

The Secret to Successful Solute–Transport Modeling

by Leonard F. Konikow

Abstract

Modeling subsurface solute transport is difficult—more so than modeling heads and flows. The classical governing equation does not always adequately represent what we see at the field scale. In such cases, commonly used numerical models are solving the wrong equation. Also, the transport equation is hyperbolic where advection is dominant, and parabolic where hydrodynamic dispersion is dominant. No single numerical method works well for all conditions, and for any given complex field problem, where seepage velocity is highly variable, no one method will be optimal everywhere. Although we normally expect a numerically accurate solution to the governing groundwater-flow equation, errors in concentrations from numerical dispersion and/or oscillations may be large in some cases. The accuracy and efficiency of the numerical solution to the solute-transport equation are more sensitive to the numerical method chosen than for typical groundwater-flow problems. However, numerical errors can be kept within acceptable limits if sufficient computational effort is expended. But impractically long simulation times may promote a tendency to ignore or accept numerical errors. One approach to effective solute-transport modeling is to keep the model relatively simple and use it to test and improve conceptual understanding of the system and the problem at hand. It should not be expected that all concentrations observed in the field can be reproduced. Given a knowledgeable analyst, a reasonable description of a hydrogeologic framework, and the availability of solute-concentration data, the secret to successful solute-transport modeling may simply be to lower expectations.

Introduction

The practice of numerical modeling of groundwater flow and transport processes is now in its fifth decade. During this time, the availability, cost, and computational power of computers have greatly evolved and improved, and so have the art, science, and practice of groundwater modeling. During the first decade or two of practice, applications were dominated by those who developed or modified the computer source code for each model application to a specific site, area, or aquifer. Today, application of groundwater models is dominated by the use of widely accepted, generic, public-domain codes, such as MODFLOW (McDonald and Harbaugh 1988; Harbaugh

et al. 2000; Harbaugh 2005) and MT3DMS (Zheng and Wang 1999), coupled with the use of pre- and postprocessing software (Graphical User Interfaces or GUIs) to facilitate model application and analysis of results for complex three-dimensional (3D) problems. In fact, GUIs make it easier for some with little experience in analyzing groundwater transport problems to apply a solute-transport model to a field problem.

Modeling groundwater flow (and head distributions) is much more common than simulating solute transport (and concentration distributions) in groundwater. Experience indicates that the latter is more difficult and less successful than the former, although “success” certainly depends on the context of the problem. One reason is that solute-transport modeling for a specific area requires an accurate model of the flow field, so the transport model must be linked to and added on to a flow model. But a flow model does not require a solute-transport model.

U.S. Geological Survey, Reston, VA 20192; lkonikow@usgs.gov
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So groundwater-flow models are inherently simpler, in that they simulate just one primary process, whereas solute-transport models are inherently more complex, in that they require simulation of both flow and transport processes. There are several additional fundamental differences between flow and transport modeling that cause the latter to be more difficult, more complex, and generally less reliable.

Because many site contamination problems involve the design of remediation plans for reactive chemicals, many recent practical applications of solute-transport models involve the use of reactive transport models. However, the consideration of geochemical and microbiological reactions adds much complexity to the numerical and data aspects of the simulations relative to simulating the advection and dispersion of a single nonreactive species, which is basic to understanding and modeling reactive transport. Similarly, if a contaminant is immiscible or partly miscible with water, the physics of the problem becomes much more complex.

The purpose of this paper is to review deterministic solute-transport modeling and discuss some fundamental differences between flow and transport modeling, the development and application of transport models to practical problems, and whether the basic processes and mathematical aspects of advective-dispersive transport of nonreactive species are adequately understood. (For brevity, we do not review stochastic approaches, which are less commonly applied by practicing groundwater scientists and engineers.) The focus is on nonreactive solutes that are miscible with water—while recognizing that many serious contamination problems involve transport of reactive species and/or fluids that may not be fully miscible with water. The issues are characterized in a framework of four major areas of concern. The first is conceptualization (how well does the classical governing equation describe the processes controlling solute transport?). The second is numerical accuracy (how well do standard numerical methods solve the governing equation?). The third issue focuses on parameter estimation (how accurately and precisely can we measure and describe the boundary conditions and properties that affect solute transport?). The final issue is a broader one that includes general concerns about model complexity and predictive accuracy.

Conceptual Issues

The theoretical basis and underpinning of a solute-transport model is the governing equation that is solved by the model. It is reasonable to ask, how well does the classical solute-transport equation describe the processes and solute behavior observed in complex field environments? As transport is dependent on flow, so the solute-transport equation is intimately linked with the groundwater-flow equation.

A general form of the equation describing the transient flow of a compressible, single-phase fluid of uniform density in a heterogeneous anisotropic aquifer

may be derived by combining Darcy's law with the continuity equation. A general groundwater-flow equation may thereby be written in Cartesian tensor notation as:

$$\frac{\partial}{\partial x_i} \left(K_{ij} \frac{\partial h}{\partial x_j} \right) = S_s \frac{\partial h}{\partial t} + W^* \quad (1)$$

where K_{ij} is hydraulic conductivity (a second-order tensor) [$L T^{-1}$]; h is hydraulic head [L]; S_s is specific storage [L^{-1}]; t is time [T]; W^* is volumetric flux per unit volume [T^{-1}]; and x_i are the Cartesian coordinates [L]. The equation can generally be applied if Darcy's law applies (and gradients of hydraulic head are the only driving force) and can be modified appropriately if fluid properties (density and viscosity) are variable.

The migration of chemicals dissolved in groundwater by advective and dispersive processes will be affected by the velocity of the flowing groundwater. Given the head distribution calculated by solving Equation 1, groundwater velocities can be calculated by dividing the specific discharge, computed using Darcy's law, by the active cross-sectional area through which flow occurs, as follows:

$$V_i = \frac{q_i}{\varepsilon} = -\frac{K_{ij}}{\varepsilon} \frac{\partial h}{\partial x_j} \quad (2)$$

where V_i is the seepage velocity (also called average linear velocity or average interstitial velocity) [$L T^{-1}$]; q_i is specific discharge [$L T^{-1}$]; and ε is the effective porosity of the porous medium.

An equation describing the transport and dispersion of a nonreactive dissolved chemical in flowing groundwater may be derived from the principle of conservation of mass (Bear 1979; Bear and Cheng 2010; Domenico and Schwartz 1998; Bredehoeft and Pinder 1973). Conservation of mass requires that the net mass of solute entering or leaving a specified volume of aquifer during a given time interval must equal the accumulation or loss of mass stored in that volume during the interval. This relation may then be expressed mathematically by considering all fluxes into and out of a representative elementary volume (REV), as described by Bear (1979) and Bear and Cheng (2010). This leads to the development of the following classical form of the advection-dispersion equation (ADE):

$$\frac{\partial(\varepsilon C)}{\partial t} = \frac{\partial}{\partial x_i} \left(\varepsilon D_{ij} \frac{\partial C}{\partial x_j} \right) - \frac{\partial}{\partial x_i} (\varepsilon C V_i) - C' W^* \quad (3)$$

where C is the concentration [$M L^{-3}$]; D_{ij} is the coefficient of hydrodynamic dispersion (a second-order tensor) [$L^2 T^{-1}$]; and C' is the concentration of the solute in the source or sink fluid [$M L^{-3}$]. Domenico and Schwartz (1998, p. 298) state, "The advection-dispersion equation is the workhorse of modeling studies in ground-water contamination."

The first term on the right side of the ADE represents the rate of change in concentration due to hydrodynamic dispersion, which includes both molecular diffusion as well as mechanical dispersion. This expression

is analogous to Fick's Law describing just a diffusive flux. This Fickian model assumes that the driving force is the concentration gradient. The relatively smooth and gradual spreading indicated by a Fickian diffusion model, however, is not always consistent with field observations and is the subject of much ongoing research and field study. The second term represents advective transport and describes the movement of solutes at the average seepage velocity of the flowing groundwater. The third term represents the effects of mixing with a source fluid that has a different concentration than the groundwater at the location of the recharge or injection. Additional terms can be added to the right side of Equation 3 to represent additional processes, such as geochemical reactions, radioactive or microbial decay/degradation, and matrix diffusion (dead-end pores or dual porosity) (e.g. Grove 1976; Domenico and Schwartz 1998; Zheng and Bennett 2002).

By assuming that the storage effects associated with transient flow are negligible and that the porosity term is constant spatially (Zheng and Bennett 2002; Goode 1992), Equation 3 can be further simplified to

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_i} \left(D_{ij} \frac{\partial C}{\partial x_j} \right) - \frac{\partial}{\partial x_i} (C V_i) - \frac{C' W^*}{\varepsilon} \quad (4)$$

Although the assumption that porosity is spatially constant may be quite reasonable and induce very little error even when it varies spatially, it tends to mask another issue of concern about porosity that is generally overlooked or ignored. Specifically, the porosity term on the left side of Equation 3 reflects the mass storage of solute within a volume of aquifer, and hence reflects the total (or bulk) porosity. The right side of Equation 3, however, reflects a porosity that is effective for the fluxes of water and solute—more a measure of mean cross-sectional area at the pore scale and interconnectedness of pores—and which will have a value less than that of the total porosity. If a single value representative of the effective porosity is used, then the solute storage capacity (and mass stored) would be underestimated; if a single value representative of the total porosity is used, then the average seepage velocity would be underestimated. Although dual porosity (matrix diffusion) models explicitly handle this, such models are typically applied to fractured rock systems and not to classical porous media.

A potentially more serious flaw in the governing ADE relates to the Fickian dispersion model in which the concentration gradient is the driving force for the spreading of solute. Bear (1979) presents a derivation of the governing partial differential equation by averaging over an REV around a point. Bear (1979, p. 232) argues that the mass flux of a solute at a point in a porous medium consists of two terms—the advective transport (solute moving with the average velocity of the water) and “an additional flux at the macroscopic scale—the dispersive flux—introduced by the process of averaging.” The latter represents the solute flux carried with the fluctuating velocity—that is, at water velocities that deviate from the mean. He further states, “This new phenomenon at the

macroscopic scale (as it does not exist at the microscopic one) represents the loss of information by the passage from one scale of description to another, larger, one.” One way to view this is that the upscaling recognizes that (1) the bulk of the dispersive flux is actually related to a variance in velocity (and advection); and (2) the actual population of velocities cannot be explicitly measured or known.

Bear (1979) then develops the classical form of the dispersion term from the statement, “As a working hypothesis, we shall assume that the dispersive flux can be expressed as a Fickian type law,” and then proceeds to develop the classical description of hydrodynamic dispersion in the governing equation. Mechanical dispersion is a function both of the intrinsic properties of the porous media (such as heterogeneities in hydraulic conductivity and porosity) and of the fluid flow. These relations are commonly expressed as:

$$D_{ij} = \alpha_{ijmn} \frac{V_m V_n}{|V|} + D_m, \quad i, j, m, n = 1, 2, 3 \quad (5)$$

where α_{ijmn} is the dispersivity of the porous medium (a fourth-order tensor) [L]; V_m and V_n are the components of the flow velocity of the fluid in the m and n directions, respectively [L T⁻¹]; D_m is the effective coefficient of molecular diffusion [L² T⁻¹]; and $|V|$ is the magnitude of the velocity vector [L T⁻¹] (Scheidegger 1961; Bear 1979; Domenico and Schwartz 1998).

From the perspective of Bear's “working hypothesis,” the Fickian model for mechanical dispersion does not have a completely rigorous mathematical derivation. Bear's assumption indicates that the spreading of solute caused by deviations in velocity from the mean can be reasonably approximated by a conceptual model in which the spreading is driven by the concentration gradient. This is the weak theoretical link because the concentration gradient, for the most part, is not the actual driving force. Does this make any practical difference? Although the Fickian model appears reasonable in some (or even many) situations, especially for relatively homogeneous media, I think that in many cases and field applications its conceptual weakness makes more of a difference than most hydrogeologists suspect.

Several key characteristics of a Fickian dispersion model are listed in Table 1. The order listed reflects a subjective assessment (and ranking from least to most) of how serious the concern is in terms of impact on the application of solute-transport models to practical field problems. First, after a solute or contaminant is introduced at a point in the porous medium, the exact solution to the equation indicates that there will be non-zero positive values of its concentration everywhere in the system. However, the values at even moderate distances may be so small as to have no practical significance. Nevertheless, the conceptual model indicates unrealistically that some molecules travel great distances in very short times.

A second issue is upstream dispersion (although this derives from the sixth issue listed). If groundwater flow

Table 1
Selected Characteristics of Fickian Model for Hydrodynamic Dispersion

1. $C > 0$ for $t > 0$ at all locations
2. Upstream dispersion
3. Irreversible spreading
4. No scale effects (macrodispersion)
5. Gaussian normal distribution
6. Dispersive flux proportional to concentration gradient

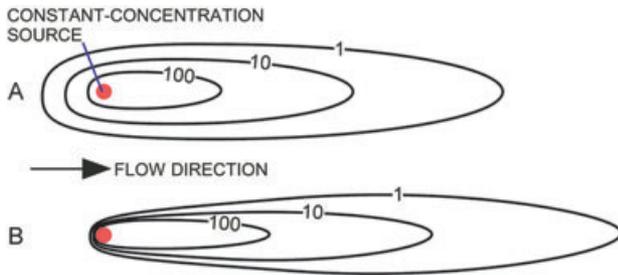


Figure 1. Schematic representation of concentration contours for a plume emanating from a hypothetical constant-concentration source in a steady uniform flow field under (A) conventional conceptualization of advection and hydrodynamic dispersion, and (B) conceptualization that mechanical dispersion is driven by variability in advective velocity with local mixing.

is toward a contaminant source (or peak concentration), then the hydraulic gradient will be in the opposite direction than the concentration gradient, which will drive a dispersive flux in opposition to the advective flux and the plume will spread in an upstream direction (Figure 1A). However, if spreading is actually related to deviations in velocity about the mean, both in magnitude and in direction at multiple scales, and all velocity vectors are in the same general (downstream) direction, then all solute will migrate downstream, though at a range in rates and with transverse spreading (Figure 1B). The velocity

distribution will yield an apparent dispersion in the downstream direction due to advective variability (analogous to one that would be driven by a concentration gradient) and local pore-scale mixing, but would not yield any upstream migration or spreading due to mechanical dispersion—contrary to what is predicted by the Fickian dispersion model—because there are no macroscale fluid velocity components in the upstream direction.

Third, the Fickian model also implies irreversible spreading. That is, if a plume evolves from a point source in a unidirectional flow field, one would expect longitudinal spreading in the direction of flow and transverse spreading perpendicular to flow. If, at some later time, the flow field is perfectly reversed, the transport direction would be in the opposite direction, but the plume should not then shrink back toward the original source; instead, it should continue to disperse relative to the new flow direction. However, consider a layered system in which each layer has a different hydraulic conductivity (Figure 2). After a solute is introduced with a step increase in concentration at a fluid source, it migrates and spreads in response to velocities that can vary substantially between layers. This would lead to a relatively small amount of local dispersion within each layer, but a much greater macrodispersion because of substantial differences in advective transport distances between layers. Mercado (1967), Matheron and de Marsily (1980), and Güven et al. (1984), among others, examined this type of stratified system and concluded that for flow and dispersion parallel to the bedding, and with little to no transverse dispersion, Fickian behavior will require very large travel distances or not occur at all and the classical ADE does not apply.

If observations of the details of the concentration distribution were limited and only available from relatively long or fully penetrating wells that sampled and mixed water from the entire thickness of the aquifer (such as well “a” in Figure 2), it would appear that the system had a relatively high dispersivity. On the other hand, breakthrough curves from point samplers would reflect smaller values of dispersivity. Ignorance of the detailed

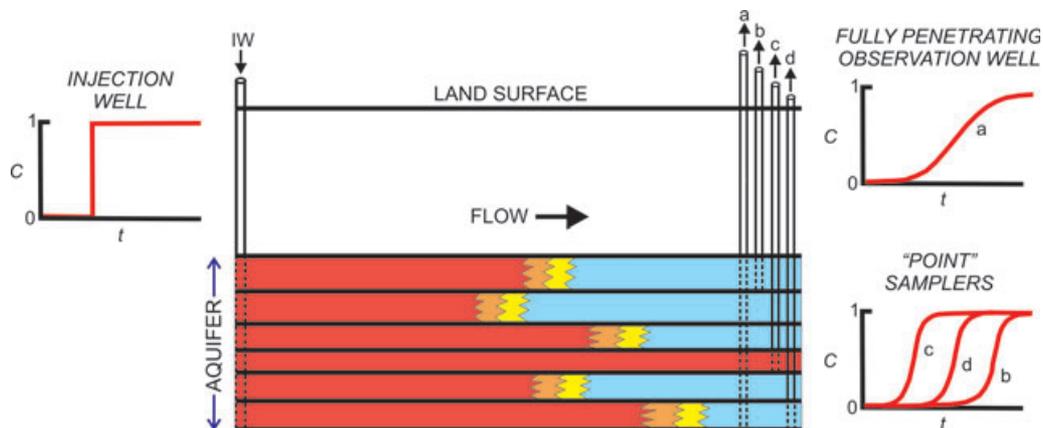


Figure 2. Illustration of solute migration in a hypothetical idealized layered aquifer system having substantial differences in average velocity among individual layers within the aquifer.

distribution of velocities (and large-scale variations in advective transport) is compensated by increasing dispersion in the framework of a Fickian model. If pumping on the left side were used to reverse the flow field, then after some time the vertically averaged solute distribution would appear to partly reverse itself—seemingly (but not actually) contradicting the principle of irreversibility. The small spreading within each layer would continue to grow after flow reverses, but the bulk of the initial spreading by differential advection seen in Figure 2 would be reversed by advection.

One might counter that this reversibility is an artifact of being ignorant of important internal structure within the aquifer and not explicitly accounting for the individual layers. That is true, and the issue of reversibility is not itself a major concern, but the point is that the governing equation is derived by averaging up from the pore scale to the microscopic scale, and our numerical models upscale further on the same principle to a macroscale and field scale. At whatever scale we operate, some internal structure will be present at the smaller scale, and we will likely be ignorant of most of it and of the true velocity distribution. Field observations of “non-Fickian” solute behavior are consistent with the notion that the Fickian process inherent in the assumed governing equation is not actually the process controlling or driving most of the observed spreading.

Stochastic theory helps resolve some of these issues. Frind et al. (1987) examined a conceptually similar but more generalized case of microstratification based on the tracer test site at Borden, Canada (Sudicky, 1986). Their results show convergence of effective dispersivity to the theoretical macrodispersivity value over a travel distance of about 50 correlation lengths of hydraulic conductivity. The transfer of mass between neighboring streamtubes of different velocity, resulting from transverse dispersion and molecular diffusion, would reduce or eliminate any apparent reversibility.

Next, the dispersivity parameter is conceived as a measurable physical parameter that primarily reflects the nature of the heterogeneity in the system. There is nothing in the classical conceptualization of the dispersivity parameter indicating that its value should be scale-dependent, yet that is what field observations tend to indicate and some stochastic transport theories allow. For example, most reported values of the longitudinal dispersivity component (α_L) fall in a range from 0.01 to 1.0 times the scale of the measurement (Anderson 1984; Gelhar et al. 1992). The apparent scale dependence of dispersivity values may simply arise from the conceptualization deficiency of mechanical dispersion being driven by the concentration gradient rather than by the velocity variability. Nevertheless, we usually estimate the value of dispersivity in a model by calibration (history matching) to observed concentrations in a tracer test or historical plume, and then expect that value to yield accurate predictions when the model is used to simulate transport over longer times and greater distances than previously experienced.

Another related issue is that the Fickian dispersion model is consistent with a Gaussian normal distribution in the spreading of solute about the center of mass (unless otherwise influenced by boundary conditions). But observed breakthrough curves across scales from lab experiments to field studies often show anomalous early arrivals and long tails. The expected normal distribution in spreading implies that the deviations in velocity about the mean across a face of the REV are normally distributed (or nearly so, or sufficient time has elapsed to damp out any extremes). This might occur in an ideally homogeneous porous medium, or otherwise if variable hydraulic properties are themselves randomly and normally distributed. But there is much hydrogeological evidence that heterogeneity is more complex and occurs across a range of scales, including microscopic and extending to very large field scales (termed “megascopic” by Bear and Cheng [2010]). Neuman and Di Federico (2003) state, “Geology is ubiquitously heterogeneous, exhibiting both discrete and continuous spatial variations on a multiplicity of scales.”

Zinn and Harvey (2003) elucidate the importance of considering the connectedness of high-conductivity materials in porous media, which can yield pathways for enhanced solute transport and overall non-Fickian transport behavior. Fractures in consolidated rock aquifers would represent one extreme example of highly connected, high-conductivity regions or zones, but similar connectedness can form by stratigraphic controls in unconsolidated sediments deposited in a variety of geologic settings. In fact, the layers represented in Figure 2 clearly have a high connectedness and spatial persistence of their hydraulic properties in the horizontal direction. Such patterns of persistence in heterogeneity are not considered in the REV-based derivation of the governing equation and would not yield a smooth and normal distribution of velocities across a face of the REV analogous to a Fickian dispersive flux. One might wonder, “How representative is the Representative Elementary Volume?”

Finally, perhaps most basic of the issues, and one which threads through the previous five, is that the Fickian model relates the dispersive flux to the concentration gradient. This is reasonable for molecular diffusion, but that is typically just a very small component of hydrodynamic dispersion. However, it may not be reasonable for mechanical dispersion in that much of the apparent dispersive flux arises from unknown variability in advective transport. The Fickian model (with or without macrodispersivity) remains a “working hypothesis” that often works well, but sometimes does not.

A better governing equation is needed, and some promising research is directed toward that goal. Multi-rate mass-transfer models account for solute flux between mobile and immobile porewater by adding additional terms to the classical transport equation, which can help explain some of the observed non-Fickian tailing phenomena (e.g., Haggerty and Gorelick 1995). Formulating the transport equation in terms of fractional derivatives can account for non-Fickian long-tailed breakthrough curves

(e.g., Benson et al. 2000; Baeumer et al. 2001). The continuous time random walk (CTRW) approach essentially accounts for non-Gaussian distributions and persistence in hydraulic properties and velocity, enabling CTRW to simulate non-Fickian transport behavior (e.g., Berkowitz et al. 2006). Cvetkovic and Haggerty (2002) develop a combined framework of CTRW with multiple-rate exchange that can simulate anomalous transport behavior in heterogeneous media. These alternative conceptual approaches, however, impose additional data requirements for estimating new or different parameters that may not be hydrologically intuitive or readily measurable.

Numerical Issues

Regardless of any flaws or weaknesses in the conceptualization of the classical solute-transport equation, for now it represents the widely accepted state of the art in practical transport modeling. Thus, it is appropriate to ask, how accurately can we solve this equation?

The partial differential equations describing groundwater flow and transport can be solved mathematically using either analytical solutions or numerical solutions. The advantages of an analytical solution, when it is possible to apply one, are that it provides an exact solution to the governing equation and is often relatively simple and efficient to evaluate. But obtaining the exact analytical solution to the partial differential equation requires that the properties and boundaries of the flow and transport system be highly and perhaps unrealistically idealized. For most field problems, the mathematical benefits of obtaining an exact analytical solution are probably outweighed by the errors introduced by the simplifying assumptions for the complex field environment that are required to apply the analytical model.

The solute-transport equation is, in general, more difficult to solve numerically than the groundwater-flow equation, largely because the mathematical properties of the transport equation vary depending upon which terms in the equation are dominant in a particular situation. When solute transport is dominated by advection, as is common in many field problems, then Equation 3 approximates a hyperbolic type of equation. But if a system is dominated by dispersive fluxes, such as might occur where fluid velocities are relatively low and aquifer dispersivities are relatively high, then Equation 3 becomes more parabolic in nature (similar to the transient groundwater-flow equation).

The numerical methods that work best for parabolic partial differential equations are not best for solving hyperbolic equations, and vice versa. Thus, no one numerical method or simulation model will be ideal for the entire spectrum of groundwater-transport problems likely to be encountered in the field. Further compounding this difficulty is the fact that in a complex field environment, the seepage velocity of groundwater is highly variable, even if aquifer properties are relatively homogeneous (because of the effects of complex boundary conditions). Thus, in low-permeability zones or near stagnation points, the velocity

may be close to zero and the transport processes will be dominated by diffusion; in high-permeability zones or near stress points (such as pumping wells), the velocity may be several meters per day and the transport processes will be advection-dominated. Furthermore, in a numerical model, local advective dominance is also affected by grid spacing and dispersivity, as reflected in the grid Peclet number (e.g., Zheng and Bennett 2002). The consequence is that for the same system, the governing equation may be more hyperbolic in one area (or at one time) and more parabolic in nature in another area (or at another time). If that is the case, no matter which numerical method is chosen as the basis for a simulation model, it will not be ideal or optimal over the entire domain of the problem, and numerical errors (in the form of numerical dispersion or oscillations) might be introduced somewhere in the solution (e.g., Bredehoeft 1971; Zheng and Bennett 2002). These numerical errors and issues typically are not so large as to impinge on the overall value and usefulness of the model, but one needs to recognize these inherent difficulties, strive to minimize and control the numerical errors, and not confuse numerical precision with predictive accuracy. These numerical issues have also led to numerous attempts to develop alternative operator splitting numerical methods that will work better for solving the transport equation than standard numerical methods (such as method of characteristics [MOC] [e.g., Konikow and Bredehoeft 1978], random walk [e.g., La Bolle et al. 1996], and Eulerian-Lagrangian Localized Adjoint Method [ELLAM] [e.g., Celia et al. 1990]).

Numerical dispersion can be controlled by reducing the grid spacing, decreasing time-step size, or adjusting the formulation of the difference equations. Unfortunately, many approaches that eliminate or minimize numerical dispersion introduce oscillatory behavior, causing overshoot behind a moving front and undershoot ahead of the front. Undershoot can result in the calculation of negative concentrations, which are obviously unrealistic. However, overshoot can introduce errors of equal magnitude that may go unnoticed because the value is positive in sign (although greater than the source concentration, so still unrealistic). Oscillations generally do not introduce any mass-balance errors (unless negative values are artificially reset to zero), and they often dampen out over time. Many approaches to reducing numerical errors require increased computational times, so tradeoffs between accuracy and efficiency may have to be assessed.

Numerical solution of the groundwater-flow equation usually is relatively efficient. Solving the transport equation for the same simulation time as the flow equation, however, may require substantially greater computational time. In practical terms, a simulation of solute transport over a period of years to decades using a fine discretization for a large region may require many hours or days to complete on a modern personal computer.

In solving the transport equation, classical numerical methods exhibit the proportionately largest numerical errors where the relative (or dimensionless) concentrations (C/C_{\max}) are lowest. Dougherty and Bagtzoglou (1993)

show that the error-to-signal (or noise-to-signal) ratio can become quite large (more than 0.1) where the relative concentrations are less than 0.01. In contaminated groundwater systems, samples from supply wells and monitoring wells frequently indicate concentrations less than 0.01 of the source concentration, though such relative concentrations are often greater than regulatory limits, so caution in accepting and assessing simulated values is warranted.

In designing and applying a solute-transport model for a field site, one must specify appropriate boundary conditions for the problem domain. Contaminants typically enter a groundwater system with a fluid source (recharge) that contains the contaminant. Consequently a third-type solute boundary condition is a logical match—in which the source concentration is specified and associated with fluid recharge but the solute concentration in the aquifer is associated with outflow. It is common to specify a first type (specified-value or constant-concentration) boundary condition to selected nodes of the grid. When a constant-value condition is imposed in a transport model, a solute flux will be forced into or out of that cell in order to maintain the specified value of concentration, and the flux can occur by both advection and dispersion processes (e.g., Konikow et al. 1997). However, it is rare that hydrogeological and geochemical conditions in the field will be such that the concentration of a solute at a point will remain fixed in time regardless of changes in the accompanying flow field or in local concentration gradients. Batu (2010) further demonstrates that the use of a constant-concentration boundary condition for a solute subject to degradation can induce mass-balance errors or lead to an overestimation of the value of the degradation rates. Thus, except perhaps to represent concentration in a large open body of water bounding an aquifer, or for a boundary far from an area containing a solute plume of interest, it would rarely be realistic or appropriate to apply a constant-concentration boundary condition to a field problem.

A numerical experiment illustrates the possible effects of the numerical solution algorithm on the accuracy of the calculated concentrations. The example implements a variety of solution algorithms used in two widely available public-domain solute-transport models (MT3DMS and MODFLOW-GWT [Konikow et al. 1996]) and compares results obtained after applying them to a hypothetical contamination problem for a nonreactive solute species.

The test problem represents an analog based on, and greatly simplified from, the groundwater contamination problem at the Rocky Mountain Arsenal, Colorado (Konikow 1977). The aquifer is a thin, gently sloping, alluvial system with moderate hydraulic conductivity. The source of contamination ($C' = 1000$ mg/L) is an unlined disposal pond, represented in the model as two adjacent injection wells. A freshwater reservoir (lake) is located on the north boundary of the model, and a river is located along the south boundary of the model. It is assumed that the aquifer receives no recharge from precipitation and has a uniform hydraulic conductivity ($K = 1 \times 10^{-4}$ m/s), an effective porosity that varies spatially in an uncorrelated

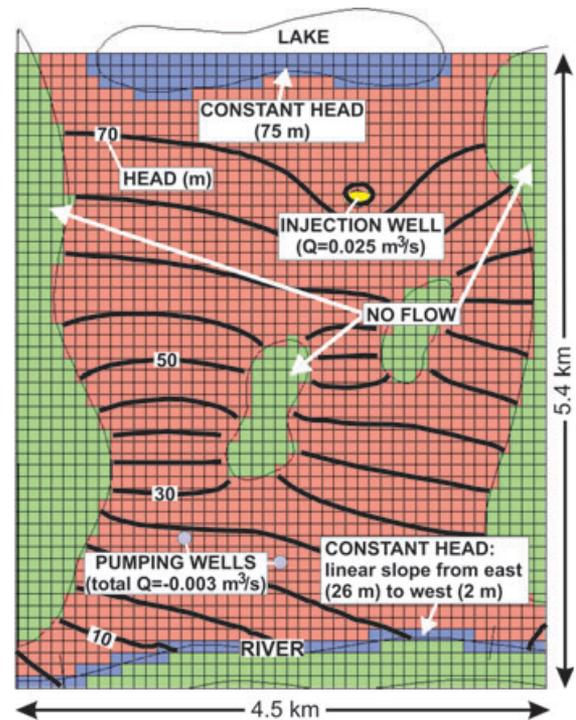


Figure 3. Boundary conditions, finite-difference grid, and calculated steady-state heads for solute-transport test problem.

random manner about a mean value of 0.20, and a steady-state two-dimensional flow field.

The boundary conditions produce groundwater flow that is generally from north to south, influenced by irregular lateral and two internal impermeable zones as well as the river acting as a sloping constant-head boundary on the southern edge of the model domain (Figure 3). Grid cells are 100 m on a side. No analytical (or “true”) solution is available for this problem, although it is presumed that the documented numerical methods will converge to the true solution as grid size and time-step size are reduced. It is not the goal of this numerical experiment to assess which model is better or best in any sense, in part because the relative strength of one method over another can change greatly depending on the characteristics of the test problem. Instead, the goal is to demonstrate possible variability in answers as affected by the choice of generic model and numerical algorithm while using typical grid spacing for the scale of the problem.

This test problem was simulated for 20 years using various solution algorithms available in both MODFLOW-GWT (Konikow et al. 1996; Heberton et al. 2000) and MT3DMS (Zheng and Wang 1999), including two finite-difference algorithms (finite-difference solution, FD and total-variation-diminishing finite-difference method, TVD) and seven different Eulerian-Lagrangian methods (ELLAM and six varieties of the MOC). Table 2 compares the results for these simulations for several measures of accuracy and efficiency. Note that the indicated run times for the MT3DMS simulations do not include the small time required to solve the groundwater-flow

Table 2

Comparison of Several Measures of Accuracy and Efficiency for Simulating the Test Problem for 20 Years Using Various Solution Algorithms Available in Public-Domain Solute-Transport Models

	MODFLOW-GWT				MT3DMS				
	MOC	MOCWT ¹	MOCIMP	ELLAM	MOC	MMOC	HMOC	FD	TVD
Transport time steps	190	99	99	99	194	194	194	433	433
Run time (s) ²	15.5	5.7	8.7	8.5	3.0	2.5	3.0	2.9	5.1
Maximum concentration (mg/L)	1009	1001	1026	1342	999	999	999	986	1053
Minimum concentration (mg/L)	-3.0	-4.0	-38	-88	0	0	0	0	-9
Mass-balance error (%)	2.3	7×10^{-6}	0.6	4×10^{-5}	2.8	5.7	3.3	2×10^{-5}	1×10^{-4}

Solution algorithms: MOCIMP = method of characteristics with implicit finite-difference solution for dispersive flux; MMOC = modified method of characteristics; HMOC = hybrid method of characteristics.

¹Spatially varying initial distribution of particles, ranging from 25 to 4 per cell, decreasing in number with distance from plume.

²MT3DMS run times are for transport only, and do not include time to solve flow equation using MODFLOW.

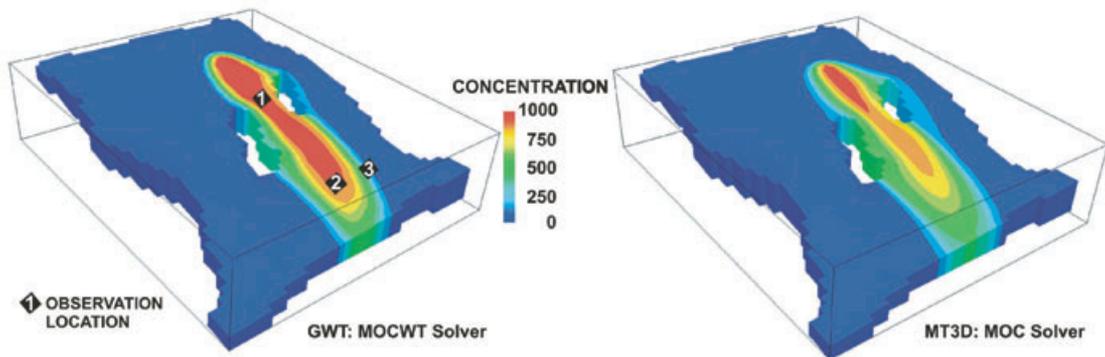


Figure 4. Calculated concentration distributions after simulating 20 years of advection and dispersion with two of the available solution algorithms.

equation using MODFLOW-2000 (approximately 0.3 s), whereas it is included in the run time for the MODFLOW-GWT simulations. Several methods produced noticeable undershoot and overshoot, especially the ELLAM results. The MOC methods (except for the weighted particle method) had notable mass-balance errors. The computational times varied by about a factor of six. Note that the relative characteristics of each algorithm listed in Table 2 are representative of the properties and characteristics of this particular test problem only.

Perhaps more importantly, the computed concentrations also can vary substantially depending on the numerical solution scheme. Figure 4 compares the computed plumes after 20 years using two different algorithms. Although the direction of plume migration is identical in all simulations, the solute distributions are not. In Figure 4, the plume computed using the method of characteristics with volume-weighted particles (MOCWT) algorithm shows much higher concentrations downgradient than does the MOC solver of MT3DMS. The variation in the results is also reflected in the range of arrival times and breakthrough curves at selected observation points (Figure 5). For example, if the detection limit or

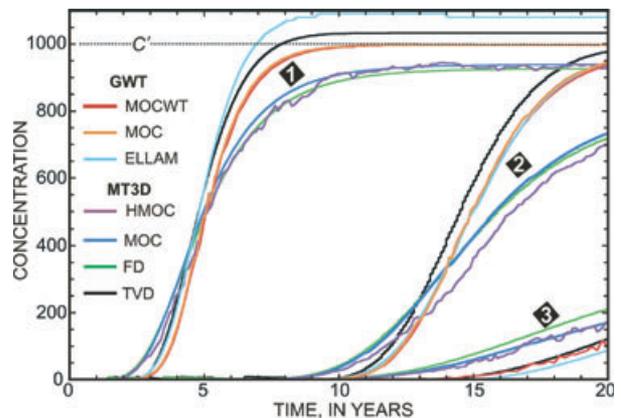


Figure 5. Comparison of simulated breakthrough curves at three observation points (locations shown in Figure 4). Source concentration (C') is 1000.

action level were at 1% of the source concentration (i.e., $C = 10$), then the first arrival at observation point 2 might have been predicted to occur between about 8.5 and 11 years, depending just on the solution algorithm (and not considering any uncertainties or errors in the model

parameters and boundary conditions). At this same observation location, the computed concentration after 20 years ranged from about 700 to 980. The differences appear large enough to be of concern, and they arise solely from the choice of numerical solution algorithm. In a regulatory or litigation arena, these differences might actually make a difference.

Some methods yield breakthrough curves at location 1 that converge to values above or below the known limit of 1000, reflecting some loss of accuracy. The several MOC solutions exhibit some jumpiness in their breakthrough curves, in which the deviations about their best-fit trend lines reflect some loss of precision. The numerical accuracy and computational efficiency of calculating concentrations in flowing groundwater are notably lower than for calculating heads. These numerical issues indicate that we need better numerical methods to solve the governing solute-transport equation.

Parameter Estimation

Regardless of weaknesses in the governing equation and our ability to solve it numerically, a number of public-domain advection-dispersion codes are widely available and commonly used for simulating solute transport. Thus, it is next appropriate to ask, how reliable and accurate will a field application of a solute-transport model be? Not only is the solute-transport modeler faced with uncertainties and errors in estimating values for the parameters that represent the coefficients in the governing ADE, but there is widespread uncertainty about what these parameters actually represent and how (or if) they can be meaningfully measured.

One critical parameter is the effective porosity (sometimes called kinematic porosity or advective porosity), which directly affects the seepage velocity, thereby affecting both mechanical dispersion and advective transport. From the perspective of Equation 2, one can view the effective porosity as simply the number by which you divide the specific discharge to obtain the average linear velocity. Interestingly, effective porosity is not in the groundwater-flow equation, so calibrating a flow model in itself may yield no information about the range of values of the effective porosity.

The total porosity represents a volumetric fraction available for fluid storage—specifically, the ratio of the volume of interstices (or voids) to the total volume of the rock or soil sample (Lohman et al. 1972). The effective porosity, on the other hand, refers to the amount of interconnected pore space available for fluid transmission relative to the total volume (Lohman et al. 1972). The degree to which effective porosity is less than total porosity will vary greatly and depend on a number of geological characteristics of the material. Domenico and Schwartz (1998, p. 14) note that “effective porosity can be over one order of magnitude smaller than total porosity, with the greatest difference occurring for fractured rocks.” The difference between the two reflects the difference between mobile and immobile water content. Immobile water may

reside stagnantly in dead-end pore space or may include slow flowing parts; the transition may be gradual and the distinction between mobile and immobile water not as sharp as the terms imply (Nimmo 2004). Regardless, the mobile water in interconnected pore spaces governs the average seepage velocity, but the immobile water is available for storage of solute—with transfer of solute between mobile and immobile water governed mostly by diffusion. If the transfer rate is relatively slow, downgradient breakthrough will exhibit tailing not consistent with a single-porosity Fickian model. The governing equation has often been modified to include terms representing this dual-porosity process, which in fractured rocks may dominantly reflect the diffusive transfer of solute between fast pathways through fractures and slow pathways in the adjacent rock matrix. Application of a solute-transport model based on the conventional advective-dispersive equation (Equation 3 or 4) to a system with a substantial fraction of relatively immobile water may represent an undue conceptual simplification that leads to substantial predictive errors, even if an observed solute distribution can be closely matched during model calibration.

Because the total porosity is a volumetric fraction, it should have no directional properties. However, the pore interconnectedness (and effective cross-sectional area for flow of mobile water) is not simply a volumetric measure, so it is possible the effective porosity can vary with direction of flow within a given volume. Neuman (2005) states that field tracer tests indicate that effective porosity may exhibit directional dependence and that this parameter is a second-rank tensor (similar to permeability). Determining such directional dependence is problematic, however, and none of the widely available solute-transport models allow directional dependence of effective porosity.

Another critical transport parameter is dispersivity. The dispersivity tensor for an isotropic porous medium can be defined by two constants—the longitudinal dispersivity of the medium, α_L , and the transverse dispersivity of the medium, α_T (Scheidegger 1961). However, in anisotropic media the number of independent dispersivity values increases (Bear and Cheng 2010). For example, in transversely isotropic porous media under nonuniform flow conditions (consistent with typical MODFLOW models where $K_h \neq K_z$), six independent dispersivities are needed. Most documented applications of transport models to groundwater problems have been based on the simpler two-value formulation for isotropic media, even for cases in which the hydraulic conductivity is assumed to be anisotropic, despite the conceptual inconsistency. SUTRA (Voss and Provost 2002) incorporates an additional level of sophistication by defining two transverse dispersivities and allowing those and α_L to vary with flow direction. Sand-box experiments (Silliman et al. 1987) on samples of anisotropic media indicated that α_L in the high- K direction can differ by as much as a factor of ten from the α_L in the low- K direction. Directionally dependent dispersivity is a useful generalization of the classical dispersion paradigm. For example, it may be expected that flow parallel to layering in a layered medium and flow

perpendicular to the layering do not necessarily have the same longitudinal dispersivity. But both MT3DMS and MODFLOW-GWT are more restrictive and only allow the specification of two unique values for α_T —one for dispersive transport in the vertical direction and one for transverse dispersive transport in the horizontal direction—following the *ad hoc* model of Burnett and Frind (1987). If advective transport of a contaminant is mostly horizontal in part of the flow system and mostly vertical in other parts of the flow system through typical stratified geological media, then use of an isotropic α_L can lead to erroneous dispersive fluxes somewhere in the system. Note that such errors would not be reflected in the numerical mass balance for the solute, as would be the case for other conceptual weaknesses in dispersion theory (that is, a perfect solute mass balance in a numerical model does not prove that you have the right answer). Any conceptual errors arising from simplification of the mathematically rigorous expansion of the dispersion term cannot be reduced by adjusting the grid spacing or time-step size.

Although conventional theory holds that α_L is an intrinsic property of the aquifer, it is found in practice to be dependent on and proportional to the scale of the measurement. Most reported values of α_L fall in a range from 0.01 to 1.0 times the scale of the measurement, although the ratio of α_L to scale of measurement tends to decrease at larger scales (Anderson 1984; Gelhar et al. 1992). Field-scale dispersion (macrodispersion) results primarily from large-scale spatial variations in hydraulic properties (and hence in velocity). Yet one can often adequately calibrate a groundwater-flow model to observed heads using a relatively coarse grid and relatively uniform hydraulic properties that do not represent heterogeneities that affect transport. If that is the framework for the transport model, calibration will likely lead to erroneously large values of dispersivity (e.g., Davis 1986). Similarly, representing a transient flow field by a mean steady-state flow field, as is commonly done, inherently ignores some of the variability in velocity and must be compensated for by using increased values of dispersivity (primarily transverse dispersivity) (Goode and Konikow 1990).

Perhaps the most important parameter controlling transport is hydraulic conductivity, through its control on velocity (Equation 2). Hydraulic conductivity can vary greatly over short distances, and heterogeneity can exhibit large spatial correlations, persistence, and connectedness. Overall, the more accurately and precisely a simulation model represents the actual distribution of K , the better it can simulate the “true” velocity distribution in space and time. Consequently, more solute spreading will be represented by differential advection, so uncertainty in estimating dispersivity and conceptual flaws in the mathematical representation of the dispersion process will matter less. Thus, much of the research in solute transport in recent years has focused on how to better describe (either explicitly or statistically) the heterogeneities in aquifer properties that control the velocity of groundwater flow and its variability.



Figure 6. Array of wells and multilevel samplers used to conduct groundwater tracer experiments at the Cape Cod, Massachusetts, research site (from LeBlanc 2006).

One research approach has been through comprehensive field “laboratories,” where test sites are comprehensively instrumented to measure heads and distributions of K and to enable detailed observations of changes in solute distribution over time for both pre-existing contaminant plumes and injected tracers in controlled experiments. One well-studied site (Figure 6) is at Cape Cod, MA, in an unconfined aquifer composed of permeable, stratified, unconsolidated, sand and gravel outwash (LeBlanc 2006; LeBlanc et al. 1991). The site has a mean K of about 0.1 cm/s and a variance of $\text{Ln}(K)$ of about 0.2 (Hess et al. 1992), so the site can be characterized as mildly heterogeneous. Comprehensively monitored tracer tests at this site and applications of numerical solute-transport models indicate that conventional numerical models based on the Fickian ADE can adequately simulate transport of non-reactive and reactive solute species in this aquifer (e.g., Garabedian et al. 1991; Zhang et al. 1998; and Parkhurst et al. 2003).

Another well-studied tracer-test site is the Macro-Dispersion Experiment (MADE) site in Columbus, Mississippi (Boggs et al. 1992). In contrast with the Cape Cod site, the aquifer at the MADE site consists of highly heterogeneous fluvial sediments (variance of $\text{Ln}(K)$ is about 4.5) (Rehfeldt et al. 1992; Molz et al. 2006). Analysis of a 20-month natural-gradient tracer test indicated “dramatically non-Gaussian behavior” (Adams and Gelhar 1992). Molz et al. (2006) report that “numerous theoretical, laboratory, and field studies have demonstrated that the classical advection-dispersion model failed to capture the key characteristics of solute-transport behavior at the MADE site.” They further state that a dual-domain model led to conceptually simple, successful simulations with “a significantly better match to the observed plumes for all three natural gradient tracer tests.”

Somewhat less heterogeneous than the MADE site is the shallow, unconfined, glacial aquifer in North Bay, Ontario, where the spatial variability of hydraulic conductivity was studied in great detail and characterized geostatistically (Sudicky et al. 2010). The mean hydraulic conductivity was about 3.5×10^{-3} cm/s (much less than at the Cape Cod site) and the variance of $\text{Ln}(K)$ was about 1.8 (less than half that at the MADE site). The

effective hydraulic conductivity and macrodispersivity tensors were calculated using the 3D stochastic theory of Gelhar and Axness (1983). Subsequent application of the HydroGeoSphere model (Therrien et al. 2005)—a 3D, variably-saturated flow, advection-dispersion transport model—reasonably reproduced “the extent and migration rates of the observed contaminant plume that was monitored using a network of multilevel samplers over a period of about 5 years” (Sudicky et al. 2010).

The overarching lessons from the several comprehensively sampled and monitored field “laboratories” include that geological heterogeneity is ever present, that heterogeneity strongly influences plume evolution, and that detailed and expensive site characterization may be required to enable the development of reliable solute-transport models. Even then, the application of classical advection-dispersion models may prove inadequate because the plume is evolving in a non-Fickian manner at the scale of the observations.

Boundary conditions required for the solution of the solute-transport equation in a complex field problem may be uncertain and difficult to define. In that sense, they can also be considered another parameter to be estimated—perhaps using automated inverse modeling tools. For example, the source term in the solute-transport equation (last term on right side of Equation 3) is the mathematically simplest part of the equation. In typical contamination problems, however, defining the source term for a solute-transport model is especially difficult because the historical timing, location, and strength of releases of solute mass into an aquifer system are rarely known or accurately reported (and, in fact, are commonly the very point of contention in litigation).

We need better methods to measure parameter values and their spatial variability and connectedness. Use of geostatistical methods of estimation and hydrogeophysical methods of measurement show great promise in the determination of hydraulic properties controlling advective transport.

The dispersivity parameters remain problematic, and the costs of field measurements (using tracer tests) are high and may yield estimates that are indicative of the scale and duration of the tracer test but that have little value for predictive purposes in modeling future responses over longer times and distances. In advection-dominated systems, such as site studies dominated by pumping wells, dispersion may not be detectable and the selected values of dispersivity may not matter much. In general, it is a good idea to use preliminary models to assess the sensitivity of the system to uncertainty in dispersivity before investing much effort in trying to measure it.

Model Complexity

As computer power has increased over the years, there seems to be a parallel trend toward developing and applying more complex models to field problems. From the simplest applications of groundwater solute-transport models some 40 years ago, various studies have added

variably-saturated flow, variable-density flow, multiphase flow and transport, reactive transport, microbial processes, ecological problems, multiscale simulations (e.g., using telescopic or adaptive meshes), finer 3D discretization, detailed deterministic or geostatistical representations of heterogeneity, dual-domain models, and consideration of stochastic processes. These add additional levels of sophistication, realism, and power to the simulation. But a downside is that they also impose additional requirements for data that may be difficult or expensive to obtain.

One advantage of a simpler model is that it is relatively easy to understand (Figure 7); as complexities are added, the model behavior becomes more difficult to understand and to explain to other interested parties, such as water managers. Model development at an optimal level of complexity (or simplicity) requires experience and good judgment, and must be done in light of model objectives. The objectives of the investigation should also influence the data-collection process, because time and effort should not be wasted on collection or analysis of data not relevant to the scale and objectives of the model.

Development and application of a groundwater model should be viewed as an evolutionary process. In developing a solute-transport model, one should start simple and add increasing degrees of complexity in small increments so that the effects of the added complexity (whether in processes, parameters, dimensionality, or boundary conditions) can be easily discerned. This will lead to a greater understanding of both the model and of the problem at hand. A model is by definition a simplification of a very complex reality, so an end point will never be reached where a model is a precise representation of reality—and that, of course, would be a futile goal to strive for. At some point during its evolution, a model will need to be accepted as usable and appropriate if it is to be of any value. Whether this “point” has arrived is a decision that should be made in light of (1) the objectives of the model study and the nature of the problem that needs to be solved; and (2) incremental costs of further improvements (and data collection) relative to benefits. Clement (in press) clearly poses this as, “When should we say enough is enough?”

Various modeling approaches for analyzing solute transport in groundwater are listed in Table 3 in order of increasing complexity of process. Various types of

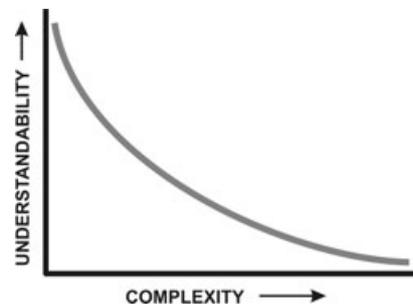


Figure 7. General hypothetical relation between model complexity and ease of understanding model behavior.

Table 3**Modeling Approaches to Solute-Transport Problems Ranked in Order of Increasing Conceptual Complexity**

Complexity Rank	Concept	Value	Example Generic Model
0	Hydrologic budget/mass balance	Mass in/mass out/mass stored	
1	Flow equation	Head/flux/direction	MODFLOW
2	Advective transport	Velocity vectors/time of travel	MODPATH
3	Advection-dispersion	Concentration distribution	MT3DMS
4A	Stochastic	Probabilities/uncertainties/nature of heterogeneity	
4B	Non-Fickian transport	Non-Gaussian distributions and breakthroughs	

lumped-parameter, black box, and mixing cell models are available. These are sometimes applied to environmental tracers used to estimate groundwater age where details of a groundwater-flow system are poorly known. They generally offer little insight into flow and transport processes except at a regional scale, and such models are generally unsuitable for small-scale site investigations. Groundwater-flow models (such as MODFLOW [Harbaugh 2005]) can yield much insight into transport directions and where solutes will migrate. Advective transport models (such as MODPATH [Pollock 1994]) go an additional step and help define both where solutes will go and how fast they will travel, but cannot predict concentrations. Given a well-calibrated flow model, advective transport models can accurately and efficiently calculate pathlines and travel times consistent with the simulated heads—generally adding substantial value for minimal additional effort. However, advective transport can be very sensitive to small errors and uncertainty in computed heads and head gradients.

The next level of complexity comes with solving the ADE, using a model such as MT3DMS (Zheng and Wang 1999). As discussed earlier, however, ADE models have some theoretical weaknesses, numerical solution errors, and practical limitations in estimating parameters. Recognition of these limitations has led to the development and application of a variety of stochastic approaches, both in terms of defining parameters and describing processes, as well as for solving equations (e.g., Dagan and Neuman 1997). For the most part, stochastic transport models, as well as other non-Fickian conceptualizations, have been limited in their use to the research community and have not yet seen widespread general application.

Some aspects of model complexity, discretization, and resolution of defining heterogeneity are illustrated with an analysis of transport from a hypothetical leaky borehole that releases contaminants into a fractured dolomite—a thin permeable unit overlying the Waste Isolation Pilot Plant (WIPP) transuranic waste repository near Carlsbad, NM (e.g., Meigs and Beauheim 2001; Altman et al. 2002).

The geostatistical characteristics of the aquifer (Table 4) provided the basis for generating one realization of a spatially correlated distribution of transmissivity

Table 4**Selected Aquifer and Model Properties for Simulation of Hypothetical Leaky Borehole in Dolomite Aquifer at the WIPP Site in New Mexico**

Property	Value
Mean transmissivity	3.4×10^{-5} m ² /s
Variance $\log_{10}(T)$	1.0
Correlation length	50 m
Thickness	4 m
Effective (fracture) porosity	0.01
Total (matrix) porosity	0.16
Dispersivities (α_L & α_T)	0.5 and 0.05 m
Regional hydraulic gradient	0.0026
Retardation factor	1.0, 12.5, and 200
Simulation time	50–10,000 yrs ¹

¹ Effective simulation time depends on assumed values for porosity and retardation factor (e.g., $t = 10,000$ yrs for $\epsilon = 0.16$ and $R_f = 12.5$; solution is identical at $t = 50$ yrs for $\epsilon = 0.01$ and $R_f = 1.0$).

on a 2-m grid spacing using the turning bands method (Zimmerman and Wilson 1990). Heads and solute concentrations were simulated in two dimensions using MODFLOW-GWT on a very fine grid ($\Delta x = \Delta y = 2$ m) over a relatively large area (1.4×3.0 km) (Figure 8A). The same system was then re-simulated on a 50-m grid spacing after upscaling transmissivity using the geometric mean transmissivity of the 625 values at the finer discretization for each coarser cell. (The boundary condition representing the leaky borehole was adjusted only to assure that an identical mass of contaminant was released into the aquifer.) The results (Figure 8B) showed that computed heads and flow directions were essentially unchanged, but that the computed plume was substantially more sensitive to the coarsening of the grid and resolution of defining heterogeneity. There was now a greater lateral spreading of the plume, particularly close to the source, and the relatively high concentrations did not migrate as far downstream as previously. To evaluate, in part, the effects of discretization and resolution of defining heterogeneity, heads for the coarser transmissivity pattern were simulated again using a 2-m grid spacing (Figure 8C).

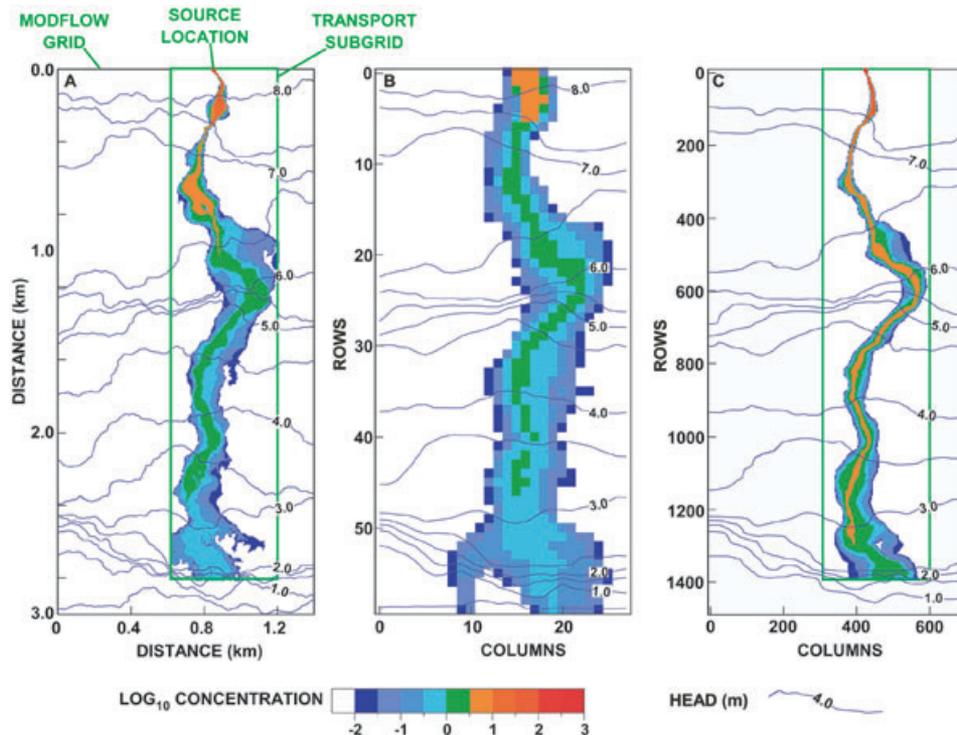


Figure 8. Effect of scale of discretization and resolution of heterogeneity on calculated heads and concentrations for case of hypothetical contaminant release from a leaky borehole in a regional aquifer: (A) grid spacing = 2 m; (B) grid spacing = 50 m; and (C) T defined on 50-m spacing from B, but numerical solution obtained using grid spacing = 2 m.

Again, the flow field and heads were insensitive to these changes, but there was a marked change in the computed plume—with the plume now much narrower and having very high concentrations migrating much further downstream, a result which can have important implications in a regulatory framework.

A similar analysis of the MADE site (Zheng and Gorelick 2003) indicated that “the relative preferential flowpaths and flow barriers resulting from decimeter-scale aquifer heterogeneity” strongly influence plume-scale solute transport and cause asymmetrical non-Gaussian solute patterns. They conclude by stating the ultimate goal is to develop a practical sound alternative to the classical ADE model.

Expectations for Transport Model Applications

In designing and applying a solute-transport model to a field problem, one should first define the questions to be asked and the purpose of the model, and develop a comprehensive conceptual model of the problem. Specify what the expectations are.

Some (if not most) of the difficulties with transport models arise from errors, inadequacies, and uncertainties in the data used to estimate parameters and to calibrate the model, and in related errors in the conceptualization of the system and problem being simulated. Groundwater-flow models are calibrated with observed head and flux data. Changes in head propagate fairly rapidly (as a pressure wave) through a porous medium, and variations between

measurement points are typically smooth and gradual. On the other hand, field measurements of concentration can change by large amounts over very small times and short distances, and solute moves slowly through a porous medium (with the speed of the water molecules), so local differences do not dampen out quickly. Concentrations in a sample are also much more sensitive to the length of the well screen (or sampling interval) than are observed heads—even within a single aquifer. Mixing within a borehole can substantially dilute the peak concentration at a sampling location, and possibly lead to overestimates of dispersivity.

Reproducing or simulating large concentration differences over small distances inherently requires the use of a finer grid than that typically needed to reproduce observed heads, and a finer grid is also important for limiting numerical errors in solving the transport equation. A large increase in the number of nodes, however, may lead to impractical computational times. Unfortunately, this tradeoff may be an incentive for some to ignore or accept numerical errors. We expect flow models to reproduce most observed heads within a few meters or less. But for transport models, we should not expect that the calculated concentrations will accurately match all variations observed in the field, or even in a single observation well. Rather, one should aim to reproduce major trends and locally averaged values.

One must be careful in specifying initial and boundary conditions. In many contamination problems, the source term (which is the mathematically simplest part

of the governing equation) is poorly known from historical records—or its definition may actually be an objective of the modeling. Inverse modeling sounds like an appealing approach. However, even with a well-calibrated flow model of a contaminated area, an inverse modeling approach to defining the transport source term may be a futile exercise because of the many complex interdependent processes affecting solute concentrations, conceptual uncertainty in the governing equation, inability to define heterogeneities, and errors in the numerical solution.

First arrivals and early detections of contaminants at relatively low concentrations are especially sensitive to numerical errors and differences among alternative solution algorithms. So one should not expect high accuracy in such calculations or predictions.

The paths and travel times of calculated plumes will be sensitive to how heterogeneity is represented in the model, and a resolution for defining heterogeneity that is adequate for a groundwater-flow model may not be adequate for a transport model (especially in the vertical direction). This is another reason that discretization scale can affect transport model calculations more than flow models—so grid size matters.

A good rule of thumb in applying solute-transport models is that the more accurately and precisely one can define the velocity field, the less one has to worry about errors and uncertainty in the dispersion process. For sites and scales beyond the strong influence of a converging flow field around a pumping well, accurately computing the velocity field will require defining hydrogeological heterogeneities. But defining heterogeneity, particularly connectedness, at relevant scales remains an expensive and challenging venture. There are practical limits to how well one can define heterogeneity and the details of the velocity field; however accurately it is defined, it can always be defined more accurately and precisely if time and funding are available to collect additional data. Good judgment and sensitivity analyses may help in balancing costs and benefits and in deciding when existing data are good enough.

Conclusions

Well-documented public-domain solute-transport models are readily available and widely used. There are a number of conceptual weaknesses and flaws in the underlying theory and classical governing equation, and the numerical solutions will inevitably contain some numerical errors. Furthermore, it is rare that sufficient reliable data will be available or obtainable to uniquely define the parameters and boundary conditions for the problem at hand. Solute-transport models should be viewed more for their value in improving the understanding of site-specific processes, hypothesis testing, feasibility assessments, and evaluating data-collection needs and priorities; less value should be placed on expectations of predictive reliability. On the basis of an improved understanding, one can presumably develop a better strategy to deal with the problem at hand.

Awareness of the conceptual weaknesses and sources of error will help the model user minimize the errors and account for them when interpreting the model results and predictions. Experience indicates that errors in the conceptual model of a flow and transport problem may be the predominant source of predictive error.

Some stochastic approaches can readily simulate both Fickian and non-Fickian transport phenomena, but stochastic theory is still evolving. Stochastic methods need to move from the realm of academic research into the domain of standard practice; this will undoubtedly occur with time.

When applying a solute-transport model to a complex field problem, one should start relatively simple and add complexities in small increments. It is typical and logical to calibrate a flow model first. It often is beneficial and cost effective to then apply an advective transport model (e.g., MODPATH) to gain insight into solute travel directions and times. For a 3D problem, the use of 3D visualization and animation tools can greatly aid the process of assessing the model and analyzing results. However, for the transport model, whether advective, advective-dispersive, or advective-dispersive-reactive, do not lock yourself into accepting all the same conceptual and numerical simplifications that were adequate and acceptable for the flow model; instead, expect that further refinement might be needed to develop a meaningful transport model.

We typically expect and achieve reasonably reliable results when developing and calibrating a groundwater-flow model. In comparison, when simulating historical concentrations or predicting future plume migration and evolution, we should not expect equivalent degrees of reliability. In that sense, the secret to successful solute-transport modeling may simply be to lower your expectations.

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