$ is its center of mass position (it may also include orientational coordinates), and U is the interaction energy potential of the entire system (including external fields). The expected value, E, is taken with respect to the phase space probability density function

$$f(\mathbf{R}_1, \ldots, \mathbf{R}_N, \mathbf{P}_1, \ldots, \mathbf{P}_N, t)$$

having $6N$-degrees of freedom where N is the number of molecules in the system. Kirkwood was a strong believer in constructing operational theories. In an operational setting, E can also be considered as a phase space average coupled with an average over space and time (coarse graining). The space-time average corresponds to the window of the experimental apparatus (see last section). The probability density is determined with respect to a set of macroscopic constraints (e.g. fixed particle number, volume, and energy). To develop the equations of motion in a porous media all one needs to do is substitute the appropriate dynamic variable in Eq. (10) and perform several standard mathematical manipulations on delta functions (cf. Kirkwood 1967, p. 59-75) and identify the molecular expressions with their hydrodynamic counterparts. This approach gives molecular expressions for all the terms in the hydrodynamic equations at the pore scale. The simplest example of the technique is given by the balance of mass. The energy and momentum conservation laws follow similarly. To develop the continuity equation for a fluid in a porous medium, one sets

$$A = \sum_{j=1}^{N} m_j\delta(\mathbf{R}_j - \mathbf{r}) \tag{11}$$

where $\mathbf{r}$ is an arbitrary point in configuration space and $\mathbf{R}_j$ is the location of the jth molecule in the pore. Defining the mass density ρ and mass-average velocity $\mathbf{u}$ as

$$\mathbf{u}(\mathbf{r}, t) = \sum_{k=1}^{N} E[\mathbf{P}_k\delta(\mathbf{R}_k - \mathbf{r})]/\rho(\mathbf{r}, t) \tag{12}$$

$$\rho(\mathbf{r}, t) = E(A) \tag{13}$$

and upon using Eq. (10) we find

$$\frac{\partial\rho}{\partial t} = -\nabla\cdot(\rho\mathbf{u}) \tag{14}$$

which is identical to the continuity equation as developed from continuum theories. Sposito and Chu would now volume average this last equation to obtain what they call a "macro" equation. Sposito's three distinct natural scales are the molecular, hydrodynamic (Classical continuum) and REV.

The major strengths and weaknesses of the statistical mechanical approach are described in the following. Like the Martingale methods discussed earlier, one of the main advantages of the statistical mechanical approach is its ability to generate molecular descriptions of the constitutive parameters. It is more general than the Martingale method in that statistical mechanics are applicable to mass, momentum, and energy. As far as developing the "form" of the transport equations, I see no advantage over the continuum approach. Sposito (1978), in his conclusion section, makes the claim that certain terms which show up on the momentum balance equations are not consistent with the continuum approach; I disagree. Using

nonlocal continuum theories (Edelen 1976), one can develop transport equations which are identical to the statistical mechanical equations. This author sees little advantage to the statistical mechanical approach for developing the form of the transport equations; the real advantage of the statistical mechanical approach is its use in examining the interactions of fluids with soil particles on the pore scale. Much information, that can not be obtained experimentally, can be obtained (Mulla et al. 1984; He et al. 1986; Schoen et al. 1987a, b; Rhykerd 1987) using numerical statistical mechanics.

In passing, I should point out that I do not see the need for an ensemble of soils, as implied by Sposito, in the statistical mechanical derivation of transport equations. All one needs is an ensemble of fluids under an external force field (the soil) to make the derivation of Sposito valid. This eliminates a major conceptual difficulty in the statistical mechanical approach.

Another approach to develop transport equations using a stochastic methodology has been put forth by Cushman (1984, 1986) and may be entitled stochastic-relativist. While this method can apply when a natural hierarchy of scales is evident, such a hierarchy is not a prerequisite for the methodology. One could adopt a purely operational methodology based on the instrument windows irrespective of any natural scales. Using this method one can either assume that there exists a natural scale hierarchy on which we take measurements, or one can take measurement on successively larger spatial and temporal scales not knowing if there are any natural scales of motion. The concepts in the latter approach are similar to those in Baveye and Sposito (1984), but they also incorporate the concept of scale of observation as in Cushman (1984). I would like to point out that Eq. (26) of Cushman (1984) is in error.

Continuum scale transport equations are assumed valid in both the void and solid space in a porous medium (the same approach may be followed on a molecular scale). Randomness enters the problem through the random geometry of the pore surfaces. Scale is introduced into the setting through a mathematical representation of the window, T, of the instrumentation. the basic continuum scale transport equations are stochastic as a result of the random boundaries. The act of convolving (denoted $*$) an instrument window with a field variable, ψ, is equivalent to taking a measurement on ψ. The relation $T*\psi$ is well defined and stochastic as a result of the stochasticity of ψ and the determinism of T. However, the quantity $T*\psi$ is on a higher scale of motion than ψ owing to the scale constraints on T (Cushman 1986). The generalized function, T, which is compact, can be viewed in many ways. It can be viewed as representing an instrument window (as already mentioned), but just as importantly, it may be viewed as a Hausdorf measure and used to compute the fractal dimension of ψ.

The development of the stochastic transport equations themselves is similar in form, but mathematically more general than the classical REV methodology and will not be summarized here. The interpretation of parameters is quite different than in the REV approach. As will be discussed in a later section, all transport coefficients can be expressed in terms of correlation functions (McQuarrie 1976). Cushman (1986) has provided the machinery necessary to take correlation functions on any scale of motion and transfer them to higher scales in an operational setting. Thus transport coefficients on higher scales of motion can be written in terms of lower scales.

The advantages to Cushman's approach are: (i) since they are defined in terms of an instrument window, all parameters have an operation meaning, (ii) the theory is developed in frequency and wave number space where scale is most easily accounted for, (iii) at every scale of motion the transport equations are random, (iv) the correlation of properties from lower scales to higher scales is easily

handled, (v) the theory is mathematically rigorous, and (vi) the method is very general in that it applies to mass, momentum, and energy. The major disadvantages to the approach are: (i) no one has taken the time to use the approach to develop operational constitutive coefficients, (ii) the mathematical form of the instrument windows are not easily determined, and (iii) a theory incorporating nonlinear windows has not been developed (Cushman 1984, has alluded to how such a theory may be developed).

Dagan (1986) has yet another interesting approach to developing stochastic transport equations when natural scales exist (see also Dagan 1981; 1982a, b, c; 1984; 1985a, b). Mathematically, Dagan's and Cushman's (1984, 1986) approaches are similar with the major exception that higher scale transport equations are developed by successive probabilistic averages in Dagan's approach. Probabilistic averaging theorems that closely resemble their volume averaging counterparts are proven, and, hence the formal structure of the averaged equations is the same in both Dagan's method and the classical volume averaging method. A typical probabilistic averaging theorem would take the form

$$E[(\nabla\psi)\chi_\alpha] = \nabla E(\psi\chi_\alpha) - \lim_{\Delta V \downarrow 0} \left[\frac{1}{\Delta V}\int_{\Delta A} E(\psi \mathbf{n})\, dA\right] \tag{15}$$

where ψ is a field variable, χ_α is the indicator function of the α-phase, ΔV is a sampling volume, and ΔA the associated surface. The assumption of stationarity is heavily relied upon to equate

$$E\langle\psi\rangle = E\psi \tag{16}$$

where $\langle\cdot\rangle$ denotes volume average. The natural scales that Dagan assumes exit include pore scale, lab scale, local scale, and regional scale. Stochasticity on the pore scale is attributed to the randomness of the indicator function χ_α of the α-phase (this is the same as in Cushman's methodology). The pore scale is defined as being small compared to the integral scale of χ_α, but large compared to the molecular scale. The fluid and solid phases at this scale are assumed to be continua and governed by classical continuum mechanics. Dagan's lab scale is on the order of $10^{-1}-10^{0}$ m and is characterized by the dimensions of common lab experiments. The local scale is on the order of the aquifer thickness ($10^{1}-10^{2}$ m). The regional scale, of the order $10^{4}-10^{5}$ m in the horizontal plane, is much larger than the acquifer thickness. On this scale, flow variables are averaged over the vertical. Randomness on the local and regional scales is characterized by the randomness in hydraulic conductivity, K, and transmissivity, T, respectively.

I find a difficult conceptual issue with the approach taken by Dagan (1986). In going from the pore scale to the lab or local scale, the stochastic equation for a fluid in the pore phase (on the pore scale recall that χ_α is random) is smoothed by taking expected values. This results in a deterministic equation on the local scale. Randomness is introduced into the local scale by assuming K is random. Now as indicated in Section 2, K can be random for either of two reasons. First, K can be assumed random owing to pore heterogeneity over the local scale. But K cannot be random for this reason in Dagan's model because K results from taking expected values of χ_α and other variables. Thus K in Dagan's model must be assumed random for the second reason, parameter uncertainty (we cannot sample K everywhere in the field). This is the only source of randomness in Dagan's model. Yet, from a physical point of view, the randomness in K and T is due not only to parameter uncertainty as discussed in Section 2, but also to χ_α on each scale of motion.

The major advantages of Dagan's approach are: (i) the method is conceptually simple, (ii) the method is consistent with existing experimental and sampling

methodology, (iii) equations developed from Dagan's approach look like those which are developed from deterministic averaging techniques, and (iv) the methodology allows one to update estimations via conditioning the equations. The major limitations of the approach are: (i) the conceptual difficulties in defining the randomness of K and T, (ii) the heavy reliance on stationarity, and (iii) conceptual difficulties with ergodicity (see Cushman 1987).

4 Solute transport without natural scales

In this section we will briefly review a technique for studying solute movement that does not explicitly require the existence of natural scales.

The methodology, called the stochastic-convective approach by Simmons (Simmons 1982, 1986, 1987; Delvary and Simmons 1987; Simmons and Kincaid 1987; Weirenga et al. 1986) is quite powerful. In passing, I should point out that the impulse-response model, as exploited by Jury (Jury 1982; Jury et al. 1982; Jury et al. 1986) is a special case of Simmons' stochastic-convective approach. Simmons' methodology is a general stochastic approach for deriving solute transport equations -- it requires no specific assumptions about natural scales of motion (e.g., existence of a REV or CLT). Much as is done in classical continuum theories, the model may be formulated in either an Eulerian or Lagrangian framework. Because of the limited distribution of Simmons' work, we will describe it in greater detail than models in earlier sections. We do not, however, go into the great detail that Simmons does.

The basic starting premise with Simmons' approach is that solute transport is described by a first order stochastic hyperbolic equation, represented in one dimension as

$$\frac{\partial C}{\partial t} + V\frac{\partial C}{\partial x} = 0 \tag{17}$$

where the velocity V is a random variable. Thus, as in several earlier models, stochasticity in C is a result of stochasticity in V. The parameter V describes the random paths, $X(t)$, which may be traversed by an individual solute molecule as it passed through the porous medium. The ensemble of random paths implicitly includes all "local-scale" dispersion mechanisms that contribute to "global-scale" dispersion. Explicit definitions of these two scales are unnecessary in Simmons' model. A notable difference between this approach and other methods mentioned earlier and those to be mentioned later, is that observables (measurable quantities) are defined in the stochastic-convective approach much as they are in quantum mechanics (Pilar 1968), i.e., observables are expected values. In all other approaches observables are realizations.

Simmons is interested in finding the observable concentration in terms of a specific velocity ensemble. This implies the need to know the relation between an observed moisture content and the velocity ensemble. Let $X(t; x_o, t_o)$ be a solution of

$$\frac{dx}{dt} = V(x, t) \tag{18}$$

$$X(t_o; x_o, t_o) = x_o. \tag{19}$$

Let $P_X(x; t, x_o, t_o)$ be a pdf for random location X which is conditioned on t, x_o, and t_o. Then P_X is given by

$$P_X(x; t, x_o, t_o) = E\{\delta[x - X(t; x_o, t_o)]\}. \tag{20}$$

Now a random concentration satisfies $C\theta = \rho$ and if C and θ are independent

(uncorrelated), then

$$EC = E\rho/E\theta. \tag{21}$$

Also, if

$$E[C(x, t_o)] = C_o(x),$$

then

$$E[C(x, t_o)] = \int_o^\infty C_o(x_o)P_{X_o}(x_o; t, x_o, t_o)\, dx_o \tag{22}$$

where

$$P_{X_o}(x_o; t_o, x, t) = E[\delta(x_o - X_o(t_o; x, t))]. \tag{23}$$

If $\theta_o(x)$ is a deterministic initial fluid content such that $\rho_o = \theta_o C_o$ holds, then for Eqs. (21) and (22) to both hold for any C_o, we must have

$$E(\theta)P_{X_o} = \theta_o(x_o)P_X. \tag{24}$$

This last equation directly ties the observed fluid content, $E(\theta)$, to a particular velocity ensemble, P_X, as required.

To show how classical solutions to the convective-dispersive transport equation are special examples of the stochastic-convective approach, one would argue as follows. Assume θ is deterministic such that $\theta = \theta_o(x)$. Further assume the dispersivity a is related to moisture content in a manner such that

$$\bar{a} = a\theta \tag{25}$$

is a constant for each flow state (for different flow states it will change). The dispersivity a is assumed stochastic. We further assume the molecular diffusion coefficient is zero (this is not a necessary requirement) and the dispersion coefficient has the form

$$D = aV. \tag{26}$$

Let

$$z(x) = \int_o^x \theta_o\, dx, \tag{27}$$

then the convective-dispersion equation may be transformed into

$$\frac{\partial C}{\partial t} + q\frac{\partial C}{\partial z} = B\frac{\partial^2 C}{\partial z^2} \tag{28}$$

where $B = \bar{a}q$ and $q = \theta V$. A stochastic Green's function solution to Eq. (28) is given by

$$C(z, t) = \int_{-\infty}^\infty C_o(z^{-1}(z_o))G(z - z_o; t)\, dz_o \tag{29}$$

where

$$G(z, t) = (4\pi Bt)^{-1/2}\exp\{-[z - qt)^2/4Bt]\}. \tag{30}$$

Thus

$$P_{X_o} = \theta_o(x_o)G(z(x; x_o); t) \tag{31}$$

where $z(x; x_o) = z(x) - z(x_o)$ for $t = 0$. Note that P_{X_o} is not stationary in space unless θ_o = constant.

As mentioned earlier, the impulse-response model as used by Jury (1982) and Jury et al. (1986) can also be viewed as a special case of the stochastic-convective approach. To see this, Simmons would argue as follows: the impulse-response

model gives the concentration observed at depth L,

$$C_L(I) = \int_o^\infty C_{in}(I - I')f_L(I')\,dI', \tag{32}$$

as a function of cumulative amount of applied water I (I = rainfall-evapotranspiration) remaining in the soil profile over a specific time t. Here $f_L(I)$ is a pdf representing the distribution of I reaching to a depth L. The input concentration, depending on the infiltration amount, is $C_{in}(I)$. If an average water infiltration rate i_o is applied over a field area, then $I = i_o T$ determines a random travel time T^o. Assuming a scaling hypothesis for the probability of reaching another depth Z expressed as

$$f_z(I) = f_L(IL/Z)L/Z, \tag{33}$$

equation (32) becomes

$$C(Z, t) = \int_o^\infty C_{in}(t - t')f_Z(i_o t')i_o\,dt' \tag{34}$$

where $C_{in}(t)$ is now a time dependent description of solute input concentration. Simmons' stochastic-convective expression for Eq. (34) would be

$$C(Z, t) = \int_o^\infty C_{in}(t - Z/V)P_Z{}^*(V)dV \tag{35}$$

where $P_Z{}^*$ is the pdf for effective uniform random velocities, $V = Z/T$, defined by travel time, T, to depth Z. Simmons' derivation of Eq. (35) does not rely on the scaling hypothesis, Eq. (33), and moreover it is easy to show that Jury's scaling hypothesis is the same as presuming that an invariant probability distribution for random velocity applies to each depth Z. It is this property of invariance that produces scale dependent dispersion (Simmons 1982).

There are a number of positive and negative aspects to Simmon's approach. Some of the advantages to the stochastic-convective approach include: (i) the method can be used to show that Eulerian and Lagrangian approaches to stochastic transport are not necessarily equivalent, (ii) the method does not require existence of natural scales of motion, (iii) the classical convective-diffusive equation is a special example of the stochastic-convective approach as is the impulse-response model, (iv) constraints involving stationarity are unnecessary, and (v) it clarifies the definitions of flux-averaged and volume-averaged concentrations. Among the disadvantages of the approach are: (i) unlike quantum mechanics where the probability density that defines the ensemble expectation is known (it results from a solution to the Schröedinger equation), there is no known method to determine the probability density for the velocity ensemble in the stochastic-convective approach, (ii) the method is only applicable to solute transport, (iii) the standard parameters measured in most transport experiments include the hydraulic conductivity, yet Simmon's method, to date, has not incorporated this parameter into the model.

5 Preasymptotic development of transport coefficients

In this section, we outline a stochastic methodology for developing time and space dependent (or equivalently frequency and wave vector dependent) transport coefficients in porous media, i.e., preasymptotic coefficients. The approach is an extension of molecular hydrodynamics (McQuarrie 1976) and will be called generalized hydrodynamics. Molecular hydrodynamics has classically been the study of the pre-continuum nature of fluids, i.e., the study of fluids which retain some detailed molecular information, but which are approaching the hydrodynamic limit (lower bound on the classical continuum scale). Many natural transport processes in porous media do not have a natural hierarchy of scales of motion and in those that do not have such a hierarchy, the asymptotic limits may not be reached owing to

time and experimental scale constraints. It thus appears that a generalization of molecular hydrodynamics should be applicable to porous media.

For instructional purposes, we will put mathematical rigor aside for the moment. Consider the following intuitive example (see also Bhattacharya and Gupta 1979, 1983). Suppose we have a system composed of N particles of mass M, suspended in a fluid composed of particles of mass m ($m \ll M$). The concentration of heavy particles is assumed dilute so that these particles do not interact with each other, but they do interact with the lighter particles which behave as a continuum. The heavy particles with velocity $\mathbf{u}$ satisfy a Langevin equation of the form

$$m\frac{d\mathbf{u}}{dt} = -\zeta\mathbf{u}(t) + \mathbf{B}(t) \tag{36}$$

where $\mathbf{B}(t)$ is a standard Brownian motion. It is well known that in the long time (asymptotic) limit (the Markovian approximation) the friction coefficient, ζ, satisfies

$$\zeta = (3k_B T)^{-1}\int_o^\infty E\,[\mathbf{B}(o)\cdot\mathbf{B}(t)]\,dt \tag{37}$$

$$\zeta^{-1} = (3k_B T)^{-1}\int_o^\infty E\,[\mathbf{u}(o)\cdot\mathbf{u}(t)]\,dt \tag{38}$$

or

$$d = k_B T/\zeta \tag{39}$$

where d is the diffusion coefficient, T is temperature and k_B is the Boltzmann constant. Many other fluctuating quantities, $\mathbf{A}(t)$, can be described by a stochastic differential equation of the Langevin type

$$\frac{d\mathbf{A}}{dt} = -\gamma\mathbf{A}(t) + \mathbf{B}(t) \tag{40}$$

If $\mathbf{A} = Md\mathbf{u}$ and $\gamma = \zeta/M$, then Eq. (40) reduces to Eq. (36). To describe the dynamics of a homogeneous system without the restriction on the time scale, Eq. (40) is rewritten as a non-Markovian generalized Langevin equation

$$\frac{d\mathbf{A}(t)}{dt} = -iF_o\mathbf{A}(t) - \int_o^t \gamma(t')\mathbf{A}(t-t')\,dt' + \mathbf{B}(t) \tag{41}$$

where F_o is real and corresponds to existence of a propagation process associated with the time evolution of the dynamic variable. The convolution in Eq. (41) accounts for memory effects.

In passing, we should note that if Eq. (40) is multiplied by $\mathbf{A}(o)/E\,[\mathbf{A}(o)]^2$ and expectations are taken (assuming $E\,[\mathbf{A}(o)\cdot\mathbf{B}(t)] = 0$) we find

$$\frac{d\psi}{dt} = -\gamma\psi \tag{42}$$

where $\psi(t) = E\,[\mathbf{A}(o)\cdot\mathbf{A}(t)]/E\,[\mathbf{A}(o)^2]$. This last equation provides the time evolution of the autocorrelation function, e.g., it may provide the time evolution of the velocity autocorrelation, which in turn gives us information on d in the preasymptotic region (as noted from Eqs. (38) and (39)). The corresponding Langevin equation for the autocorrelation in Eq. (41) takes the form

$$\frac{d\psi}{dt} = -\int_o^t \gamma(t')\psi(t-t')\,dt' + iF_o\psi(t). \tag{43}$$

Equation (43), without the second term on the right hand side, is called the memory function equation for the autocorrelation and γ is called the memory function or kernel. It can be shown (McQuarrie 1976) that all autocorrelation functions satisfy an equation of this form. The entirety of the physics of any transport

process is implicitly contained in the memory function $\gamma(t)$ and consequently a knowledge of $\gamma(t)$ allows us to study the preasymptotic nature of transport processes. Equation (43) is often generalized to allow for a heterogeneous medium by replacing $\gamma(t)$ with $\gamma(\mathbf{x}, t)$.

Let $G(\mathbf{r}, t)\, d\mathbf{r}$ be the conditional probability of finding a molecule in $d\mathbf{r}$ at $\mathbf{r}$ at time t given it was initially located at the origin and let $F_s(\mathbf{k}, t)$ be its space Fourier transform. It can be shown (McQuarrie 1976) that F_s is a correlation function and hence satisfies Eq. (43). It can be shown further that the memory function γ takes the form

$$\gamma(\mathbf{k}, t) = \mathbf{k} \cdot E\{\mathbf{u}\exp(-\mathbf{k}\cdot\mathbf{r})\exp[it(I-P)L]\mathbf{u}\exp(i\mathbf{k}\cdot\mathbf{r})\}\cdot\mathbf{k} \tag{44}$$

where P is a projection operator (Schoen et al. 1987) and L is the Liouvillian. The expected value of the function in braces in Eq. (44) is identified with a time-wave vector dependent preasymptotic diffusion tensor $\mathbf{d}(\mathbf{k}, t)$,

$$\gamma(\mathbf{k}, t) = \mathbf{k} \cdot \mathbf{d}(\mathbf{k}, t) \cdot \mathbf{k} \tag{45}$$

If we now take the time-Fourier transform of Eq. (45), then

$$\gamma(\mathbf{k}, \omega) = \mathbf{k} \cdot \mathbf{d}(\mathbf{k}, \omega) \cdot \mathbf{k} \tag{46}$$

is a generalized preasymptotic wave vector and frequency-dependent diffusion coefficient satisfying (Zwanzig 1964)

$$\mathbf{j}(\mathbf{k}, \omega) = -\mathbf{d}(\mathbf{k}, \omega) \cdot (\nabla C)_{\mathbf{k},\omega} \tag{47}$$

which is a generalized Fick's law (with scale-dependent diffusion) valid at all wave numbers and frequencies. In the above, the subscripts $\mathbf{k}$ and ω on the gradient indicate Fourier component. It can be shown rigorously (Schoen et al. 1987) that

$$\lim_{\substack{|\mathbf{k}| \to 0 \\ \omega \to 0}} \mathbf{d}(\mathbf{k}, \omega) = \mathbf{d} \tag{48}$$

where $\mathbf{d}$ is the classical diffusion tensor.

Cushman (1986) has provided the necessary machinery to extend these concepts to porous media. Cushman, working in wave vector and frequency space, shows how correlation functions on higher scales of motion can be written in terms of lower scales of motion. On any scale of motion the Liouville equation is valid and thus a memory function equation will exist for the autocorrelation functions in a porous media. The memory function equation can be written in terms of autocorrelations as a function of scale of observation. It is this last point that gives a rigorous operational derivation of, for example, a preasymptotic scale-dependent dispersion coefficient in porous media.

6 Asymptotic development of dispersion tensors -- assuming the validity of the dispersion equation

Several researchers have assumed the correctness of the stochastic conservation equations (at least on a "local" scale) and have gone on to use stochastic approaches to develop the form of the asymptotic constitutive coefficients. In this section, we will concentrate on two stochastic asymptotic approaches to developing the dispersion coefficient. I will try to highlight all assumptions and their justification. From the classical theories of diffusion (cf. Einstein 1956) one should not at all be surprised that the dispersion tensor in porous media theories is directly related to the velocity autocorrelation function, or equivalently, its Fourier transform. Because the velocity autocorrelation is directly related to the correlation of hydraulic conductivity through Richard's equation and because the hydraulic conductivity is a parameter which is usually measured in the field, we

find that many authors tend to write the dispersion tensor in terms of the conductivity correlations. In the previous section we illustrated how one may rigorously derive preasymptotic information on transport parameters. For this reason, I will focus attention in this section on the asymptotic limit.

Classically (Bear 1972), the dispersion tensor has been written in the form

$$\mathbf{D} = d_L \delta_{11} + d_T(\delta_{22} + \delta_{33}) \tag{49}$$

where the tensor is expressed in the principal coordinate system with directions parallel and normal to the Darcy velocity, $\mathbf{v}$. It is assumed that the dispersion coefficient is isotopic in directions normal to $\mathbf{v}$. The general form of the transverse, d_T, and longitudinal, d_L, components is given by

$$d_L = d_m + a_L \mid \mathbf{v} \mid \tag{50}$$

$$d_T = d_m + a_T \mid \mathbf{v} \mid$$

where d_m is the effective coefficient of porous diffusion. It is generally assumed that d_m is smaller than the molecular scale diffusion coefficient and that d_m is a scaler. Recent molecular dynamics simulations (Schoen et al. 1987a) indicate this need not necessarily be true in layered colloidal systems (e.g. clays).

When soil samples are fairly uniform, researchers have found that the above expression for the dispersion tensor holds in many laboratory experiments (Bear 1972; Fried 1975). Because the classical developments do not account for evolving heterogeneities, one would not expect the model to hold for more complicated systems. In fact, what one finds is that in nonuniform media a_L and a_T turn out to be scale-dependent (cf. Martin 1971; Lawson and Elrick 1972; Dieulin et al. 1981).

One of the most straightforward and informative methods for developing asymptotic dispersion coefficients was proposed by Winter (1982) (see also Winter et al. 1984a, b; Neuman et al. 1987). The basic approach of Winter is to assume the Darcy velocity is random on a local scale (on the order of a meter) and that a local convective-dispersion equation is obeyed. The local velocity, $\mathbf{v}$, is assumed weakly stationary and of the form

$$\mathbf{v}(\mathbf{x}) = \boldsymbol{\mu} + \varepsilon \mathbf{u}(\mathbf{x}) \tag{51}$$

where $\boldsymbol{\mu}$ is the mean local velocity and ε is a small perturbation parameter. One would like to show that the asymptotic average concentration also satisfies a convection-dispersion equation. This has only been accomplished in a few very select cases (incompressible velocity fields in 3-D). Winter and co-workers assume the asymptotic average concentration satisfies a convective-diffusion equation and go on to find explicit expressions for the asymptotic velocity, $\mathbf{V}$, and asymptotic dispersion coefficient, $\mathbf{D}$, via a perturbation expansion of the form

$$\mathbf{V} = \mathbf{V}_o + \varepsilon \mathbf{V}_1 + \varepsilon^2 \mathbf{V}_2 + \cdots \tag{52}$$

$$\mathbf{D} = \mathbf{D}_o + \varepsilon \mathbf{D}_1 + \varepsilon^2 \mathbf{V}_2 + \cdots \tag{53}$$

where ε need not tend to zero as $\mid \mathbf{x} \mid \to \infty$ or $t \to \infty$. Specifically, if C is the local scale concentration, then it is assumed

$$\tilde{C}(\mathbf{x}, t) = \lim_{\lambda \to \infty} E[C_\lambda(\mathbf{x}, t)] = \lim_{\lambda \to \infty} \lambda^{n/2} E[C(\sqrt{\lambda}\mathbf{x} + \lambda t \boldsymbol{\mu}, \lambda t)] \tag{54}$$

satisfies

$$\frac{\partial \tilde{C}}{\partial \tau} = [1/2 \ \nabla \cdot \mathbf{D} \cdot \nabla] \tilde{C}, \ \tilde{C}(\mathbf{x}, o) = \delta(\mathbf{x}) \tag{55}$$

where $t = \tau/\lambda$ and n is the dimension of the system.

Because C satisfies

$$\frac{\partial C}{\partial \tau} = [1/2\ \nabla\cdot\mathbf{d}\cdot\nabla - \mathbf{v}\cdot\nabla]C,\ C(\mathbf{x}, o) = C_o(\mathbf{x}) \tag{56}$$

we have the semigroup equation (cf. Goldstein 1985)

$$\lim_{\lambda\to\infty} Ee^{t(A+\varepsilon B)}e^{-\sqrt{\lambda}t(\mathbf{V}\cdot\nabla)} = e^{t(\nabla\cdot\mathbf{D}\cdot\nabla)/2} \tag{57}$$

where the infinitesimal generator is given by

$$A = (\nabla\cdot\mathbf{d}\cdot\nabla)/2 + \sqrt{\lambda}\boldsymbol{\mu}\cdot\nabla \tag{58}$$

and

$$B = \lambda\boldsymbol{\mu}(\sqrt{\lambda}\mathbf{x})\cdot\nabla \tag{59}$$

The expectation E is taken with respect to the distribution of $\mathbf{u}$ which is assumed weakly stationary. Using a Kubo type formal expansion (Kubo 1963) for $e^{\varepsilon Bt}$, using Eqs. (52) and (53), and equating powers of ε, we may explicitly determine the $\mathbf{V}_i$ and D_i, $i = 0,1,\ldots$. The results are (Winter 1982) $\mathbf{V}_o = \boldsymbol{\mu}$, $\mathbf{D}_o = \mathbf{d}$, $\mathbf{V}_1 = 0$, $\mathbf{D}_1 = \mathbf{0}$ and

$$(\mathbf{V}_2)_\ell = (2\pi)^{-n}\sum_{m=1}^{n}\int_{R^n} i\frac{k_m\hat{\rho}_{m\ell}(\mathbf{k})}{F(\mathbf{k})}\,d\mathbf{k} \tag{60}$$

where

$$F(\mathbf{k}) = \mathbf{k}\cdot\mathbf{d}\cdot\mathbf{k} - i\boldsymbol{\mu}\cdot\mathbf{k}, \tag{61}$$

$$\hat{\rho}_{m\ell}(\mathbf{k}) = \int_{R^n} E[\mathbf{v}_\ell(\mathbf{x})\mathbf{v}_m(\mathbf{x}) - \mu^2]e^{i\mathbf{k}\cdot\mathbf{x}}\,d\mathbf{x}, \tag{62}$$

$$\mathbf{D}_2 = (2\pi)^{-n}\int_{R^n}\frac{1}{F(\mathbf{k})}\hat{\rho}_{m\ell}(\mathbf{k})\,d\mathbf{k}$$

$$- 2(2\pi)^{-2}\sum_{p=1}^{n}\sum_{r=1}^{n}\int_{R^n}\frac{k_p\hat{\rho}_{p(m}(\mathbf{k})\,d_{\ell)r}\,k_r}{[F(\mathbf{k})]^2}d\mathbf{k},\quad n \geq 2 \tag{63}$$

where $(\cdot)$ on subscripts denotes symmetric part. If one assumes $\nabla\cdot\mathbf{v} = 0$, then it follows that

$$\mathbf{D} = \mathbf{d} + (2\pi)^{-n}\int_{R^n}\frac{\mathbf{k}\cdot\mathbf{d}\cdot\mathbf{k}}{(\mathbf{k}\cdot\mathbf{d}\cdot\mathbf{k})^2 + (\boldsymbol{\mu}\cdot\mathbf{k})^2}\hat{\rho}_{m\ell}(\mathbf{k})\,d\mathbf{k},\quad n \geq 2. \tag{64}$$

Neuman, et al. (1987) present a number of special cases and approximations to the last result. The major strengths and weaknesses of Winter's approach are summarized below.

The major advantages to Winter's approach are the ease with which explicit forms of the dispersion tensor may be developed from Eqs. (63) and (64) and the ability to analyze dispersion in terms of Peclet numbers. The major limitations are the large number of assumptions required to develop various forms of the tensor, many of which are very difficult (if not impossible) to justify experimentally. Assumptions used to derive the general form of the dispersion tensor include: (i) at least two distinct scales exist, the local and asymptotic, (ii) stochasticity of the concentration profile results strictly from stochasticity of the local velocity field, (iii) the local velocity and log hydraulic conductivity are weakly stationary, (iv) the asymptotic expected value of the concentration satisfies a dispersion-convection equation, (v) the asymptotic expected value of the concentration equals the field realization (see Sposito et al. 1986 for a discussion of this), (vi) the Kubo formalism of the semigroup problem can be justified rigorously, (vii) the second order

approximation adequately describes dispersion, (viii) various mathematical constraints apply to $\hat{\rho}$ (e.g., bounded at the origin), and (ix) boundary conditions are unimportant (see Schoen et al. 1987 for problems that may be encountered with this assumption). As currently applied, the method is nonoperational in the sense that the measurement process is not accounted for. Constraints imposed to get practical forms of the dispersion tensor include: (i) $\nabla \cdot \mathbf{v} = 0$, (ii) isotropy on a local scale, (iii) various special forms of the log-conductivity, and (iv) the local dispersion tensor is constant (see Gupta and Bhattacharya 1986 for a discussion of this).

An alternative method for developing asymptotic dispersion tensors from a stochastic point of view is due to Gelhar and co-workers (Gelhar et al. 1979; Gelhar and Axness 1983). Gelhar's method is analogous to those used in classical turbulence theories (Monin and Yaglom 1973). While the basic ideas behind Gelhar's technique are correct, there are several inconsistencies in his perturbation approach (Cushman 1983; Neuman et al. 1987). To illustrate the method and point out these inconsistencies, we will go into some detail in the following.

Assume steady state conditions with the log of the local hydraulic conductivity, $Y = \ln K$, weakly stationary. The local mass balance takes the form

$$\nabla \cdot \mathbf{v} = 0 \tag{65a}$$

or

$$\nabla \cdot (\mathbf{k} \nabla h) = 0 \tag{65b}$$

where h is the local hydraulic head. Equation (65b) may be equivalently written as

$$\nabla Y \cdot \nabla h + \nabla^2 h = 0 \tag{66}$$

Setting $Y = E(Y) + \tilde{Y}$ and $h = E(h) + \tilde{h}$ we find

$$\nabla \tilde{Y} \cdot \nabla E(h) + \nabla \tilde{Y} \cdot \nabla \tilde{h} + \nabla^2 E(h) + \nabla^2 \tilde{h} = 0 \tag{67}$$

or equivalently (Cushman 1983)

$$\nabla \tilde{Y} \cdot \nabla E(h) + (\nabla \tilde{Y} \cdot \nabla \tilde{h})\tilde{} + \nabla^2 \tilde{h} = 0 \tag{68}$$

where

$$(\nabla \tilde{Y} \cdot \nabla \tilde{h})\tilde{} = \nabla \tilde{Y} \cdot \nabla \tilde{h} - E[\nabla \tilde{Y} \cdot \nabla \tilde{h}] \tag{69}$$

If we assume the second term in Eq. (68) is small, then we arrive at

$$\nabla \tilde{Y} \cdot \nabla E(h) + \nabla^2 \tilde{h} = 0. \tag{70}$$

This approach is equivalent to a first order perturbative analysis which has been criticized by Cushman (1983). Cushman, with a trivial example, shows that even if Y deviates from its mean only slightly, $\nabla \tilde{Y}$ may be extremely large. Hence, it is conceivable that $\nabla \tilde{Y} \cdot \nabla \tilde{h}$ is non-negligible. In fact, the assumption that $\nabla \tilde{Y}$ is small is equivalent to the assumption that $\tilde{Y}$ has only short wavelength components -- hence, it is an asymptotic approach much as is Winter's.

Upon inserting the spectral integral representations (Gihman and Skorohod 1980) for $\tilde{Y}$ and $\tilde{h}$, we find (in terms of the spectral densities $\phi_{\tilde{Y}}(\mathbf{k})$ and $\phi_{\tilde{h}}(\mathbf{k})$ of $\tilde{Y}$ and $\tilde{h}$, respectively)

$$i\mathbf{k} \cdot \nabla E(h) \phi_{\tilde{Y}}(\mathbf{k}) = \mathbf{k} \cdot \mathbf{k} \phi_{\tilde{h}}(\mathbf{k}) \tag{71}$$

(Note: For those unfamiliar with Lebesgue-Stieltjes measures, we have used spectral densities in Eq. (71) rather than these measures. In fact, in all practical applications to date, the measures have been assumed absolutely continuous. Hence, we have lost no generality with this simplification.)

Now if we set $K_m = \exp(EY)$, then

$$K = K_m \exp\tilde{Y} = K_m(1 + \tilde{Y} + \frac{\tilde{Y}^2}{2} + \cdots) \tag{72}$$

Upon setting the Darcy velocity

$$\mathbf{v} = E(\mathbf{v}) + \tilde{\mathbf{v}} \tag{73}$$

Darcy's law takes the form

$$\mathbf{v} = -K\nabla h = -K_m(1 + \tilde{Y} + \frac{\tilde{Y}^2}{2} + \ldots)\nabla(E(h) + \tilde{h}) \tag{74}$$

which upon taking expected values up to second order gives:

$$E(\mathbf{v}) = -K_m[\nabla E(h) + 1/2E(\tilde{Y}^2)\nabla E(h) + E(\tilde{Y}\nabla\tilde{h}) + 1/2E(\tilde{Y}^2\nabla\tilde{h})]. \tag{75}$$

To first order we have

$$E(\mathbf{v}) = -K_m \nabla E(h). \tag{76}$$

Also, to first and second order we have, respectively,

$$\tilde{\mathbf{v}} = -K_m[\nabla\tilde{h} + \tilde{Y}\nabla E(h)] \tag{77}$$

$$\tilde{\mathbf{v}} = -K_m[\nabla\tilde{h} + \tilde{Y}\nabla E(h) + 1/2(\tilde{Y}^2)\nabla E(h) + (\tilde{Y}\nabla\tilde{h})\tilde{} + 1/2(\tilde{Y}^2\nabla\tilde{h})]. \tag{78}$$

It was pointed out by Newman et al. (1987) that Gelhar and Axness (1983) keep the first two terms in Eq. (75) and that is neither a first or second order approximation. Newman et al. point out that to be consistent, either Eqs. (75) and (78) or Eqs. (76) and (77) should be used in any perturbative analysis. I will go even further and suggest that since Gelhar uses Eq. (70), he should use Eqs. (76) and (77). If, on the other hand, Eqs. (75) and (78) are used, then to be consistent, Eq. (68) should replace Eq. (70). As suggested by Newman et al. (1987), if we proceed with the first order analysis, then we get

$$\boldsymbol{\phi}_{\tilde{\mathbf{v}}}(\mathbf{k}) = -K_m \nabla E(h)\phi_{\tilde{Y}}(\mathbf{k}) - K_m i\mathbf{k}\phi_{\tilde{h}}(\mathbf{k})\phi. \tag{79}$$

Substituting $\phi_{\tilde{h}}(\mathbf{k})$ in Eq. (71) we find

$$\boldsymbol{\phi}_{\tilde{\mathbf{v}}}(\mathbf{k}) = [\mathbf{I} - \frac{\mathbf{kk}}{|\mathbf{k}|^2}]\cdot E(\mathbf{v})\phi_{\tilde{Y}}(\mathbf{k}) \tag{80}$$

where $\mathbf{I}$ is the identity matrix. If we now take the scalar product of Eq. (80) with itself and take expected values, we find

$$\hat{\boldsymbol{\rho}}_{\tilde{\mathbf{v}}} = [\mathbf{I} - \frac{\mathbf{kk}}{|\mathbf{k}|^2}][\mathbf{I} - \frac{\mathbf{kk}}{|\mathbf{k}|^2}]\hat{\boldsymbol{\rho}}_{\tilde{Y}} \,|\, E(\mathbf{v}) \,|^2 \tag{81}$$

which is the relation between velocity perturbations and hydraulic conductivity perturbations expressed in terms of their respective autocorrelations.

Gelhar and Axness (1983) next proceed to expand Eq. (56) for the steady state problem in terms of perturbations and spectral densities. Let $C = E(C) + \tilde{C}$, then Eq. (56) with $\partial C/\partial t = 0$ and $\mathbf{d}$ assumed constant gives

$$\nabla\cdot[E(\mathbf{v})\tilde{C} + \tilde{\mathbf{v}}E(C) + \tilde{\mathbf{v}}\tilde{C} - E(\tilde{\mathbf{v}}\tilde{C})] = \mathbf{d}\cdot\nabla^2\tilde{C}. \tag{82a}$$

The equation for means is

$$\nabla\cdot[E(\mathbf{v})E(C)] + \nabla\cdot E(\tilde{\mathbf{v}}+\tilde{C}) = \mathbf{D}\cdot\nabla^2 E(C). \tag{82b}$$

The first order approximation to Eq. (82a) is

$$\nabla \cdot [E(\mathbf{v})\tilde{C} + \tilde{\mathbf{v}}E(C)] = \mathbf{d} \cdot \nabla^2 \tilde{C}. \tag{83}$$

Again, if Eq. (83) is used, then to be consistent, Eqs. (76) and (77) should be used. If Eq. (82a) is used, then Eqs. (75) and (78) should be used.

Gelhar calls $E(\tilde{C}\tilde{\mathbf{v}})$ the macroscopic dispersive flux and writes it as

$$E(\tilde{C}\tilde{\mathbf{v}}) = |\mathbf{v}| \mathbf{D} \cdot \nabla E(C) \tag{84}$$

where $\mathbf{D}$ is again the macroscopic (asymptotic) dispersion tensor. This macrodispersion tensor is now easily found by aligning the coordinate axis so that Eqs. (50a) and (50b) hold, assuming $\tilde{C}$ is weakly stationary and using the spectral representation of $\tilde{C}$. The result is

$$\mathbf{D} = \int_{R^3} \frac{\hat{\boldsymbol{\rho}}_{\tilde{\mathbf{v}}}(\mathbf{k})\, d\mathbf{k}}{[ik_1 + a_L k_1^2 + a_T(k_2^2 + k_3^2)]\,|\mathbf{v}|^2}. \tag{85}$$

As this brief outline indicated, there are a number of strengths and limitations to this approach. The major strengths are similar to those of Winter, although Winter's approach is more general (Gelhar's approach relies on the stationarity of C). The limitations to Gelhar's approach are also similar to those of Winter's. The major assumptions used by Gelhar include: (i) at least two distinct scales exist; the local and the mean (it is not completely obvious how the mean scale of Gelhar relates to the asymptotic scale of Winter), (ii) it is not clear to me how stochasticity arises in Gelhar's approach; I assume, like Winter, it arises from stochasticity of the local velocity, (iii) the local velocity, concentration and log hydraulic conductivity are weakly stationary; (iv) the mean concentration is measurable (see Sposito et al. 1986 for a discussion of this), (v) a first order approximation is sufficient to give reasonable results, (vi) various mathematical constraints apply to $\hat{\rho}$ to assure its existence, (vii) boundary conditions don't affect dispersion (see Schoen et al. 1987 for problems with this assumption), and (viii) the local dispersivity is constant (see Gupta and Bhattacharya 1986 for a discussion).

7 Summary

In this article, we have made an attempt to review a number of stochastic methodologies which are in use to develop subsurface transport equations and constitutive parameters. We have not been directly concerned with stochastic methodologies which are in use solely to solve stochastic PDE's. We have been somewhat selective in our choice of methodologies; choosing only those approaches that the author perceives as having the greatest ramifications and with which the author is familiar. An attempt has been made to outline the major assumptions associated with each methodology, and to highlight each method's strengths and weaknesses. The list of topics includes: Martingale techniques (e.g., Brownian motion), stochastic-convective methodologies, stochastic-relativist methodologies, spectral-integral approaches, perturbative analysis, statistical-mechanical techniques, and generalized hydrodynamics. Each method has its own advantages and disadvantages. The choice of methodology is dictated by the problem at hand. Most often one looks for a technique that is applicable and which minimizes the assumptions necessary to be consistent with experimental data.

We illustrated why the scale of observation is a key to our understanding of stochasticity. Any stochastic technique must adequately handle the concept of measurement scale. It was pointed out that only Clifton and Neuman (1982), Neuman and Depner (1987), Dagan (1986) and Cushman (1984, 1986) have accounted for the measurement process in stochastic subsurface transport.

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