Advection, Dispersion, and Confusion

by Fred Molz

Introduction

This commentary is motivated by a recent paper (Hadley and Newell 2014; “The New Potential for Understanding Groundwater Contaminant Transport”) along with the comment by Shlomo Neuman and the response by Hadley and Newell (Neuman 2014). After reading and thinking about this material, it is evident to me that Neuman and Hadley & Newell (H&N) are not talking about the same thing. Based on field experience, H&N are describing a perceived inconsistency in the macrodispersion concept that dates back to Gelhar et al. (1979) and Gelhar and Axness (1983). In contrast, Neuman does not answer this concern. Instead, based on his statements and references, he describes ongoing extensions of the classical macrodispersion theory that mostly date back only to 2006. This leaves unanswered the question that H&N pose: What is the physical meaning and basis for the classical concept of macrodispersion, and how should one visualize contaminant transport in groundwater? The intention of this commentary is to clarify this core question that has been a bone-of-contention for more than 30 years, since Anderson (1979).

Fundamentally, in any fluid flow, including groundwater flow, there is only advection and diffusion. So if we could reproduce the true fluid velocity field in a porous medium, all we would need in order to model solute transport would be molecular diffusion. The problem is that the true velocity field is often difficult or essentially impossible to calculate or measure, so we have to introduce an approximation—hence the Darcy velocity in porous media. The Darcy velocity is a fictitious velocity that is viewed as the average of the true velocity over a porous volume that is small relative to the problem dimensions but large relative to the individual pores—the so-called representative elementary volume (REV). (The word “fictitious” is used, because the concept of a unique “REV” in natural heterogeneous sediments has not been conceptualized clearly or verified experimentally. Nevertheless, it appears approximate in a useful sense.) The Darcy velocity concept has been valuable, especially for groundwater supply calculations and estimates, but a problem arises when reactive solute transport is included. If a groundwater solution moves from point A to point B at a constant Darcy velocity, the actual fluid velocity will not be constant. Below the REV scale, the solution will still be moving in a complex, tortuous fashion, and this will cause a type of transport interacting with that caused by diffusion. The phenomenon behind this additional mixing is called mechanical dispersion, and in the direction of flow it is commonly approximated as being proportional to the velocity, with the proportionality constant being called the longitudinal dispersivity. The combined effect of molecular diffusion and mechanical dispersion is typically represented mathematically by Equation 1, so in a mathematical sense the combination, called hydrodynamic dispersion, is equivalent to enhanced diffusion, and diffusion causes irreversible mixing in the transport equation (more about this later).

$$D_L = \delta_L V_L + D_* \quad \text{and} \quad J_C = -D_L \frac{\partial C}{\partial L}$$

(1)

where $D_L$ is the longitudinal dispersion coefficient ($L^2/T$), $\delta_L$ is the longitudinal dispersivity ($L$), $V_L$ is the Darcy velocity in the $L$ direction ($L/T$), $D_*$ is the effective molecular diffusion coefficient in the porous medium ($L^2/T$), $J_C$ is the solute flux in the $L$ direction ($M/L^2/T$) and $C$ is the solute concentration ($M/L^3$). Dispersivities transverse to the Darcy velocity may also be defined.

In fluid mechanics, the mathematical concept of dispersion, as distinct from diffusion and advection, dates back to Taylor (1953, 1954) and Aris (1956). As these classical papers show, there is one key question that has to be answered before the mathematical concept of dispersion takes on physical meaning. What is the travel distance required before asymptotic or Fickian conditions occur, and Equation 1 becomes meaningful physically? In the following discussion, I will review the data and field experiments performed decades ago indicating that large travel distances are required before Fickian (or
asymptotic) conditions in a contaminant plume would be expected to occur, if ever.

The Hadley & Newell (2014) Paper

Under “Key Points and Concepts Going Forward,” H&N state that (1) “diffusion is big” (several colleagues and I have made the same point in numerous previous publications); (2) traditional “advection and dispersion modeling may be ‘OK’ for some applications” (if by traditional dispersion the authors mean traditional macrodispersion, we do not expect many valid applications); and (3) “we need new models” and “a major paradigm shift” (previous data, field studies, and new models are already available, but not in the references of H&N, and I agree that the thinking in a lot of the professional journals and textbooks is confused—thus an attempt at clarification).

Molz et al. (1983) and Güven et al. (1984) applied the “Aris moment method” to understand full aquifer dispersion in a typical aquifer assuming stratified, horizontal flow. (The late Oktay Güven worked out the mathematical details in both papers.) As a sample aquifer, the formation studied by Pickens and Grisak (1981a, 1981b) was utilized. This aquifer was 8.2 m thick, and a REV dispersivity of 0.7 cm was measured, along with the vertical variation of horizontal hydraulic conductivity using laboratory, single-well and two-well tracer tests. Using conservative estimates for the necessary parameter bounds, travel distances of 5.4 to 109 km were required before full aquifer macrodispersions of 50 to 990 m would have meaning. The implication is that the full aquifer macrodispersion approach would have little applicability anywhere near a source of contamination, and the fact that we assumed local (REV) dispersivities to be applicable meant that our huge travel estimates were conservative. An additional major practical conclusion of the Molz et al. (1983) and Güven et al. (1984) studies was that the measured variation of the horizontal hydraulic conductivity with depth, K(z), at one or more locations was of major practical importance in understanding contaminant transport in groundwater, and this result still stands.

About this time, Anderson (1983) warned model developers to be cautious, and Gillham et al. (1984) published a paper entitled, “An Advection-Diffusion Concept for Solute Transport in Heterogeneous Unconsolidated Geologic Deposits,” an idea elaborated by Sudicky (1983). So the advection-diffusion concept along with serious questions about the practical meaning of dispersion due to K variations above the REV scale (macrodispersion) were out there in the early 1980s. In 1985, Güven et al. (1986) submitted a comment on the Gillham et al. (1984) paper that was complimentary to most of the points raised in the paper, with the main concern being that insufficient weight was being placed on the two-dimensional (2D) or three-dimensional (3D) velocity variations that occur in real aquifers. (In the same issue of Water Resources Research, Sposito et al. [1986] warned of the uncertain physical basis for the stochastic convection-dispersion equation.) In their reply, Sudicky and Gillham (1986) did not disagree with this, but they felt that the measurement problem was too great, so they suggested that the stochastic, macrodispersion approach of Gelhar et al. (1979) and Gelhar and Axness (1983) was the way to go, the main potential advantage being less computation required on mid-1980s computers. So by 1985, a bifurcation point was reached: The scientists more inclined to deterministic concepts had a measurement problem on their hands, whereas the stochastic hydrologists were inclined to assume that the development and application of Gaussian geostatistical concepts to an analysis of (assumed) stationary K distributions (macrodispersion above the REV scale) was the way to go. There was uncertainty at the time, but I feel confident now in saying that stochastic macrodispersion theory was overemphasized relative to other approaches. As evidence, we point to the paper published by Gelhar et al. (1992, Figure 1) listing dispersivity values varying from 1 or 2 cm up to 7000 or 8000 m, over travel distances of 1 m to 100 km. The values associated with contamination events at scales of 100 to 1000 m vary from about 12 to 40 m, with an average of about 22 m. Based on the evidence discussed so far, there is little doubt that over such travel distances contaminant spreading will be typically dominated by advection, so this list of scale-dependent values related to the concept of macrodispersion would have little physical meaning—that’s why it is “scale-dependent—and the use of such numbers in models would magnify irreversible mixing way beyond what is seen in the field.” There are additional field tests that support this. Güven et al. (1985) analyzed a single-well tracer test performed by Pickens and Grisak (1981a, 1981b). The purpose of the tracer test was to study and model scale-dependent dispersion. Pickens and Grisak measured the hydraulic conductivity distribution [K(z)] in the vicinity of their tracer test, but then did not use this fundamental information in the analysis of their data. Consequently, based on a one-dimensional (1D) radial advection-dispersion model, they calculated a scale-dependent dispersivity given on average by $\delta_L = 0.18 \text{ L}$, with $L$ the travel distance in meters. They also estimated the REV dispersivity and arrived at a value of 0.7 cm. To show that the scale-dependent (macro) dispersivity was an artifact of the 1D analysis, Güven et al. (1985) simulated the experiment in detail with 2D flow (cylindrical coordinates) and a constant REV dispersivity of 0.7 cm. The result was a near perfect fit to the experimental data, showing that the idea of scale-dependent macrodispersion was indeed a conceptual artifact in this case.

Over several following years, these workers and others were able to simulate additional single-well and two-well tracer tests on much larger scales with either a very small REV dispersivity or no dispersivity. All that was needed was one or more measurements of K(z)—horizontal hydraulic conductivity as a function of elevation “z” (Molz et al. 1989). The final two-well tracer test at the Mobile Site was performed and reported by Güven et al. (1992). An injection and withdrawal well were located in a confined granular aquifer that was about 21 m thick. The wells were located 40 m apart, and K(z)
was measured at 14 nearby locations using observation wells and a borehole flowmeter to measure \( K(z) \) (Molz et al. 1990). Fluid injection and simultaneous withdrawal took place for about 33 d at the rate of 1397 m\(^3\)/d, and for the first 96 h a sodium bromide tracer was added to the injection water. Tracer concentrations vs. time were monitored in multilevel sampling wells and in the recovery well. After the data were analyzed, a 3D advection-only model was used to simulate the results. The noncalibrated model results presented in the paper are excellent representations of the data, showing that over the 33 d-test period, advection dominated all scales of dispersion.

There are additional field studies by others that support the contention that advection dominates REV dispersion over considerable travel distances and times in granular aquifers (Barker et al. 1987; Ronen et al. 1987a, 1987b; Smith et al. 1987). As much as possible, all of these studies made careful point measurements of dissolved oxygen or nitrate concentrations as a function of elevation, \( C(z) \), in order to better understand microbial activity occurring in the study aquifers. All of the studies documented the persistence of large vertical concentration gradients after large travel distances. Molz and Widdowson (1988) simulated the overall microbial activity and showed that the transverse REV dispersivities required to maintain such vertical gradients had to be less than 1 mm. At a horizontal groundwater velocity of 10 cm/d, this would yield a vertical mechanical dispersion coefficient of 1 cm\(^2\)/d or 1.2 \( \times 10^{-5} \) cm\(^2\)/s—comparable to the molecular diffusion coefficient of salt in water. The main conclusion of Molz and Widdowson (1988) was that one must use advection-dominated transport models to simulate chemical and microbial activity in granular aquifers. Otherwise, true mixing and biochemical kinetics are seriously overestimated.

What about the field experiments at Borden and Cape Cod that were interpreted at least partly as supporting the near-source validity of the macrodispersion concept (Freyberg 1986; Sudicky 1986; Garabedian et al. 1991). In fact, Fickian conditions did not occur in the Borden experiment. According to theory, such conditions would occur when the second moments of the concentration distribution about the mean become constant. To obtain that result in the Borden experiment (Freyberg 1986, Figure 10), the data point at about 1040 d had to be ignored. This is important because the second moment variations with time (Freyberg 1986, Equation 5) are supposed to converge to a constant, not diverge. This led Freyberg (1986) to conclude that the evidence of scale-dependent dispersion was detected, which resulted in the factually true but contradictory statement that, “The asymptotic (Dagan 1984) longitudinal dispersivity obtained from the calibration is 0.49 m, although asymptotic conditions were apparently not reached (italics mine).” This, along with the layered horizontal sediment structure of the Borden aquifer and the plume-splitting result in a previous Borden tracer test (Sudicky et al. 1983), led my colleagues and me to suspect that the Borden dispersivity was probably a nonphysical, scale-dependent result that would continue to grow with time or travel distance if the experiment had been continued.

The Cape Cod experiment (Garabedian et al. 1991) is more difficult to elaborate, because the experimental results conform almost perfectly with classical macrodispersion theory: good mass balance (their Figure 4), nearly constant velocity (their Figure 6), and highly linear longitudinal bromide concentration variance with travel distance, almost from the very beginning of the experiment (their Figure 10). This led to the authors’ major conclusion that: “longitudinal mixing was the dominant dispersion process, with dispersivity reaching a limiting value of 0.96 m after 26 m of travel distance of the center of mass” (italics mine). It was also noted that the transverse vertical dispersivity was very small, less than 1.5 mm based on multiple estimates.

Probably, the main characteristic of many aquifers that inhibits dispersive mixing is the tendency for horizontal layering having variable hydraulic conductivity. If these layers then have sufficient horizontal connectivity, water in the high K layers can spread rapidly from the center of mass with minimal diluting. Advection literally “runs away” from diffusion, which is the main motivation for the original advection-diffusion concept of Sudicky (1983) and Gillham et al. (1984). Looking at the sediment photograph in Figure 3 of LeBlank et al. (1991), complex layering is evident and called “stratified.” I am not sure how many \( K(z) \) measurements were made at the actual Cape Cod test site. Garabedian et al. (1991) mention an average estimated porosity of 0.39 and a mean hydraulic conductivity of about 110 m/d. Also mentioned are nearby flowmeter tests and measurements on cores indicating that “the outwash (K) varies by about one order of magnitude.” Based mainly on experience, I would say that if there is much lateral continuity of the K layers at the Cape Cod site, then asymptotic behavior of the tracer plume would be unexpected with a travel distance of only 26 m and a vertical dispersivity of less than 1.5 mm. There was also a significant vertical component of plume velocity, and the glacial outwash sediments were exceptionally clean of clay particles. From my perspective, the Cape Cod tracer test may be the “exception that proves the rule,” and if so, it would be valuable to understand the reason. It is also possible that given the complex, stochastic structure of natural sediments, that we are only beginning to understand (Meerschaert et al. 2013; Dogan et al. 2014), a linear concentration variance with time is not a sufficient condition for asymptotic conditions to occur.

Last but not least, the MADE study: three large-scale natural gradient tracer tests (macrodispersion experiment—MADE) were conducted in sediments underlying the Columbus Air Force Base in Columbus, Mississippi, from the late 1980s to late 1990s with funding support from the Electric Power Research Institute (EPRI) and the U.S. Air Force. The value of these experiments was that the aquifer selected was typical of many natural aquifers comprised of unconsolidated fluvial deposits. The naming of the experiment (Macrodispersion) suggested a certain degree
of expectation concerning the dominant transport process, but the various participants demonstrated integrity and continued working carefully with the data even when the expected results were not obtained. Ultimately, this led to the development of some real knowledge.

Some of the most important lessons from over 25 years of research at the MADE site have been articulated by Zheng et al. (2011). A major insight from the MADE research is that the advection-macrodispersion model (AMDM) failed to capture key characteristics of plume-scale transport. The work by Harvey and Gorelick (2000), Feehley et al. (2000), and Julian et al. (2001) presented a clear illustration of the mismatch between the field-measured and model-calculated plumes for the three natural gradient tracer tests (MADE 1, 2, and 3). In more recent forced-gradient tracer tests, the same conclusion was reached (Liu et al. 2010; Bianchi et al. 2011a).

The failure of the AMDM to represent plume-scale transport at the MADE site can be attributed to the fact that spatial heterogeneity and resulting solute transport is not stochastically randomizing, that is, leading to an anisotropic random walk, as assumed in the AMDM. Rather, structured heterogeneity enhances connectivity of high-conductivity aquifer sediments (Fogg et al. 2000; LaBolle and Fogg 2001; Zheng and Gorelick 2003), which leads to “dual-domain” behaviors, that is, preferentially faster flow paths where transport is predominantly advective, and the relatively impermeable aquifer matrix where transport transverse to the main flow is dominated by dispersion. As stated by Zheng et al. (2011), “if microscale variations of water flux due to aquifer heterogeneity could be precisely described, the natural model would be based solely on a combination of advection and molecular diffusion; macrodispersion would not be a dominant process.” Indeed, with the “dual-domain” transport behaviors described above, modeling analysis of all five previous tracer tests uniformly demonstrated that the dual-domain mass transfer model was able to fit the experimental data much better than the AMDM, in terms of both plume geometries and solute mass distributions (Feehley et al. 2000; Harvey and Gorelick 2000; Julian et al. 2001; Salamon et al. 2007; Liu et al. 2010; Bianchi et al. 2011a). Zheng and Gorelick (2003) and several follow-up studies (Liu et al. 2004, 2007; Bianchi et al. 2011b) have developed the concept of connected heterogeneities at decimeter and smaller scales as the explanation for “anomalous” transport at the MADE site. Thus, in field practice, the dual-domain mass transfer model is likely to be more feasible than a detailed advection-diffusion model. However, to our knowledge a rigorous method for measuring or estimating the required first-order rate coefficient is not currently available (Haggerty and Gorelick 1995).

### Closing Thoughts

I hope that the review and observations presented in this commentary have removed some of the confusion related to advection and dispersion in groundwater, but I do not want to give the impression that the problem is solved. However, using modern techniques such as borehole flowmeters, direct push, and other geophysical techniques, it is becoming increasingly practical to obtain detailed hydraulic conductivity “K(z)” measurements in aquifers (Liu et al. 2009). This, in turn, provides more insight into how hydraulic heterogeneity is structured in natural sediments, and motivates a more promising type of stochastic hydrology. One insight based on small-scale K measurements has been called the fractal-facies concept (Lu et al. 2002). Here it was shown that if K measurements were analyzed stochastically on a facies basis, the ln(K) distribution in each facies, but not in all facies combined, displayed properties of the stochastic fractal known as fractional Brownian motion (Molz et al. 2004). It makes sense that potential statistical order in aquifer physical properties should be analyzed on a facies basis, because each facies can represent totally different sedimentary environments, often separated by geologic time intervals. Very recently, detailed K measurements made by Bohling et al. (2012) followed by a theoretical analysis (Meerschaert et al. 2013) showed that to a good approximation the fractal-facies concept could be applied to the MADE aquifer. Dogan et al. (2014) then built on this information to show that if high-resolution K data are used with adequate connectivity, a conventional advection dispersion model can be used successfully to capture the MADE transport behaviors. In their study, the flow and transport model had a grid spacing of 0.25 × 0.25 × 0.05 m in 3D. The longitudinal REV dispersivity used in the model was 5 cm, with horizontal and transverse dispersivities of 0.5 and 0.05 cm, respectively, and the senior author reported that they got good results even with a smaller longitudinal dispersivity (Dogan, personal communication, 2014). With such a fine grid resolution and small dispersivities, this study provides compelling evidence that H&N’s approach “exacerbates systematic errors inherent in standard deterministic models of groundwater flow and transport.” Then, without discussing Fickian or asymptotic requirements, he outlines a new, highly mathematical, transport theory that he and his colleagues are developing. Although they might well formulate a basis for more general future work, the new papers have not presented any field verifications and are limited to nonreactive transport. So it would appear that their results are not yet of interest to field workers who need a realistic concept of transport in groundwater, have powerful computers but often limited data and commonly must deal with biochemical reactions. The very fact that an extended stochastic theory is in the early stages of being developed implies that the existing stochastic, macrodispersion theory is misleading, and this is what I think H&N are trying to say in an honest manner.

### Neuman (2014) Comment on Hadley & Newell (2014)

For the most part, Neuman (2014) did not address the concerns of H&N directly. He starts by saying
evidence for the argument made in this commentary and in previous work. Advection dominates in the broader source area, and K(z) measurements with proper statistical analysis are the key to performing realistic transport simulations in natural aquifers, or in evaluating the hydrogeology even if simulations are not performed. Horizontal connectivity of K variations should be assumed until proven otherwise. Plumes can maintain a discrete identity over long travel distances, and in the presence of natural ambient flow, sampling wells can produce highly mixed and ambiguous data even if flushed before sampling (Elci et al. 2001). The concept of macrodispersion requires an unrealistically long travel distance or travel time before so-called (theoretical) Fickian, or asymptotic, conditions are reached, and they are probably never reached in many natural heterogeneous systems, because the plume would be expected to separate into subplumes that no longer interact, as observed by Sudicky et al. (1983). According to Fischer et al. (1979), a similar phenomenon occurs in meandering (but not straight and uniform) rivers where turbulent diffusion is analogous to mechanical macrodispersion, and subplumes get captured in the various meanders and become separated. Whatever it is precisely, matrix diffusion is going to be of great importance, because it will always be much slower than advection, and it will therefore dominate the long aquifer reclamation time scales being observed in the field. Advection, however, will determine first arrival times in almost all cases, with much less dilution than expected based on macrodispersion concepts.

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