Distinguishing Advection, Dispersion, and Diffusion in Fractured Bedrock

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Abstract

Mass spreading of non-reactive tracers in fractured bedrock can be described by a combination of molecular diffusion, hydrodynamic dispersion, and heterogeneous advection. These spreading mechanisms are fundamentally different in their behavior and, therefore, must be measured separately for reliable predictions of transport to be made. To separate the influence of these mechanisms, to the extent possible, specially designed tracer experiments can be conducted. To isolate the effect of molecular diffusion, tracers of different diffusion rates can be compared. Because molecular diffusion occurs at a rate independent of fluid velocity, repeating experiments at different velocity will also highlight the influence of molecular diffusion. Distinguishing hydrodynamic dispersion and molecular diffusion is best accomplished by isolating molecular diffusion and heterogeneous advection. Heterogeneous advection is conceptualized here as spreading caused by variable transport rates in media of different hydraulic conductivity. Heterogeneous advection is distinguished from hydrodynamic dispersion, therefore, by reversing the flow field during a tracer experiment. This should result in the nullification of the influence of heterogeneous advection and leave only the influence of dispersion and diffusion on the breakthrough curve. A flow-field reversal results during a push-pull tracer experiment, for example. It is important to realize, however, that advection, dispersion, and diffusion are coupled processes so that experimental separation is never complete and is meaningful only with respect to a particular theoretical transport model.

Introduction

The spreading of a dissolved mass as it moves with ground water has been traditionally ascribed to three different mechanisms: molecular diffusion, hydrodynamic dispersion, and macrodispersion. This description of mass spreading has served well in relatively homogenous unconsolidated media because usually one mechanism dominates over the other two, depending upon the length scale of investigation. At the pore scale, for example, diffusion is the most important mechanism of mass spreading, at local scales (1-10 m) hydrodynamic dispersion is usually most significant, and at larger scales (>10-100 m) spreading is largely controlled by the variable advection through sediment facies with varying hydraulic conductivities, often called macrodispersion but termed here as heterogeneous advection. In this article, the meaning of these terms in fractured rock are explored, along with empirical approaches to separating the corresponding mechanisms using tracer experiments.

Fractured rock is set apart from heterogeneous unconsolidated media by both the magnitude and the geometry of the hydraulic conductivity distribution. Slug-test measured transmissivity in a single fractured rock formation may vary over seven orders-of-magnitude (Shapiro and Hsieh, 1998). This is a much larger range than is normally encountered even in highly heterogeneous glacial deposits. More importantly, these transmissivity variations can occur over the scale of millimeters. A fracture in granite, for example, may have a hydraulic conductivity many times greater than the adjoining rock matrix. The implication of these hydraulic conductivity contrasts is that the fluid velocity field is also extremely variable. Mass-spreading mechanisms that are important at one location, may be insignificant in the immediate vicinity. Coupling of one or more of these mechanisms may lead to unexpected transport behavior. For example, *Wood et al*. (2004) recently showed how diffusion of X to fracture surfaces is coupled with advection in fractures to yield radon transport well beyond what would be expected if only one mechanism dominated transport.

Spreading in ground-water has traditionally been predicted using the advection-dispersion equation parameterized with composite terms that quantify both diffusion and hydrodynamic advection (see for example (Freeze and Cherry, 1979)):

$$D_L = D^* + \alpha_L v \text{ and } D_T = D^* + \alpha_T v . \quad (1)$$

In Equation 1, D^* is the coefficient of molecular diffusion, v is the average linear velocity, D_L , D_T are the coefficients of longitudinal and transverse dispersion, respectively, and α_L and α_T are the longitudinal and transverse hydrodynamic dispersivities, respectively. This approach requires an assumption of "Fickian" transport behavior, meaning spreading that resembles, mathematically, molecular diffusion. Although there have been recent attempts to generalize the advection-dispersion equation for application in fractured rock (see the review by *Berkowitz* (2002)), the more serious problem is that diffusion, dispersion, and heterogeneous advection are physically very different transport mechanisms. Unless they can be separated, any analysis based upon interpretation of transport using the advection-dispersion equation is suspect.

Experimental Separation of Mass Spreading Mechanisms

The most direct way to measure mass transport in the field is to conduct an artificial tracer experiment. An artificial tracer experiment is conducted by injecting a known mass of tracer into a forced or natural hydraulic gradient and measuring the concentration of tracer as it arrives at one or more detection points. The history of concentration at a detection point is the breakthrough curve, from which transport parameters can be derived by matching the breakthrough data to a transport model. As multiple and distinct transport mechanisms prevail in fractured rock multiple transport parameters must be derived from such tests. The key to separating the influence of diffusion, dispersion, and heterogeneous advection, is to design the tracer experiment in such a way that a unique fit of the transport model to the breakthrough curve can be obtained. Tracer experiments must be designed such that the physical uniqueness of various mass-spreading mechanisms can be exploited.

Molecular Diffusion

Molecular diffusion is the most straightforward spreading mechanism to discern as it is independent of fluid velocity. As diffusion is caused by the random kinetic motion of jostling water molecules, it is truly isotropic. Compared to most natural water velocities in permeable geologic media, molecular diffusion works very slowly. For this reason, the later-time portion of the breakthrough curve (the breakthrough tail) has been considered indicative of diffusive mass exchange between fractures and the surrounding rock matrix. *Maloszewski and Zuber* (1983; 1985; 1990; 1993), for example, have examined a number of tracer breakthrough curves in fractures to derive rates of matrix diffusion. Such an exercise generally consists of fitting at least three model parameters, so that the results may be ambiguous when based upon a single breakthrough curve. More certain results are obtained when tracers of varying diffusivity (different molecular size) are combined in a single experiment. If all tracers are chemically non-reactive and are otherwise transported identically (i.e. by advection, hydrodynamic dispersion, heterogeneous advection), then differences in breakthrough may be attributed to the different diffusion rates. *Garnier et al.* [1985] conducted field experiments in a fractured chalk using fluorescein, iodide, and deuterium as tracers. A clear separation in the three breakthrough curves was observed, and was later explained using a matrix-diffusion model [*Maloszewski and Zuber* 1990; *Moench*, 1995]. *Sanford et al.* [1996] observed similar breakthrough separation of gas tracers in a fractured saprolite.

Caution is warranted in interpreting such experiments, however, because diffusion cannot be decoupled from other spreading mechanisms in fractured rock. Under a relatively homogeneous flow field, diffusion will have little impact on hydrodynamic dispersion or heterogeneous advection because each streamline is relatively similar to the neighboring streamline. In fractured rock, however, velocity fields can be so heterogeneous that neighboring streamlines may have vastly different velocities. As a result, diffusion from one streamline to another may have an important impact on tracer transport. Figure 1 presents a tracer breakthrough from a weak-dipole tracer experiment conducted in a fractured crystalline rock near Mirror Lake, New Hampshire (Becker and Shapiro, 2000). The diffusivities of deuterated water (HDO), bromide (Br^-), and pentafluorobenzoic acid (PFBA) are $2.3 \cdot 10^{-5}$, $2.0 \cdot 10^{-5}$,

$0.66 \cdot 10^{-5}$ cm^2/sec, respectively. Although no difference in late-time breakthrough was observed because diffusion rates are insignificant in the matrix over the duration of the experiment (about 1 week), some separation was observed at early time in spite of careful mixing of the tracers prior to injection. This breakthrough separation is possibly due to the interaction of molecular diffusion and advection near the well bore where injection causes extremely heterogeneous velocity fields.

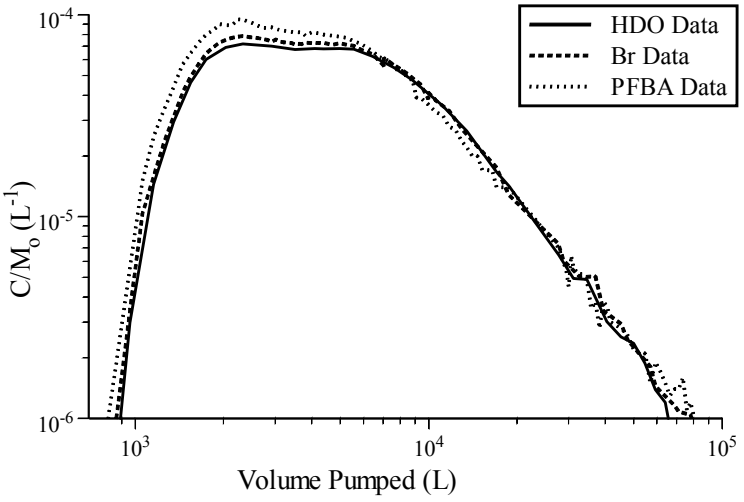

Figure 1. Normalized breakthrough of tracers with different diffusivity, in a tracer experiment conducted in fractured crystalline bedrock.

Hydrodynamic Dispersion

The term hydrodynamic dispersion is used primarily in association with the advection-dispersion equation. It accounts for diffusion-like "Fickian" spreading of mass during transport. The coefficient of hydrodynamic dispersion is generally expressed as the combination of spreading in the direction of average linear velocity (longitudinal component) and perpendicular to the direction of average linear velocity (transverse component). Although there is some evidence that small amounts of dispersion can occur upstream in rock fractures (Dijk and Berkowitz, 1999), hydrodynamic dispersion acts primarily in the direction of flow. This is a fundamental difference between hydrodynamic dispersion and molecular diffusion, as diffusion acts in all directions simultaneously.

Efforts to measure hydrodynamic dispersion in fractured rock using tracer experiments have met with limited success. Breakthrough curves obtained in fracture rock systems tend to exhibit strong tailing. Such behavior is observed in Figure 1. Pure advection dispersion predicts that a breakthrough curve should appear approximately parabolic when characteristic length scale is a factor of ten or more larger than the dispersivity. Thus, the advection-dispersion equation does not well represent most breakthrough curves obtained in fractured rock systems. While most researchers have explained breakthrough tailing by diffusion of mass into the rock (Birgersson and Neretnieks, 1990; Grisak and Pickens, 1980; Maloszewski and Zuber, 1993; Neretnieks et al., 1982; Novakowski et al., 1998), others have conjectured that diffusion into inter-channel small-aperture spaces in the fracture itself serves as a diffusive sink for mass in the fractures (Abelin et al., 1994; Dykhuizen, 1992; Johns and Roberts, 1991; Rasmuson and Neretnieks, 1986). Still others have proposed models by which mass is exchanged with stagnant fluid along the fractures walls (Coats and Smith, 1964; Raven et al., 1988) or by diffusion into an unspecified distribution of stagnant fluids associated with the fracture and matrix (Haggerty and Gorelick, 1995; Haggerty et al., 2000). Prediction of these coupled transport processes require the extension of the advection dispersion equation by the

introduction of additional transport parameters. Often these additional parameters have similar effects on the breakthrough curve as hydrodynamic dispersion, making it nearly impossible to separate hydrodynamic dispersion from other transport mechanisms based upon a single breakthrough curve.

Because hydrodynamic dispersion is a somewhat loosely defined concept, it is best to distinguish diffusion from dispersion by measuring the impact of diffusion. As stated previously, one way to do this is to employ tracers of varying diffusivity. Another approach is to take advantage of the fact that hydrodynamic dispersion is a function of velocity, whereas diffusion is not. Forced gradient tracer experiments conducted at different velocities, i.e. different pumping rates, should express different mass spreading behavior in the breakthrough curves. Figure 2. presents breakthrough curves collected from the same wells at Mirror Lake shown in Figure 1 (Becker and Shapiro, 2000). Figure 2 compares weak-dipole tests, where 5% of the withdrawn fluid was constantly reintroduced at the injection well with a radially convergent tests where tracer mass was introduced as a slug, with no constant injection. The pumping rates for Tests D, A, B, C, were 9.8, 8.3 , 5.2,and 2.9 liters per minute, respectively. The pumping rate for the radially convergent experiment was 4.5 liters per minute. The breakthrough curves for all experiments are quite similar in late time. It is impossible to distinguish, within measurement error, the difference in breakthrough tails of these experiments. It is only the early time behavior that varies, and it varies in a limited way. The lowest rate experiment, Test C, exhibited a very different breakthrough behavior than the other experiments. There appears to be a threshold velocity below which early breakthrough changes markedly. The advection-dispersion equation would predict a gradual change in breakthrough curve shape with velocity that is not observed in these experiments. It is possible that this is related to tracer density, but comparisons with tracers of varying density suggest this is not the case (Becker, 2003). It is likely, therefore, that transport was not dominated by hydrodynamic dispersion in these experiments. Transport mechanisms other than local mixing and diffusion appear to dominate transport.

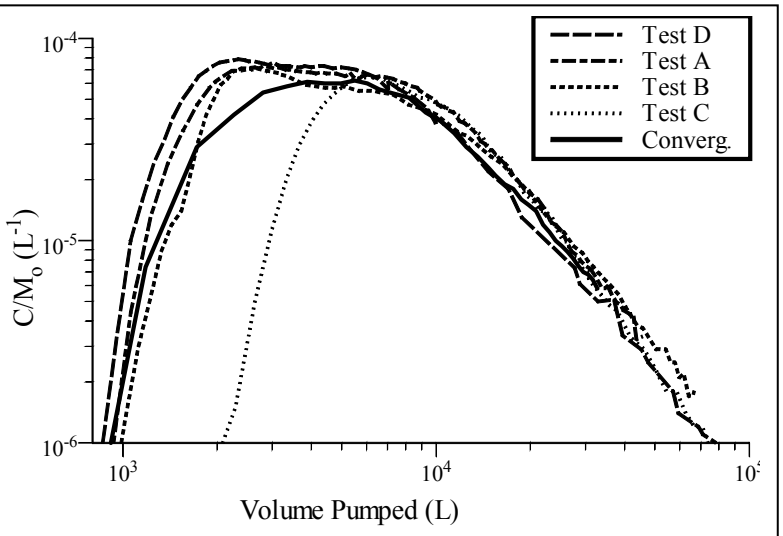

Figure 2. Tracer experiments repeated between the same two wells, but a different pumping rates and hydraulic configuration.

Heterogeneous Advection

Spreading of dissolved mass in ground-water can occur at macro-scales, i.e. at scales well beyond the pore scale. Whereas the characteristic length scale predicted from relatively homogeneous unconsolidated geologic media is expected to be on the order of grain size, macro-dispersion is expected to have a length scale on the order of meters to tens of meters. At this scale, mass spreading is not easily conceptualized as a "mixing" phenomenon.

Rather, it is most often depicted as the summed effect of mass traveling through multiple layers or facies of varying hydraulic conductivity. Unless these layers can be individually monitored downstream, mass arrival is dispersed by the staggered arrival of multiple mass transport pathways. Consequently, the term "heterogeneous advection" is preferred here over macrodispersion, and a clear distinction is made between heterogeneous advection and hydrodynamic dispersion, at all scales.

We define the difference between diffusion and heterogeneous advection as follows: heterogeneous advection is reversible, whereas hydrodynamic dispersion is not. Consider the illustration (Figure 3) of three slugs of tracer injected into a homogeneous flow field applied to three layers of differing hydraulic conductivity (time t = 0). A slug introduced into a higher conductivity layers will move faster under the same hydraulic gradient as a slug introduced to a lower hydraulic conductivity layer. As it moves forward, hydrodynamic dispersion will spread the slug or plume in both longitudinal and transverse directions (times t = 1, t = 2). If the flow field was instantaneously reversed at time t = 2, the tracer plumes would travel in the opposite direction. Again the more permeable layers will more quickly advect the tracer, now reversing the separation made when the flow field was in the initial direction. Hydrodynamic dispersion, however, always acts in the direction flow and continues to increase local spreading. The individual plumes disperse, while the overall divergence of mass actually contracts. At the final time, t = 4, the influence of heterogeneous advection is completely reversed, while the influence of hydrodynamic dispersion remains.

Reversing a uniform flow field in situ is impractical. Reversing a radial flow field is done routinely,

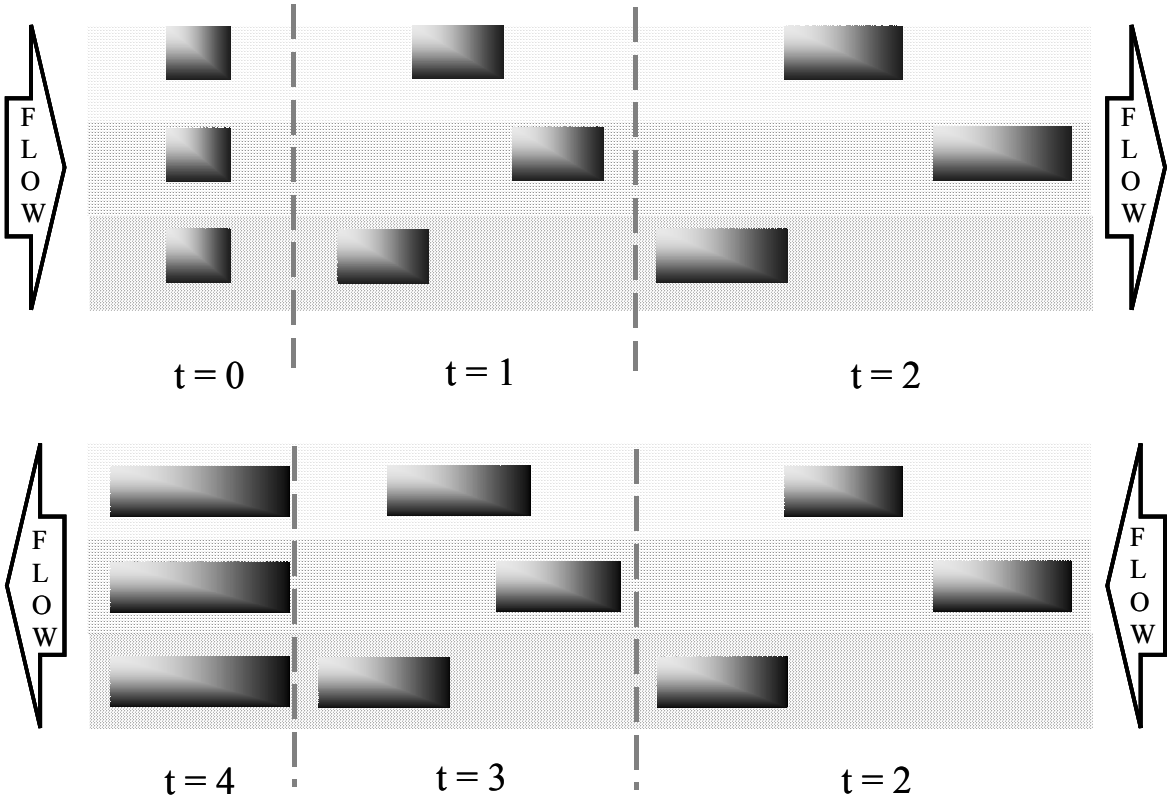

Figure 3. Illustration of the difference between hydrodynamic dispersion and heterogeneous advection.

however, in "push-pull" tracer experiments. In a push-pull experiment, a known mass (slug) of tracer is injected into a formation sometimes followed by a known volume of tracer-free water (a chaser). The slug and chaser are then withdrawn from the same well, usually at approximately the same rate the fluid was injected. Under these conditions the flow field is reversed from radially divergent to radially convergent. Consequently, any heterogeneous advection caused by tracer flowing along disconnected flow paths should be entirely reversed during the withdrawal phase. Push-pull tracer experiments should, in theory, measure only hydrodynamic dispersion and matrix diffusion.

In practice, however, heterogeneous advection and hydrodynamic dispersion cannot be entirely separated. In fractured rock, water is thought to move in a channeled manner through essentially disconnected narrow flow paths. Each of these channels are likely to have a different effective hydraulic conductivity dependent upon the interconnection of these flow paths through the varying aperture field. The pathways, therefore, experience a variety of average linear velocities. A variance in average linear velocity will lead to a variance in the effect of hydrodynamic dispersion along each of the flow paths, as the coefficient of hydrodynamic dispersion is a function of average linear velocity. Figure 4 shows the result of a push-pull experiment conducted again in the crystalline rock formation near Mirror Lake, New Hampshire. Concentrations are normalized to the injected concentration and time is normalized to the duration of the push phase of the experiment. The circles are PFBA breakthrough during withdrawal phase. The dashed lines represent predicted breakthrough according to a radial formulation of the advection-dispersion equation (Becker and Shapiro, 2003). The mass under each of these breakthrough curves is a function of the flow rate into individual channels which is assumed to be a function of the cube of the mean aperture according to the so-called "cubic-law" (Witherspoon et al., 1980). The velocity at which mass moves in these channels is a function of the square of the mean aperture, again according to the cubic-law. Regardless of the assumed distribution of channel aperture in the formation, the sum of these individual breakthroughs resemble the solid black line in Figure 4. Only the largest channel, which controls the first arrival of tracer, relates the total breakthrough behavior to the specific attributes of the formation. The reader is referred to *Becker and Shapiro* (2003) for details of the this so-called "multipath" model of transport.

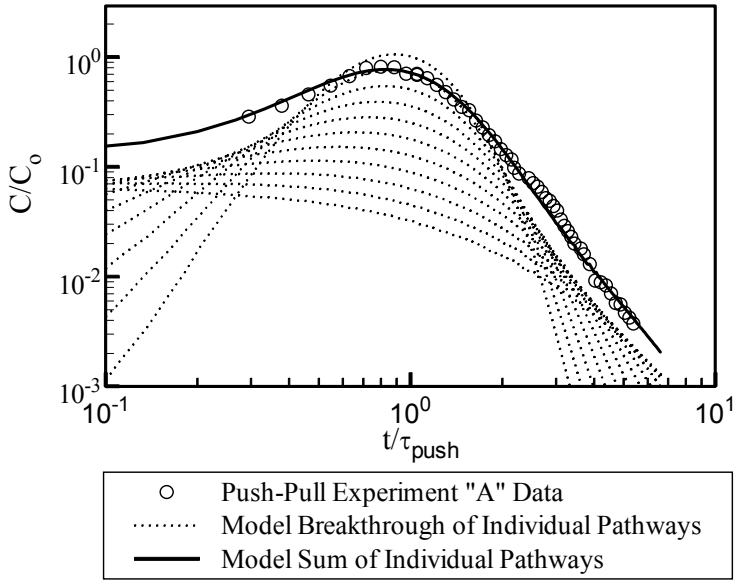

Figure 4. Comparison of PFBA breakthrough from a push-pull experiment conducted to a theoretical "channel" model.

The relevant point here is that, although heterogeneous advection is not strictly expressed in the breakthrough curve because the flow field was reversed, the interactions between heterogeneous advection and hydrodynamic dispersion are evident. Breakthrough tailing still occurs, but is much less extended than in experiments conducted in unidirectional flow fields. The power-law slope of the breakthrough tails in Figure 2 is –2, for example, while the power-law slope of the push-pull breakthrough curve in Figure 4 is –4.5. Breakthrough tailing in the push-pull experiment is thought to be due entirely to hydrodynamic dispersion. This formation is too impermeable to allow significant matrix diffusion over the several hour duration of this push-pull experiment (Becker and Shapiro, 2000). In more porous formations push-pull breakthrough curves may relate the combined impact of both hydrodynamic dispersion and matrix diffusion.

Conclusions

Non-reactive tracers tend to be much more dispersed during transport in fractured rock than in unconsolidated geologic media. Mass spreading is due to a combination of diffusion, hydrodynamic dispersion, and heterogeneous advection. These spreading mechanisms are fundamentally different in their behavior and, therefore, must be measured separately for reliable predictions of transport to be made. Such a separation may be made if specifically designed tracer tests are conducted. To isolate the effect of molecular diffusion, tracers of different diffusion rates should be compared. Repeating experiments at different velocity will also highlight the influence of matrix diffusion as molecular diffusion occurs at a rate independent of fluid velocity. Distinguishing hydrodynamic dispersion and molecular diffusion is best accomplished by using these methods to isolate the effects of molecular diffusion and heterogeneous advection. Heterogeneous advection is conceptualized here as spreading caused by variable transport rates in media of different hydraulic conductivity. Heterogeneous advection is distinguished from hydrodynamic dispersion, therefore, by reversing the flow field during a tracer experiment. This should result in the nullification of the influence of heterogeneous advection and leave only the influence of dispersion and diffusion on the breakthrough curve. It is important to realize, however, that advection, dispersion, and diffusion are coupled processes so that experimental separation is never complete and is meaningful only with respect to a particular theoretical transport model. Here, it has been assumed that the classic advection-dispersion equation is valid for fractured media. This assumption is a matter of some debate.

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