

Conservative Transport in Multi-Scale Media

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Abstract

Development of computer codes for groundwater modeling requires significant advances in our understanding of fundamental physical and chemical processes to account for the multitude of length scales found in natural media. Flow and transport are fundamental to all models and understanding how to account for scale in these processes is a prerequisite for making significant advances in groundwater modeling. Transport based on the advection–diffusion equation is readily shown by practical example to be inadequate for making fate–transport predictions. The theoretical deficiencies of the ADE are well known although alternative theories tend to involve complicated non-local formulations. A review of basic assumptions of transport theory show that the dominant process in transport through porous media is differential advection, which can be characterized statistically through spatial and temporal correlations in the velocity field. Differential advection reduces to the ADE only as an asymptotic case that is rarely valid in real media. A particle tracking scheme is introduced that contains appropriate velocity statistics and is shown to predict realistic contaminant plumes. A particular generating scheme for random correlated velocities is introduced that can be mathematically transformed to a system of transport equations for parallel media whereby each equation transports a fraction of the media with its characteristic advection velocity. The rate of transport among the parallel media determines the spectrum of length scales. The formulation of a continuum multi-scale transport model thus follows naturally from the particle tracking scheme. A cursory analysis indicates the transport scheme has the potential for non-conservative transport whereby diffusion-limited processes, sorption–desorption, and reactions can be included in the scheme by adding parallel media with zero advection velocity.

Introduction

Since its introduction in the 1990s, the DoD Groundwater Modeling System has become the standard tool for assessment and design of groundwater cleanup schemes [6]. In the initial phase of its development, the GMS was intended as a stable user interface from which to launch groundwater simulations using a variety of models. At the same time, research efforts were initiated to develop computer codes better suited for High Performance Computing resources with more realistic physical and chemical models [19]. Part of this development requires advances in numerical and computational technique, but a part requires significant advances in our understanding of fundamental physical and chemical processes, including how these processes are expressed compactly in mathematical form. In this latter effort, a key impediment to understanding physical processes is the inability to make sufficiently detailed observations in laboratory and field experiments. The difficulty derives from the role of scale. All observations require sampling a finite volume over a finite time interval. Most of these observations quantify the constituents of the aquifer and pore fluid in terms of concentration, which carries the connotation that all parts of the volume “see” the same proportions of constituents. Such may be the case in bench-scale batch test, but is far from the reality of conditions within an actual aquifer. As a result, the kinetic effects derived from diffusion or the partitioning effect of the pore space topology is lost in such tests. Column tests are more realistic but the effectiveness of diffusion in the transport process is still overstated by the limited size of the specimen. The result is a spurious scale effect in which physical parameters appear to depend on the size of the problem [16]. Numerical computations similarly involve averaging over discrete volumes. Thus, application of fundamental observations to *in situ* conditions involves a more formal role of scale in groundwater theory than presently exists.

This paper summarizes the development of a practical theory for contaminant transport that is free from spurious scale effect. An exciting aspect of the theory is the role of HPC in its evolution. Through access to DoD HPC resources it was possible to construct high resolution models that were instrumental in first, understanding the role of scale in observations and computations and second, devising process descriptions in which scale plays an explicit role. Peters et al [17] provide a general overview of this research and how it relates to the GMS development. In this work the computer serves as an experimental device whereby a process is simulated in detail at a scale consistent with the physical process. Numerical simulations of this sort yield accurate results when sufficient resolution is applied but are impractical for large-scale computations. The goal is to develop low-resolution computations that describe the processes statistically but with coupling among processes that correctly accounts for their characteristic time and length scales. Measurements in the virtual experiments can be made at fundamental scales or averages can be computed that match the observation scale. Such statistical data are not available from actual

physical experiments. The link between the virtual and real laboratories is made by the ability of the virtual experiments to produce the averaged results obtained from the real experiments.

Study Focus

The focus of this work is the mechanics of *conservative* transport, where conservative implies that only the transport processes of advection, diffusion, and dispersion need to be considered in the mass balance relationship. Constituents separated by length greater than those associated with diffusion do not interact. Dispersion is, physically, differential advection at the fine scale, which has the same effect on representative elemental volume (REV) concentrations as diffusion. However, at scales on the order of the diffusive length the two processes are quite different because differential advection does not mix constituents within the REV. Where differential advection is the dominant transport process, the traditional advection–dispersion equation (ADE) vastly overestimates the mixing of constituents giving rise to an apparent scale effect in kinetic rates. At present, the only “practical” way to account for these differences is to explicitly model the transport as advection, which implies that all computations be performed at the pore scale. Such computations are, of course, practical only for very small volumes of porous media.

Conservative transport has received considerable attention from the research community and there is general agreement that the fundamental problem is understood in principle; dispersion can be computed from the advection velocities [16, 22, 9, 4, 5, 3, 15, 8]. The practical inability to model advection at the fundamental pore scale poses the real difficulty. A theory that is practical at the coarse scale where engineering computations are made, requires a statistical description of the problem. The ADE gives a statistical description to this process, which essentially assumes that there is no correlation between velocities from one time to the next or between two particles separated by an arbitrarily small distance. Unfortunately, the structure of porous media involves a multitude of length scales that creates significant correlations among velocities in both time and space. As a result, the ADE suffers from significant scale effect. It follows that two elements are needed to make transport computations practical. First, multi-scale data on advection is required to quantify the statistical descriptions. Second, a mathematical vehicle is needed that is as straight forward as the ADE but capable of capturing the correlation structure of velocity statistics. In Part I, the first problem is addressed through the application of HPC to create a virtual laboratory in which velocity measurements are possible. The second problem is shown in Part II to have a simple solution which has significant potential for application to more complex processes of reactive transport.

A Practical View of the Scale Problem

In conservative transport, contaminant particles move by net translation with the fluid and by random molecular motion (molecular diffusion). For transport through porous media, the translational motion (advection) is generally much larger than the molecular component and is highly variable in space. This differential advection tends to separate contaminant particles in a way that, when viewed from a distance, resembles diffusion. In fact, the concentration distribution measured at the end of a laboratory column of one soil may be fit nicely to a diffusion model. Unfortunately, natural soils are highly variable and exhibit structure at many (some say all) scales [14]. This makes application of traditional transport models difficult.

The traditional approach for simulating transport is built around the advection-dispersion equation (ADE). The ADE simply is a mass conservation statement for the constituent stating that the change in mass within a control volume equals the net flux through its boundaries by advection and dispersion, or

$$\frac{\partial c}{\partial t} = -\nabla \cdot (\mathbf{vc}) + \nabla \cdot (\mathbf{D} \cdot \nabla c). \quad (1)$$

where c is concentration, t is time, \mathbf{v} is transport velocity, and \mathbf{D} is the dispersion coefficient. This equation lumps the net translation into an advection term and the net spreading into the dispersion term. To apply the ADE, we must choose appropriate coefficients for the advective and dispersive terms of the equation. Consider this traditional, “lumped” ADE approach to the conservative transport of a constituent at a heterogeneous field site.

It is well established that the apparent dispersivity, used to compute the dispersion coefficient in the ADE, exhibits a scale effect (Figure 1). The apparent dispersivity grows with the plume size until the size of the plume reaches 10-15 times the size of the largest scale of heterogeneity [7]. For this application with a plume size of roughly 2500 ft., an appropriate choice for dispersivity would seem to be about 250 ft. However, this philosophy of selecting dispersivity based on plume size has several flaws. First, the dispersivity, theoretically a constant, must be adjusted as the plume grows. Second, the observed trend in apparent dispersivity was derived from data on impulse sources of constituent. For a plume emanating from a nearly constant source (like the source in this application), the inadequacy of this Fickian model for dispersion is apparent. Contaminant particles that entered the flow field early may have experienced much of the velocity spectrum and may behave much like an asymptotic, Fickian plume. But, those contaminant particles entering the flow field at a later time retain much of their original velocity character. Thus, the apparent dispersivity should be larger for the leading edge of the plume than nearer the source. Consequently, a large value of dispersivity represents the leading edge well, but

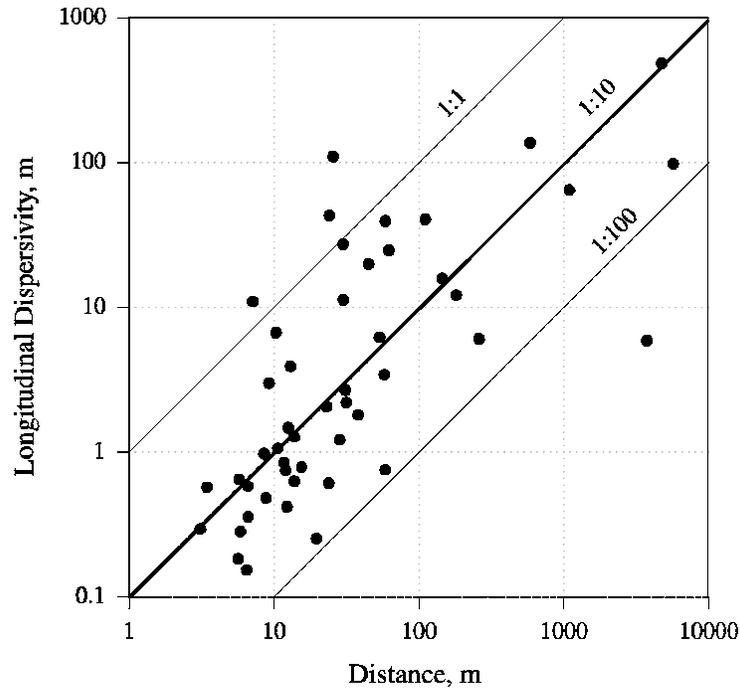


Figure 1: Apparent scale effect in dispersivity (*modified from [1]*).

overpredicts spreading of the plume near the source. Significant, artificial upstream spreading is possible.

This problem can be alleviated somewhat with the inclusion of geostatistical simulation within the flow prediction, thereby explicitly capturing as much differential advection as possible. But, we cannot afford to represent all scales of advection contributing to the spreading of constituents at the field scale and an improvement in the description is required.

Solution of the equation for flow through porous media provides a Darcy flux, which is the volume of water passing through a unit of cross-sectional area per unit time. The material contained within the control volume over which the equations are applied is not uniform. That is, water, air, and soils exist in each of our elements and the soil is considered stationary. Thus,

Darcy flux represents only the volumetric movement of the water, and not the speed at which the water particles move. This is relevant for conservative transport because a conservative tracer migrates within the water, and not the solids. Therefore, the Darcy flux always underestimates the speed of fluid motion and transport. For example, if half the porous medium were solid and half water, to achieve a Darcy flux of 1.0 ft/day, the average water velocity would need to be 2.0 ft/day. This water velocity is called many names including the seepage velocity, linear pore velocity, or transport velocity. As a first estimate, the transport velocity may be computed by dividing the Darcy flux by the total volume fraction of the pore space, or total porosity.

As stated earlier, the primary mechanism for constituent migration through porous media is advection with the moving water. However, all water in a porous medium is not uniformly mobile. Some of the water lies in dead-end pores. Some is adhered tightly to the soil grains. So, instead of the total porosity that represents all of the pore space, the mobile water content (or effective water content) is a better estimate of that fraction of the volume transporting constituents in the pore water. Effective porosity is the term used to describe the fraction of the porous medium that actually participates in the transport. Therefore, to convert the Darcy flux to a transport velocity, it must be divided by the effective porosity.

$$\mathbf{v} = \frac{\mathbf{q}}{\phi_e} \quad (2)$$

where v is the seepage or transport velocity, q is the Darcy flux, and ϕ_e is the effective porosity. In saturated porous media, the effective water content is the specific yield, which is often recommended as an estimate of effective porosity. Specific yield is measured as the volume fraction of water produced by gravity drainage. Unfortunately, specific yield may be a poor estimate of the effective porosity in heterogeneous media. The disconnect arises because specific yield is a steady-state concept. Transport is often highly transient. Constituents move first through the less resistive media or preferential flow paths.

Numerical experiments were conducted on a block of soil composed of two materials. Ninety-five percent of the block is made of a lower permeability material, and 5 percent is a higher permeability material contained in a strip that traverses the entire block. The conductivity ratio between the two materials is 20. The specific yield of each material is set to 0.2. Thus, the specific yield of the composite block should be about 0.2. A uniform flow field was established parallel to the bedding. A conservative tracer was introduced as a pulse at the block boundary. Neglecting diffusion, the higher conductivity material (5 percent of the domain) carries over 50 percent of the tracer mass. Thus, the center of mass of the tracer is heavily weighted toward the faster paths and the effective porosity required to produce this transport velocity is about one

sixth the specific yield.

When transport occurs slowly and diffusion has time to involve the lower conductivity material, specific yield becomes a better estimate of the effective porosity. Thus, in heterogeneous media, the effective porosity is a function of the Peclet number (ratio of advection to diffusion). For this computational example, an infinite Peclet number reduces the effective porosity to one-sixth the specific yield and a small Peclet number (large diffusion) produces an effective porosity much closer to the specific yield. This dependence on Peclet number is shown here with a mild degree of heterogeneity. The effect would be more pronounced with more contrasting conductivities. For the field application problem, the specific yield was estimated at about 0.22. But, to match plume migration would require an effective porosity in the range of 0.03. This ratio of 7:1 required for the field study compares well with our computational example.

The issue here is one of homogenization. The control volume is defined at a scale that encompasses multiple velocities. But the traditional “lumped” ADE requires that a single transport velocity be provided that applies to all constituent particles within the volume. Again, some relief may be found by decreasing the size of the control volumes (computational elements) and including geostatistical representation of the material. But, the problem is a fundamental one and may not be addressed practically with increased resolution.

This multiple-velocity effect in transport has been recognized for many years. Dual porosity models with mobile and immobile fluids have been used to represent fractured rock systems. However, in fractured rock, the effective porosity and specific yield may not differ significantly because the rock matrix contributes little to the specific yield. In heterogeneous porous media, significant transport may occur in soils of several different conductivities.

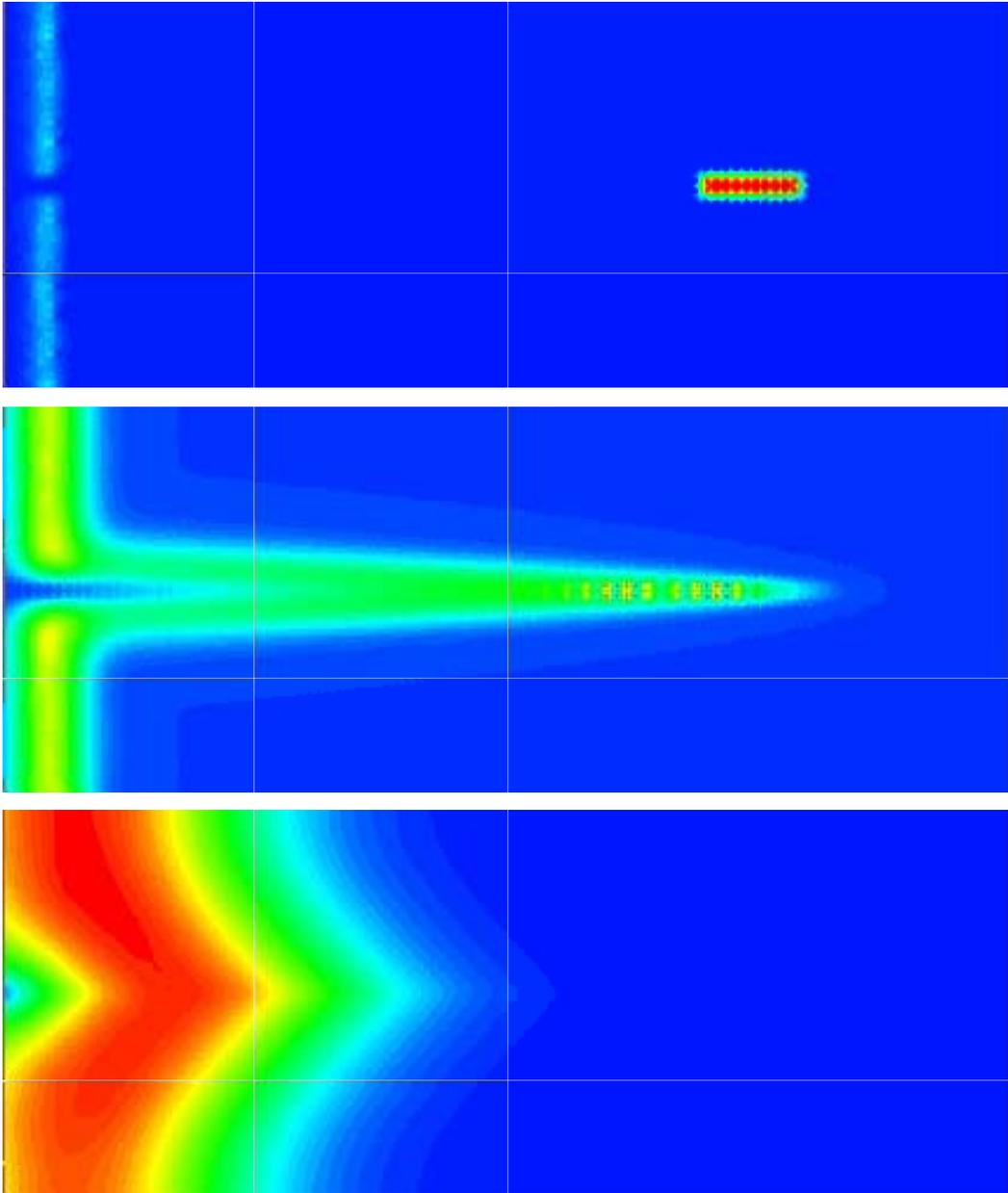


Figure 2: Computational example showing transport of a pulse of tracer through a block of porous media containing two materials. Diffusion is smallest at the top and largest at the bottom.

Part I: The Virtual Laboratory

Direct measurement of particle velocities is restricted to spatially and temporally averaged measurements in a few simple experimental configurations. By contrast, the simulated velocity data obtained from computer experiments provides detailed information at a resolution limited only by the capacity of the computer. The two types of experiment are verifiable by comparisons of average quantities measurable in the physical experiments. Thus, the virtual laboratory provides the detailed observations needed to quantify the statistical picture of dispersion while the physical laboratory serves as a link to reality.

Three types of analyses were used in this work. The first analysis consisted of pore-scale computations in which the flow field was modeled by the Navier-Stokes equation using the Lattice-Boltzmann method coupled with a particle tracking method to model transport. These simulations were verified using velocity measurements from NMR experiments [13, 11]. The second type of simulation was a Darcy-scale computation in which the flow field was computed as a potential flow problem in a heterogeneous porous media. Tompson and his colleagues [21, 20] pioneered highly resolved Darcy-scale simulations as a means to create realistic dispersive effects. In their method, random permeability fields are generated with spatial correlation matching that of the target medium. In the simulations reported here, advection is modeled using particle-tracking methods to limit numerical diffusion. While a small dispersivity is included to capture sub-grid scale variability, dispersion primarily results from differential advection that arises from the random permeability distribution. Like the pore-scale simulations, the random field simulations provide a means to study the time-correlation of individual particles.

The third type of simulation performed with a network flow model in which the porous media is idealized by a very dense network of flow paths. The network model was verified by Howington et al [16, 7] to realistically simulate contaminant plumes for a number of physical situations including density driven flow and immiscible fluid flow. The network model is especially useful in understanding the statistics of velocity and sub-scale concentration distributions. Its accuracy depends primarily on the resolution of the network. While the network is an abstract idealization of real porous media, it is no more so than a continuum model based on Darcy permeability. The key point is that contaminant plumes simulated by the network are comparable to those observed in actual media; the network provides the statistics of advection velocities that produce those plumes.

The Statistics of Dispersion at the Pore Scale

A number of pore-scale simulation models have been used in academic research but are limited in size and scope. What is needed for the present research is a pore-scale model that spans between pore scale and REV scale whereby fluid flow within the pores is governed by the Navier-Stokes equations but the total problem domain defines a REV. Otherwise, boundary conditions dominate the simulations or larger-scale structures do not develop in the flow field. As shown by Maier and Bernard [10], the smallest discrete scale must resolve features such as re-circulation zones near which diffusion dominates transport. The simulation domain should contain travel paths having highly correlated advection velocities that persist over many pore sizes. The computer resources needed to achieve such a span of scales is very large. Computer memory must be allocated to resolve the pore spaces sufficiently to approximate the flow. The domain size must allow the plume to grow large enough that it can serve as the initial distribution for Darcy-scale model. Such a pore-scale model would have linear dimensions of several hundred particles. The smallest element in this domain provides for a sufficient number of lattice cells to resolve circulation fields at the pore level. Considering a minimal requirement to be 10 lattice points for the nominal particle size, LB simulations with approximately 1 billion lattice sites are needed. Some leverage is obtained from periodic media whereby the boundary conditions of the flow field at the “exit” boundary match those at the “entrance” boundary. A periodic array of identical flow fields combine to create an infinite array of periodic cells. Contaminant transport through the array is conducted using only the flow information contained within one cell. Large flow fields can be created from periodic cells, although there is evidence that velocity auto-correlations over distances greater than a unit cell are artificial and do not reflect the behavior of real porous media [12].

Despite limitations, results were obtained from pore scale computations from which definitive conclusions can be made. Detailed comparisons with NMR spectroscopy show qualitative agreement on the spatial distributions of the tracer at both short and long time intervals. This amounts to a verification of sorts to the computational results at the microscopic level. However, the NMR methods entail a number of limitations in accuracy and quantitative comparisons are still restricted to macro-scale measurements. The pore scale computations agree with a number of theoretical and empirical results on the scaling of asymptotic longitudinal and transverse dispersion with Peclet number.

An interesting and potentially significant finding from the pore scale transport studies is the character of velocity correlation as the asymptotic state is approached. Correlation may be viewed from either a temporal or spatial viewpoint. Quantities are, of course, perfectly correlated with themselves when samples are compared at zero separation distance and time. When measurements

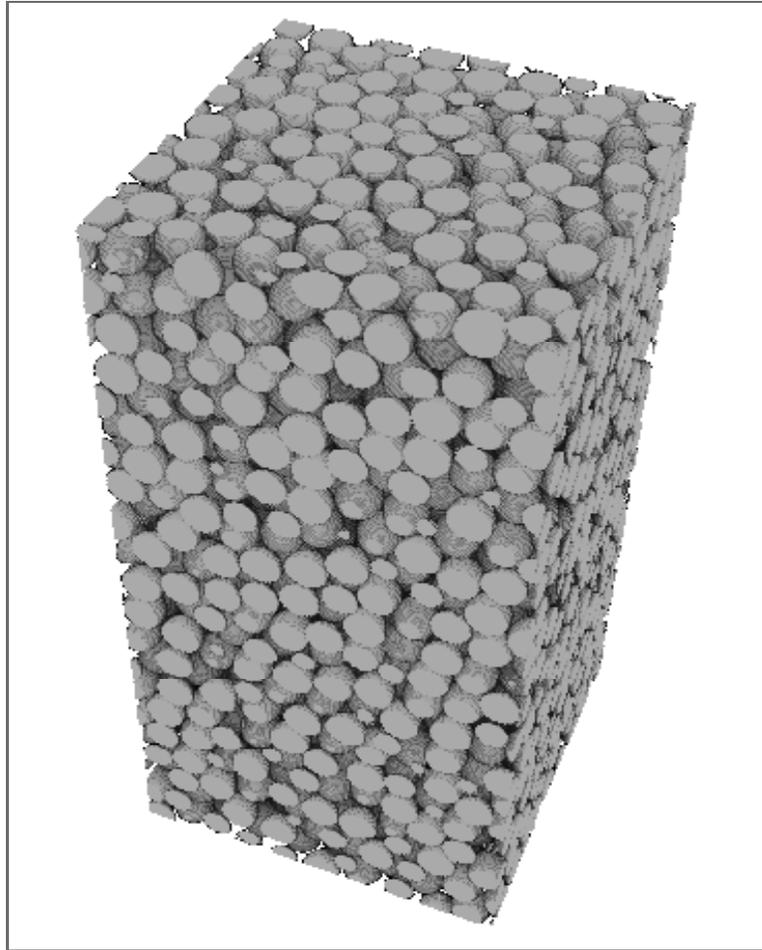


Figure 3: An example pore-scale domain used in the virtual laboratory.

are separated by time and distance, correlations diminish such that there is zero correlation among quantities after long time intervals when the sampling points are separated by significant distance. Here, time and distance are related by velocity history of the particle and correlations in time and distance are interchangeable. The auto-correlation function, that quantifies the decay in correlation, is generally assumed to be a smooth function that asymptotically approaches zero. Simulated auto-correlation functions are neither smooth nor asymptotic. The functions do display the expected smooth decay over a characteristic time τ , but become periodic as time increases. In some cases the periodicity is clearly related to the structure of the pore space, as in the case of cubic bead packs, but in other cases appear to have no relationship to an obvious spatial structure in the pore space. The limitations on domain size limits investigation on the long-range correlation but as discussed in the subsequent sections, the non-asymptotic nature of auto-correlation functions appear to be a characteristic feature of porous media.

The Statistics of Dispersion at the Darcy Scale

Darcy-scale simulations consist of flow and transport computations for randomly generated permeability fields. The permeability fields are created on a high-resolution grid where each grid cell represents the smallest discrete feature in the field. The aim of the study was to describe the velocity correlations that are produced in random media. The principal descriptive feature of the permeability is the correlation length. The correlation length is a rough measure of the persistence of a feature in space; the greater the correlation length the greater the spatial correlation. Spatial features comprising many grid cells emerge from the *turning band procedure*, which imparts spatial correlations to the permeability [21, 20]. The correlation of the velocity field is a combined product of the random permeability field, the flow equations, and the boundary conditions.

The velocity field was characterized by the time correlation of velocities as seen by particles transported through the medium. The auto-correlation was computed as the covariance of velocities at times separated by a time lag τ . The result is a covariance function $C(\tau)$. The covariance is computed from an ensemble of 100 particles to ensure a statistically significant result. Simulations were performed for two permeability correlation lengths corresponding to 5 and 10 grid cells. The time lag τ was expressed in a dimensionless unit computed from the ratio of time to time it takes to cover the correlation length. Two features emerged. First, in the initial phase corresponding to a time lag of 10 correlation lengths, the correlation diminishes rapidly. Second, the correlation does not reach an asymptotic value but begins to fluctuate around zero. The period of this correlation oscillation appears to be about 75 correlation lengths, which means that the fluctuation is more frequent for short correlation lengths. Thus, $C(\tau)$ for the velocities derived from a random permeability field include negative values and oscillate about zero with a

well-established period of oscillation. This finding is at odds with the typical view of covariance behavior but fully consistent with the results of the pore-scale simulations. It should be noted that the permeability field itself is constructed with a covariance that decreases monotonically with lag distance. The oscillatory behavior is a product of the flow field that emerges from the random permeability field.

The Statistics of Dispersion from Network Simulations

The network model is used to describe the scale effect in dispersivity [16] and is described in detail by [7]. The network model consists of a dense three-dimensional array of “throats” that meet at “connections” (Figure 4). The network throats represent flow (advection) pathways and the connections represent points of mixing. Flow through the network is controlled by total pressure at the connections as in a network of pipes. The fluid may be of variable density, which contributes a density-driven component of head. Generally, the head loss within a throat follows a linear relationship between flow velocity and total gradient, although capillary forces can be included to model multi-phase flow. The throats include a compressibility to model storage in transient flow problems.

Transport of contaminant through the network consists of advection through throats and mixing at the connections. At present, there are no provisions for diffusion in the throats. However, the assumption of perfect mixing in the connections is partly based on the idea that diffusion within a throat randomizes the streamline by which a particle will enter a connection. The scale of the problems considered by the network is such that diffusion plays a limited role.

The relationship between the network and a real porous medium is based on the fact that if an observation is a spatial average of detailed quantities then many fine-scale systems can produce the same coarse-scale result. In the case of a linear flow network this fact can be demonstrated formally based on the ability to construct a network that will produce the same coefficient matrix as a given discrete system produced from a finite element or finite volume representation. The transport problem is more involved, but comparison to experiments shows that the network produces plumes having growth rates similar to real porous media. The realism of the network simulations appears to be tied primarily to the resolution (fineness) of the connections. Howington [7] devised an empirical calibration procedure that related the parameters for the statistical description of throat properties to the longitudinal and transverse dispersivity. The key generalization gained from this procedure is that longitudinal dispersivity depends on the variance of throat conductivity whereas transverse dispersivity depends on the mean length of throat. It follows that the network is most effective in modeling real media at high resolution when the number of network connections is on the order of millions.

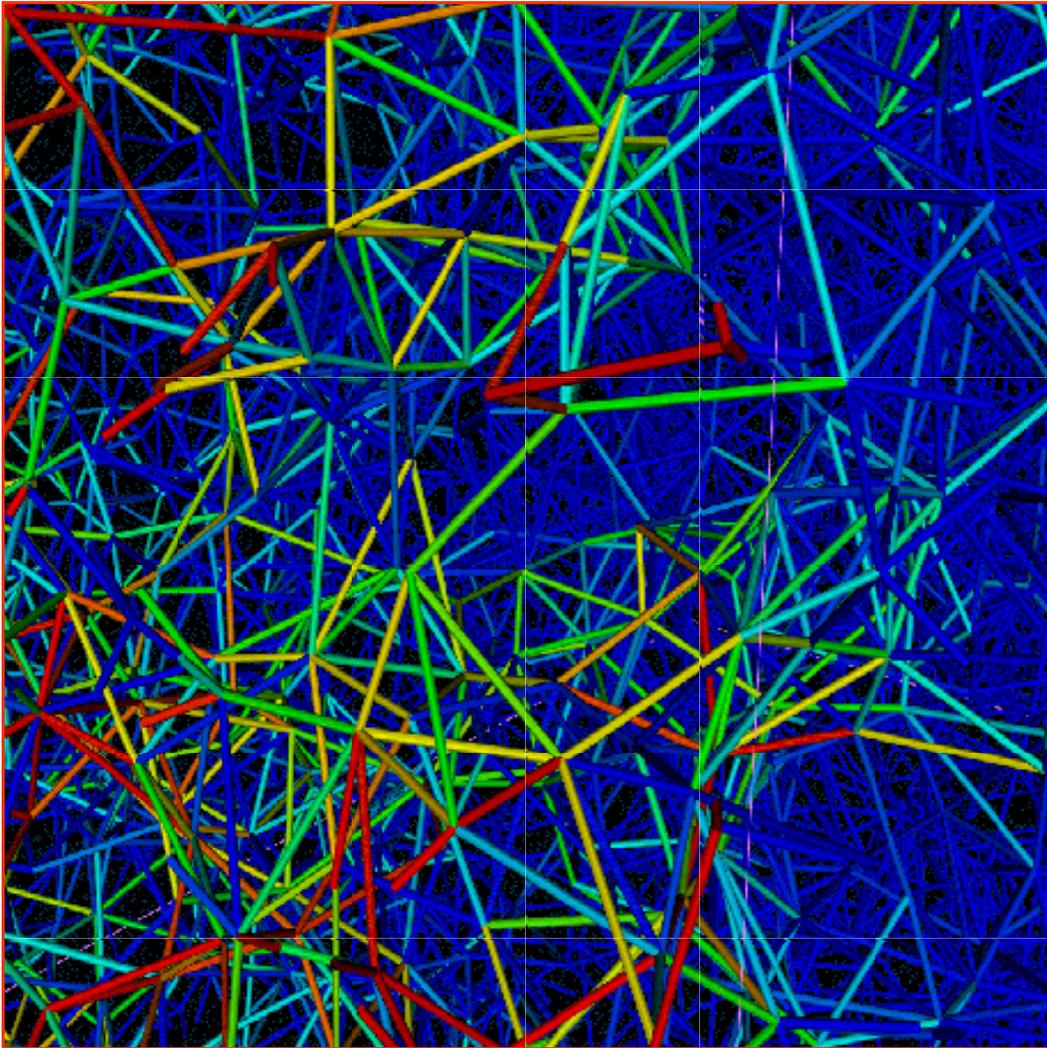


Figure 4: A schematic of a network used in the virtual laboratory.

The network does not use particle tracking, making time correlation impractical. Instead, an equivalent spatial correlation, ρ_{xv} is employed [16]. Additionally, the network analysis focused on the apparent dispersivity, α , and its growth with increasing plume travel distance. The apparent dispersivity measures the rate of plume growth from a point source, a rate that is constant for the ADE. Therefore, the growth in α is a direct measure in the deviation from traditional transport theory. The apparent dispersivity is proportional to the spatial correlation ρ_{xv} and is the integral over time of the function $C(\tau)$. It has been established from numerous simulations that the relationship between spatial correlation and time is similar to the time correlations derived from the pore scale and Darcy scale simulations. The correlation persists for a plume travel distance of approximately 10 times the correlation length. Beyond this length, the correlation does not asymptotically go to zero but rather oscillates around zero. The long term oscillation is reflected in the changes in α which also oscillate about an apparent asymptote.

Part II: A Theory for Dispersive Transport

The difficulty in applying results of velocity measurements, either from physical or virtual experiments, is deriving a compact mathematical formulation of transport for general velocity statistics. Very realistic contaminant plumes can be created from high resolution random field models. Unfortunately, computational requirements of these models increase exponentially with the range of scales represented (as measured by smallest element relative to domain size). The key to extending the range of the computation lies in the observation that high resolution is required to create the variable (random) flow that drives dispersion. Peters and Howington [16] noted that the averaged flow field is smooth, even for highly heterogeneous networks, and can be reproduced by a traditional flow model using less computational resources. Similarly, a comparison by Ashby et al [2] of flow systems in heterogeneous versus homogeneous media based on total head showed that the pressure field is not greatly affected by the very large permeability variation at the finest scale; the effect of the fine scale permeability variability is to create variability in the fine scale velocity field. The implication of this observation is that a pressure solution is not needed on a highly-resolved heterogeneous medium, provided the velocity statistics that drive the dispersive transport can be replicated by other means. If the velocity statistics can be modeled as a random process that is driven by the average flow field, the high-resolution flow computations can be avoided. The classical advection-dispersion equation (ADE) attempts this through a Fickian description of dispersion. Unfortunately, the Fickian model lacks a correlation structure found in

real media. Non-local models of dispersion [9, 3, 15] provide correlation structure but, to date, have not provided versatile, practical transport models.

Time-Correlated Models of Dispersion

A promising approach for bridging the model scales for contaminant transport and remediation processes is correlated particle tracking [17, 18]. Particle tracking is a numerical technique that produces the solution to the ADE by assigning to each particle the mean advection velocity and a random component having a velocity variance equal to the square root of the diffusivity. In correlated particle tracking, velocities are generated randomly but with correlation in time. In effect, the particle tracking approach directly simulates the behavior that non-local models attempt to capture more compactly by their integral formulations. Dispersion is controlled primarily by deviations from mean flow, which necessarily display a history dependence that can be expressed through a velocity correlation structure. It follows that an equivalent medium can be created that has the same dispersive effects as the actual heterogeneous medium.

The statistical components of the particle tracking velocities are time-correlated such that they replicate those of the medium. As for the traditional random walk model, the advection velocity v_i consists of a mean flow \bar{v}_i and a random deviation from the mean v'_i .

$$v_i = \bar{v}_i + v'_i. \quad (3)$$

By definition, the mean of v'_i is zero. If v'_i has a Gaussian distribution, Fick's law can describe the dispersion created by the random component, resulting in the traditional ADE. However, in a medium having a finite length scale, $v'_i(t)$ is correlated with $v'_i(t - t')$, for all $0 < t' < t$. This correlation is readily seen for transport in a network because once a "particle" of mass enters an advection path, its velocity remains constant until it exits the path. If the plume of particles is small relative to the sizes of the advection paths, the advection velocities display high correlation. This correlation persists as long as a significant portion of particles retain the same velocities. If the pathways are short relative to the plume size, the time required for particles to enter and exit pathways is short compared to the time required for the plume to move a significant distance and the correlation effect is lost. Once correlation is lost, a diffusion model for dispersion is appropriate for defining general plume size and movement. Hence, as a plume grows, an asymptotic state is obtained for which the traditional ADE approximates the growth rate.

A potential problem with this approach is that the dispersivity in a correlated system is time dependent, requiring the history of concentrations to be maintained. For long simulations, any

HPC resource would be swamped by the required calculations. The alternative to a memory-based system is one based a set of state variable that control dispersive flux that evolve in time. The procedure begins with a generator of random correlated statistics based on a convolution integral

$$v'_i(t) = \int_0^t \phi(l-l')\beta_i(l')dl' \quad (4)$$

where $l = l(t)$ is the path length followed by the particle and β_i is an random function that satisfies for all $\Delta l > 0$

$$\int_0^l \beta_i(l')\beta_i(l'-\Delta l)dl' = 0. \quad (5)$$

Note that the physical basis of the correlation is length scale, not time scale. The relationship between the plume position and its size should be independent of mean seepage velocity (provided molecular diffusion is small). Accordingly, l is the independent variable and dependence on t is purely parametric.

The statistical correlation structure is embedded in the kernel $\phi(l)$ which can be related to the auto-correlation function $R(l, \Delta l)$ given by

$$R(l, \Delta l) = \frac{1}{l} \int_0^l v'_i(l')v'_i(l'-\Delta l)dl' \quad (6)$$

For long times it is assumed that an asymptotic state is reached whereby for some l_o

$$\frac{1}{l} \int_0^l v'_i(l')v'_i(l'-\Delta l)dl' = \frac{1}{l-l_o} \int_{l_o}^l v'_i(l')v'_i(l'-\Delta l)dl', \quad (7)$$

At the asymptotic condition, $R(l, \Delta l) = R(\Delta l)$.

For N discrete intervals δl_n , the values of $R(n\delta l)$ can be arranged into a $N \times N$ correlation matrix $[R]$. A correlated sequence of velocities can be generated from

$$\{v'_i\} = [L_R]\{\beta_i\} \quad (8)$$

where $[L_R]$ is a lower triangular matrix obtained from a $L-U$ decomposition of $[R]$ and $\{\beta_i\}$ is a vector of uncorrelated random numbers. Equation (8) can be viewed as a discrete approximation of Equation (4), thereby establishing a relationship between the generating kernel and the correlation function defined by Equation (6).

The State Description of History Dependence

The disadvantage to Equation (4) as a generating model becomes apparent. Each random number in the sequence is a weighted sum of all previous entries in the random sequence. The random sequence $\beta_i(l)$ grows large as time progresses and soon makes the computation impractical. The required sequence is shorter if the “memory” of the correlation is short. However, the sequence length grows as the time step becomes smaller. The storage problem can be avoided if the history description can be replaced by a state description. To do this first note that the correlation kernel is typically a decaying function that can be represented by a finite Dirichlet series

$$\phi(l) \approx \sum_{r=1}^M A_r e^{-\alpha_r l}. \quad (9)$$

This, in turn, leads to

$$v'_i(t) = \sum_{r=1}^M \mathcal{V}_i^r(t) \quad (10)$$

where

$$\mathcal{V}_i^r(l_o + \Delta l) = \mathcal{V}_i^r(l_o) e^{-\alpha_r \Delta l} + \int_0^{\Delta l} A_r e^{-\alpha_r(l-l')} \beta_i(l') dl'. \quad (11)$$

Note that if $\beta_i(l')$ is taken as a constant over the interval, Equation (11) reduces to

$$\mathcal{V}_i^r(l_o + \Delta l) = \mathcal{V}_i^r(l_o) e^{-\alpha_r \Delta l} + \beta_i \frac{A_r}{\alpha_r} (1 - e^{-\alpha_r \Delta l}). \quad (12)$$

For small $\alpha \Delta l$, $\mathcal{V}_i^r(l_o + \Delta l)$ is weighted toward $\mathcal{V}_i^r(l_o)$ creating a correlation in time. For large values of $\alpha \Delta l$, $\mathcal{V}_i^r(l_o + \Delta l)$ is weighted toward the uncorrelated random number β_i . The parameters A_r and α_r describe the spectrum of correlation lengths present in the heterogeneous flow field. Importantly, in this state-based approach, the M values of $\mathcal{V}_i^r(l_o)$ replace the sequence of random velocities, thus making the method computationally feasible. The number of state terms retained depends on the number of scales to be represented in the correlation.

An interesting aspect of the particle tracking technique is the ability to model length scales much smaller than are obtainable with direct simulation by high resolution models. The dispersion rate of a direct simulation code is limited by the smallest spatial element in the grid. In particle tracking, the dispersion rate is limited numerically by the smallest step taken by a particle. In principle, the particle tracking method can capture dispersion at the smallest scale. However, to achieve such results, the numerical time step must be small. In dispersive transport problems, the time and length scales are related. To capture small dispersivity, a fine grid with an appropriately small time step is needed. For a particle tracking method, the spatial element of dispersion is no longer tied to the grid, and the time step, alone, is the limiting factor.

An advantage of the correlated particle tracking is that it is easily adopted into finite element/finite volume engineering codes such as those routinely used in field-scale cleanup. Particle tracking offers particular advantage for parallel processing because each particle is independent. The independence of the particle motions simplifies implementation for the random walk routines. Inter-processor communication comes about only when computing mean flow velocities, data collation for post processing graphics, or calculating concentrations for chemistry computations.

Thus, a traditional subsurface model combined with the computationally efficient particle-tracking model can replicate the physical realism of the high resolution model but with much less computational burden. The important feature of the random walk model is the time-correlated velocity statistics created by heterogeneity. These statistics do not need to be obtained from a single high-resolution simulation that spans all scales, but can be assembled from several simulations that collectively span the scales. The current HPC Challenge Project combines data from the network, pore-scale lattice-Boltzmann, and continuum-based finite element models to quantify velocity statistics of over a very large range of scales. These statistics are the basis of a procedure that permits relatively lower resolution flow computations, consisting of a few hundred thousand elements, to replicate the transport behavior of high-resolution analyses that contain billions of elements. The particle models are ideal to take advantage of HPC resources because particle tracking methods parallelize well and have large memory requirements.

A Simple Generator of Correlated Velocity Statistics

The preceding section gives a formal procedure for creating an efficient generator of correlated random numbers that honor a specified auto-correlation function. It is useful to consider an alternative generator based on a heuristic understanding of dispersion. The network model suggests a mathematically unsophisticated procedure that captures the correlation structure of the network well, using very few state variables. The algorithm begins assigning each particle a velocity v_i and a time $t_o = l_o/|v_i|$. The length l_o is a randomly generated advection length. The particle retains the velocity v_i until $l > l_o$, at which time a new random velocity is computed as $v_i = \bar{v}_i + \beta_i$ and a new random l_o is computed. Thus the statistical “state” is given by v_i and t_o . This “tube” algorithm produces plumes statistically identical to the high resolution model without performing a detailed high-resolution flow analysis. In the high resolution model velocity statistics are generated from the highly heterogeneous distribution of material properties. In the particle model the statistics are generated randomly by the algorithm described above.

Velocity auto-correlation functions based on samples of 100 particles were constructed for the tube model for three different values of correlation length. An interesting observation of the tube algorithm is the similarity between the auto-correlation functions produced by high resolution

models and the tube model. As noted previously, all of the simulations produced an auto-correlation relationship that decays rapidly into a long-term oscillation having a frequency that is proportional to the correlation length. The tube model produced auto-correlation functions that are identical to those of the high resolution models. The fact that long-term oscillatory behavior is produced by all simulations including the simplistic tube model implies that an oscillating auto-correlation function is a fundamental property of discrete systems.

The State Transition Concept

The statistical generator discussed in the previous section contained two key ideas. First, the history dependence in the velocity field can be cast as a function of state rather than a function of history, thus achieving a large economy in computation and storage. Second, velocity can be generated from procedures, such as the tube model, that inherently produce the correct history dependence including effects not explicitly introduced into the model. Such behavior has a predictive quality not necessarily found in methods based on prescriptive velocity correlation functions. In this section, a model is described that is simple to implement, more compact than even the tube model, and potentially applicable to a wider range of processes than conservative transport.

Consider the contaminant plume as a mixture of particles carried along with the water. Both the dispersive and diffusive effects of travel through the porous medium are the result of the velocity statistics. Rather than generate these velocities from random numbers sampled from a continuous distribution, a discrete distribution will be used. At any instant in time a particle will be selected from a velocity “bin” or group. The instantaneous rate of plume growth depends on how these velocities are distributed spatially, for the rate may vary even if the percentages of particles within each group remain the same over time. The evolution of plume growth depends on how membership of each particle to the various velocity groups changes with time. If each particle retains the same membership for long times, the plume grows at an increasing rate. If particles change membership at a high rate, uncorrelated motion results and the plume grows at a constant rate. The particle state is described completely by its membership, which is denoted by a single integer.

The dispersion is captured by the transition of particles from one group to another over an interval of time. The time can be represented as a sequence of finite steps during which the particles change randomly from one group to another. The probabilities of transition are represented by a Markov matrix such that the fraction of particles within a group that are transitioned into another group are given as

$$\{\mathbf{f}\}_{i+1} = [\mathbf{M}]\{\mathbf{f}\}_i, \quad (13)$$

where $\{\mathbf{f}\}_{i+1}$ is the vector of particle fraction in each velocity class at t_{i+1} , $\{\mathbf{f}\}_i$ is the fraction at t_i , and $[\mathbf{M}]$ is the transition matrix. For each state k , the transition cumulative probability distribution is computed from the k th column in $[\mathbf{M}]$.

The transition model produces realistic plumes, is easy to incorporate into existing particle tracking transport models and is easy to calibrate. Non-symmetric entries in the transition matrix reflect different rates for sorption and de-sorption or irreversible reaction paths. Thus, complicated geochemical pathways can be modeled by a simple robust scheme that does not depend on sophisticated solutions to the advective transport equations. Conceivably, all processes could be dealt with as part of a first-order Markov transition process, giving rise to a single efficient algorithm that covers all transport and fate processes. Such a model could make particularly efficient use of HPC resources.

A Continuum Formulation

The representation of differential advection as a collection of velocity groups is effectively a multiple porosity model. The continuum is envisioned as multiple parallel continua, each having assigned one of the group velocities. The continua interact through exchange of mass. The coefficients defining the rate of exchange among the continua are derived from the transition matrix. The multiple porosity concept is amenable to traditional numerical implementations of the ADE by recognizing that the particle tracking process for a given velocity group is a approximation of the advection-diffusion with source-sink terms. In effect, the mass within a velocity is transported as though it is a distinct species. Thus, the concentration $c(t)$ is the sum of concentrations $\sum_r c_r(t)$ where

$$\frac{\partial c_r}{\partial t} = -\nabla \cdot \mathbf{v}_r c_r + \nabla \cdot \mathbf{D}_r \cdot \nabla c_r + \sum_i A_{ri} c_i. \quad (14)$$

The transfer coefficients A_{ri} define the rate at which mass from each group is transferred to other groups. These coefficients are related to the matrix $[\mathbf{M}]$ by

$$[\mathbf{A}] = \frac{1}{\tau} \log[\mathbf{M}], \quad (15)$$

where $[\mathbf{M}]$ is defined for a time interval τ . The reasoning behind Equation (15) is as follows. First note that the concentration is a fraction of the total mass such that

$$c_r = f_r c. \quad (16)$$

Then for some fundamental increment of time, τ , the distribution of concentrations among the groups is $\{\mathbf{c}\}_{t+\tau} = [\mathbf{M}]\{\mathbf{c}\}_t$. After a time of 2τ the fraction is $[\mathbf{M}]^2\{\mathbf{c}\}_t$, and so on. In general, after some Δt ,

$$\{\mathbf{c}\}_{t+\Delta t} = [\mathbf{M}]^{\Delta t/\tau}\{\mathbf{c}\}_t. \quad (17)$$

Note further that the solution to $\{\dot{\mathbf{c}}\} = [\mathbf{A}]\{\mathbf{c}\}$ is $\{\mathbf{c}\} = [\mathbf{M}]^{\lambda t}\{\mathbf{c}\}_o$ where $[\mathbf{A}] = \lambda \log[\mathbf{M}]$. By applying this result to Equation (17), it follows that $\lambda = 1/\tau$. It is seen that $[\mathbf{A}]$ is the seed matrix from which the transition matrix for a particular time interval can be obtained.

A Non-Local Formulation

While of limited practical value, the tie between the multiple-porosity continuum and non-local models is of academic interest nonetheless. A coupled non-local model follows formally from Equation (14) through application of Laplace transforms in time and Fourier transforms in space by noting the solution for c_r is a convolution integral in time and space. By invoking $c = \sum_r c_r$ along with the property $\sum_r A_{ri} = 1 \forall i$ a non-local“advection” equation is obtained.

$$\frac{\partial c}{\partial t} = \int_0^t \int_{\mathbf{x}} \phi(t - t', \mathbf{x} - \mathbf{x}') \nabla c(t', \mathbf{x}') \, d\mathbf{x}' dt'. \quad (18)$$

Conclusions

From a practical example, the traditional ADE formulation for Groundwater transport models was shown to be inadequate for real geologic media. The example also showed that the failure of the ADE was tied to its assumption that advection velocities could be described by an asymptotic statistical state. The correct view of advection-driven transport, which can be traced from the early work of Taylor to non-local diffusion laws proposed by a number of researchers in recent years, clearly shows that dispersion is controlled by spatial correlations in the velocity field that

are not represented by the Fickian dispersion model on which the ADE is based. The difficulty is devising a model that is practical in its implementation and calibration.

A particle-based transport scheme was devised that replicated the spatial correlations found in geologic media. Particle-tracking schemes have a long track record in groundwater modeling, whereby the random velocity generator is derived from the statistical interpretation of the ADE. The introduction of correlated velocity statistics in the discrete particle model obviated the need to build from a mathematically compact continuum relationship such as the ADE, by directly generating velocities with the requisite correlations. Instead, a continuum model was derived from a particular velocity generating scheme that was consistent with a multiple-porosity interpretation of the flow field. This interpretation amounted to assuming that a contaminant is transported through parallel interacting media such that the ADE was valid within each media. The multi-scale character of plume growth results from the characteristic scale associated with each medium. It followed that the coupled equations can be coerced into the form of a non-local equation in which the dispersion is defined by a convolution integral over time and space. Thus, a sequence of equivalent interpretations of the transport process are presented from which a particular model can be constructed to deal with the particular features of the problem at hand.

The critical role of HPC in this development cannot be overstated. In the absence of observational data from the pore-scale and network models, key insights may have been difficult to obtain. Also, the ability to compare simulations from distinctly different conceptual models helped separate basic principles from artifacts associated with the simulation process. It is expected that use of computer simulation as virtual laboratory will be met with scepticism by many in the experimental sciences. However, when the computer simulation is used in conjunction with physical experiments as link to reality, the scientist obtains an unparalleled view of microscopic processes.

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