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IMPACT OF MEASUREMENT INSTRUMENT AND CONCEPTUAL
MODEL ON ANALYSIS OF SUBSURFACE HETEROGENEITY**

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ABSTRACT

The initial focus of this research was on particle transport. Results included investigation of two important influences on particle transport: (i) the presence of preferential hydraulic pathways and (ii) the presence of contact surfaces. Based on these early results, the project was redirected into analysis of measurement scale and scaling, with emphasis on microbial transport and characterization of hydraulic heterogeneity. Efforts have revolved around experiments within scaled and random porous media, numerical studies, and stochastic theory. A number of significant findings in terms of the measurement, microbial transport, and the development of sampling strategies have been accomplished. This work has resulted in 17 journal publications, a large number of conference presentations, two Ph.D. dissertations, two Master's theses, and two manuscripts upon which undergraduate students have been co-authors.

With respect to microbial transport, we have collaborated with Dr. Madilyn Fletcher, University of South Carolina, in the design and performance of a series of hydraulic, chemical, and microbial experiments. Results from these experiments provided the first experiments of which we are aware that include monitoring the movement of microbes in a statistically stationary, heterogeneous porous medium. Results from this work include the observation that the microbial transport, as observed in breakthrough curves at a well, is relatively independent of the individual realization of the random field. While this type of result was anticipated for chemical transport in these stationary media, it was not initially anticipated for microbial transport. This result implies that there is the possibility of defining, for at least some microorganisms and porous media, a large-scale transport behavior that is dependent only on the statistics of the hydraulic heterogeneity (and not the detailed distribution of sediments in a particular realization). We consider these results to be among the most significant of our project.

With respect to hydraulics, we pursued a series of efforts involved in development of stochastic theory and application of Monte Carlo analysis. In terms of stochastic theory, we developed and tested a technique where by the variance and integral scale of the transmissivity distribution (or distribution of hydraulic conductivity in three-dimensions) could both be estimated based on local measurements of the hydraulic gradient. This method was tested against controlled laboratory measurements of the hydraulic gradient (in the stationary random fields mentioned above) and available field data. This work led directly to publications on this method, but also led to consideration of the design of data sampling networks for systems in which the sampling instrument had significant impact on network design. Further, this work led to the observation that our current field methods for measuring hydraulic head in wells is generally inadequate to provide reliable measures of the hydraulic gradient (particularly in the vertical direction).

In terms of Monte Carlo analysis, we were able to study, in detail, the structure and uncertainty in capture zones for municipal pumping wells in both confined and unconfined systems. This work led to the suggestion, in one of our recent papers, that capture zones (for wellhead protection) not be considered as deterministic entities but, rather, random surfaces defined by the degree of heterogeneity of the sediments and the degree of confidence required in the final solution.

With respect to chemical transport, we ran a series of chemical transport experiments on our two-dimensional, stationary, random conductivity fields. These results were used both to verify available stochastic theory for ergodic transport, and also to provide data sets sufficient for use by others in considering non-ergodic transport. These results were also used to demonstrate the dependence of support of a chemical sample at a well on the rate of pumping from that well.

RESEARCH PURPOSE, CENTRAL HYPOTHESES, AND OBJECTIVES

Authors such as Cushman have argued that development of a consistent model of flow or transport in a heterogeneous medium requires incorporation of the measurement instrument into the model development. Review of the literature, however, indicates that there has been little in the form of detailed study of the interaction of measurement scale, measurement instrument, the form of the heterogeneity, and the information gained from a sampling plan.

Intermediate-scale experiments (experiments conducted on heterogeneous media under controlled conditions) seem to be a natural tool to apply to the problem of analyzing existing theories of scaling and extending our knowledge into consideration of processes and mechanisms not currently included in our theories (e.g. flow / transport to wells, microbial transport). An example of the use of intermediate-scale experiments was provided in the early work from this project where the focus was on the processes and mechanisms influencing particle transport in saturated porous media (e.g., Silliman, 1995; Silliman, 1996). This early work allowed detailed study of the movement of particles in saturated, heterogeneous media, including demonstration of the importance of continuous paths of large grain-sized sediments to the transport of particles and the impact of contacts between sediments of different grain size.

Following this early work, the project was refocused on scaling of a number of mechanisms (flow, chemical transport, and microbial transport) and analysis of a number of measurement instruments. Specifically, the project was focused on the interaction of scale of measurement, scale of a particular process (e.g. fluid flow, chemical transport, microbial transport), and the form of the heterogeneity. The primary hypothesis addressed in the majority of the latter half of the research project were:

The degree to which physical, chemical, and/or microbial heterogeneity in a saturated porous medium will translate into a measured heterogeneity in a response variable is a function of the measurement instrument utilized, the conceptual model underlying the analysis of collected data, and the scale of the analysis.

This hypothesis was addressed through a series of experiments performed on constructed, heterogeneous media, as well as stochastic and Monte Carlo simulation. The constructed media consist of mixtures of sands with the distribution of hydraulic properties guided by numerical simulation of real systems.

SUMMARY OF SCIENTIFIC ACCOMPLISHMENTS

Scientific accomplishments over the history of this project may be broadly characterized into four areas: (i) Equipment and technique development (including sampling strategy), (ii) Analysis of particle transport as a function of dimension of heterogeneity, (iii) Assessment of hydraulics and hydraulic measurement in heterogeneous media, (iv) Assessment of chemical transport under mean-uniform and non-uniform flow fields within heterogeneous media, and (v) Characterization of microbial transport under mean uniform and non-uniform flow fields within heterogeneous media. Primary accomplishments, as represented in the published literature derived from this project, are summarized below with reference to appropriate publications (attached).

Equipment and Technique Development

This project has involved several layers of development of equipment and techniques, including consideration of the interaction of statistical theory and experimental design. Among the preliminary efforts required for this effort was the design of the experimental tanks used throughout this project. These tanks allowed control of mean-horizontal flow, changes in input concentration (of chemicals and microbes), monitoring both tank outflow and at internal sampling points, and visual monitoring of chemical migration. Examples of the design and construction of these tanks are given in Silliman et al. (1994), Silliman (1995, 1996), Silliman and Caswell (1998), Silliman et al. (1998), and Silliman and Zheng (2001).

An important accomplishment of this portion of our research has been the development of the conceptual basis for experimental design based on consideration of scale of measurement and the inclusion of measurement instrument in the design of sampling strategy. A significant portion of our work in this area was presented in Silliman et al. (1998), Conwell et al. (1997) and Zheng and Silliman (2000a). An application of this work in a different setting was presented in Silliman and Berkowitz (2000). Silliman et al. (1998) presents our efforts in designing experiments based on the relative scales of action of the different mechanisms investigated within an individual experiment (e.g., hydraulics versus chemical transport). This led to the design of the extremely complex porous media (porous media designed to mimic the behavior of a second-order stationary random field with a large number of integral scales in the longitudinal and transverse dimensions) used in our most recent experiments on hydraulics, chemical transport, and microbial transport (e.g., Zheng and Silliman, 2000; Silliman and Zheng, 2001; Silliman, 2001; Silliman et al., 2001). Conwell et al. (1997), Zheng and Silliman (2000a) and Silliman and Berkowitz (2000) provided fundamental statistical theory delineating the impact of measurement instrument and measurement design on the spatial statistics estimated from the resulting data sets.

Particle Transport as Function of Dimension of Heterogeneity

Among the earliest work accomplished within this research project was the study of the role of dimension of heterogeneity on transport processes, with particular emphasis on particle transport. This work resulted in two manuscripts that provide a comparison of particle transport in two-dimensional versus three-dimensional media (Silliman, 1995; Silliman 1996). In the first of

these two manuscripts, we demonstrated that particles (latex spheres) were irreversibly trapped within sands, with trapping preferentially occurring along borders where flow moves from coarse grained sediments into fine-grained sediments. More significantly, the second manuscript demonstrated that added degrees of freedom provided by flow through a three-dimensional (versus two-dimensional) medium substantially changed the outlook on particle transport. Specifically, the three-dimensional medium provided a higher probability of encountering flow routes along which particles could transport across the experimental medium without encountering either the fine-grained sediments or critical contacts between grain sizes.

Results Involving Hydraulics

A substantial portion of our research effort was focused on hydraulics in heterogeneous porous media. Hydraulics were approached through three foci: (i) prediction of hydraulic behavior using stochastic theory and analysis of field data sets, (ii) application of Monte Carlo simulation, and (iii) laboratory study of hydraulic response for heterogeneous media.

Stochastic Theory: In our work on Monte Carlo simulation of wellhead capture zones (below), we determined the importance of estimation of the local hydraulic gradient in the presence of limited data. This observation led to publication of a technical discussion regarding estimation of the hydraulic gradient (Cole and Silliman, 1996) and a second publication noting uncertainties in the estimated gradient within heterogeneous media (Silliman and Frost, 1998). This work led to a new research project (supported by funding outside of DOE) on uncertainties in estimating groundwater gradients due to heterogeneity and measurement error.

This study of the hydraulic gradient also led to extension of our work into the realm of stochastic hydrology. The hydrologic literature has seen a significant growth in the development and application of stochastic theory over the past several years (see reviews of this literature in Silliman, 1995; Zheng and Silliman, 2000b; Silliman and Zheng, 2001). We extended this theory via a stochastic method that allowed estimation of the variance and integral scale of the transmissivity field. These two parameters are critical to a number of analyses, including estimation of the dispersive properties and variability of hydraulic response.

It is significant that this new theory is based on the analysis of the hydraulic gradient (versus the hydraulic head considered in a number of other theories). This provides a stationary random field for the analysis and, more importantly, the hydraulic gradient produces a semivariogram that has finite range and a well defined sill value (thus substantially reducing uncertainty related to the process of fitting the sample semivariogram to the theoretical model). The details of this method are contained in Zheng and Silliman (2000) and in the dissertation of Dr. Li Zheng (University of Notre Dame, Ph.D. Dissertation, "Statistical characterization of the transmissivity distribution in groundwater aquifer using hydraulic head measurements", 1998).

Monte Carlo Analysis of Wellhead Capture Zones: A significant application of the consideration of hydraulic heterogeneity has been the analysis of the shape and size of capture zones to both active and passive wells in unconfined aquifers. Based on second-order stationary random transmissivity fields, our simulations demonstrated uncertainties in both the mean flow direction and the geometry of capture zones leading to wells in unconfined aquifer systems. In our early work in this regard (Cole and Silliman, 1997), we demonstrated the significant uncertainty that

can occur in estimating wellhead capture zones, particularly if uncharacterized anisotropy may be present in the *spatial structure of the random variation in the transmissivity*. In our latter efforts in this regard (Cole and Silliman, 2000), we discuss one approach to characterizing this uncertainty through use of confidence intervals for capture zones.

Laboratory Experiments: These theoretical studies were carried into the laboratory through construction of a series of laboratory flow cells allowing analysis of heterogeneous porous media. While our early work was based on glass beads and relatively arbitrary forms of heterogeneity (e.g., Silliman 1995, 1996), our more advanced work involved construction of statistically-based random porous media. These included a scaled medium (based on a finite replication of the Sierpinski carpet from fractal theory), a bimodal medium representing a second-order stationary random field, and a series of media replicating log-normally distributed, second-order stationary random fields (e.g., Silliman and Caswell, 1998; Silliman et al., 1998; Silliman and Zheng, 2001).

In Silliman and Caswell (1998), we demonstrated that the existing stochastic theory for the effective hydraulic conductivity matched well with our experimental data when the porous medium reasonably fit the assumptions underlying the theory (i.e., when applied to the medium representing a second-order stationary random field), but was dramatically in error when applied to a porous medium for which the assumptions underlying the theory did not apply (e.g., the scaled medium). This result is of significance for a number of reasons, not the least of which is the observation that a number of recent studies are leading to the conclusion that the distribution of the hydraulic conductivity may be scaled under a number of field conditions. The implication here is that our body of stochastic theory (based on second-order stationary random fields) may have limited applicability under a variety of natural conditions.

In Silliman et al. (1998 - and the accompanying Master's thesis by P. Conwell, 1997), we utilized visualization to monitor local water velocities. Using this approach, we were able to demonstrate that the water velocities in our experiments closely replicated the variograms predicted using available stochastic theory. Perhaps more significantly, this work demonstrated the extreme difficulty and uncertainty that will result if these same variograms are estimated from physical measurements of hydraulic head (and, through taking differences in space, the hydraulic gradient). This is particular true for situations in which the variation in the hydraulic head is expected to be small relative to the precision of the measurement instrument.

In Silliman and Zheng (2001), we replicated a log-normally distribution transmissivity field with approximately 60 integral scales in the direction of mean flow and 25 integral scales in the direction perpendicular to mean flow. Through an elaborate instrument array designed to be optimal for this particular random field, we were able to monitor both the hydraulic head and the hydraulic gradient at various measurement scales. This unprecedented data array / controlled porous medium combination allowed us to verify stochastic theories (both for hydraulics and transport) using experimental data. The agreement between theory and our experiments was quite satisfactory, and led to a number of observations regarding the source of uncertainty in this form of analysis (see manuscript for complete discussion).

Results Involving Chemical Transport

A substantial portion of our research effort was focused on chemical transport in heterogeneous porous media. The bulk of this work was completed through the analysis of data

collected in laboratory experiments. Chemical transport was monitored both visually and electronically and was investigated in a number of heterogeneous media. In several cases, the results from the laboratory were compared with predictions derived from stochastic theory for ergodic systems. In one case, results from a prior experiment were compared with a new theory for transport known as the Continuous Time Random Walk (CTRW) model (Berkowitz et al., 2001).

In Silliman et al. (1998), we created a bimodal permeability field that reproduced an exponential variogram structure with an integral scale on the order of 1/15 of the horizontal scale of the entire porous medium. This work allowed us to provide a first view of the potential for laboratory studies to support verification efforts on stochastic theory. Unfortunately, the length of the porous medium (relative to the integral scale) was insufficient in this case to achieve ergodic results. Hence, there was only limited agreement between observation and theory. Further, these experiments were limited to one-dimensional averages as the chemical tracer was introduced over the entire depth of the medium. Hence, two-dimensional theories could not be compared with these results.

As discussed in Silliman and Zheng (2001), the length limitation was corrected through construction of a porous medium in which there were approximately 60 integral scales of variability along the line of the mean hydraulic gradient (25 integral scales perpendicular to this line). The limitation to one-dimension was eliminated through construction of a central reservoir (on the inflow side of the experimental apparatus), allowing injection of finite sized plumes of tracer. Finally, two detailed experiments were run on this medium using two significantly different mean velocities (a difference of a factor of 25) to check for anomalies related to low or high velocity effects.

Results on this more sophisticated porous medium showed substantial agreement between the two runs and excellent agreement with theory in terms of the first moment of spatial location of the center of mass of the plume (in both the longitudinal and transverse directions). These results also demonstrated relative agreement with the second spatial moment in the longitudinal direction, but general disagreement with the second moment perpendicular to the mean flow direction. This latter result was anticipated and is directly related to the assumption of ergodicity (which does not apply at these length scales). Other authors have a comment in press in the journal titled *Transport in Porous Medium*, demonstrating stronger agreement with these results based on non-ergodic theory. Finally, this paper discussed the extreme sensitivity of these results to the instrument used to measure concentration. This final result has significant implications for field applications in which sensitivity in terms of detection limits on field tracer chemicals may dramatically impact the interpretation of a field result (relative to theory).

Results Involving Microbial Transport

The latter portion of our research effort was dedicated to experiments on microbial transport through heterogeneous porous media. These experiments were designed to address the paucity of theory and field data describing the details of microbial transport in saturated, heterogeneous media. To our knowledge, our experiments on two replications of a log-normally distributed random field remain the only experiments published involving microbial transport in second-order stationary random media. These final experiments built upon an extensive series of prototype experiments that allowed us to perfect our procedures in terms of packing and running

these experiments. These prototype experiments were discussed in the progress reports associated with this project.

The final experiments involved the transport of bacteria through porous media designed to allow the hydraulic conductivity to vary as a two-dimensional, log-normally distributed, second-order stationary, exponentially correlated random field. The bacteria used were *Pseudomonas fluorescens* R8, a strain demonstrating appreciable attachment to surfaces, and strain M1, a transposon mutant of strain R8 with reduced attachment ability (both strains were obtained through our cooperation with Dr. Madilyn Fletcher at the University of South Carolina). In benchtop, sand-filled columns conducted in the laboratories of Dr. Fletcher, transport was determined by measuring intensity of fluorescence of stained cells in the effluent, or by measuring radio labeled cells that were retained in the sand columns. Results demonstrated that strain M1 was transported more efficiently than strain R8 through columns packed with either a homogeneous silica sand or a more heterogeneous sand with iron oxide coatings.

Two experiments conducted in the tank involved monitoring transport of bacteria to wells via sampling from wells and sample ports in the tank. Bacterial numbers were determined by direct plate count. At the end of the first experiment, the distribution of the bacteria in the sediment was determined by destructive sampling and plating. The two experiments produced bacterial breakthrough curves that were quite similar even though the similarity between the two porous media was limited to first and second-order statistical moments. This result appears consistent with the concept of large-scale, average behavior such as has been observed for the transport of conservative chemical tracers. The transported bacteria arrived simultaneously with a conservative chemical tracer (although at significantly lower normalized concentration than the tracer). However, the bacterial breakthrough curves showed significant late-time tailing. The concentrations of bacteria attached to the sediment surfaces showed considerably more spatial variation than did the concentrations of bacteria in the fluid phase. This contrast between behavior in the fluid phase and on the solids is consistent with field observations by other authors and initial modeling of these heterogeneous media.

These experiments on microbial transport are among the most exciting results of our research project as they demonstrate a number of interesting features, including:

- Microbial results that are reproducible over multiple realizations of a random field, implying the potential for a large-scale averaged behavior for bacteria similar to that observed for transport of conservative chemicals.
- Consistency between the observed bacterial breakthrough and those predicted through the results of preliminary modeling using a kinetic attachment / detachment model.
- Large spatial variability in attached bacteria counts versus relatively uniform (spatially) bacteria counts in the fluid phase.
- Long-term, relatively uniform concentration of bacteria in the fluid phase following the initial breakthrough of the bacteria.

The results of these experiments are reported in Silliman et al. (2001)

JOURNAL PUBLICATIONS DERIVED FROM THIS PROJECT

The following journal publications resulted from this research project:

- Silliman, S.E., R. Dunlap, M. Fletcher and M.A. Schneegurt, "Bacterial transport in heterogeneous porous media: Observations from laboratory experiments", *Water Resources Research*, 37(11), 2699-2708, 2001.
- Silliman, S.E., "Laboratory study of chemical transport to wells within heterogeneous porous media", *Water Resources Research*, 37(7), 1883-1892, 2001.
- Silliman, S.E., and L. Zheng, "Comparison of observations from a laboratory model with stochastic theory: Initial analysis of hydraulic and tracer experiments", *Transport in Porous Media*, 42(1/2), 85-107, 2001.
- Cole, B.E., and S.E. Silliman, "Utility of simple models for capture zone delineation in heterogeneous unconfined aquifers", *Ground Water*, 38(5), 665-672, 2000.
- Zheng, L., and S.E. Silliman, "Estimating the variance and integral scale of the transmissivity field using head residual increments", *Water Resources Research*, 36(5), 1353-1358, 2000.
- Berkowitz, B., H. Scher, and S. Silliman, "Anomalous transport in laboratory-scale, heterogeneous porous media", *Water Resources Research*, 36(1), 149-158, 2000. {Correction appeared 36(5), 1371, 2000.}
- Zheng, L. and S. Silliman, "Estimating the theoretical semivariogram from finite numbers of measurements", *Water Resources Research*, 36(1), 361-367, 2000.
- Silliman, S.E., L. Zheng, and P. Conwell, "The use of laboratory experiments for the study of conservative solute transport in heterogeneous porous media", *Hydrogeology Journal*, 6, 166-177, 1998.
- Silliman, S.E., and S. Caswell, "Observations of measured hydraulic conductivity in two artificial, confined aquifers with boundaries", *Water Resources Research*, 34(9), 2203-2213, 1998.
- Silliman, S.E., and C. Frost, "Monitoring of the hydraulic gradient using a three-point estimator", *Journal of Environmental Engineering*, 124(6), 517-523, 1998.
- Conwell, P.M., S.E. Silliman, and L. Zheng, "Design of a piezometer network for estimation of the variogram of the hydraulic gradient: The role of the instrument", *Water Resources Research*, 33(11), 2489-2494, 1997.
- Cole, B., and S. Silliman, "Capture zones for passive wells in heterogeneous unconfined aquifers", *Ground Water*, 35(1), 1997.
- Cole, B. and S. Silliman, "Estimating the horizontal gradient in heterogeneous, unconfined aquifers: Comparison of three-point schemes", *Ground Water Monitoring and Remediation*, Spring, 84-91, 1996.
- Silliman, S.E., "The importance of the third dimension on transport through saturated porous media: Case study based on transport of particles", *Jour. of Hyd.*, 179, 181-196, 1996.
- Silliman, S.E., "Sample support in a single fracture: Considering the definition and control of the support of a water sample", *Geophys. Res. Letters*, 22(11), 1145-1147, 1995.
- Silliman, S.E., "Particle transport through two-dimensional, saturated porous media: influence of physical structure of the medium", *Jour. of Hyd.*, 167, 79-98, 1995.
- Silliman, S.E., C.C. Cady, and K. Snyder, "Application of a UV-Curing resin to hydrodynamic studies in porous media", *Ind. Eng. Chem. Res.*, American Chemical Society, 33(8), 1997-2001, 1994.

STUDENTS PARTICIPATING IN THIS PROJECT

Dr. Li Zheng, Completed Ph.D. under this project

Dr. Bryce Cole, Completed Ph.D. under this project

Mr. Peter Conwell, Completed M.S. under this project

Ms. Gloria Mantz, Completed portion of M.S. degree under this project

Ms. Antonia Dey, Pursued graduate studies under this project

Mr. Stephen Caswell, Completed undergraduate research project (and paper) under this project

Mr. Craig Frost, Complete undergraduate research project (and paper) under this project

Ms. Karen Snyder, Completed undergraduate research project (and paper) under this project

Application of a UV-Curing Resin to Hydrodynamic Studies in Porous Media

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Laboratory techniques which have been applied to visualization of flow and transport in porous media include light transmission, matched index of refraction methods, NMR techniques, and use of epoxies. Within the present study, a commercially available UV-curing resin is applied to capturing transport behavior in porous media. This technique provides several capabilities including rapid solidification of soil structure/fluid distribution at preselected locations without stopping flow and the ability to cure the structure of fluid interfaces. The resin cures rapidly, exhibits minimal change in volume during curing, and does not show substantial local flow alteration during curing (despite relatively large increases in temperature during the curing process). This resin holds promise to enhance our ability to study flow and transport through porous media, including the study of particle transport, hydrodynamic dispersion, fluid distributions in multiphase flow, and fingering during infiltration.

Introduction

Within the hydrologic community, increasing attention is being paid to the details of flow and transport within saturated and partially saturated porous media. Subjects of interest include scaling of measurements, flow in unsaturated and/or multiphase systems, hydrodynamic dispersion, and colloid/microbial transport (e.g., through geological sediments). Interest in the details of flow and transport through porous media has led to a variety of experimental efforts addressed to the problems of recording precise behavior within controlled media. Included among these are light transmission techniques, matching of index of refraction of the medium and carrying fluid, and the use of fluids which polymerize *in situ*.

A number of authors have suggested techniques for visually inspecting experimental media in order to record transport mechanisms. These techniques range from simple photographic records of experiments to light transmission through glass bead media (Hoa, 1981; Glass et al., 1989; Norton, et al., 1993). Light transmission techniques (e.g., Glass et al., 1989) allow measurement of flow and transport properties over large regions.

Recent work by Conrad et al. (1992) and Wilson et al. (1993) has led to the development of a technique in which artificial slices of a porous medium are created through etching of glass plates. This technique has allowed these authors both to create intricate, two-dimensional examples of porous media and to carefully examine a variety of phenomena including blob formation in residual saturation (Conrad et al., 1992) and the distribution of bacteria in multiphase systems (Wilson et al., 1993).

Other authors have created artificial media for the specific purpose of visualizing flow. Mannheimer (Richard Mannheimer, Southwest Research Institute, personal communication, 1993) has worked with transparent slurries to create transparent porous media through which the movement of chemicals can be traced using visible light. In a similar approach, researchers such as Peurrung and Kulp (1992) and Montemagno (Wang, 1994) are using media and fluids with matched indices of refraction to allow visual analysis of flow and transport through porous

structures. These techniques hold significant promise in allowing continuous recording of such phenomena as the development of a contact surface in multiphase flow (e.g., between a solid and a fluid or between two fluids), the movement of color tracers through a porous medium, and the movement/trapping of particles within a porous medium.

Mayer and Miller (1992, 1993) review a number of efforts in which a styrene liquid is utilized as an agent which will polymerize after removal of an inhibitor and/or addition of an initiator. This approach has been used extensively by a number of authors including Wilson et al. (1989), Wardlaw and Yu (1988), and Chatzis et al. (1984). Mayer and Miller (1992) extend the use of the styrene polymerization process through curing of the polymer using γ -radiation.

In addition to the methods described above, several authors have attempted to reproduce fracture patterns and pore microstructure through use of Wood's metal (e.g., Cody and Davis, 1991; Pyrak-Nolte, 1991). By injecting molten Wood's metal into a sample (e.g., a piece of coal) and allowing it to harden, the internal pore structure of the sample may be examined by dissolving the surrounding rock matrix with acid and/or examining the distribution of the hardened woods metal through CAT (computer-aided tomography)-scan methodologies.

The present project was motivated by a desire to capture geometry and transport characteristics of a porous medium subject to the following restrictions:

- (i) No addition of secondary fluids to the flow field is permitted after the start of an experiment.
- (ii) The technique should allow local measurement without requiring that the entire flow field be cured or destroyed.
- (iii) Local curing should not dramatically alter the flow field.
- (iv) The technique should be applicable to three-dimensional media.

The technique developed involves application of a commercially available resin which is cured through exposure to ultraviolet (UV) light. We have applied this resin to a number of investigations including examination of particle transport, hydrodynamic dispersion, and multiphase flow. It provides several new capabilities which, when combined with the existing tools described above,

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Figure 1. Cured sample of UV resin in no. 10 mesh glass beads after 10 s of curing.

allow for expanded flexibility in the design of experiments for the study of microscopic and macroscopic behavior in porous media.

UV-Curing Resin

Developed for application in the coating industry, the resin used was UV-2035-clear, deep-curing UV resin purchased from the Polychem Corporation (Cranston, RI). The resin has a reported viscosity of 200 cP at 24 °C and a specific gravity at 20 °C of 1.10. The resin is also reported by the company to act as a Newtonian fluid. The boiling point is reported as 400 °F, the vapor pressure is 0.01 mmHg, and the solubility in water is 0.055% by weight at 20 °C. Curing time is reported as 5 s under a UV source measuring 10 000 mW/cm². Based on the Material Safety Data Sheet supplied by the manufacturer, there are no significant health concerns in handling the resin unless there are prolonged or repeated exposures to vapors generated at high temperatures (not anticipated in the present application). [All values reported are supplied by the manufacturer. Several of these parameters have been verified within our laboratory.]

During our work, we have used a focused source of UV light with a strength of approximately 1000 mW/cm² (UVEX Model SCU110, UVEX, Sunnyvale, CA). The wand through which the UV light passed was 8 mm in diameter. Curing has been performed by exposing the resin through a variety of media, including glass, 1.2-cm-thick Plexiglas, and glass rods with a 1-mm o.d. As would be expected, curing time and curing depth are functions of the area of exposure and the type of medium through which the UV light must pass. Figure 1 shows the cured sample obtained through 10 s of direct exposure of no. 10 sieve glass beads saturated with the resin.

The resin cures to a transparent solid with little distortion with respect to light transmission. This transparent cure, as represented in Figure 1, provides a significant advantage in the application to porous media as it provides the potential to examine the details of flow within individual pores. Microphotographs of cured specimens can be accomplished relatively easily (e.g., see later section on particle transport).

Curing of the resin leads to a slight reduction in volume. During our tests, the volume reduction during curing was approximately 1.4%. During curing through Plexiglas, the resin does appear to separate slightly from the Plexiglas. However, the separation must be minimal as no liquid resin is observed to flow between the Plexiglas and cured resin following the termination of local curing. The resin is easily removed from Plexiglas following cure.

Figure 2. Temperature of resin during curing showing difference of open container of resin and resin cured in the pores of no. 10 sieve glass beads.

Liquid resin remaining within the medium can be removed using methanol.

The curing process is strongly exothermic. Figure 2 shows the results of two experiments involving the curing of resin in the vicinity of a thermistor embedded in the resin. In the first experiment, the resin was cured in an open container (containing nothing but resin). The second experiment involved curing the resin which filled the pore spaces of large-diameter glass beads (no. 10 sieve). For both experiments, the UV light was maintained for approximately 60 s (substantially longer than required to achieve local curing). The temperature increased substantially in both experiments. The lower increase in temperature in the presence of the glass beads may be anticipated from the reduction in total volume of resin present and the scatter of the UV light during the curing process. The cured volume for the resin in the glass beads (volume of the resin plus the volume of the entrapped glass beads) was larger than the cured volume in the open container (pure resin). From these results, it is estimated that temperature increase during a short-duration curing event (e.g., 5–10 s) within a glass bead medium will be on the order of 5–10 °C.

Despite the high temperatures generated during curing, the resin does not demonstrate any strong thermal conduction cells near the cure point, even in an open container of resin with a concentrated region of curing. This lack of formation of convection cells was evident in a series of experiments in which an emulsion of water and the resin was cured. No increased motion in the emulsion was observed in the vicinity of the cure area during the curing process.

Application to Particle Transport

The resin was applied initially to the study of particle transport through heterogeneous media. The experimental setup is shown in Figure 3 and involves a Plexiglas tank packed with a heterogeneous medium consisting of various sized glass beads [the region containing the medium has dimensions 35 cm (length parallel to flow) by 13.5 cm (height) by 4.8 cm (width)]. The objective of the research was to record the location of latex particles, at specific measurement times, being transported through a heterogeneous, porous medium. The experiments are performed in both two and three dimensions, and we wished to employ a method for "freezing" the particle

Figure 3. Flow cell design used for transport experiments. Flow is from left to right. The medium is packed in the center of the cell. The length of the flow cell ranges from 40 to 120 cm.

Figure 4. Photograph of cured resin and glass beads at tip of glass rod removed from flow cell after particle transport experiment.

Figure 5. Microphotograph of latex sphere captured within cured resin sample shown in Figure 4. The diameter of the latex sphere is approximately 10 μm . The glass beads surrounding this pore are seen as the dark bodies near the edges of this photograph.

location at preselected times and locations within the medium. The frozen samples containing the porous medium and the pore fluid were then to be studied to determine the location, within individual pores, of the latex particles.

In order to complete these experiments, the glass medium is saturated with the resin. Using a peristaltic pump, additional resin is added to the inflow reservoir (Figure 3) to force flow through the medium. Typical flow rates utilized were in the range of 1.0–10.0 mL/min. Although pressure across the pump was not measured, there was no obvious strain on the standard (Masterflex) peristaltic pumps utilized. Latex spheres with diameters ranging from 2 to approximately 90 μm are then added to the inflow reservoir. Outflow from the downstream reservoir is collected at regular intervals to monitor the breakthrough of the particles at the outflow from the medium. In addition, a series of 1-mm-diameter glass rods (approximately 10 cm in length) are inserted through the wall of the flow cell (utilizing compression fittings customized to fit the glass rods) with the tips located at various positions within the heterogeneous medium. As the experiment progresses (and prior to the arrival of the particles at the outflow reservoir), the UV light source is used to locally cure the resin surrounding individual glass rods. This allows the position of the particles to be locally frozen at the time of exposure.

Following completion of the experiment, the glass rods are carefully removed from the medium and the cured resin samples surrounding the tips of the glass rods are removed and analyzed. Figure 4 is a photograph of one of these cured samples. Figure 5 shows a microphotograph of a latex sphere as it was frozen during transport through the sample shown in Figure 4. Within this figure, the latex particle is the sphere apparent in the photograph. The nearest glass beads comprising the porous medium are in the dark regions near the edges of the photograph. The latex particle is approximately 10 μm in diameter. Identification of the particles is enhanced through the use of fluorescent particles and color photography, but Figure 4 illustrates the potential for identification. In this situation, the particle is seen to be located at the center of the pore (a substantial distance from the surrounding glass beads).

Thus the resin provides the potential to allow the researcher to identify both the location of selected particles during flow and the interaction of multiple particles in the flow field (note that the particle in Figure 4 is moving as an individual particle rather than as a particle cluster). By curing different rods at different times, a history of particle location is obtained. It is noted that because sample size can be controlled by adjusting the period of UV exposure, the influence of curing a local sample on the

Figure 6. Contact surface between air, resin, and glass bead after curing during an infiltration experiment.

flow through the medium can be predicted and controlled through adjustment of exposure time.

Although all images shown here involve use of the resin with glass beads, we have also utilized the resin in relatively high organic content silty sand (a dark, muddy sediment which did not allow significant light penetration). Although the radius of cure was reduced in this low-light-transmittance medium, curing was still effective. As would be expected, the volume of cure is closely related to the depth of light penetration. Hence, curing is most rapid and deep in glass beads and clean quartz sands, and is reduced in both speed and depth in media containing a significant opacity.

Extension of the Application of the Resin to Additional Research Questions

Although the UV-curable resin was investigated primarily for our work in particle transport, it has been shown to have significant potential for other applications of interest to those working in porous media. The cured resin sample shown in Figure 6, for example, has captured the shape of the contact surface and the contact angle of an interface between air, resin, and glass beads during a transient infiltration event. The experiment involved introduction of the resin at a point source into an initially "dry" glass bead medium. Although the details of this experiment are beyond the scope of this paper, this figure shows the potential for capturing detailed interface structure during transient events.

A similar application involving characterization of a water/resin emulsion (water and the resin are immiscible) has proven quite successful. Our initial results indicate that if curing is accomplished carefully (to avoid overexposure which might lead to significant thermal effects), then the contact surfaces do not show significant deformation during the curing process, thus allowing analysis of emulsion distribution and surface characteristics. Certainly, this must be examined in greater detail prior to basing measurements on this technique. However, this application may prove useful both in understanding the evolution of the transient behavior of emulsions and in characterizing the transient behavior in immiscible displacement experiments in porous media.

Finally, the manufacturer has provided a number of dyes which can be utilized with the resin. Although our initial efforts with these dyes indicates that great care must be taken to avoid modification to the resin viscosity, these dyes have shown significant promise for detailing hydrodynamic dispersion in porous media. Through displacement of clear resin with dyed resin, we have been

able to rapidly cure regions of the invading front. Examination of the cured samples allows analysis of the hydrodynamic aspects of dispersion at a variety of scales including detailed distribution of dyes at the pore scale, distribution of the invading front at the scale of several pore lengths, and observation of the behavior of the front at macroscopic and/or megascopic scales. As such, this resin has the potential to provide the detailed data sets required for consideration of the scaling and averaging theories recently appearing in the literature [refer, for example, to the review paper by Cushman (1986) and more recent work on dimensional dispersion theory such as that by Wheatcraft and Tylor (1988) with further discussion in Tylor and Wheatcraft (1992)].

Discussion

Within this paper, a number of applications of a commercially available UV-curing resin to experimental characterization of flow and transport in porous media have been discussed. The applications include detecting particle locations during transport through porous media, capturing contact angles and surface features in unsaturated and immiscible displacement experiments, and characterizing hydrodynamic dispersion at a number of scales. As such, this resin, developed for a significantly different application, shows significant promise as a tool in visualizing mechanisms of flow and transport in porous media.

Advantages of the use of this resin include the clarity of the resin upon curing and the speed of curing. The clarity allows visual analysis of medium structure, particle locations, etc., throughout relatively thick cured samples. The speed of curing allows the development of an experimental design in which samples are collected at essentially instantaneous points in time for analysis of particle transport or hydrodynamic dispersion. The resin also can be cured through glass rods, allowing point curing deep within a medium. This increases the potential for examination of structures in three-dimensional media. Further, the resin cures without requiring addition of initiators or secondary chemicals.

In our initial studies of multiphase and unsaturated systems, it appears that the cured resin maintains the surface and contact angle characteristics present prior to cure. This conclusion is supported by the minimal change in volume observed during cure. If true, this technique could provide a means of obtaining critical data for the development of the theory of multiphase (and unsaturated) flow through porous media. However, additional work is required to verify this hypothesis.

The resin also demonstrates several limitations and/or disadvantages. Substantial heating has been observed during extended exposure to UV light. The viscosity of the resin is substantially greater than that of water, thus increasing both viscous stress and the pressure gradients present within an experimental design as compared to the stress and gradients present when using water at the same flow rate. As discussed above, the cure of the resin is also limited to applications in which UV light may pass into the porous medium. Thus curing was limited in the silty sand medium to approximately 0.5 cm. Perhaps most significantly, the resin appears dramatically different than water in terms of chemistry. As a result, its application may be limited for problems involving adsorption and chemical reactions. Further, it is doubtful at the present time that this resin will have significant use in experiments involving live bacteria or viruses (the resin is reported on the MSD sheet to be difficult to biodegrade).

It is anticipated that this resin would work well in combination with a number of the other visualization techniques described earlier. Direct visual recording of transport through porous media (e.g., through the side wall of an experimental flow cell) is possible through use of the dyes available. Further, it is anticipated that light-transmittance methods (e.g., Glass et al., 1989) could be easily adapted to the use of the resin (requiring only recalibration of the instrument and characterization of changes in light transmittance during curing). It is also conceivable that an index of refraction matched medium could be located to allow velocimetry techniques to be applied prior to curing (e.g., Peurrung and Kulp, 1992).

Conclusions

In conclusion, an application has been discussed where a commercially available UV resin has been applied under conditions not originally intended in the product development. However, the resin represents a significant tool for researchers to utilize in the characterization of flow and transport through porous media. As a stand-alone tool, the resin allows detailed, three-dimensional characterization of various parameters including the structure of the medium, the physical characteristics of internal interfaces, distribution of an invading front, and distribution of particles during transport. In combination with some of the other visualization techniques described by other authors, the resin increases the flexibility and capability of the researcher interested in collection of data on mechanisms influencing flow and transport.

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Particle transport through two-dimensional, saturated porous media: influence of physical structure of the medium

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Abstract

Recent work in the transport of particulate matter (e.g. bacteria, viruses, colloids, mineral grains, etc.) through saturated porous media has indicated that the physical structure of the porous medium plays an important role in both the ability of the particles to be transported and the distribution of the particles deposited within the medium. The present paper addresses the influence on particle transport within saturated porous media of high-permeability pathways and contacts between different grain size distributions. The experiments discussed in the present paper were conducted in two dimensions. The porous media were constructed from glass beads and the tracer utilized consisted of latex spheres in the size range 2–90 μm (2–30 μm in most experiments). Among the media designs investigated were simple layers in which flow was parallel to layers, simple layers in which flow was at an angle to layers, a simple form of heterogeneity including non-parallel layers and disjoint inclusions, and a complex heterogeneity involving three interconnected permeabilities. Results from these experiments demonstrated that significantly greater numbers of particles were deposited at media contacts where water moved from larger to smaller diameter glass beads. Further, it was observed that outflow concentrations of particles for media containing continuous flow paths within the coarse glass beads were substantially higher than outflow concentrations of particles for media containing a number of interfaces between different-sized glass beads. Specifically, a comparison of the results for a medium in which flow was parallel to layers with results from a medium in which flow was at an angle to the layers led to the observation of higher concentrations of latex particles in the outflow from the former medium and high concentrations of particles remaining in the region of layer contacts within the latter medium. Similar observations were made in a comparison of the simple and complex heterogeneities. Based on these results and prediction of straining based on the filtration literature, it is argued that the enhanced deposition observed near contacts results from sorption leading to increased straining in regions of lower uniformity in the glass bead distribution. These results demonstrate the importance of particle transport along high-permeability pathways and the enhanced deposition which may occur at contact surfaces within heterogeneous porous media.

1. Introduction

The movement of particles through a saturated porous medium is of interest in a number of fields including groundwater flow and transport, transport in petroleum reservoirs, analysis of filtration systems, and design of column reactors. A number of mechanisms influence particle transport through porous media, including reaction to physical heterogeneities (e.g. spatially varying hydraulic conductivity and/or pore geometries), adsorption, motility, and changes in the flow field owing to particle-induced changes in the permeability distribution (see, e.g. McDowell-Boyer et al., 1986; McCarthy and Degueudre, 1993). Within the present study, the influence of physical heterogeneity on particle transport through saturated, two-dimensional media is studied experimentally. Although applicable in a number of fields involving various types of porous media, the following literature review is focused primarily on the literature involving particle transport in groundwater systems, as this was the work which motivated the present study.

Over a number of years, substantial work has been presented in the groundwater literature demonstrating that advective-dispersive transport of conservative, dissolved chemicals in porous media is strongly dependent on the distribution of the hydraulic conductivity (e.g. Schwartz, 1977; Gelhar et al., 1979; Smith and Schwartz, 1980; Silliman and Simpson, 1987; Neuman, 1993). From this work, the critical effect of physical heterogeneity on transport was identified and, for conservative, dissolved chemicals, theoretical models were developed describing this transport based on statistical or fractal descriptions of the hydraulic conductivity or velocity fields.

More recently, researchers have applied the general concepts derived from the study of the transport of conservative chemicals to the study of the major mechanisms which influence particle transport in porous media (e.g. Mills et al., 1990; Harvey and Garabedian, 1991; McCarthy and Degueudre, 1993; Harvey et al., 1993;). In the work by Mills et al. (1990), for example, a column consisting of concentric cylinders of two different permeability porous media was used to study the movement of bacteria in the presence of a preferred hydraulic pathway. These workers showed that the presence of a high-permeability finger substantially aided in the transport of the bacteria.

Harvey et al. (1993) completed a detailed study of the simultaneous transport of a conservative chemical (bromide), latex particles, and bacteria through a natural sediment (in the field) and a reconstructed medium (in the laboratory). These workers concluded, in part, that despite different mechanisms influencing transport of latex particles in the field as compared with mechanisms influencing transport in laboratory columns, one major influence on transport was the heterogeneity of the medium through which the particles were transported. Further, there was some suggestion by these workers that early arrival of their microorganisms was related to the presence of extended preferred pathways consisting of horizontally extensive zones of media with similar conductive properties.

Powelson et al. (1993) also documented the importance of preferred pathways for virus migration in a study of the infiltration of viruses with wastewater. It was also

suggested in that study that interfaces within the medium between different grain sizes may play an important role in the distribution of viruses within the subsurface.

Based on the work of Mills et al. (1990), Harvey et al. (1993), and Powelson et al. (1993), it can be argued that physical heterogeneity (i.e. spatial variations in the size distribution and hydraulic conductivity of the medium) plays a significant role in the transport and deposition of particles within a porous medium. The present work contributes to this study of particle transport through a series of controlled laboratory experiments in which the movement and deposition of latex particles are monitored in media for which both the degree of continuity of high-permeability zones and the dominance of intermedia contacts are varied. Efforts are made in the present work to reduce the relative influence (relative to the hydrodynamic influences) of mechanisms such as motility, growth, reproduction, and mechanical sticking to surfaces (e.g. through biological filaments), through the use of relatively large latex particles (2–90 μm) to be transported and relatively simple media (glass beads). As is demonstrated below, sorption remained a significant mechanism in these experiments. Although the use of relatively large latex particles reduces the direct transference of these results to much smaller colloids, bacteria, and viruses, these experiments serve to illustrate the strong influences which preferred flow pathways and media contacts may have in the generic particle transport problem. Application to other media and other particles will require consideration of the relative length scales involved.

2. Equipment and experimental design

All experiments described in this paper were run on two-dimensional flow cells as shown in Fig. 1. Constructed of Plexiglas and sealed with silicon glue, these flow cells allow flow across the experimental medium to be maintained at a constant rate (constant flux inflow and constant hydraulic head outflow). The fluid was added to the inflow reservoir using a peristaltic pump, and outflow was maintained at a constant hydraulic head through use of a constant head overflow. The inflow reservoir was mixed during tracer introduction, whereas the outflow reservoir was mixed throughout the experiment. All sampling of outflow was performed from the outflow mixing loop. The combined volume of the outflow reservoir and mixing loop was minimized to minimize dilution within this reservoir. Tests of the mixing within this reservoir demonstrated that it had minimal effect on the recorded outflow concentrations.

The experimental media were constructed between the soil screens. All experimental media were constructed of technical quality glass beads. Glass beads were selected for this initial study to allow control over placement and characterization of hydraulic parameters and to reduce uncertainties in the interpretation of results which might be related to irregularities in the geometry of individual grains. Three size ranges were used for most experiments: (1) 5–7 mesh (2.8–4.0 mm), (2) 8–12 mesh (1.7–2.36 mm), and (3) 16–20 mesh (0.85–1.00 mm). The saturated hydraulic conductivity of each of these bead sizes was measured through a simple Darcy experiment and determined to

Fig. 1. Flow cell design. As shown, flow goes from right to left. Inflow is controlled through constant rate injection. Outflow is controlled through constant head overflow. The inflow reservoir has a mixing loop to allow introduction-mixing of tracer within this reservoir. The outflow reservoir has a mixing loop to allow collection of sample with average outflow concentration. Interior dimension of the soil compartment is approximately 4.8 cm (width) by 13.5 cm (height) by 35 cm (length parallel to flow).

be 1.20 cm s^{-1} , 0.90 cm s^{-1} , and 0.54 cm s^{-1} for the 5–7 mesh, 8–12 mesh, and 16–20 mesh, respectively. Henceforth in this paper, these media will be referred to as coarse (5–7 mesh), medium (8–12 mesh), and fine (16–20 mesh). It is noted that these three sizes would all be considered relatively coarse in normal soil classification schemes (i.e. all would be considered sands or gravels).

Packing was performed with the top wall of the cell removed. The glass beads were added in thin (less than 1.0 cm) layers with the glass beads poured through approximately 1.0 cm of water. Packing was guided by predesigned arrangement of the media, to produce a heterogeneous porous medium with appropriate flow properties. Where the medium was complex, MODFLOW (McDonald and Harbaugh, 1988) was used to analyze the flow field, thus guiding medium design and post-experiment sampling. Short circuiting along the top wall of the cell was prevented through addition of a square plastic lattice along this wall (see Fig. 1).

Except where noted, the fluid used for packing was reverse osmosis (RO) water modified with sodium chloride to match the density of the latex particles used as tracer (density of latex particles, 1.05 g cm^{-3}). All water required for an experiment was prepared ahead of time, to insure uniform density throughout the experiment.

As noted in the previous paragraph, the primary tracers used in this work were latex particles in various size ranges between 2 and approximately $30 \mu\text{m}$. This size range was selected both to reduce the influence of diffusion on the particle path and to allow samples to be analyzed with the equipment available in our laboratory (currently limited to a $400\times$ trifocal microscope). Mixtures of these particle sizes

were suspended in a small quantity of the previously prepared water and then injected-mixed into the inflow reservoir. Prior to introduction of the latex particles, a dye tracer was used for each packing to allow visual inspection of the transport routes taken by the flow and to check for any 'short circuits' along the walls of the flow cell. The dye tracer also allowed visual inspection of mixing in the inflow and outflow reservoirs.

A number of different media packings were investigated. Four packings, as shown in Fig. 2, were selected for presentation here, to offer insight into different transport pathways. The first design involves a simple layered system with flow parallel to layers. The second design contains approximately the same percentage of coarse, medium and fine glass beads as in the first packing, but packed in slanted layers. The third design consists of a variation on a simple layer containing a non-horizontal layer and two inclusions. Only the coarse and medium glass beads were used in this design. The final design involved a relatively complex heterogeneity containing a number of contacts between glass bead sizes. This design resulted in a flow field for which no flow line could cross the medium entirely within the coarse glass beads.

Each experiment was run through the following steps:

- (1) The flow cell was packed and sealed.
- (2) Flow was established by injecting into the inflow reservoir at a constant rate and maintaining the outflow reservoir at a constant head.
- (3) A short pulse of dye was introduced to the inflow reservoir. If the dye indicated short circuiting around the edges of the flow cell, the experiment was terminated and the cell repacked.
- (4) After the dye was flushed from the entire length of the experimental medium, particles were added to the inflow reservoir and allowed to enter the medium for approximately 20–30 min (varied by experiment). The inflow reservoir was then flushed with water without particles, thus creating a 'pulse' of particle tracer.
- (5) After a number of pore volumes had passed through the experimental medium, flow was terminated.
- (6) The medium was then destructively sampled (cores of glass beads and the pore fluid were collected). The volume of an individual core was approximately 8 cm^3 .

Outflow and destructive samples were counted using a standard trifocal microscope at magnification $400\times$. Counting was performed in Sedwich-Raptor cells. To reduce the time required to complete individual counts, the counts were divided into two discrete size ranges (rather than attempting to create histograms over a number of size ranges). The two sizes were particles less than $7.0 \mu\text{m}$ ($7.5 \mu\text{m}$ in two of the experiments) and particles greater than $7.0 \mu\text{m}$. This size (7.0 or $7.5 \mu\text{m}$) was selected based on a natural break in the size distribution of the source of particles used.

One concern which was noted with respect to the outflow counts was that the outflow reservoir had a non-negligible volume. As a result, outflow from the medium was mixed (and diluted) with the water in the outflow reservoir. The influence of mixing within the outflow reservoir can be estimated through a simple mixing model.

Media Pattern #1: Simple Layers

Media Pattern #3: Simple Heterogeneity

Media Pattern #2: Slanted Layers

Media Pattern #4: Complex Heterogeneity

Fig. 2. Four heterogeneous distributions of glass beads discussed in the present paper. Although additional media have been examined, the results from these four designs provide insight into the general results obtained to date. The white regions represent the large glass beads ($K = 1.20 \text{ cms}^{-1}$), the dotted regions represent the medium glass beads ($K = 0.90 \text{ cms}^{-1}$), and the dashed regions represent the small glass beads ($K = 0.54 \text{ cms}^{-1}$). In all packings shown, flow went from left to right.

For the experimental design utilized and the flow rates used in each of the experiments discussed below, the difference between outflow counts from the medium and outflow counts from the reservoir were shown to be insignificant. As a result, all counts reported below are outflow counts from the reservoir without correction for the mixing volume.

3. Results

3.1. Packing 1

The particles appearing in the outflow reservoir for the experiment involving flow parallel to layers resulted in a relatively smooth breakthrough curve with little evidence of tailing. Further, there was little separation by size observed for the outflow results in this experiment, and the percentage of particles greater than $7.5\ \mu\text{m}$ in the outflow was approximately equal to that in the tracer source. The outflow breakthrough curve is shown in Fig. 3(a). The breakthrough curve separated by particle size is shown in Fig. 3(b). The volumetric flux for this experiment was $3.6\ \text{ml min}^{-1}$ and the source concentration was approximately $400\,000\ \text{particles ml}^{-1}$.

A number of particles were deposited within the medium during this experiment, with the concentration of deposited particles declining with distance from this reservoir. Fig. 4(a) shows the particle counts per milliliter for several locations within the glass beads. Further, although there was no discernible separation of particle size in the outflow counts (as noted above), there was a distinct pattern of separation within the destructive samples. Specifically, as shown in Fig. 4(b), the percentage of particles less than $7.5\ \mu\text{m}$ increased with distance from the inflow reservoir.

3.2. Packing 2

The outflow concentrations for the slanted layers were significantly less than those observed when flow was parallel to layers. As illustrated in Fig. 5, the concentration never exceed $700\ \text{particles ml}^{-1}$. The flow rate through the medium was approximately $2.9\ \text{ml min}^{-1}$ with an inflow particle concentration of approximately $250\,000\ \text{particles ml}^{-1}$. Nearly all particles observed in outflow samples were of a size less than $7\ \mu\text{m}$.

As the number of particles appearing in the outflow reservoir was relatively low for the second packing, it was not surprising that the number of particles deposited within the medium was high relative to the first packing. In addition to higher numbers of deposited particles, there was also a significant change in the distribution of the deposited particles for this experiment (as compared with the first packing). As shown in Fig. 6, there is a distinct increase in the concentration of particles at several locations along the contacts between the coarse and the medium glass beads, and again along the contact between the medium and the fine glass beads. Further, the concentrations of particles along these contacts are significantly higher than

(a)

(b)

Fig. 3. Particles per milliliter recorded in samples collected from the outflow reservoir for the first medium design (simple layers with flow parallel to layers). (a) Total particle counts; (b) counts separated by size (over $7.5 \mu\text{m}$ and under $7.5 \mu\text{m}$).

those observed at any location in the first packing despite the lower source concentration.

As with the result for Packing 1, the percentage of particles which are considered small (in this case less than $7 \mu\text{m}$) increases significantly with distance from the inflow reservoir. As shown in Fig. 6(b), these results indicate that the deposited particles are

Fig. 5. Particles per milliliter recorded in samples collected from the outflow reservoir for the second medium design (slanted layers). Owing to low number of counts, outflow counts are not separated by size.

present, the reason for this high concentration has not been determined. However, experimental error is a possibility.

For this experiment, the percentage of deposited particles with a size less than $7.5\ \mu\text{m}$ tended to increase with distance from the inflow reservoir. As shown in Fig. 8(b), the increase is particularly apparent in regions where flow passes from the large into the medium glass beads. The deposited particles are generally underrepresented in particles less than $7.5\ \mu\text{m}$.

3.4. Packing 4

Particle counts in the outflow reservoir for the fourth packing were generally low. As shown in Fig. 9, the counts were consistently less than $1000\ \text{particles ml}^{-1}$ (for a flow rate of approximately $2.7\ \text{ml min}^{-1}$ and an inflow particle concentration of approximately $350\,000\ \text{particles ml}^{-1}$). As with the slanted layer medium, the majority of the outflow counts were of size $7.0\ \mu\text{m}$ or less.

The distribution of deposited particles within this last packing was relatively complex. As shown in Fig. 10(a), the particle counts tended to reflect the structure of the medium. Particular details of interest in this rather complex result are the high particle counts in the region near the inflow (labeled 'A' in the figure) where flow went from medium glass beads into the fine glass beads, and in region near the bottom (labeled 'B' in the figure) where flow went from the coarse into the medium and fine

Fig. 6. Particles in samples removed from glass beads after termination of slanted layer experiment. Counts are in the pore fluid after the sample was vigorously shaken in sample bottle. (a) Total counts (hundreds of particles per milliliter); (b) Per cent counts less than 7 μm (source contained approximately 72% particles less than 7 μm).

glass beads. In both of these regions, high particle counts coincide with regions where water flows across a contact from larger glass beads to smaller glass beads.

Despite this significantly more complex structure of the distribution of counts of deposited particles, the general trend in particle size distribution is once again for the percentage of small particles to increase with distance from the inflow reservoir and for the counts generally to be underrepresented in small particles near the inflow reservoir. These results are shown in Fig. 10(b).

4. Analysis of results

4.1. Preferred pathways

Analysis of the results for the simple layers and the simple heterogeneity indicates that the observed outflow curves are consistent with particle transport occurring primarily within the coarse glass beads. For the experiment in which the medium was layered parallel to flow, one can estimate the volume of flow moving through each of the three layers (assuming no vertical communication between layers). Using the dimensions for the system shown, it is estimated that approximately 0.93 ml min^{-1} is being transported through the coarse glass beads. Given the layer thickness of 1.5 cm, width of the flow cell of 4.8 cm, a flow distance from inflow to outflow of

Fig. 7. Particles per milliliter recorded in samples collected from the outflow reservoir for the third medium (simple heterogeneity). (a) Total counts per milliliter; (b) counts separated by size (over 7.5 μm and under 7.5 μm).

Fig. 8. Particles in samples removed from glass beads after termination of experiment on simple heterogeneity. Counts are in the pore fluid after the sample was vigorously shaken in sample bottle. (a) Total counts (hundreds of particles per milliliter); (b) per cent counts less than 7.5 μm (source contained approximately 50% particles less than 7.5 μm).

approximately 35 cm, and an assumed porosity of 30%, the travel time from inflow to outflow is estimated to be approximately 84 min. This agrees well with the calculated center of mass of approximately 80 min based on the observed outflow curve. Travel time in the medium glass beads is estimated to be approximately 110 min and travel time in the fine glass beads is estimated to be 400 min. No obvious secondary peaks were observed at these later times.

A similar analysis on the simple heterogeneity provides an estimate of travel time in the coarse zone of approximately 130 min as compared with an observed center of mass of slightly greater than 120 min. As shown in Fig. 7(a), there is evidence of a lower secondary peak after 150 min for this experiment. Based on the distribution of particles remaining within the medium after the experiment, it is believed that this secondary peak is related to flow around and through the inclusions within the coarse layer rather than flow directly through the layer of medium glass beads.

Based on these two analyses, it would appear that the primary route for particles from the inflow reservoir to the outflow reservoir is through the coarse glass beads. This hypothesis is further supported by the results from the complex heterogeneity and the slanted layers. In the slanted layers, few particles traversed the medium despite a direct path through the fine glass beads. In the complex heterogeneity,

Fig. 9. Particles per milliliter recorded in samples collected from the outflow reservoir for the fourth medium design (complex heterogeneity). Owing to low number of counts, outflow counts are not separated by size.

few particles reached the outflow reservoir despite the presence of direct flow routes through both the medium and fine glass beads.

4.2. Deposition at contact surfaces

The results presented indicate that deposition of particles is enhanced in regions surrounding contacts between different-sized glass beads. For example, particle counts in the slanted layers (Fig. 6(a)) indicate that the concentration of particles deposited within the medium in the regions immediately surrounding the contacts between layers is significantly higher than the concentration observed in situations where flow is parallel to layers (e.g. the results for Packing 1 shown in Fig. 4(a)). Further, these counts are higher than those observed immediately down flow from these contacts and in the regions nearer the inflow reservoir. This result implies concentrations of the particles at the coarse-medium glass bead contact and at the medium-fine glass bead contact greater than would be expected in any of these media in the absence of a contact. This increase occurs despite the fact that some particles are capable of moving beyond these contacts (as evidenced both by the particle deposited downstream of this contact and the particle counts in the outflow). It is clear that this contact has a dramatic effect on the number of particles remaining within the glass beads in this region. It may also be significant that the

Fig. 10. Particles in samples removed from glass beads after termination of experiment on complex heterogeneity. Points A and B illustrate regions of high counts associated with flow crossing a contact between different-sized glass beads. Counts are in the pore fluid after the sample was vigorously shaken in sample bottle. (a) Total counts (hundreds of particles per milliliter); (b) per cent counts less than 7 μm (source contained approximately 55% particles less than 7 μm).

concentration of deposited particles is not uniform over the contacts between glass beads.

Similar observations regarding the influence of contacts are supported by the two heterogeneous media. Review of the results for the simple heterogeneity medium (Fig. 8(a)) shows an increase in particle counts upstream of both inclusions (consisting of medium glass beads). Review of the results for the more complex heterogeneity (Fig. 10(a)) also shows significant increase in particle counts at contacts between glass bead sizes. In particular, the region labeled A in Fig. 10(a) shows high particle counts in a region where flow moves from the medium-sized glass beads into the small glass beads. A similar observation is made for the results from the region labeled B where flow moves from the coarse glass beads into a region consisting of medium and fine glass beads.

It is interesting to consider whether these results are consistent with mechanisms of adsorption and/or straining. As noted by Harvey and Garabedian (1991) and Harvey et al. (1993), an inverse relationship between the immobilization of a particle and the size of the particle is indicative of sorptive behavior, whereas a direct relationship is indicative of a straining type of behavior. The results obtained when flow was parallel to layering (Packing 1) and when flow was allowed to pass through a large region of coarse glass beads (Packing 3) showed little separation by particle size in the outflow

counts (Figs. 4(b) and 8(b)). Hence, it is difficult to differentiate between the two possible outcomes for homogeneous regions.

The outflow counts for Packings 2 and 4 were too low to determine reliably whether the percentage of large vs. small particles had shifted from the percentage in the inflow source. However, the destructive samples demonstrate variation in particle size distribution across these two media. In particular, the small particles were under-represented (i.e. the large particles were preferentially retained) in destructive samples collected from regions surrounding contacts where flow passed from larger to smaller glass beads (the percentage of small particles increases with distance from the inflow reservoir, owing to the loss of the large particles at shorter distances). Applying the arguments of Harvey and Garabedian (1991) and Harvey et al. (1993), these results indicate that straining is an important mechanism influencing these experiments.

In arguing for straining as a significant mechanism, it is appropriate to consider whether the pore sizes present in these media are of appropriate size to provide conditions for straining of the latex particles used. Review of the literature provides a means of estimating when a particle should be removed from a flow field via straining within the matrix of the medium. Matthess and Pekdeger (1985) presented a formula for the geometrical suffusion security, or η_{SG} , by which to gauge whether straining should be significant in particle transport through a porous medium. These workers indicated that η_{SG} should be greater than 1.5 for straining to be significant, where η_{SG} is given by

$$\eta_{SG} = d_m / (F_S d_K)$$

where F_S is an empirical transit factor for suffusion, which is assumed by these workers to be equal to 0.6 (this parameter is presented as a measure of the heterogeneity of the medium) and d_m is the diameter of the particle. The parameter d_K is given by

$$d_K = 0.455 U^{1/6} e d_{17}$$

where U is the uniformity coefficient (d_{60}/d_{10}) and e is the void ratio. The three parameters d_{10} , d_{17} , and d_{60} represent the grain sizes for which 10%, 17% and 60%, respectively, of the mass of a sample of the sediment is of smaller grain diameter.

In applying the numbers observed in this experiment, this equation provides η_{SG} values which are substantially too small relative to the criterion of 1.5. That is, this equation predicts that straining should not, in fact, be significant in this situation. For example, the contact between the coarse and the medium-sized glass beads would lead to a value for $F_S d_K$ on the order of 500–600 μm . For the contact between the medium- and fine-sized glass beads, the value for $F_S d_K$ would be on the order of 100–200 μm . In both cases, particles substantially greater than the 2–90 μm particles used here would be necessary to derive an η_{SG} greater than 1.5.

In attempting to interpret these results, therefore, one must look further than simple sorption (which would not be predicted to increase substantially at the contact) or simple straining. One hypothesis which appears consistent with these results involves a combination of sorption and straining in which sorption in the pores in the

mixed zone of the media contacts leads to physical straining within these pores. Specifically, it is envisioned that sorption may lead to deposition of particles along the surface of the glass beads. In regions where the pore throats are relatively small, such sorptive behavior may lead to significant reduction in the effective pore diameter (equivalently, the sorbed particle effectively becomes a solid within the medium). Because of the reduced pore diameter, the probability of bridging across the pore (or straining of a single particle) is increased, thus leading to enhanced deposition.

This conceptualized explanation of the behavior at the interfaces appears to be consistent with arguments in filtration theory. Rajagopalan and Tien (1979), for example, demonstrated that the filter coefficient (effectively the rate constant for the reduction in concentration of particles in the fluid phase) increases with the specific deposit, σ , for small values of σ . The specific deposit, σ , was defined by these workers as the ratio of the volume of particulate matter deposited within a sediment sample to the volume of the sample. As the volume of the latex particles used in the present work represents only a small fraction of the total volume of the core samples, the result reported by Rajagopalan and Tien would indicate that the deposition rate will increase as the volume of deposited particles increases. As a result, in a region where sorption might lead to increased straining (such as the layer contacts), one could argue that the local increase in σ will lead to an increased filter coefficient (which, in turn, will further increase σ).

This hypothesis leads to a secondary hypothesis regarding reversal of the flow field. Given the present media and particle sizes, one could argue that sorption is likely to be the primary mechanism of deposition in a homogeneous packing of the coarse beads. Further, no noticeable release of particles occurred following passage of the tracer front in the previous experiments. Therefore, one could argue that if flow is reversed in a homogeneous packing after the completion of the breakthrough of the particles to the outflow reservoir, few particles will be released to the inflow reservoir. If, however, the medium consists of flow across a simple series of layers (as shown in Fig. 11), then sorption and straining should occur in the region of the layer contacts. In this case, reversal of flow after completion of the breakthrough curve to the outflow reservoir might lead to transport of particles held by straining back to the inflow reservoir.

To check this second hypothesis, a series of experiments in homogeneous and layered media was completed. In each of these experiments, a standard particle tracer test was conducted to the point where no additional particles were observed in the outflow reservoir. At this point, the flow direction was reversed (the magnitude was held constant) and samples from what had been the inflow reservoir were taken. No destructive samples were collected. In the case of flow through a homogeneous medium consisting of the coarse beads, very few particles were observed in the inflow reservoir following reversal of flow. On the other hand, when the experiment was conducted on the medium shown in Fig. 11, reversal of flow led to observation of a number of particles in the inflow reservoir. Although this experiment was run qualitatively rather than quantitatively (hence actual numbers are not reported), it was felt that this outcome provided support of the argument that straining contributed to particle deposition at the contacts.

Fig. 11. Experimental medium in which flow moves perpendicular to layers. Experiment involves running a particle tracer test from left to right across the medium. Upon completion of the experiment, flow is reversed to move from right to left and the reservoir on the left is sampled for particles being transported back to this location. The white regions represent the large glass beads ($K = 1.20 \text{ cm s}^{-1}$), the dotted regions represent the medium glass beads ($K = 0.90 \text{ cm s}^{-1}$), and the dashed regions represent the small glass beads ($K = 0.54 \text{ cm s}^{-1}$).

5. Conclusions

In this paper, a series of two-dimensional experiments have been presented which demonstrate the roles played by high-permeability zones and media contacts in particle transport and particle deposition. It is argued that particles which successfully crossed these experimental media were transported primarily along flow paths passing entirely through the coarse glass beads. Further, although particles were deposited generally throughout the experimental media, the concentration of deposited particles was significantly higher in the region of contacts where flow moved from a region consisting of one size glass bead into a region dominated by smaller glass beads. It is anticipated that this increased rate of deposition at the contacts results from a combination of sorption and straining.

These results are in general agreement with previous efforts by such workers as Harvey et al. (1993) and Powelson et al. (1993) in that preferred (high-permeability) pathways played a key role in advancing particles through the flow field and that media contacts represented regions of increased deposition. In comparing our efforts with those reported by Harvey et al. (1993), it would appear that our results are more consistent with the laboratory portion of that study. In particular, the relative retention of particles in these experiments (large particles being retained in preference to small particles at short flow distances in the complex heterogeneity and slanted layers) were consistent with the laboratory results of Harvey et al. (1993), but inconsistent with their field results. As suggested by those workers, the laboratory results are more indicative of straining as a primary mechanism. This suggestion is consistent with the argument presented here that straining becomes a significant mechanism at the contacts between different-sized glass beads. That straining could be significant for

bacteria under certain field conditions has been suggested by Harvey and Garabedian (1991).

It must be stressed that these results should be extended to particles of other sizes only with caution. As noted by other workers (e.g. Rajagopalan and Tien, 1979), the mechanisms governing transport of latex spheres of size 2–90 μm are different from those governing transport of latex spheres of size less than 1.0 μm . Further, as noted in the work of Bower (1986) and Harvey et al. (1989, 1993), latex spheres will interact with the medium in a different fashion than will bacteria. Hence, the relative magnitude of the different mechanisms influencing particle transport (e.g. sorption, straining, motility, density segregation, etc.) and the characteristics of the specific particle being transported (relative to the surface chemistry of the medium) must be considered in extending the results of this (or other) studies to new media and/or other particles.

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Sample support in a single fracture: Considering the definition and control of the support of a water sample

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Abstract. Defining and controlling the support of a field measurement (the volume or area over which the measurement is averaged) are critical components of statistical and/or geostatistical analysis of a data base. For the present study, the seemingly simple task of collecting a water sample from a single fracture is used to illustrate potential difficulties in defining and controlling the support of that sample. Two definitions of sample support are considered for this problem. First, one could argue that the support is equal to the volume of water collected. Second, one could argue for a support defined in terms of the area of the fracture surface contacted by the water collected. Collection of water samples with constant volume is relatively straight forward in a field situation. Collection of a water sample which involves contact with a specified area of the fracture, however, is not as straight forward. In the case of negligible regional flow, a constant sample area can be approximated by requiring that the sample volume collected be linearly proportional to the locally averaged aperture (thus requiring an independent measure of the local aperture). In the case of regional flow with continuous sampling, a constant area within each sample can be approximated by matching the up gradient width of the capture zone of the sampler (thus requiring knowledge of the local aperture and hydraulic gradient). These examples illustrate how difficult it may be to define and control the support for even these simple measurements. Further, they illustrate that the choice of support may be closely related to the purpose for which the measurement is collected. Additional attention to the problem of defining / controlling sample supports has the potential to pay substantial dividends in the areas of scaling and field design / analysis.

Introduction

Knowledge of the support of an individual measurement (where support refers to the volume or area over which the measurement is averaged) within a sampling protocol can be important for full characterization of the data set generated. While techniques are available in the geostatistical literature for dealing with variable support, collection of samples with consistent support often simplifies the generation of spatial and/or temporal statistics. Further, recent work by other authors has shown the importance of interpreting the support volume of individual samples in order to correctly interpret

model development (e.g., Cushman, 1986) or scaling relationships (e.g., Neuman, 1990).

In the present paper, the variation in support volume for two simple field methods is presented. These two case examples are utilized to illustrate some of the difficulties one might encounter in defining and controlling the sample support of a particular measurement. Specifically, focus is placed upon how one might define different supports for the same measurement and how, through manipulation of field techniques, one might reduce variability in the support for a set of measurements.

These two case studies are intended to be simple examples of the complex problem of defining / controlling sample support. It is anticipated that consideration of the support will become increasingly important as studies of scaling relations in fracture media, similar to those considered by Neuman (1990) and others for porous media, are pursued.

The Support of a Chemical Sample

Both case studies discussed below are based on the sample support of a water sample collected from a single fracture for the purpose of characterizing some aspect of chemical quality or chemical interaction (e.g. delineation of a contamination plume or characterization of adsorptive behavior). In both cases, it is assumed that the water sample is collected via fluid withdrawal from two or more wells and/or piezometers. Further, it is assumed that the rock matrix is effectively impermeable such that the water sample is derived entirely from the fracture. The primary question addressed via these case examples is whether the support of the chemical sample can be defined and adjusted through consideration of the field procedure.

The support of a water sample, in this situation, may be viewed via at least two definitions. In the first, the support is defined as being equal to the volume of water withdrawn from the fracture. This is the support which is commonly utilized in field procedures based on sampling either at a constant rate of withdrawal (for continuous sampling) or with a constant volume sample collected at each discrete sampling period (for sampling at discrete time intervals). This support may be justified, for example, by arguing that the each sample should represent a similar percentage of the regional flow.

In the second definition, the support is equal to the area within the fracture plane from which the sample was withdrawn (herein termed "area of coverage"). This support might be justified, for example, through an argument that this support will provide consistent numbers in terms of water / rock interaction.

Certainly, one could argue with the details of either of these definitions. The point here is not to select one over the other, but to illustrate the ramifications of having more than one

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possible definition of support and the need to adjust field procedures to select between these definitions.

Practical Aspects of Sampling: Negligible Regional Flow

The difference between these two definitions, and the impact of the definition selected on sampling strategy, are developed through two case examples. The first case considered is the problem of collecting a series of discrete chemical samples from boreholes penetrating a single fracture. For this case, the regional flow rate is considered negligible and the aperture is assumed to vary spatially.

The technique employed to collect these samples will depend on a number of factors such as the diameter of the boreholes and the equipment present at the field site. For the present discussion, it is assumed that straddle packers are used to isolate the fracture within a borehole with a 5 centimeter radius. It is assumed further that the separation distance between the straddle packers is 25 centimeters. Under these circumstances, there is approximately two liters of dead volume between the straddle packers (Figure 1).

Prior to collecting a sample, one might argue that the dead volume between the straddle packers needs to be removed. Assuming for the sake of this example that removal of a single dead volume is sufficient, it is concluded that this field set up requires a minimum of two liters to be removed from the fracture prior to collecting the chemical sample.

The two supports discussed above are now considered for two locations within this fracture. In the first location, it is assumed that the local aperture is approximately constant with an average of 0.1 mm. In the second location, it is assumed the local aperture is once again approximately constant, but the separation between the walls of the fracture is reduced to an average of 0.05 mm.

Figure 1: Simple schematic of a straddle packer system for sampling from a single fracture. The volume between the packers is here considered "dead volume".

When the first definition of the support (the support is equal to the volume collected) is used, the sampling strategy at these two locations within the fracture are identical. Specifically, a constant volume (with a minimum of two liters) is pumped for each sample collected.

When the support of the sample is based on the area of coverage, sampling strategy varies with the local aperture. To simplify the presentation of this example, the assumptions that regional flow is negligible and the fracture aperture is constant are invoked. Under these assumptions, flow is symmetric around the borehole from which the sample is withdrawn, and the area of fracture from which the sample is collected will be a circle centered on the borehole. For the smaller aperture (0.05 mm) with a pumped volume of two liters, the area of coverage is 400,000 square centimeters. This provides a radius of coverage of approximately 360 cm (3.6 meters). If the same quantity is withdrawn from the borehole in the region of the larger aperture (0.1 mm), the area of coverage would be reduced to approximately 200,000 square centimeters (radius of 2.5 meters). In order to obtain the same radius of coverage (3.6 meters), four liters would need to be removed from this borehole.

For this case, then, variation in the definition of the support leads to variation in sampling strategy in the field. Specifically, utilizing the area of coverage as the measure of the support, one would need to adjust the sample volume as a linear function of the average aperture in the immediate vicinity of the sampling borehole. As this adjustment requires an estimate of the local aperture, utilizing area of coverage as the support will lead to the requirement that the aperture be estimated (e.g. through a slug test) prior to conducting sampling.

Practical Aspects of Sampling: Uniform Regional Gradient

The second case study, shown in Figure 2, involves a fracture with a spatially constant hydraulic conductivity (and aperture). A borehole is installed at the center of this flow

Figure 2: Schematic representation of regional flow in a single fracture subject to continuous local pumping. For this study, it is assumed that the left and right boundaries of the fracture are maintained at constant heads and that the upper and lower boundaries are subjected to no flow boundary conditions.

field and is pumped at the constant rate of 0.1 liters per minute. The borehole is open only to the fracture. The flow field is considered to be at steady state with respect to the boundary conditions shown in Figure 2. In this case, the relationship between the width of the capture zone (for the borehole) and the fracture aperture is considered.

The width of the capture zone at the upstream boundary is a function of the transmissivity of the fracture and the boundary conditions. The problem shown in Figure 2 is analyzed through use of MODFLOW (McDonald and Harbaugh, 1988) in combination with MODPATH (Pollock, 1989). In the case where the support is based on a constant volume of fluid removed and the boundary conditions are as shown in Figure 2, the capture zone will vary with transmissivity (or aperture) within the fracture in a fashion similar to that shown in Figure 3. As expected, an increase in the transmissivity leads to a reduction in the width of the capture zone. It is of interest to note that the width of the capture zone is negligible for transmissivities above approximately $10^{-8} \text{ m}^2/\text{s}$.

When the support volume is defined via the area of contact, it is clear, based on figure 3, that a single pumping rate from the borehole will not provide the same contact area for different apertures. Adjustment of the sampling design to account for this difference in the sample geometry is not as simple as in the case where regional flow was negligible. The problem becomes one of matching the ratio of the regional flow to the pumping rate at the borehole for every sample location. In this case, then, maintaining a constant support area will require knowledge of the transmissivity (or aperture) and the hydraulic gradient in the vicinity of the borehole.

It might be noted that this case study becomes significantly more complicated if the aperture varies as a random field. In

this case, it is likely that a constant area of coverage could be realized only in a statistical sense (e.g., a constant expected value for the area of coverage).

Discussion

These two case studies are intended to demonstrate the difficulty one might encounter in defining the support of a sample and then ensuring that the support among a group of samples is nearly constant. While one might argue as to which, if either, of these supports (volume versus area) is more reasonable for a particular sampling scenario, the point here is that consideration of the support of a given measurement (both in terms of defining the most appropriate support and in determining the support for a given measurement) should receive increasing consideration.

While these examples involved relatively simple analyses of support, such characterization techniques as hydraulic testing, tracer testing, ground penetrating radar, and interhole geophysical methods will involve supports which are more difficult to define and which involve relatively complex modeling efforts to quantify. In these situations, as in all field sampling, consideration of support should be directly related to the potential application of the data. Significant efforts at defining and quantifying sample support may pay substantial dividends in terms of reduced uncertainty within a data base, simplification of data analysis, and, potentially, increased utility of a data analysis. Such efforts are particularly important when issues of scaling are addressed through use of field data.

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Figure 3: Width of capture zone at the left boundary versus transmissivity of fracture for a single fracture subject to the boundary conditions shown in Figure 2. Pumping at the borehole is at a rate of 0.1 liters per minute.

The importance of the third dimension on transport through saturated porous media: case study based on transport of particles

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Abstract

The present study involves transport experiments in a three-dimensional, heterogeneous porous medium. Latex particles and chloride are utilized as tracers in an artificially constructed porous medium consisting of a heterogeneous mixture of glass beads. Data collected include chloride breakthrough, particle breakthrough, and particle concentrations remaining within the medium (determined through destructive sampling at the end of the experiment). The chloride breakthrough curve showed substantial dispersion, an indication of mixing among a number of different flow lines (the mixing occurred in the outflow reservoir). The outflow counts of latex particles showed that particles were successfully transported across the length of the medium. The destructive samples showed a complex distribution of particles remaining within the medium which was correlated with the structure of the medium, particularly with the high-permeability pathways. Comparison of the results from this experiment with results from previously published results from two-dimensional media demonstrate that the successful transport of particles across the medium is a direct result of connectivity of high-permeability paths in the third dimension. This observation is consistent both with the distribution of trapped particles and with numerical analysis of the flow field.

1. Introduction

A number of researchers have demonstrated the important role of hydraulic heterogeneity on chemical and/or particle transport in saturated porous media. Schwartz (1977), for example, utilized numerical analysis of a random conductivity field to demonstrate the relationship between mechanical dispersion and spatial variations in hydraulic conductivity. This early work of Schwartz has been extended by a number of workers to provide a framework from which to understand the

relationship between spatial variation (e.g. correlation structure) in the hydraulic conductivity and/or pore water velocity and the rate of spread of a tracer moving with the pore fluids (e.g. Gelhar and Axness, 1983; Dagan, 1988; Neuman, 1993).

These theoretical efforts have been supported by a number of experimental studies, including work with both chemical and particulate tracers (bacteria, viruses, colloids, and synthetic particles). Silliman and Simpson (1987), for example, completed laboratory efforts on sand media modeled after the work of Schwartz (1977). These workers showed the dependence of the dispersivity on the pattern of the heterogeneity present, and documented that a scale effect on the dispersivity could occur owing to physical heterogeneity. Sudicky (1986) and Mackay et al. (1986) discussed transport studies at the Borden site. Similarly, LeBlanc et al. (1991) provided data from a natural gradient test at the Cape Cod site. In both of these significant field efforts, experiments led to the observation that hydraulic heterogeneity had a major influence on the transport process. Harvey et al. (1993) worked under both laboratory and field conditions to compare the transport of a conservative chemical tracer, injected bacteria and latex spheres. Among the results discussed by Harvey et al. was the important influence of physical heterogeneity on the transport processes governing the movement of these tracers. Similarly, Powelson et al. (1993) demonstrated the importance of heterogeneity on particle migration. Following the transport of viruses during infiltration experiments, these workers demonstrated that viral transport was enhanced by the presence of preferential pathways, and further suggested that these pathways are related to the layered structure of the porous medium under study. Finally, Silliman (1995—henceforth termed the 'earlier paper') completed a series of experiments on two-dimensional heterogeneous media utilizing latex particles as the tracer, and demonstrated close correlation between the form of the physical heterogeneities and the transport-trapping behavior of the particles.

These theoretical and experimental studies have clearly illustrated the importance of physical heterogeneity in the transport of chemicals and/or particles. In addition to this focus on heterogeneity, there has been growing interest in the role which the physical dimension of the medium plays in the flow and transport processes. A number of workers have provided theoretical insight into the influence of dimension on fluid flow. Bakr et al. (1978), for example, compared fluid flow in one-dimensional media and three-dimensional media, demonstrating that the variance of the predicted hydraulic head at a point is strongly dependent on the dimension of the medium (specifically, it is substantially less in a three-dimensional medium than in a one-dimensional medium). In a more recent study of the effective properties of heterogeneous media, Dagan (1986) demonstrated that the effective conductivity in a two-dimensional, isotropic random field was equal to the geometric mean of the local conductivities. In comparison, Dagan argued that the effective conductivity in three dimensions increased to the geometric mean multiplied by $1 + (\sigma_L^2/6)$, where σ_L^2 is the variance of the natural log of the hydraulic conductivity (this result was also discussed by Gelhar and Axness (1983)). These results illustrate the importance of the third dimension in defining the flow field through a heterogeneous medium.

Based on this theoretical work, the observations of Powelson et al. (1993) on preferred pathways, the results reported in the earlier paper, and generic study of

percolation processes in two and three dimensions (Silliman, 1990), the present study examines the transport of chloride and latex particle tracers within a medium with variations in the hydraulic conductivity in all three physical dimensions. The medium is designed such that relatively complex preferential pathways are present within the medium.

2. Equipment and experimental design

The experiment presented below was run in the flow cell shown in Fig. 1. Constructed of Plexiglas and sealed with silicon glue, the cell allowed flow to be maintained at a constant rate (constant flux inflow and constant hydraulic head outflow). The fluid was added to the inflow reservoir using a peristaltic pump, and the outflow was maintained at a constant hydraulic head through use of a constant head overflow. The inflow reservoir was mixed during tracer introduction, and the outflow reservoir was mixed throughout the experiment. All sampling of outflow was performed from the outflow mixing loop.

The experimental medium was constructed of technical quality glass beads packed between the soil screens (Fig. 1). The glass beads selected for this study were identical

Fig. 1. Schematic diagram of the flow cell utilized for these experiments. Dimensions of the soil compartment in this design were 53 cm (parallel to mean flow) by 38 cm (vertical dimension) by 20 cm (width of flow cell). The inflow loop was utilized to introduce tracer to the inflow reservoir. The outflow loop was used to maintain a well-mixed outflow reservoir.

to those discussed in the earlier paper. Three size ranges were used: (1) 5–7 mesh (2.8–4.0 mm diameter); (2) 8–12 mesh (1.7–2.36 mm diameter); (3) 16–20 mesh (0.85–1.0 mm diameter). The saturated hydraulic conductivity of each of these bead sizes was measured through a simple Darcy experiment and determined to be 2.50 cm s^{-1} , 1.90 cm s^{-1} , and 0.51 cm s^{-1} for the large, medium, and small beads, respectively (it should be noted that these conductivities were incorrectly reported by Silliman (1995)).

Packing was performed with the top of the cell removed. The glass beads were added in thin (less than 1.0 cm) layers with the glass beads poured through approximately 1.0 cm of water. Packing was guided by predesigned arrangement of the medium. The fluid used for packing was water with a density increased to 1.05 g cm^{-3} (utilizing sodium chloride) to match the density of the latex particles which were used as a tracer. After the entire cell was filled with glass beads, the upper surface of the flow cell was installed. The final packing was visually tested for short-circuit flow paths along the surfaces of the flow cell through use of a red dye; after inspection, the dye was completely flushed from the medium before the start of experiments.

Two tracers were utilized in this work. The first (non-conservative) tracer consisted of latex spheres without special surface coating. Two diameters of spheres were added. Approximately 70% of the spheres were in the size range of $4\text{--}7 \mu\text{m}$, and the remainder were in the size range of $20\text{--}25 \mu\text{m}$. Concentration of the spheres entering the medium was approximately $50\,000 \text{ particles ml}^{-1}$ and all spheres had a density estimated by the manufacturer to be 1.05 g cm^{-3} . The second (conservative) tracer was the chloride ion at a concentration approximately 400 mg l^{-1} above background concentration (as noted below, the flow cell had been flushed extensively with reverse osmosis water before using the chloride tracer). The chloride ion was added in the form of sodium chloride.

The experiments were conducted as follows:

- (1) flow was established by injecting water at a density of 1.05 g cm^{-3} into the inflow reservoir. The flow rate into the medium was constant at approximately $78 \text{ cm}^3 \text{ min}^{-1}$. Outflow from the medium was allowed to come to steady state through a constant elevation overflow.
- (2) The latex particles were introduced to the medium via mixing into the inflow reservoir. The particles were allowed to enter the medium for 30 min and then the inflow reservoir was flushed with high-density (1.05 g cm^{-3}) water without particles, thus creating a 'pulse' of particle tracer. Concentration within the inflow reservoir during the pulse was approximately 50 000 particles per milliliter.
- (3) Upon completion of the particle tracer experiment (e.g. particle concentrations in the outflow had returned approximately to zero), the medium was flushed with reverse osmosis water to allow running of the chloride tracer experiment.
- (4) When the low-density water had displaced the majority of the high-density water (determined through monitoring of the chloride concentration in the outflow from the medium), a steady-state flow rate was established. The chloride tracer was then added to the inflow reservoir as a step change in

concentration. The rate of injection of fluid into the medium during this tracer test was approximately $84 \text{ cm}^3 \text{ min}^{-1}$. The concentration of the chloride was monitored at the outflow reservoir.

- (5) When the outflow chloride concentration had reached (approximately) the concentration in the inflow reservoir, flow was terminated and the medium was destructively sampled (cores of glass beads and the interstitial fluid were collected) to determine the concentration of particles remaining within the medium. The volume of an individual core was approximately 8 cm^3 . Concentrations were determined by vigorously shaking each sample and then sampling the pore fluid. Counts were conducted with a trifocal microscope utilizing Sedwish-Raptor cells.

It is noted that the major changes in the ionic strength of the pore fluid in steps (3) and (4) have the potential to redistribute sorbed latex particles within the medium. In prior experiments in homogeneous and heterogeneous media utilizing the same type and size of glass beads and latex particles, however, we have not observed any additional movement of the latex particles during a reduction in ionic strength. Further, during the present experiment, no additional particles were observed in the outflow from the reservoir after the change in ionic strength. We take this evidence to indicate that the latex particles trapped within the medium are not remobilized during the reduction in the ionic strength. Hence, it is assumed that these destructive samples fairly represent the distribution of trapped particles before change in ionic strength.

All experiments were conducted with a distribution of glass beads as shown in Fig. 2. Fig. 2(a) shows vertical cross-sections through the front and rear halves of the flow cell parallel to the mean flow direction ($53 \text{ cm} \times 38 \text{ cm}$ face in Fig. 1). Fig. 2(b) shows a series of horizontal layers which compose the medium. Careful comparison of Figs. 2(a) and 2(b) allows one to see that a number of connections have been established among the large glass beads in the front and back cross-sections shown in Fig. 2(a). Of interest is the continuous, although tortuous, path of large glass beads which crosses the medium within the lowermost four layers. These layers are separated by a continuous layer of small glass beads from the seventh layer, where a continuous, although curved, path is available within the large glass beads. Another continuous layer of small glass beads separates the seventh layer from the ninth layer, where a large region of large beads is subdivided by a narrow, vertical zone of small glass beads. Hence the overall medium provides several potential transport routes entirely within large glass beads, and selected routes (e.g. Layer 9 and the layers consisting of small glass beads) where there is no direct route entirely within large glass beads. Results of numerical modeling (discussed below) supported our empirical interpretation that this medium contained several flow paths by which particles could travel the length of the medium entirely within the coarse glass beads.

3. Results

Because the outflow reservoir had finite volume (estimated to be approximately

Mean Flow
Direction →

FRONT WALL

(a) 5-7 Mesh 8-12 Mesh 16-20 Mesh

Mean Flow
Direction →

BACK WALL

(a - cont'd) 5-7 Mesh 8-12 Mesh 16-20 Mesh

Fig. 2. Details of the distribution of glass beads within the experimental medium: (a) vertical cross-sections along the front and back walls parallel to the mean flow direction; (b) horizontal layers composing the medium. Comparison of distributions in (a) and (b) will demonstrate that a number of connected paths exist within the large glass beads in this medium.

2000 cm³), outflow concentration of the chloride and the latex particles was not equal to the concentration leaving the medium at the downstream screen. As a result of mixing within the outflow reservoir, the concentration in the outflow from the reservoir, $C_r(t)$, is related to the outflow from the medium, $C(t)$, through a standard mixing equation. Assuming that the outflow from the reservoir is sampled at discrete time intervals, Δt , the mixing equation is given by

$$C' = [C_r(t + \Delta t) - C_r(t) \exp(-Q\Delta t/V)] / [1.0 - \exp(-Q\Delta t/V)] \quad (1)$$

where C' is the average of $C(t)$ over the period $(t, t + \Delta t)$, V is the volume of the outflow reservoir, and Q is the volumetric flux leaving the porous medium. This equation provides an estimate of the concentration of the fluid entering the outflow reservoir based on the measured concentration of the fluid leaving the outflow reservoir. All outflow data reported have been adjusted for mixing in the outflow reservoir via Eq. (1).

As shown in Fig. 3, the adjusted, normalized chloride breakthrough recorded at the outflow reservoir demonstrated significant dispersive spread. Efforts to fit a single velocity or dispersivity to this curve were conducted utilizing the standard solution to the one-dimensional advection-dispersion equation for a step increase in

Fig. 3. Chloride breakthrough curve observed in the outflow reservoir. This curve has been adjusted to account for mixing in the reservoir and has been normalized to the inflow concentration. The continuous line represents a fit to this curve utilizing the advection-dispersion equation with $v = 0.55 \text{ cm min}^{-1}$ and $\alpha = 9.1 \text{ cm}$ (α is the dispersivity where it is assumed that $D = \alpha v$).

the concentration (e.g. Freeze and Cherry, 1979), as

$$C'/C_0 = \frac{1}{2} \{ \text{erfc}[(x - vt)/(2(Dt)^{0.5})] + \exp(vx/D) \text{erfc}[(x + vt)/(2(Dt)^{0.5})] \} \quad (2)$$

where C_0 is the concentration at $x = 0$, v is the average pore water velocity, x is the distance from the inflow reservoir (here set equal to the length of the flow), and D is the dispersion coefficient. The primary reason for attempting to fit the advection-dispersion equation to the results was to provide an indication of the magnitude of dispersion represented by this outflow curve. Discussion of whether this curve represents a Fickian process and/or asymptotic behavior within this system is beyond the scope of this paper. As shown in Fig. 3, however, a reasonable fit between the observations and the solution given in Eq. (1) can be obtained with an average pore velocity of 0.55 cm min^{-1} and a dispersivity of 9.1 cm . Assuming a porosity between 20 and 25%, this velocity is consistent with the velocity estimated from the volumetric flux ($84 \text{ cm}^3 \text{ min}^{-1}$ over an area of approximately $20 \text{ cm} \times 38 \text{ cm}$). In comparing this dispersivity with previously published results (e.g. Silliman and Simpson, 1987; Neuman, 1990), it is apparent that this is a relatively large dispersivity

for this scale of transport and is consistent with a heterogeneous medium with high variability in pore water velocities.

The results for the outflow particle counts, shown in Fig. 4, demonstrate that particles were successfully transported across the medium and that there was little difference in terms of the breakthrough curves between the small particles and the entire particle population. Two qualitative observations are made regarding these results. First, the travel time for the particles is very similar to the travel time for the chloride tracer. Second, the breakthrough results shown in Fig. 4 show multiple peaks of breakthrough. These multimodal results are interpreted as being indicative of particle transport via a number of transport paths through the medium. Such an interpretation is consistent with the numerical modeling, which demonstrated multiple flow pathways through this medium (discussed below).

An indication of the route taken by the particles is provided by the particle counts in the destructive samples. The results, summarized for each horizontal layer in Fig. 5, provide significant insight into the flow routes taken by the particles. As discussed in the previous paper, the medium and fine glass beads are relatively effective at removing particles from the transport fluid (apparently related to a combination of adsorption and straining). This can be observed by the large number of particles remaining within the fine glass beads in the various layers (e.g. Layers 2, 3, 4, 6, 8, and 10) and the rapid decline in particle counts both with distance into fine bead

Fig. 4. Particles per milliliter observed in the outflow reservoir (corrected for mixing in the reservoir).

Fig. 5. Concentration of particles trapped within the experimental medium following the experiment. Concentrations are normalized to the inflow concentration of particles.

regions (e.g. Layers 6, 8 and 10) and downstream of regions where flow is forced across fine glass beads (e.g. Layer 9). The observation that a relatively large number of particles were transported to the outflow reservoir (mass recovery is estimated at approximately 17%) is a strong indication that preferential flow pathways exist within this medium.

MODFLOW (McDonald and Harbaugh, 1988) and MODPATH (Pollock, 1989) were used as corroborative tools to both design and analyze these experiments. For this modeling, the medium was subdivided into eight vertical slices aligned as shown in Fig. 2(a). Each slice was subdivided into over 500 elements. Although further refinement of this grid might provide greater detail in terms of the flow field, the specific purpose of the modeling was not to provide a detailed simulation of this medium. Rather, it was intended to provide a basic analysis of the geometry of flow lines.

The details of the flow lines generated in this simulation are complex, and are beyond the scope of this paper. Therefore only basic results are presented here.

Fig. 6 shows the variation in transport time vs. elevation in the flow cell for particles leaving the medium. The figure was generated by uniformly distributing approximately 900 particles on the inflow face of the medium and following these particles to the outflow boundary. First arrivals (approximately 35–40 min) occur in the coarse beads composing Layers 7 and 9 and in the medium and coarse beads in Layers 2 and 3 (see Fig. 2(b)). These transport times coincide well with the observed breakthrough of particles at approximately 50 min (Fig. 4). A second cluster of transport times is observed in Fig. 6 around 65–75 min. In this case, the exit location coincides with the coarse bead region in Layer 4. The timing once again coincides with an observed peak in the breakthrough curve at 70–80 min (Fig. 4). Significant later arrival times are predicted, based on Fig. 6, from the layers consisting of finer glass beads. The failure to observe particle breakthroughs after approximately 150 min is indicative of the failure of the particles to be successfully transported through these regions.

In terms of following flow lines through the medium, the modeling demonstrates the three-dimensional character of this medium. Fig. 7 provides an illustration of the complexity of two flow lines originating near the top of the coarse beads in Layer 1.

Fig. 6. Distribution of travel time across medium vs. location of exit from the medium. Results are derived from MODFLOW (McDonald and Harbaugh, 1988) and MODPATH (Pollock, 1989) numerical code and are based on 896 particles distributed uniformly over the inflow face of the medium. These results demonstrate the significant difference in travel times for different flow paths to the outflow.

Fig. 7. Two flow paths identified through the numerical modeling: (a) three-dimensional view of the flow paths; (b) view of the flow paths looking down on the top of the flow cell; (c) view of the flow paths looking at the front of the flow cell; (d) view of the flow paths looking backwards from the outflow reservoir. Estimated travel time across the medium for Line i was approximately 130 min. Estimated travel time for Line ii was approximately 53 min.

Fig. 7(a) shows a three-dimensional perspective of these two flow lines, where the mean flow is in the direction of increasing x . To aid in interpreting this figure, Figs. 7(b), 7(c), and 7(d) respectively show top (x, z), front (x, y), and outflow end (y, z) perspectives of these lines. The first flow line follows, approximately, the geometry of the coarse glass beads of Layers 1 and 2 to the back of the flow cell (x, y and z all increasing). It then rises to approximately the elevation of Layer 4 (to $x = 28$ cm, $y = 8.5$ cm, and $z = 13$ cm) from where it moves back towards the front of the flow cell (y remains constant and x increases whereas z decreases). In comparison, the second flow line starts from a similar location in the flow cell. Although this flow line mimics the behavior in the first two layers of the medium by traveling towards the back of the flow cell and rising to Layer 2, it does not move back towards the front of the flow cell. Rather, it follows the coarse and medium glass beads along the rear of the flow cell to the outflow (z remains approximately constant as x increases from 25 cm to 53 cm).

These two flow lines illustrate the complexity of the flow field and the full three-dimensional nature of the flow. The numerical results, in general, led to identifying at least three routes by which flow would probably cross this medium entirely within the coarse glass beads. These included flow lines following the curve path in Layer 7, flow

lines leading to the outflow reservoir in the coarse region of Layer 2 (e.g. similar to Fig. 7, Line ii), and flow lines leading to the outflow in the coarse region of Layer 4 (e.g. similar to Fig. 7, Line i). Clearly, a two-dimensional model would be inadequate to provide the details of the last two flow lines.

4. Discussion of results: the role of the third dimension

If one were to consider either vertical cross-section shown in Fig. 2(a) independently, one would conclude, based on the results from the earlier paper, that very few particles would be successfully transported across the medium. The reason for this conclusion is that two-dimensional transport in either of these cross-sections would require water either to flow across contacts between different-sized glass beads or to flow within regions consisting of small or medium-sized glass beads. In fact, the most likely route for successful transport within either of these cross-sections would be across Layer 9 (Fig. 2(b)), where flow is within coarse glass beads except for a very narrow layer of fine glass beads. That the number of particles being transported through the fine glass beads in Layer 9 is reduced substantially by the presence of that layer is indicated by the relatively low particle counts observed downstream of this layer in Fig. 5.

Based on this reasoning, the large number of particles in the outflow curve is interpreted as a strong indication that the connectivity between the two vertical layers shown in Fig. 2 was of critical importance in allowing particles to be successfully transported across the porous medium. Specifically, it is suggested that the particles arriving at the outflow reservoir were transported primarily within the tortuous, large glass bead flow paths.

Further, the multiple-peaked nature of the particle breakthrough curve (Fig. 4), in combination with the numerical results, provides evidence that the particles were traveling via multiple flow routes. Specifically, these multiple breakthroughs are interpreted as being related to the three primary pathways discussed in the previous section.

This interpretation (that a number of particles are transported across the medium along these three flow routes) is supported by the results from the destructive samples. By reviewing the particle counts from destructive samples for Layers 4, 7, and 9, it is observed that relatively high destructive counts are observed in the coarse bead regions near the outflow reservoir in Layers 4 and 7 (unfortunately, owing to a sampling error, the counts near the outflow in Layers 2 and 3 were not recorded). These high counts are similar to those observed in the coarse bead regions near the inflow reservoir (e.g. Layers 1 and 9), but are higher than observed in most layers at significant distance from the inflow boundary. Particle concentration near the outflow reservoir in Layer 9, for example, is lower than both those observed near the outflow in Layers 4 and 7, and those observed upstream of the small bead zone within Layer 9. Given the effectiveness of the narrow fine glass region in Layer 9 and the low counts at a significant distance from the inflow boundary in all layers except Layers 4 and 7, the high counts in these two layers imply that substantial numbers of particles were

transported to these regions without passing through the fine glass beads or through significant lengths of the medium glass beads. Given the relatively high count in the coarse region of Layer 2, it is anticipated that a similar observation (high counts near the outflow) would have been observed in this layer as well.

For Layer 7, it is relatively easy to argue that the flow route is through the curved region of the coarse glass beads, a route that could be considered two-dimensional in the horizontal plane. The flow routes leading to Layers 2 and 4, however, are more complex. Analysis of the distribution of glass bead sizes in combination with the general results from the numerical modeling leads to the conclusion that neither of these flow routes could be considered two-dimensional with respect to any plane passing through the experimental medium. In fact, for particles to be transported successfully to the outflow in Layer 4, flow will move essentially within a horizontal plane near the inflow reservoir in Layer 1, nearly vertical through the coarse bead regions of Layers 2 and 3, and once again within a nearly horizontal plane within Layer 4 (see Fig. 7). As a result, the full three-dimensional connectivity of this medium was critical in determining both the transport of particles to the outflow reservoir and the distribution of particles remaining within the medium.

5. Conclusions

The results presented demonstrate the importance of considering the connectivity of high-permeability regions within saturated porous media. Although the particular medium utilized is limited in terms of the use of artificial solids (glass beads) and a relatively contrived distribution of permeabilities, these results represent one of the first controlled laboratory experiments of which we are aware in which the role of the third dimension is specifically considered. The results are interpreted as indicating that the third dimension has the effect of enhancing particle transport across the porous medium. Specifically, it is concluded that this enhancement is related, in large part, to the connectivity of the high-permeability pathways crossing the medium. This conclusion is supported by (1) the high degree of mixing observed in the chemical tracer experiment, (2) the mass recovery and multiple-peaked shape of the particle breakthrough curves, (3) the correlation of the particle concentrations in the destructive samples with the physical heterogeneity, and (4) the numerical analysis of the flow field.

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Estimating the Horizontal Gradient in Heterogeneous, Unconfined Aquifers: Comparison of Three-Point Schemes

by Bryce E. Cole and Stephen E. Silliman

Abstract

At least two approaches may be used to estimate the horizontal components of the hydraulic gradient based on measured heads from three observation points. First, the gradient may be estimated by passing a plane through the measured heads (h-method). Second, if the elevation of the base of the aquifer is known to be spatially constant, an estimate of the gradient may be obtained using the squares of the measured heads (h^2 -method). In the present study, these methods are examined in application to a heterogeneous system. Using Monte Carlo analysis, we demonstrate that the magnitude of the gradient estimated via the h-method involved significant bias, which increased when the distance separating the wells increased. In contrast, bias in the estimated magnitude of the gradient based on the h^2 -method decreased with increasing separation among the wells. Estimation variances for both the magnitude and orientation of the gradient also decreased with separation distance. The variance in the orientation was observed to remain relatively high, however, even at relatively large separations among the wells (e.g., 10 integral scales). These results are interpreted as implying that the best estimate of the gradient for steady flow in an unconfined aquifer is derived from the h^2 -method with the wells separated by significant distances. These results also demonstrate the uncertainty inherent in estimating the gradient based on limited field data.

Introduction

Determination of the magnitude and orientation of the gradient of the water table is a critical step in many ground water investigations. In wellhead protection studies, for example, proper estimation of the gradient is critical to the delineation of the capture zone of a wellfield. Similarly, in design of remediation strategies, proper estimation of the direction and rate of migration of contaminant plumes is crucial to the siting of remediation wells. Finally, in the design of field experimental efforts, accurate estimation of the direction and rate of ground water flow is important in designing sampling strategies. For many field applications subject to limited investigation costs, however, it may be necessary to estimate the gradient based on a limited number of observation wells. The smallest number of wells needed to estimate the gradient from hydraulic head measurements is three wells. At least two questions arise, however, from the use of only three wells:

1. Does the approximation of the water table as a plane introduce unacceptable errors to the analysis?
2. What influence does the separation distance between the wells (relative to the length scale of the hydraulic heterogeneity) have on the accuracy and precision of the gradient estimate?

The first question results from analysis of the Boussinesq equation for steady-state flow in an unconfined aquifer. The equation is non-

linear in h , but is linear in h^2 under appropriate conditions (Bear 1979). Consequently, the hydraulic head for steady-state flow between two parallel, constant head boundaries does not vary linearly in space (e.g., Figure 1). Thus, a planar approximation to this curve, based on observed heads at A, B, and C, introduces error into the estimate of the hydraulic gradient. The magnitude of this error will depend on both the separation among the wells and on the curvature of the surface. As noted by Bear (1979), however, this error is commonly small, since the change in hydraulic head, and therefore the curvature of the water table, is typically small.

The second question is warranted based on review of the significant literature examining flow in heterogeneous systems. As early as the work of Warren and Price (1961), authors have recognized the role that spatial heterogeneity in the hydraulic conductivity plays on the distribution of hydraulic head within an aquifer. A number of authors have examined the statistical description of hydraulic head for flow in one, two, and three dimensions (Freeze 1975; Bakr et al. 1978; Smith and Freeze 1979) and the development of effective hydraulic parameters (e.g., Dagan 1979). Two conclusions may be drawn from these efforts (see Dagan 1986 for a relatively complete review). First, hydraulic heterogeneity leads to variability in hydraulic head and, thereby, to variability in the estimate of the hydraulic gradient. Second, little work has been completed for unconfined aquifers illustrating the magnitude of the potential errors in the estimate of the hydraulic gradient.

The present study focuses specifically on estimation of the hydraulic gradient using three observation wells in an unconfined aquifer. This focus was selected both for relevance to field problems commonly encountered and to demonstrate a "worst case" study. It is anticipated that the uncertainty in the estimation of the hydraulic gradient will decrease as the number of wells used in the estimation procedure increases. Statistics regarding the magnitude and direction of the estimated hydraulic gradient are generated through use of Monte Carlo simulation.

It should be noted that the questions raised in this paper reflect only a portion of the overall problem of estimating hydraulic gradient. Additional difficulties such as temporal transients in the measured head due to nonsteady flow or variations in barometric pressure during the period of sampling (e.g., Crawford and Rasmussen 1994) will also directly influence the quality of the estimate of the gradient.

Mathematical Problem Statement

The problem addressed involved two-dimensional, horizontal flow in an unconfined aquifer. The base of the aquifer was at constant elevation, and this elevation served as the reference datum for the hydraulic head. Under these conditions, the saturated thickness of the aquifer was everywhere equal to the hydraulic head. Figure 2 shows the mathematical representation of the physical problem. The predominant flow direction was parallel to the x -axis due to constant hydraulic heads

Figure 1. Three-point planar approximation of water-table surface. Curvature of the water table leads to bias in the estimate of the hydraulic gradient.

Figure 2. Flow domain with boundary conditions. Gradient estimated at Well A. Upstream well pair shown. Point A is centered in the flow domain from top to bottom and located 11,500 feet (3513 m) from the upstream boundary.

assigned along the left and right boundaries. For all cases, the gradient was estimated at Point A (Figure 2).

The governing equation for flow in the aquifer is the linearized, steady-state Boussinesq equation (Bear 1979):

$$\frac{\partial}{\partial x} \left(K(x,y) \frac{\partial h^2}{\partial x} \right) + \frac{\partial}{\partial y} \left(K(x,y) \frac{\partial h^2}{\partial y} \right) = 0 \quad (1)$$

where K is the hydraulic conductivity, and h designates the hydraulic head. To study the influence of water table

curvature (hereafter termed Case 1), the hydraulic conductivity was assumed homogeneous and isotropic (thus, reducing Equation 1 to Laplace's equation in h^2). To study the influence of spatial variation in the hydraulic conductivity (hereafter termed Case 2), the hydraulic conductivity was assumed to be locally isotropic but no longer homogeneous (i.e., varied with x and y). For both cases, the left and right boundaries were assigned constant heads, h_u and h_d , respectively. The upper and lower boundaries of this system were specified through forcing the hydraulic head to vary linearly in h^2 (equation shown in Figure 2).

Case 1 — Water Table Curvature Consideration

Since K does not vary spatially for Case 1, Equation 1 reduces the Laplace equation in h^2 . Solution of the Laplace equation subject to the stated boundary conditions yields the following expression for the hydraulic head:

$$h(x,y) = \left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x}{L} \right]^{0.5} \quad (2)$$

where L is the distance between the upstream and downstream boundaries. Since the expression for the hydraulic head is dependent only on its x -coordinate, the hydraulic gradient can be estimated from the hydraulic heads at Point A and at a second point, either upstream or downstream of A, with abscissa value x_p . Using these values, the estimate of the gradient based on h (the h -method) is

$$\frac{\left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x_p}{L} \right]^{0.5} - \left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x_a}{L} \right]^{0.5}}{(x_a - x_p)} \approx \frac{dh}{dx} \quad (3)$$

where x_a is the distance from the upstream boundary to Point A. Similarly, the estimate of the gradient based on h^2 (the h^2 -method) can be stated as:

$$\frac{\left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x_p}{L} \right] - \left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x_a}{L} \right]}{2(x_a - x_p) \left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x_a}{L} \right]^{0.5}} \approx \frac{dh}{dx} \quad (4)$$

In the following sections, Equations 3 and 4 are compared with the analytical derivative of Equation 2 for a number of values for x_p and h_u . This derivative is given by:

$$\frac{dh}{dx} = \frac{h_d^2 - h_u^2}{2L \left[h_u^2 - \left(h_u^2 - h_d^2 \right) \frac{x}{L} \right]^{0.5}} \quad (5)$$

Case 2 — Heterogeneous Aquifer

For the second case study, the hydraulic conductivity was assumed to vary spatially as a correlated, second-order stationary random field. The hydraulic conductivity fields are produced via discrete analysis (Silliman and Wright 1991). Through use of this algorithm, the square domain shown in Figure 2 was discretized into 201×201 elements (of constant size), and each element was assigned a hydraulic conductivity value.

As a result of the random variation in K , an analytical solution was not available for this problem, and all derivatives were estimated via numerical solution of Equation 1 using MODFLOW (McDonald and Harbaugh 1988) with the MICCG solution package (Hill 1990). MODFLOW was applied to the hydraulic conductivity field through further discretization of the grid in the vicinity of A to allow noninterpolated gradient estimates at small separation distances. This refined mesh had more than 150,000 elements.

Due to the two-dimensional nature of the flow field, three points were required to estimate the magnitude and orientation angle of the water table. For this study, the three points used were Point A and a pair of points equidistant from Point A along the primary flow direction (see Figure 2).

When the estimate of the gradient was based on the h -method, the formulas providing estimates of the magnitude of the gradient in the x and y directions were:

$$Mag_x = \left| \frac{h_a - \frac{(h_1 + h_2)}{2}}{x_a - x_p} \right| \quad (6a)$$

$$Mag_y = \left| \frac{h_1 - h_2}{y_1 - y_2} \right| \quad (6b)$$

where h_a denotes the head at Point A; h_1 is the head at the observation point located at (x_p, y_1) ; and h_2 is the head at the observation point located at (x_p, y_2) . Alternatively, the gradient estimate based on the square of the hydraulic head (h^2 -method) is:

$$Mag_x = \left| \frac{h_a^2 - \frac{(h_1^2 + h_2^2)}{2}}{2h_a(x_a - x_p)} \right| \quad (7a)$$

$$Mag_y = \left| \frac{(h_1^2 - h_2^2)}{2h_a(y_1 - y_2)} \right| \quad (7b)$$

From these components of the gradient, the magnitude and orientation angle of the resultant gradient could easily be computed and compared with the equivalent

homogeneous gradient, the magnitude of which is given in Equation 5, and the orientation angle of which is zero degrees (parallel to the x axis).

Monte Carlo Runs for Case 2

A number of distributions of the hydraulic conductivity were considered using Monte Carlo simulation. For each distribution, the hydraulic conductivity was log-normally distributed with an exponential correlation structure given by:

$$R_{\lambda} = 1 - \exp\left(\frac{-\lambda}{\alpha}\right) \quad (8)$$

where λ is the separation distance between two points, and α is the integral scale. The integral scale was anisotropic with α being four times as large in the x direction as in the y direction. Each Monte Carlo run consisted of 250 realizations. Parameters for all sets are listed in Table 1. Figure 3 shows a typical distribution of hydraulic conductivity based on the algorithm used. The fact that α was four times larger in the x direction than in the y direction is reflected in this figure by the oval (elongated) shape of the conductivity regions. The average size of each of the conductivity zones is a function of the magnitude of α .

Table 1
Parameter Values Used for Each Set
of Monte Carlo Simulations Completed

Simulation Set	Mean K	Var{log K}	Integral Scale (x-direction)
	K in ft./day		feet
1	28.3	0.444	410
2	28.3	0.111	410
3	28.3	1.000	410
4	28.3	0.444	160
5	28.3	0.444	1160
6	28.3	0.444	3730

Results

Case 1 — Homogeneous Medium

The results for the homogeneous medium were consistent with previous discussions of the linearization of the Boussinesq equation. Specifically, as shown in Figure 4, the gradient estimate determined from the h^2 -method was equal to the true gradient at Point A regardless of the location of the second observation well. In contrast, the gradient estimate determined from the h-method was consistently biased except when the second observation point was located immediately next to Point A. This bias increased as the distance from Point A to the second observation point increased. The bias also increased as the head difference, $h_u - h_d$, increased.

Figure 3. Distribution of Log(K) in one region of one realization of the Monte Carlo simulation. The shapes of the individual contours are a result of the particular algorithm used. The conductivity values are in log meters per second. The integral scale (α) influences the average length of the individual contours.

Figure 4. Gradient estimates in homogeneous aquifers. Solid lines represent results for the h-method for different values of $h_1 - h_r$ ($h_1 = 230$ feet for all three lines — $h_r = 180$ feet, 131 feet, and 82 feet for the three lines, respectively). The dashed line represents results from the h^2 -method. This method is insensitive to the difference, $h_1 - h_r$.

Heterogeneous Medium

The base set of results for the heterogeneous case involved Monte Carlo simulations for a log K variance of 0.444 and an integral scale of 410 feet (125 m) in the x direction. Figures 5a and 5b show the mean and variance of the estimated magnitude of the gradient (normalized with respect to the magnitude at Point A for a homogeneous system) vs. the distance in the x-direction (in terms of number of integral scales) between Point A and the pair of observation points. These results show that the magnitude estimate was biased for both estimators at short separation distances. The magnitude derived from the h-method remained biased for all well separations shown (although the curve passes through a single point where it is unbiased). In contrast, the bias

Figure 5a. Average estimate of magnitude of the hydraulic gradient (heterogeneous medium) as a function of x-separation between wells. Solid line represents the h-method, dashed line the h^2 -method.

Figure 6a. Mean estimate of the orientation angle (in degrees) of the gradient as a function of x-separation between wells. Solid line is the h-method, dashed line the h^2 -method.

of the estimate based on the h^2 -method consistently decreased as separation distance increased. In both methods, the variance of the estimate was high for short separations, but declined with distance after one integral scale. The observed decrease in variance for separations larger than one integral scale corresponds with prior work in flow through heterogeneous media (e.g., Dagan 1986).

The statistics for the orientation angle of the gradient are shown in Figures 6a and 6b. The mean orientation angle appeared essentially unbiased for both estimators. The variance of the orientation angle, however, was quite large for both estimators through several multiples of the integral scale. Due to this large variation, sensitivity studies were conducted on the variance of the orientation angle.

Figure 7 shows the dependence of the orientation angle variance on the variance of the hydraulic conductivity. The integral scale was held constant for these

Figure 5b. Variance of the estimate of the magnitude of the hydraulic gradient as a function of x-separation between wells. Solid line is the h-method, dashed line the h^2 -method.

Figure 6b. Variance of the estimate of the orientation angle as a function of x-separation between wells. Solid line is the h-method, dashed line the h^2 -method.

calculations at 410 feet (125 m). Since there was little difference in the orientation angle statistics shown in Figure 6b, only statistics derived from the h^2 -method are shown in Figure 7. These results indicate that increases in the log K variance lead to increased variance of the estimated orientation angle for all separation distances among the wells.

Figure 8 shows the variance of the calculated orientation angle for various integral scales (variance of log K equal to 0.444). This figure demonstrates two interesting features. First, for separation distances less than approximately one integral scale, the variance of the orientation angle did not vary significantly with integral scale. Second, for the shorter integral scales, the variance remained as high as 100 even for large separation distances (e.g., 10 integral scales). Further, it may be inferred that the variance for the larger integral scales was lower than otherwise might be anticipated due to the influence of the boundaries of the numerical model (i.e., the proximity of the constant head boundaries).

Figure 7. Comparison of the variance of the estimate of orientation angle (angle in degrees) for various values of log K variance. Variance (log K) values are: solid - 0.111; dashed - 0.444; dotted - 1.00.

Discussion

These results provided insight into the use of three observation points to estimate the horizontal components of the hydraulic gradient. One major conclusion indicated by the results presented in Figures 5a and 6a is that unbiased estimates of both the magnitude and orientation of the hydraulic gradient are possible using three observation points. In particular, unbiased estimates of the orientation angle can be obtained using either the h -method or the h^2 -method at nearly any separation distance. In contrast, and under the conditions of this analysis, only the h^2 -method will provide unbiased estimates of the magnitude. This is an important result as it demonstrates that estimation of the hydraulic gradient in an unconfined aquifer using the h -method (i.e., through use of hydraulic head rather than the square of the hydraulic head) will produce estimates of the magnitude of the gradient that are consistently in error. Even with the h^2 -method, unbiased estimates are obtained only when the wells are separated by significant distances (i.e., several integral scales). Further, although not covered in this paper, the h^2 -method remains unbiased at large distance only if the base of the aquifer is at constant, known elevation.

The results presented also provide insight into the variability of the estimates of the magnitude. The results presented in Figure 5b indicate that substantial estimation variance will be present for the estimate of the magnitude when the three observation points are closely spaced (i.e., separated by less than one integral scale). The variance in the magnitude drops rapidly when the observation points are separated by more than one integral scale, resulting in a relatively reliable estimate of the magnitude for large separations.

Similar confidence cannot be expressed for estimates of the orientation angle. While the orientation angle was found to be unbiased, the results presented in Figures 6b, 7, and 8 show that the variation in the orientation angle may be large, even at large separation dis-

Figure 8. Comparison of variance of the estimate of the orientation angle (angle in degrees) for different integral scales. Lines represent integral scales of the following: solid - 160 feet; dashed - 410 feet; dash-dot - 1160 feet; dotted - 3730 feet.

tances (i.e., relatively large multiples of the integral scale). At short separation distances, the standard deviation in the orientation angle may be as high as 25 to 30°. While the standard deviation declines with separation distance, it was observed to be as high as 10° at separation distances of 10 integral scales. Such a large standard deviation may be significant in many applications. For example, if the observation wells are close together such that the standard deviation of the orientation angle is 30° (see Figure 6b), a 95 percent confidence interval on the orientation angle would include an approximately 120° arc (i.e., the angle estimated from the three-point scheme ± 2 standard deviations). Even at large separations, the 95 percent confidence interval covered an approximately 40° arc in our results. Clearly, such a large uncertainty could have significant impact on a site characterization, assessment of the movement of a chemical species, or design of a monitoring network.

For each of the results presented, it is apparent that the separation distance is a critical parameter in determining the bias and variance of the estimate of the hydraulic gradient. Unfortunately for field hydrologists, review of the recent literature points to two difficulties in estimating the integral scale from limited site-specific data. First, there is significant variation in the integral scale from one site to another. Hoeksema and Kitanidis (1985), for example, found variation of the integral scale of more than one order of magnitude when comparing results for unconsolidated sediments. Hence, there does not appear to be a "generic" integral scale for a given geologic setting. Second, the integral scale appears to be dependent on the scale of measurement. The comments of Anderson (1991) and the reply of Neuman (1991) to Neuman's scaling paper (Neuman 1990) provide an interesting introduction to the scaling issue.

Fortunately, the literature also provides some evidence that this problem is tractable in the general sense. As mentioned by Anderson (1991), the changing scales of heterogeneity are most likely related to changes in

facies. Hence, prior knowledge of the geology of region may aid in estimating integral scales appropriate in a given setting (see, for example, Davis et al. 1993). Hence, the field hydrologist may be able to glean information from the local sedimentary geology regarding the length scales of the local facies, and these facies scales, in turn, may be used as initial estimates of the local integral scales.

In a separate study, Silliman and Frost (1995) have looked at the three-point gradient estimate for three field sites at which multiple wells were present. Using measured heads in these wells, it was observed that the variance of the estimate of the magnitude became negligible for well separations between 220 feet (for a field site covering approximately 500 feet $\times$ 1300 feet) and 5000 feet (for a field site covering approximately 0.5 miles $\times$ 2 miles). The distances required for the variance of the estimate of the orientation to become negligible were larger by a factor of approximately two. While the initial investigation of these field sites does not provide a definitive measure of the scale of measurement necessary to obtain low variance estimates of the gradient, it does provide an initial indication of the order of magnitude of the separation distances required.

Conclusions

The results presented demonstrate that at least two factors influence our ability to estimate the magnitude and orientation of the horizontal components of the hydraulic gradient in unconfined aquifers. These are the nonlinear head distribution inherent in unconfined aquifers and the variation in the distribution of hydraulic head caused by heterogeneity in the hydraulic conductivity. The nonlinearity of the head distribution leads to the observation, as noted in this paper and previously by other authors, that a three-point scheme based on the h-method will lead to biased estimates of the magnitude of the gradient. Heterogeneity leads to bias in the estimated magnitude even when using the h^2 -method for small separation distances. This bias in the h^2 -method declines, however, with increasing separation distance.

The observation that hydraulic heterogeneity leads to uncertainties in the estimated orientation of the hydraulic gradient is considered the most significant result presented. With standard deviation of the estimate as high as 10° even when the observation wells are separated by large distance (e.g., as large as 10 integral scales in this study), this result implies that orientation angles determined from three-point schemes should be used only with great care and with consideration of the potential ramifications of error in this parameter.

One might conclude from this discussion that the best estimates of the hydraulic gradient, using only three wells, will be obtained when the three wells are separated by large distances. From the viewpoint of estimating regional flow in the absence of local sources and sinks, this conclusion is believed to be correct. This general conclusion is tempered, however, by realities

such as the impracticality of separating monitoring points by distances on the order of thousands of feet, the additional uncertainties associated with data errors or hydraulic transients, and the difficulty associated with avoiding local sources or sinks (wells, creeks, wetlands, etc.). We conclude, therefore, that uncertainty is inherent in the estimation of the hydraulic gradient (particularly in the estimation of the orientation of the gradient) and that this uncertainty should be factored into the analysis of a field investigation.

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Biographical Sketches

Bryce E. Cole received his Ph.D. from the Department of Civil Engineering and Geological Sciences at the University of Notre Dame (Notre Dame, IN 46556) in 1996 and is currently teaching at Walla Walla College, Walla Walla, Washington. He

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Capture Zones for Passive Wells in Heterogeneous Unconfined Aquifers

by Bryce E. Cole^a and Stephen E. Silliman^b

Abstract

Many piezometers/wells produce at such low rates and frequency that their impact on local hydraulics is negligible, e.g. monitoring piezometers or homeowner wells installed in highly permeable soils. Defining a deterministic capture zone for such wells is often of limited utility as the capture zone is effectively a single flow line directed upgradient. In order to provide more insight into the capture zone of such wells, a statistically based capture zone, termed a "percentile capture contour" (PCC), is introduced and analyzed. The capture zone is defined by quantifying, for a given travel time, the variation of the length and orientation of the flow line emanating upgradient from the well. Capture zone variation herein depends on second-order stationary random hydraulic conductivity fields and is calculated using Monte Carlo analysis.

Monte Carlo analysis yielded increases in mean travel distances as the variance and/or integral scale in log K increased, but decreases as the angle between the principal direction of the correlation structure and the regional flow increased. The average travel distance exceeded the travel distance estimated by a homogeneous solution using the geometric mean hydraulic conductivity. Transverse variation depended both on the variance and integral scale of log K, but was insensitive to the orientation of the principal correlation direction. The mean orientation of the flow path varied with the principal direction of the correlation structure, deviating up to 20° from the orientation of the hydraulic gradient. These observations are consistent with flow following preferential pathways and indicate that significant uncertainty exists for source prediction of water feeding passive wells.

1.0 Introduction

A number of authors have studied the influence of spatial variation of the hydraulic conductivity on prediction of ground-water flow and transport processes. These studies have included consideration of effective hydraulic properties (e.g., Indelman and Dagan, 1993; Neuman and Orr, 1993), dispersive spread (Russo, 1993; Neuman, 1993; Schwartz, 1977), particle transport (Silliman, 1995 and 1996; Harvey et al., 1993), and distribution of hydraulic head (Bakr et al., 1978; and others). From these studies, it can be concluded that regional flow within a heterogeneous aquifer will involve significant local variation in the pore-water velocities. As a result, significant uncertainty exists regarding the flow path followed by ground water subjected to a regional hydraulic gradient.

In the present study, we consider the problem of predicting the source region of water arriving at an effectively passive ground-water well. The term passive refers here to a well from which water is produced at such a low rate that no noticeable impact occurs to the distribution of hydraulic head within the aquifer. Such a well might represent a monitoring well which is sampled over time in order to monitor the movement of a plume (i.e., a sampling design problem) or a well drilled for a single-family dwelling which is producing from an aquifer with high hydraulic conductivity and specific yield (a wellhead protection application).

The confidence associated with prediction of the geometry of the flow line supplying water to the passive well depends on

the uncertainty present in our knowledge of the spatial distribution of the hydraulic conductivity. This uncertainty is addressed herein through generation of multiple realizations of the spatial distribution of hydraulic conductivity, numerical solution of the flow equation for steady-state flow in an unconfined aquifer, and reverse particle tracking to delineate the flow line passing through the center of the well. Variation of the flow line is quantified (for a set transport time) in terms of the distance traveled by the particle both upgradient of the passive well and transverse to the path predicted for a homogeneous, isotropic medium. The distribution of travel distance is based on a five-year travel time. The variation in transverse location of the flow line is based on 50-year travel times to ensure that most particles travel to the upstream boundary of the flow domain.

Applications of this study fall within the realm of both physical situations mentioned above. In the case of a monitoring well, field design is often based on centering a monitoring well network along the centroid of the plume in a direction predicted from analysis of the regional gradient. If a monitoring well shows negative for the contaminant, it is normally assumed that any contamination which has occurred directly upgradient (based on regional analysis) has not yet reached the well's location. If variation in the flow field led to contamination upgradient bypassing this monitoring well, the conclusion that the contamination remains upgradient may be in error. Of interest, then, is the reliability of the monitoring well in detecting chemicals emanating from a source location which is upgradient as estimated from a regional analysis of flow.

For single-dwelling wells in which contamination occurs, investigations may be undertaken to delineate potential sources of the contaminant so as to plan for short- and long-term solutions, as well as to determine liability. Source identification efforts will be based, in part, on determination of the flow lines which are likely to pass near the well. Similar concerns regarding identification of potential sources will also arise should wellhead protection efforts be pursued prior to contamination.

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2.0 Problem Statement

In the present study a flow line extending upgradient from a passive well is considered within a heterogeneous, unconfined aquifer. Flow in the aquifer is driven solely by the regional gradient between regions of constant potential (see Figure 1) and is assumed to be both two-dimensional (i.e., in the horizontal plane designated by Points A, B, and C) and at steady state. This square aquifer has a side dimension of 5025 meters. For mathematical solution of this problem with spatially varying hydraulic conductivity, the flow domain is divided into 201 cells in both the x- and y-directions. Hydraulic conductivity values are assigned to each of the cells as described below and K is assumed everywhere to be locally isotropic. The well is located at (3513, 2513). The regional gradient is aligned, on average, with the x-axis from left to right (Figure 1) between constant head boundary values of 70 and 45 meters. First type boundary conditions are utilized along the upper and lower edges as well, with linear variation for the square of the hydraulic head of 4900 m² and 2025 m² (i.e., the squares of the boundary values).

The upper surface of the (nonleaky) confining layer underlying the aquifer is presumed to be at constant elevation, which for steady, two-dimensional flow renders the flow equation linear (using the base of the aquifer as datum) in terms of the square of the head as follows:

$$\frac{\partial}{\partial x} \left(K(x, y) \frac{\partial h^2}{\partial x} \right) + \frac{\partial}{\partial y} \left(K(x, y) \frac{\partial h^2}{\partial y} \right) = 0. \quad (1)$$

Solution of the flow equation is accomplished using the method of finite differences as represented by MODFLOW (McDonald and Harbaugh, 1988). Hydraulic conductivities are assigned to the cells as discussed below. Due to the linearity of equation (1) in terms of the square of the hydraulic head, the confined aquifer option was employed in MODFLOW using the hydraulic conductivities as input in place of the transmissivities and the square of the fixed boundary heads in place of the boundary head values. The result of this substitution provides a solution in terms of h^2 , thereby requiring that the square root of the output be calculated in order to provide the distribution of h . The advantage of this implementation, as compared to utilizing the unconfined aquifer option in MODFLOW, was that solution of the matrix via the Modified Incomplete Cholesky Conjugate Gradient package (Hill, 1990) required only one half the computation time required of the nonlinear, unconfined aquifer option.

A further consequence of linearization is that the inter-cellular flows calculated by MODFLOW are incorrect. The ground-water velocity, however, can be estimated from Equation (41) in McDonald and Harbaugh (1988, pp. 5-8) combined with the predicted distribution of head. From nodal velocities, reverse particle tracking is accomplished using the interpolation scheme proposed by Prickett et al. (1981) and a fourth-order Runge-Kutta integration method (Shafer, 1987).

Since the well pumping rate is taken as zero, no singularity occurs in the two-dimensional representation of the hydraulic head. The capture zone for the well is herein represented by a single flow line (as shown in Figure 1), which can be estimated using reverse particle tracking from the well.

3.0 Monte Carlo Simulation

The generation of statistics regarding the distribution of flow lines is accomplished through Monte Carlo simulation. As

applied here, one realization refers to generation of correlated, random hydraulic conductivities for each of the cells in the mesh used in MODFLOW, followed by solution of the flow field as described in the previous section and reverse particle tracking from the well. One set of Monte Carlo simulations refers to hydraulic conductivity generation through flow line determination for 250 realizations.

Two random field generators, turning bands (Tompson et al., 1989), and discrete analysis (Silliman and Wright, 1991), were utilized to generate realizations of log K (log of the hydraulic conductivity). Since the two generators led to similar results, only the discrete analysis results are presented herein. The random fields were constrained by requiring that the well be located in a region for which the hydraulic conductivity was greater than the mean log K. This condition provided continuity with additional studies which were performed with actively pumped wells (Cole, 1995). In those studies, constraining the well to regions of high conductivity was justified based on the observation that most large capacity wells will be located in high conductivity units. For the present study, an arbitrary cutoff value for the hydraulic conductivity at the well was set as the geometric mean used for all the random fields, i.e., all realizations analyzed had a hydraulic conductivity for the cell containing the well which exceeded the geometric mean.

Eleven sets of simulations were completed with random field statistics shown in Table 1. The correlation structure of the conductivity field is anisotropic (with a ratio between the maxi-

Fig. 1. Translation of the physical, hypothetical unconfined aquifer into its mathematical representation which is shown with one possible flow line to the well.

Table 1. General Parametric Description of the 11 Cases Used

Case	$\mu_{\text{Log } K}$	$\sigma_{\text{Log } K}^2$	λ_x/l_x^a	$\theta_{\text{Pri. Cor.}}^b$
1	-4.0	4/9	0.01	0
2	-4.0	4/9	0.03	0
3	-4.0	4/9	0.075	0
4	-4.0	4/9	0.239	0
5	-4.0	4/9	0.378	0
6	-4.0	1/9	0.03	0
7	-4.0	1	0.03	0
8	-4.0	4/9	0.03	30
9	-4.0	4/9	0.03	60
10	-4.0	4/9	0.03	90
11	-4.0	1	0.378	0

^aRatio of the integral scale to flow domain length in the x-direction.

^bOrientation of the primary correlation axis relative to the mean region gradient direction (the x-axis).

imum and minimum integral scales of 4:1) in all cases. Cases 1-7 were run with the hydraulic gradient parallel to the principal axis (large integral scale axis) of the correlation structure. Cases 8-10 involve rotation of the correlation anisotropy relative to the mean regional gradient direction.

For each set of simulations, flow lines emanating from the well were followed upgradient (i.e., via reverse particle tracking) for periods ranging from 5-50 years depending on the particular statistic sought. The statistics recorded from these simulations included: (1) mean, standard deviation, and skew of the distance between the upstream end of the flow line and the well for five years of travel, and (2) contour regions (termed percentile capture contours and discussed below) indicating the uncertainty in the path of the flow line from the upstream point to the well. Of interest in the present paper is the variation of these statistics with changes in the log K variance, the integral scale of the hydraulic conductivity, and the orientation of the principal axis of the correlation structure.

Measurement of the travel distance from the end of the flow line to the well is straightforward. Reverse particle tracking is utilized to follow the flow line upgradient from the well for the specified time period, five years here. Defining the uncertainty in the path taken by that flow line is less precise. In the present paper, a measure of this uncertainty is derived through definition of "percentile capture contours" (PCCs) for both finite travel time and infinite travel time. PCCs are geometric constructs which provide an estimate of the size and shape of the region of the aquifer containing a set percentage of the particle traces.

To illustrate construction of a PCC for both finite time and infinite time, consider a set of 12 realizations of flow lines emanating (upgradient) from a nonpumping well in which the mean regional flow is as shown in Figure 2. The well is located at $x = 50$. For each realization, the flow line is tracked as it moves upgradient over a period of 50 years. The y-location of the flow line is recorded at a number of distances upgradient from the well (in this case, at $x = 49, 47, 45, 40, 35$, and 30). After completion of all M realizations in a simulation set ($M = 12$ in this case), the set of y-ordinates recorded at each x-location are ranked. The resulting data appear as the dots shown in Figure 2, and amount to histograms of y-locations for various distances upgradient of the well.

To derive the Qth PCC for finite time, a closed curve is drawn through these histograms such that, at each x-location, the curve excludes a constant number of the dots, N , where N is

chosen by maximizing $N/M \leq (1.0 - Q/100)$. The PCC is closed at the furthest distance upgradient for which at least N dots are present. For example, a 50th PCC estimated from the data shown in Figure 2 is constructed by excluding 6 particles at each upgradient location (i.e., $\max\{N/12 \leq (1.0 - 50/100)\}$ provides $N = 6$). The points excluded from the PCC are divided equally above and below the PCC, such that the PCC contains the median behavior of the realizations. [Note: if the number of excluded points had been odd, the extra point is arbitrarily placed below the PCC, thus leading to a bias. This bias decreases as the total number of realizations increases.] In the example in Figure 2, all 12 realizations produced flow lines which crossed $x = 49$. Hence, the PCC at $x = 49$ encloses the innermost 6 dots. At $x = 40$, only nine realizations provided flow lines which crossed this x-location (the remaining three flow lines did not reach this location within the allotted time period). Therefore, the PCC is defined by the innermost 3 dots (again, the outermost 6 dots are always excluded). At $x = 30$, only five realizations produced flow lines which crossed this x-location. As a result, it is impossible to exclude 6 dots and the PCC is closed at the previous location (in this case, $x = 35$). Figure 2 shows the final estimated PCC for this case as a dashed line.

The infinite time PCC is defined by constructing a curve for which a percentage of the particle traces reaching a particular x-location are included. That is, rather than excluding a set number of dots, one includes a set percentage of the dots at a given x-location. To ensure that the PCC contains at least $Q\%$ of the flow lines, the number of points excluded is always rounded down to the next integer number [i.e., at most $(100-Q)$ percent of dots are excluded]. The excluded points are once again divided equally above and below the PCC, with odd numbers arbitrarily being added below the PCC. These construction statements are once again illustrated in Figure 2 for the 50th PCC. As noted above, all 12 flow lines crossed $x = 49$ within the allotted time. As a result, 6 dots were excluded (and 6 included) in the PCC. This results in the PCC being defined by the central 6 dots. Only 11 flow lines crossed $x = 47$ within the allotted time. As a result, 5 dots are excluded and 6 dots are included (i.e., at least 50% of the dots are included in the PCC). The 5 excluded points are divided as 3 dots below the PCC and 2 above the PCC. Similar reasoning applies for each lateral location through $x = 30$ where only 5

Fig. 2. Example of particle trace ordinate sorting for various x-locations. Delineation of the 50th PCC is completed for infinite time (solid line) and finite time (dashed line).

realizations produced flow lines which had travelled this distance. In this case, 50% of 5 leads to the exclusion of 2 dots (2.5 rounded down), yielding the PCC as the central 3 dots.

This construction method for the PCC is based on two general assumptions. First, with a finite number of realizations, the histogram of locations where flow lines cross a particular x-coordinate represents only an approximation of the true distribution of flow lines which could be produced by a particular set of random field statistics. This problem can be minimized by utilizing relatively large numbers of realizations (see discussion in the next paragraph). Second, because the simulations are limited to finite transport times, the definition of the infinite time PCC is based on fewer data points as the distance upstream of the well increases (e.g., less than half the original 12 particles cross $x = 30$ for the problem illustrated in Figure 2). Fewer flow lines at a given x-location imply a poorer representation of the true distribution of flow lines at that location. This problem can be minimized by using large enough transport times that the majority of the flow lines cross the furthest upstream location for which the PCC is to be defined.

Examples of 20th, 50th, and 90th PCCs generated for Case 2 of the present study are shown in Figure 3. This figure contains only the finite time PCCs corresponding to a 50-year travel time (both finite and infinite PCCs are shown in subsequent comparisons). The maximum widths range from 100 meters for the 20th PCC to 500 meters for the 90th PCC. As expected, a substantially larger uncertainty exists for the 90th PCC than the 50th PCC, since approximately five times fewer "extreme paths" are excluded.

It is important to verify that the contours and statistics derived from 250 realizations approximate the population statistics to a reasonable degree. This assertion is tested by comparing the 99th PCC based on 250 realizations with that for 500, 1250, and 2500 realizations. This should be a very conservative benchmark, since all results discussed below are based on the 50th PCC. As shown in Figure 4, the estimated PCC does not vary significantly as the number of realizations increases. Larger numbers of simulations serve primarily to smooth the boundary as opposed to drastically relocating it. Hence, it is concluded that 250 realizations provide reasonable statistics for the present study. (There is some indication that 250 realizations are inadequate for very long integral scales—see Cole, 1995).

4.0 Results for Five-Year Travel Distance

Table 2 shows the statistics generated for the travel distance between the well and the upstream distance to the start of the flow line over a five-year period utilizing Cases 1-5, i.e., those involving variation in the integral scale. These results indicate that regardless of correlation scale, the average travel distance is at least 40 percent larger than the distance predicted by a homogeneous solution (251 meters) based on the geometric mean hydraulic conductivity. Further, the average travel distance increases as the integral scale increases. The standard deviations are on the order of magnitude of the mean distance travelled and the skew is large for the longer integral scales. For the integral scale ratio of 0.38 (Case 5), three percent of the realizations led to particles travelling further than the distance from the left edge of the grid to the well (approximately 3500 meters).

Large standard deviations and a positive skew are indicative of flow following preferential pathways in a number of the realizations. Specifically, as the integral scale increases, the

Fig. 3. Example 50-year (finite time) PCCs for: 20 percent—dash-dot line, 50 percent—dashed line, and 90 percent—solid line. Note: y-axis is exaggerated.

Fig. 4. Convergence of the infinite travel time 99th PCC for Case 11. Contours correspond to: 250—dotted line, 500—dot-dash line, 1250—dashed line, 2500 realizations—solid line.

probability that a particle travels predominantly within a high permeability zone increases. Thus, preferential flow through high permeability regions appears responsible for the increased mean travel distance for the larger integral scales.

The consistently large mean travel distance (as compared to that predicted using the geometric mean) for all integral scales is

Table 2. Five-Year Travel Distance Statistics Generated from Cases 1-5

Case	λ_x/l_x	Mean ^a	Stand. dev.	Skew
1	0.010	352 m	163	0.650
2	0.030	512 m	275	0.729
3	0.075	625 m	353	0.863
4	0.239	757 m	626	2.24
5	0.378	875 m	739	1.97

^aThe travel distance using a homogeneous geometric mean K was 251 meters.

thought to be related to two effects. First, as just discussed, preferential pathways lead to a skewed distribution of travel distances and an increase in the mean. This argument appears consistent with theoretical derivations showing effective hydraulic conductivity to be anisotropic in conductivity fields for which the correlation structure is anisotropic (e.g., Indelman and Dagan, 1993). In such studies, it has been shown that the conductivity parallel to the primary axis of correlation (in Cases 1-5, parallel to the regional gradient) is greater than the geometric mean conductivity, leading to larger travel distances under the flow conditions modeled here.

Second, as noted previously, the realizations were limited to random fields in which the well was located in a region containing higher than average hydraulic conductivity. As a result, each realization is biased towards longer travel distances. This second argument was investigated utilizing realizations without restricting K at the well for a limited number of realizations. These efforts demonstrated that conditioning did increase mean travel times, but not to the degree required to explain the difference between the observed results and the travel time predicted based on the geometric mean.

When the log K variance is increased while the integral scale is held constant, the mean and standard deviation of the travel distance increased. As shown in Table 3 for Cases 2, 6, and 7, the skew also increases as the variance increases. Thus, similar to an increase in the integral scale, an increase in the variance of log K results in further concentration of flow along selected paths, due to the larger differences between low and high K regions.

The influence of rotating the correlation structure relative to the regional hydraulic gradient is illustrated via the results reported for Cases 2, 8, 9, and 10 in Table 4. $\theta_{\text{Pri Corr}}$ is defined as the angle between the x-direction of the grid and the direction of the largest integral scale (for the hydraulic conductivity). It is observed that the mean and standard deviation of travel distance decrease as $\theta_{\text{Pri Corr}}$ increases. No clear pattern is apparent for the skew, which had a maximum at a 60° angle. Finally, the average direction of travel was not aligned with the x-axis for either the 30° or 60° results. Such deviation from the x-axis is predicted by theoretical consideration of flow in statistically anisotropic media, e.g., Gelhar (1993).

5.0 Fiftieth Percentile Capture Contours

Construction of the PCCs was indicated in Figure 2, and their general shape in Figure 3 was seen to consist of elongated

Table 3. Five-Year Travel Distance Statistics for Cases 2, 6, and 7

Case	$\sigma_{\text{Log K}}$	Mean	Stand. dev.	Skew
6	1/9	346 m	106	0.343
2	4/9	512 m	275	0.729
7	1	660 m	412	0.735

Table 4. Five-Year Travel Distance Statistics for Cases 2, 8, 9, and 10

Case	$\theta_{\text{Pri. Corr.}}$	Mean	Stand. dev.	Skew
2	0 degrees	512 m	275	0.729
8	30	440 m	228	0.673
9	60	290 m	137	0.978
10	90	266 m	108	0.536

Fig. 5. Fiftieth PCC for Cases 1-5 which involve various integral scales of the hydraulic conductivity using the (a) 50-year travel time estimate, and (b) infinite travel time estimate. Line types utilized are: Case 1—x, Case 2—dotted line, Case 3—dashed line, Case 4—dash-dot line, Case 5—solid line. Note: y-axis is exaggerated.

regions whose transverse width decreased as one approaches the well. The variability of the PCC shape is considered for the cases outlined in Table 1.

5.1 Integral Scale

The dependence of the transverse width of the PCC on the integral scale was investigated. This investigation was based on analysis of Cases 1-5 and the 50th and 99th PCCs. For the present paper the 50th percentile contours were chosen, since less noise is apparent for them than for the 99th PCCs. Figure 5 shows a comparison of the 50th PCCs for 50-year travel times. Figure 5a shows the results for a finite travel time and Figure 5b shows the results for infinite travel time, indicating a widening of the PCC as the integral scale increases. Further, based on Figure 5a, an increase in the integral scale leads to an elongation of the PCC (consistent with the travel distance statistics reported in Table 2). Due to variability in the results for the larger integral scales, it is difficult to state with confidence how the PCC varies for Cases 3, 4, and 5, except to note that these PCCs are significantly larger than those delineated for Cases 1 and 2. In all

cases, the lateral spread, even for the 50th PCCs, was quite substantial, amounting to approximately 500 meters at maximum in this particular situation.

It is noted in Figure 5b that the widths of the PCCs for infinite time increase with distance upstream from the well. However, for shorter integral scales (Cases 1 and 2) the width remains approximately constant for distances greater than 15 integral scales upstream from the well. Due to the proximity of the upstream boundary, it cannot be determined whether the same behavior would be observed for the larger integral scales.

5.2 Variance of the Hydraulic Conductivity Logarithm

The maximum transverse width of the 50th PCCs increased for an increase in the variance of $\log K$ as shown in Figure 6 for Cases 2, 6, and 7, but to a lesser degree than in the previous comparison. These data suggest that a larger $\log K$ variance increases the uncertainty of the flow path position near the well and the absolute distance travelled, while the transverse width of the PCC is dictated more by the integral scale.

5.3 Maximum Correlation Direction

The hydraulic conductivity statistics in the preceding examples have been evaluated for simulations where the maximum integral scale was aligned with the regional flow direction. In Cases 8-10, the alignment of the statistical anisotropy is rotated relative to the regional flow. Figure 7 indicates that the primary result of rotating the correlation anisotropy is a rotation of the mean flow direction, up to 20° from the regional flow direction. Despite this significant deviation in the mean flow direction, however, the width of the 50th PCC remains approximately constant for all orientations of the anisotropy. The upstream extent of the contours also supports the earlier observation that the mean travel distance decreases with rotation of the anisotropy (as reflected here in the shortening of the length of the 50th PCC progressively from Case 2 through Cases 8, 9, and 10).

The deviation in the mean direction of transport with rotation of the anisotropy of the hydraulic conductivity correlation structure is a vivid illustration of the large scale anisotropy in effective parameters predicted by other authors through theoretical analysis (e.g., Gelhar, 1993; Indelman and Dagan, 1993). In these previous theoretical studies, it has been demonstrated that statistical anisotropy in a random field which is physically isotropic at each point in space will lead to overall behavior of the flow field which is consistent with an anisotropic effective hydraulic conductivity tensor. Although comparing the exact angles observed in our work with the angles predicted theoretically is beyond the scope of the present paper (see Cole, 1995), the results shown in Figure 7 are clearly more consistent with an anisotropic hydraulic conductivity field (for the 30° and 60° rotations) than one which is isotropic.

6.0 Conclusions

Through examination of the variability of flow lines intersecting a passive well in a heterogeneous, unconfined aquifer via Monte Carlo analysis, it has been shown that travel distance predictions based on homogeneous, isotropic K (specifically, predictions based on the geometric mean value for the site) may lead to substantial errors in terms of actual travel distance and direction of a conservative contaminant in a heterogeneous, statistically anisotropic system. Among the errors noted was the

dependence of the mean travel distance on the correlation structure of the conductivity field (with the mean travel distance for a correlated field larger than the distance for a homogeneous field for all integral scales tested). Further, the mean orientation of travel deviated from that predicted for an isotropic medium when the correlation structure of the hydraulic conductivity was anisotropic. This latter observation is potentially of major significance as it implies (consistent with earlier theoretical work by other authors) that even in media which are locally isotropic (such as coarse sands or gravels), the mean direction of water movement may not be perpendicular to regionally derived contours of hydraulic head if the correlation structure of the medium is anisotropic.

Through this study of the variability in the flow line feeding a passive well, it was observed that in addition to variability in the mean transport properties (mean distance and mean orienta-

Fig. 6. Fiftieth PCC for Cases 2, 6, and 7 involving variation in the $\log K$ variance and a 50-year travel time. Line types are: Case 2—dashed line, Case 6—dash-dot line, Case 7—solid line. Note: y-axis is exaggerated.

Fig. 7. Fiftieth PCC for Cases 2, 8, 9, and 10, involving rotation of the correlation anisotropy relative to the x-direction in the model, and a 50-year travel time. Line types are: Case 2—dotted line, Case 8—dot-dash line, Case 9—dashed line, Case 10—solid line. Note: y-axis is exaggerated.

tion), substantial variability around these means was observed. In our effort to characterize this latter variability, the percentile capture contour (PCC) was defined as a graphical representation of the uncertainty in the location of the travel path to the well. In the present study, the 50th PCC was the primary PCC used to investigate spatial variability in the flow line. Based on the 50th PCC, it was observed that the primary influences on the PCC were the integral scale and the primary axes of anisotropy of the correlation structure. The variance in log K also influenced the PCC to a lesser degree. Again, it is stressed that the mean orientation was aligned with the mean regional gradient only when one of the two primary correlation axes was aligned with the mean regional gradient. As noted above, these results correspond with theoretical arguments regarding effective hydraulic parameters in hydraulic conductivity fields characterized by anisotropic correlation structure. Significantly, these results (and the theoretical results presented in the literature—e.g., see Gelhar, 1993, for a summary) indicate that the parameters to which the variability in the flow line is most sensitive (i.e., the integral scale and anisotropy of the correlation structure of the hydraulic conductivity) are perhaps the most difficult parameters to estimate on a limited budget at a field site.

The result of this study, then, leads to the observation that there is significant uncertainty in defining the flow path to a passive well in an unconfined aquifer. This uncertainty can only be reduced through collection of additional field data via carefully determined sampling, discussion of which is beyond the scope of the present paper. In the absence of information beyond a general knowledge of the mean hydraulic conductivity and the mean gradient direction, these results along with previous theoretical studies lead to the conclusion that the field hydrologist should add a degree of safety (e.g., a safety factor) to calculations of both the expected distance of travel and the expected orientation of travel. First, based on the results presented, uncertainty in the flow direction may be as high as ± 30 degrees if no information on anisotropy of the conductivity structure is available. This uncertainty is in addition to the uncertainty in the estimate of the regional gradient due either to temporal changes in the gradient or estimation of the gradient based on limited data. These latter uncertainties have been shown to be quite large in the absence of reliable field data sets (e.g., Cole and Silliman, 1996). Further, other authors have observed, or their data have suggested, significant deviations between expected direction of chemical transport and observed chemical transport (see, for example, the comments of Gelhar, 1993, p. 156).

Secondly, uncertainty in the mean distance of travel appears to be generally covered, in the present study, by a safety factor between 2 and 3 relative to the geometric mean. This second safety factor is more difficult to define from the present work, however, as two competing limitations of the present work directly influence this number. First, as noted above, the mean travel distance is artificially high here as the realizations are limited to those with high hydraulic conductivity in the region surrounding the well. Second, an additional uncertainty is inherent here as it is assumed that the geometric mean conductivity is known for the specific aquifer. Uncertainty in the estimate of the geometric mean contributes directly to a larger uncertainty in the ratio of the mean travel distance to the travel distance estimated using the approximate mean conductivity. Despite these latter uncertainties, however, it is felt that a factor of safety between 2

and 3 will cover the mean behavior under most situations involving nearly steady flow in an unconfined aquifer.

In summary, it is concluded that prediction of transport routes to a passive well without consideration of a PCC or safety factors (or collection of substantially greater amounts of data) does not accurately reflect the uncertainty inherent in characterizing flow/transport in the presence of hydraulic heterogeneity. Hence, it is recommended that practicing hydrologists present field analyses in terms which recognize this uncertainty through inclusion either of factors of safety or another means of expressing this uncertainty to the client.

7.0 References

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Design of a piezometer network for estimation of the sample variogram of the hydraulic gradient: The role of the instrument

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Abstract. The process of designing a piezometer network for estimating the sample variogram of the hydraulic gradient is used to illustrate one strategy for designing a sampling network which is both appropriate for the estimation of spatial statistics and efficient in terms of the measurement instrument. Constraints which impacted this design can be broken into three categories: (1) constraints discussed by previous authors and related to increasing confidence in the estimate of the sample variogram, (2) new constraints related to the spatial distribution of sample location for the purpose of estimating the sample variogram, and (3) constraints related to the measurement instrument. After assessing a number of sampling strategies, including uniform grids, randomized grids, various geometric patterns, and scaled sampling patterns, an acceptable design for the present application was developed from a scaled sampling pattern which was modified to utilize the instrument more efficiently. In this situation the final design could not have been established except through consideration of both the statistics to be derived from the sampling effort and the instrument used to obtain the measurements.

1. Introduction

The covariance and semivariogram (henceforth called the variogram) are important functions in groundwater hydrology, and the estimation of the sample variogram remains a fundamental process in the analysis of geostatistical structures. The development of an efficient sampling design has long been recognized as a key step in this process, the goal of which is a quality estimation of the sample variogram. *Armstrong* [1984] provided an outline of the difficulties associated with variogram estimation from sampled data. Among the problems noted in that paper were (1) choice of distance classes which produced widely varying numbers of data pairs in each class, (2) sampling from mixed populations without separation of data by population, (3) the presence of outliers or highly skewed distributions, and (4) artifacts due to user errors (such as use of artificial values to account for missing data or adjustment of zero and negative values prior to taking the logarithm of the data).

On the basis of the recognition of the importance of the spatial distribution of sample data as noted by *Armstrong* [1984] and others, *Russo* [1984] and *Warrick and Myers* [1987] have discussed algorithms whereby appropriate sample locations may be identified for estimating the sample variogram. Focusing on the distribution of data pairs used in the estimation of the sample variogram, *Warrick and Myers* [1987, p. 497] formalized the analysis of sampling networks using an optimization scheme based on the following five constraints: (1) For each distance-angle class (used in estimating the sample variogram), the number of pairs should be as large as possible, particularly for short distances; (2) the average of the distances in each class should be close to the plotted distance; (3) the variance of the distances in each class should be small; (4) the average of the angles in each class should be close to the

plotted angle; and (5) the variance of the angles in each class should be small.

In the present study the constraints specified by *Warrick and Myers* [1987] were investigated for application to a design problem involving placement of piezometers for estimation of the variogram of the hydraulic gradient. This design required that three piezometers be utilized for each gradient estimate to be collected (following on the work of *Silliman and Frost* [1996] and discussed below). In placing the piezometers it was desired to maximize the efficiency of use of each of the piezometers while providing adequate gradient data to estimate the structure of the sample variogram over a wide range of distance classes. It was determined that in addition to the constraints for selection of the sampling locations as presented by *Warrick and Myers* [1987], a modified set of constraints on the sampling design was required to explicitly include consideration of increasing the uniformity of sampling in space and the inclusion of the measurement instrument in the design process.

2. The Experiment and Motivation

The design problem discussed below is based on a series of ongoing experiments being conducted on constructed aquifers within the groundwater laboratory at the University of Notre Dame. The motivation for these experiments is the assessment of first-order stochastic theory for flow and chemical transport as applied to random fields subject to high-density sampling. In particular, recent developments [*Zheng et al.*, 1996] have led to the observation that the integral scale and variance of the hydraulic conductivity field may be estimated directly from analysis of the variogram of the hydraulic gradient. The present discussion is centered on the design of the piezometer network used in these experiments to estimate the sample variogram of the hydraulic gradient.

Within the work presented by *Zheng et al.* [1996], local estimates of the hydraulic gradient, termed the "samples" in the present paper, are obtained from head measurements collected from sets of three piezometers arranged in right trian-

Figure 1. Basic geometry of instruments and samples (samples shown as asterisks, piezometers shown as circles).

gles, as illustrated in Figure 1 and following the work presented by Silliman and Frost [1996]. The arrangement of the three piezometers (both the placement as right triangles and the separation among the three piezometers) is herein termed the "instrument." The sample variogram of the hydraulic gradient is therefore estimated from a series of samples (i.e., gradient estimates), each measured with the same instrument (a cluster of three piezometers arranged in the prescribed geometry). The design problem for obtaining the sample variogram of the hydraulic gradient consists of two components. The first is the placement of sample locations so as to allow estimation of the sample variogram. The second is the efficient use of a set of instruments which will produce the desired samples. In this particular case the least efficient use of the instrument would require three new piezometers per sample location. Efficiency can be increased by considering sampling designs in which the head measured at one piezometer is used in the estimation of the gradient at two or more locations. Figure 1 illustrates one design in which individual piezometers contribute to more than one instrument.

3. Constraints for the Sampling Design

As noted in the previous paragraphs the design problem addressed involves placement of piezometers such that the variogram of the hydraulic gradient may be estimated. This design problem involves a number of constraints which can be broadly classified into three groups: (1) constraints common with the work of Warrick and Myers [1987] and related to the use of sample values in the estimation of the sample variogram, (2) constraints which are in addition to those of Warrick and Myers [1987] and which are related either to long lag distances on the variogram or to developing wider coverage of the sam-

ple space, and (3) constraints related to the measurement instrument. These three groups of constraints are summarized in Table 1 and are discussed below.

The three constraints in the first group are similar to constraints listed by Warrick and Myers [1987] and are necessary constraints on sample location for any effort involving estimation of the sample variogram. In addition to this group, the fourth and fifth constraints are identified as a second group relating to distribution of sample pairs.

The fourth constraint was established through consideration of two interests: (1) the intended use of the results of this sampling to assess both the short- and long-scale behavior of the variogram of the hydraulic gradient and (2) the lack of knowledge, a priori, of the length scales of interest for the present application. As a result, it is not possible to state, prior to the experiment, the distance classes which will be of primary importance to the final analysis of the variogram. Because of these competing interests, there is substantial motivation to maintain essentially constant numbers of sample pairs in each distance class of interest.

The fifth constraint is related to ergodicity. From ergodicity one can argue that a spatial statistic will converge to an ensemble statistic only when the spatial statistic is averaged over a large region in space (see, for example, the discussion of ergodicity by Karlin and Taylor [1975]). In practical terms the variability within a random field leads to a concern that reliance on tight groupings of sample pairs within one region of the field may lead to sample statistics (in this case, individual points on the sample variogram) which do not reflect the statistics of the entire field. Corsten and Stein [1994], for example, attribute failure of a nested sampling scheme to reliance on sampling from only limited regions of the field. In the present design effort the fifth constraint is used to give preference to those designs which provide data pairs from multiple regions of the sample space over those designs for which all pairs in a particular distance class of the sample variogram are from a local, clustered region in space.

As noted above, the efficient use of these instruments is an important consideration in this network design. As a result, a third category of three additional constraints is specified which are related to the consistent and efficient use of a limited number of piezometers. As an upper bound to the amount of data which may be gathered, considerations related to the laboratory facilities and the structural integrity of the container housing the experiment led to the assessment that it would be

Table 1. Summary of Design Constraints

Category	Constraint
Constraints discussed in previously published papers	(1) for each distance class of interest, obtain a maximum number of sample pairs (with a minimum of 30 sample pairs, as derived from the suggestion of Journel and Huijbregts [1978]) (2) for each distance class, minimize the difference between the average separation distance and the midpoint of the distance class (the plotted distance) (3) for each distance class, minimize the variance of the distance between sample pairs
Constraints addressing long separation distances and the spatial distribution of data pairs	(4) obtain approximately uniform numbers of sample pairs in all distance classes of interest (5) distribute the sample pairs such that the information contained in each distance class is sampled from multiple regions of the field of interest
Constraints addressing measurement instrument	(6) limit the number of piezometers used to 130 (7) maximize the number of samples obtained per piezometer installed (8) maintain a constant (right triangle) geometry and enclosed area for each sampling instrument

difficult to install and handle more than 130 piezometers (constraint 6). Given that the number of piezometers available for the design is limited, there is an obvious desire to use each piezometer efficiently. This involves obtaining as many gradient samples per piezometer as possible and leads to constraint 7. Finally, as mentioned earlier, to ensure that each sample used to calculate the variogram is obtained using a consistent instrument, only right triangles of constant area will be considered, yielding constraint 8.

The present discussion is limited to analysis of isotropic sample variograms (as evidenced by the lack of discussion of the angle of separation between data pairs). Extension to anisotropic variograms would involve the addition of constraints related to division of sample pairs by direction as discussed by Warrick and Myers [1987]. Discussion of anisotropy is omitted from the present manuscript solely to simplify the following discussion of the impact of instrument on the sampling design.

Finally, it is noted that an analysis of whether these constraints lead to designs which can be shown mathematically to lead to optimal parameter fits to the sample variogram (e.g., using the methods of Kitanidis [1987] or similar parameter estimation techniques) is beyond the scope of the present paper. While these issues are of extreme importance, the present discussion is limited to selection of a sampling design based on the listed constraints. Hence the recommendation of Journel and Huijbregts [1978] of using at least 30 pairs per distance class and the initial constraints presented by Warrick and Myers [1987] are accepted without further analysis.

4. Evaluation of Sampling Designs

The strategy used for the evaluation of sampling designs relative to the constraints outlined above involved modification of the sampling design through a controlled trial-and-error approach. The general steps involved in this approach were (1) selection of a pattern for the locations of samples, initially based upon a uniform grid, a randomized distribution, a geometric design, or scaled pattern; (2) placement of piezometers (three piezometers per sample) to produce the desired pattern of samples; (3) assessment of the adherence of the design to the eight constraints listed above; and (4) modification of the sampling pattern to increase adherence to the constraints.

Various sampling strategies were investigated including a uniform grid, a randomized grid, a geometric pattern approach, a scaled pattern, and a modified scaled pattern. In each case it was assumed that the sampling was performed in a field with dimensions of 1.0 by 1.0 arbitrary units. For each sampling design, pairs of sample locations were subdivided by separation distance, as would be performed for estimation of the sample variogram, and grouped into distance classes for analysis. The adherence to the eight criteria specified above was then assessed for each sampling pattern. For the sake of brevity, only the uniform grid and modified scaled pattern are discussed in detail.

4.1. Uniform Grid

Consistent with the findings of other authors [e.g., Russo, 1984; Warrick and Myers, 1987], it was observed that the use of uniform grid sampling patterns (e.g., rectangular and hexagonal) led to several difficulties. First, the distribution of sample pairs by distance is quite uneven for the uniform grid. For example, a square grid consisting of 144 points, shown in Fig-

Figure 2. Uniform grid pattern for sample locations using 144 sample points.

ure 2, leads to a distribution of sample pairs by distance as shown in Figure 3. It is clear that this uniform grid fails to provide uniform numbers of pairs in each distance class. Further, as the minimum distance achievable with a regular grid is equal to the separation distance between neighboring sample locations, the only way to gain information for the variogram at small distances without increasing the number of sample locations is by reducing the distance between samples. This leads to a reduction of the total area covered by the sampling design and minimizes the information gained at larger separation distances.

Despite these difficulties, the uniform grid sampling scheme fulfills many of the previously stated constraints. For example, information for each of a wide range of distance classes is gained from multiple regions of the field of interest. Further, the sample pairs used in estimating the sample variogram may be separated into discrete distance classes (as shown in Figure 3), thus providing strict adherence to constraints 2 and 3 since all distances within the class are equal and therefore show zero variance and zero bias. Finally, this distribution of sample points also performed relatively well with respect to constraint 7 as this grid of samples may be estimated using only 169 piezometers. In this manner the uniform grid yields 0.85 samples (gradient estimates) per piezometer. It should be noted, however, that this number of piezometers violated criterion 6 (maximum number of piezometers equal to 130).

In an effort to gain a better distribution of sampling pairs by distance class and/or a wider range of distance classes covered by the grid, randomized grids based on an initial uniform grid (following on the discussion of Warrick and Myers [1987]) were investigated. In all cases involving randomized grids, greater adherence to the criteria based on distribution of sample pairs was accompanied by a significant decrease in the efficiency of use of the piezometers. In the limit of a completely random grid of sample locations, the efficiency of the grid decreases to approximately 0.33 samples per piezometer (i.e., three piezometers for each sample). Hence it was concluded that randomized grids would not lead to adequate sampling designs for the present application.

Figure 3. Number of sample pairs per discrete separation distance for uniform grid sampling design (shown in Figure 2). The number of discrete separation distances is 77.

4.2. Modified Scaled Pattern

Scaled sampling patterns were also investigated in an effort to distribute sample pairs more evenly in space and among distance classes. On the basis of a review of common fractal patterns, a two-dimensional extension of the Cantor set [Mandelbrot, 1983] was arbitrarily chosen as the starting point for this effort (Figure 4). This particular design appeared to work quite well in terms of many of the constraints. There were discrete distance classes into which the sample pairs fall. As a result, constraints 2 and 3 were met exactly. Also, there were significant and relatively uniform numbers of sample pairs in distance classes from relatively small separation through relatively large separation. Hence constraints 1 and 4 were also fulfilled by this sampling scheme.

There were, however, two major drawbacks to this scaled sampling scheme. First, the sampling pattern shown in Figure 4 is reflective of a nested sampling scheme as discussed by Corsten and Stein [1994]. As noted by these authors, such a sampling scheme may lead to relatively sparse sampling and thereby to relatively poor statistics. In the case shown in Figure 4 the short distance information was derived primarily from the outer four sampling regions within the sample scheme. Little short-length scale information was obtained along the vertical and horizontal lines bisecting the sample space. Second, the irregular spacing of the sample locations was such that it was difficult to make efficient use of the piezometers in generating these estimates. To obtain the 225 samples shown in Figure 4, a total of 435 piezometers was needed, violating constraint 6 and resulting in an instrument efficiency of 0.52.

In order to address these two problems the sampling scheme was modified through a series of steps, including the following: (1) The Cantor set was truncated so that only selected sample points were included in the fourth generation, reducing the number of points at which samples were to be collected; (2) the sample points were shifted slightly to increase the efficiency of piezometer placement; (3) this increased efficiency allowed

piezometers to be added to increase the density of sampling along the vertical and horizontal centerlines; and (4) eight additional piezometers were added to select locations to improve overall piezometer efficiency. Figure 5 shows the final design of the piezometer network (consisting of 125 piezometers) and the resulting locations of hydraulic gradient samples.

In reviewing the design constraints the sampling network shown in Figure 5 appears to be efficient in terms of each constraint. First, as shown in Figure 6, the number of sample pairs exceeds 30 in a large number of discrete distance classes. In fact, a significant number of classes contain 48 or more data pairs. Hence constraint 1 is satisfied. Because these discrete distances classes may be used to estimate the variogram, the plotted distance for each distance class is unbiased and has zero variance, satisfying constraints 2 and 3. Further, the num-

Figure 4. Scaled pattern of sample locations (includes four generations of Cantor set).

Figure 5. Modified scaled sampling pattern using 125 piezometers to give 148 individual samples (samples shown as asterisks, piezometers shown as circles).

bers of pairs per class is relatively uniform over a wide range of distances, from substantially less than 0.1 to greater than 1.0, which satisfies constraint 4. In terms of the efficiency of the use of the sampling instrument (constraint 7), this design provides for 1.2 samples per piezometer, a substantial improvement in efficiency over the previous designs. Finally, the number of piezometers required in this design is 125, satisfying constraint 6, and the 148 resulting samples are relatively uniformly distributed in space, as required in constraint 5.

5. Discussion

The development of the sampling design for this particular application provides a number of important observations re-

garding the general design of sampling patterns. First, these results are consistent with observations by other authors [e.g., Russo, 1984; Warrick and Myers, 1987] in noting that a uniform rectangular grid leads to two significant problems in terms of variogram estimation: (1) the number of sample pairs per class varies dramatically within the different distance classes, and (2) it is expensive (in terms of number of measurements required) to acquire information at both the small and the large scale utilizing a uniform grid. In contrast, the use of the scaled sampling pattern appears to provide relatively uniform distribution of sample pairs over a wide range of distances. Assuming that care is taken to avoid the sparsity discussed by Corsten and Stein [1994] for nested sampling networks, a scaled sampling design represents a reasonable starting point for design of sampling networks.

Second, and perhaps most significant, these results show the important relationship between sampling instrument and sampling design. In cases where the instrument requires combining measurements from more than one spatial location, the statistics derived from the sampling design will be impacted both by the desired distribution of samples and by the required distribution of measurement locations. In the present case study, practical considerations regarding the number of piezometers which could be used placed an indirect upper limit on the number of samples which could be obtained. As a result, criteria in addition to those discussed by Russo [1984] and Warrick and Myers [1987] were necessary in order to achieve efficient use of the instrument. Consideration of instrument efficiency made random sampling patterns undesirable for the present application, despite their enhanced performance with regard to uniformity of sample pairs per distance class. In contrast, through consideration of the efficiency of the use of the instrument and the additional constraints, a final sampling pattern was chosen which is consistent with all constraints and leads to a final instrument efficiency of 1.2 gradient samples per piezometer. It was only through consideration of constraints on

Figure 6. Number of sample pairs per discrete separation distance for final sampling design. The number of discrete separation distances is 411.

both the instrument and the sample design that the final design for placement of piezometers could be chosen.

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MONITORING HYDRAULIC GRADIENT USING THREE-POINT ESTIMATOR

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ABSTRACT: A new method is discussed for garnering information from water level data. Using measured water level data, a series of local estimates of the hydraulic gradient are generated. By combining these estimates through a series of plots, significant insight can be gained regarding the regional gradient and local variability in the gradient. Utility of this approach for monitoring gradients and temporal changes in the gradient is discussed through application to experimental results from a constructed aquifer, field results from a site in northern Indiana, and numerical simulation. In each case, the estimator provided a measure of the regional gradient and local deviation from this regional gradient. It is shown that the shape of the plots generated is related to errors in measurement and local anomalies in the hydraulic gradient. It is suggested that the shape of the plot also provides insight into the relative scales of the measurement and the integral scale of the underlying transmissivity field.

INTRODUCTION

Monitoring/remediating a contamination event requires estimation of the hydraulic gradient at the scale of the contaminant plume or remediation effort. While the horizontal components of the gradient can be estimated from as few as three points in space, estimation from only three points is subject to significant error (Cole and Silliman 1996; Sudicky 1986). It is therefore recognized that the gradient estimates derived from depth to water measurements collected at only three wells should be used with extreme caution.

When a larger number of wells are available at a field site, the classic approach to utilizing water level data is to contour the hydraulic head measurements [e.g., Fetter (1994), p. 114]. While contouring commonly provides a good overall view of the regional variation in hydraulic head, it can lead to uncertainty in the estimate of the regional hydraulic gradient (in terms of both the direction and the magnitude) and loss of detail regarding local fluctuations in the hydraulic head. The question therefore arises as to whether there are alternative approaches to interpreting hydraulic head measurements that provide additional information for the same effort in terms of data collection and analysis.

A number of authors have addressed this, and similar, questions related to hydraulic heterogeneity through Monte Carlo studies and theoretical analyses of heterogeneous conductivity fields [e.g., Freeze (1975); Gelhar and Axness (1983); Dagan (1986); Neuman and Orr (1993); and a number of other authors]. Among the results from this significant body of literature is the observation that the variability in the gradient will decline as the scale of measurement increases. That is, deviations in the hydraulic gradient tend to be local and related to the length scales of the hydraulic heterogeneity. Further, a number of authors have demonstrated that the variations in the hydraulic head can be used to gain information regarding the distribution of the hydraulic conductivity. The inverse method, in particular, has found wide application to the problem of parameter identification in ground-water projects [e.g., Poeter and McKenna (1995); Harvey and Gorelick (1995); Yeh (1986)].

Under a variety of field conditions, the data requirements and effort required for inverse modeling may not be justified for the application at hand. For example, one might be interested in determining whether the distribution of the hydraulic gradient at a field site might be varying with time through analysis of data collected at the same site on two different dates. Alternatively, one might be interested in estimating whether a local surface water body represents a significant hydrologic feature in an unconfined aquifer.

For these situations, it is suggested that additional insight can be gained beyond simple contouring of the data without necessitating additional data collection. By estimating the hydraulic gradient locally (utilizing subsets of the entire data set), significant insight can be generated in terms of the characterization of both the regional gradient and local variations in the gradient. While built on classic procedures for estimating the hydraulic gradient from depth-to-water data, this approach differs from previous approaches in that it specifically recognizes and makes use of the local variations in the gradient estimate to increase the information available for design of monitoring and remediation systems.

METHOD

The method discussed here is based on estimating the horizontal components of the hydraulic gradient for each possible triplet of wells (excluding those which are colinear). Given three wells with screens at similar depths, the magnitude and orientation of the horizontal components of the local gradient (relative to the local coordinate system used in locating the wells) can be determined through calculation of the coefficients in one of the following equations (assuming steady-state flow with no local sources/sinks):

$$H(x, y) = Ax + By + C \text{ (for a confined aquifer)} \quad (1)$$

$$H(x, y) = (Ax + By + C)^{0.5} \text{ (for an unconfined aquifer)} \quad (2)$$

where x and y = spatial coordinates relative to some established coordinate system; H = measured hydraulic head in the wells; and A , B , and C = constants to be determined. Specifically, given measured hydraulic heads at each of three wells [i.e., measuring $H(x, y)$ at each of these three locations] in combination with the appropriate equation [(1) or (2)], the problem of determining the constants A , B , and C reduces to a problem of solving three equations for three unknowns (see example given here).

From these equations, the magnitude of the gradient is estimated as

$$\text{Magnitude} = (A^2 + B^2)^{0.5} \text{ (confined aquifer)} \quad (3)$$

or

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TABLE 1. Hypothetical Well Data for Example Provided in Text

Well (1)	X-coordinate (m) (2)	Y-coordinate (m) (3)	Measured water level elevation in well (m) (4)
1	534.12	134.37	132.37
2	439.43	236.34	131.86
3	422.13	162.33	132.01

$$\text{Magnitude} = 0.5 [(A^2 + B^2)/(Ax + By + C)]^{0.5} \quad (4)$$

(unconfined aquifer)

The orientation for both confined and unconfined aquifers is

$$\text{Orientation} = \arctan(B/A) \quad (5)$$

It is noted that the formulas for the unconfined aquifer [(2) and (4)] are based on the assumptions that the elevation of the base of the aquifer is approximately constant in space and that the datum used in the measurement of the hydraulic head is the base of the aquifer.

As a simple example of the application of these equations, consider three wells located as shown in Table 1 and with measured hydraulic heads as shown in the table. Assume that these wells are screened at equivalent depths in a confined aquifer and that there is reason to believe that the flow field is at steady state. The three equations derived from these data [(1)] are

$$132.37 \text{ m} = (534.12 \text{ m})A + (134.37 \text{ m})B + C \quad (6a)$$

$$131.86 \text{ m} = (439.43 \text{ m})A + (236.34 \text{ m})B + C \quad (6b)$$

$$132.01 \text{ m} = (422.13 \text{ m})A + (162.33 \text{ m})B + C \quad (6c)$$

Note that these data provide for three equations [(6a)–(6c)] and three unknowns (A , B , and C). This set of simultaneous equations can be solved by hand, through simple formulas, or through simple matrix solution methods to provide unique solutions for A , B , and C . For the numbers given in (6a)–(6c), A is estimated as 0.002559 (m/m), B as -0.002625 (m/m), and C as 131.3558 m. Thus the magnitude of the gradient is estimated as

$$[(0.002559)^2 + (-0.002625)^2]^{0.5} = 0.003667 \text{ (m/m)}$$

The orientation is given by

$$\arctan[(-0.002625)/(0.002559)] = -45.72^\circ$$

or approximately 45° below the positive x -axis.

By repeating this procedure for every possible triplet of wells at a field site (i.e., measuring H at each of three wells and solving for the magnitude and orientation of the gradient), a number of estimates of the magnitude and the orientation are produced. Three plots have been found useful in interpreting these estimates. The first, the logarithm of the magnitude of the gradient versus the area enclosed by the three observation wells, provides direct evidence of the variability of the magnitude of the gradient, the regional magnitude (if one exists within the length scales tested), and the rate of convergence of the magnitude to the regional value (related to the integral scale of the conductivity distribution as discussed below). The second plot, the orientation of the gradient versus the area enclosed by the three observation wells, provides similar information for the orientation of the gradient. The third, a plot of the orientation versus the logarithm of the magnitude, provides a rapid means of identifying outliers in the original data and evidence of regions where the gradient deviates from the regional gradient due to local zones of low permeability.

The utility of each of these plots is illustrated through the case studies presented later.

CONSTRUCTED AQUIFER

A confined "aquifer" was constructed in a laboratory flow cell for the study of hydraulic heterogeneity and chemical transport. The aquifer was constructed as a two-dimensional, stationary, correlated random field utilizing two well-sorted sands with two distinct hydraulic conductivities. Fig. 1 shows the random field (the black region shown in the figure is a dye moving from left to right across the aquifer). The lighter regions consisted of a fine sand with a measured hydraulic conductivity of approximately 0.057 cm/s. The darker regions consisted of a coarse sand with a measured hydraulic conductivity of approximately 0.5 cm/s. The overall dimensions of the aquifer were 0.92 by 0.45 m. The aquifer was everywhere approximately 0.1 m in depth. The correlation structure was modeled after an anisotropic exponential correlation structure. The integral scale in the 0.92 m direction in the flow cell was approximately 0.06 m. The integral scale in the 0.45 m direction was approximately 0.03 m. The conductivity was uniform over the aquifer thickness (i.e., the conductivity varied in two dimensions). The pattern for the random field was generated using the discrete analysis field generator (Silliman and Wright 1991) and the correlation structure was modeled after an exponential correlation function.

Flow was initiated through injection of water into a reservoir at one end of the medium (Fig. 2) while the hydraulic head was held constant at the opposite end of the medium. Hydraulic head was measured at 25 piezometers distributed as shown in the figure. The impact of the distribution of measurement points on the results of the analysis of the gradient is discussed, in part, in the following using the numerical results and is the focus of continuing research. For the distribution of piezometers shown, approximately 2,300 estimates of the gradient were obtained.

As demonstrated in Fig. 3(a), the estimated magnitude of the gradient converged with increasing area enclosed by the three piezometers to a value of approximately 0.045–0.05. As demonstrated in Fig. 3(b), the estimated angle of orientation of the gradient also appeared to converge with increasing enclosed area, but at a slower rate than observed for the magnitude. The angle to which the orientation converged was estimated at approximately 170° relative to the positive x -axis (the x -axis was aligned parallel to the 0.92 m dimension of the tank with x increasing from left to right). Given the constant influx on the left boundary and the constant heads maintained at the right boundary, it was anticipated that this angle would converge to 180° (the orientation that would have been observed for a homogeneous medium with the given boundary conditions). The discrepancy in orientation is related to the large low permeability region in the upper right portion of the porous medium. Due to this low permeability region, observed flow in this tank demonstrated a consistent slope toward the lower right.

The variation in the orientation was observed to be greater than $\pm 90^\circ$ at small enclosed areas. One interpretation of this variability may be derived through consideration of the location of the piezometers in combination with review of the data shown in Fig. 3(c), a plot of the variation of the estimated orientation as a function of the estimated magnitude. The data in Fig. 3(c) indicate that there was a close relationship between the magnitude and the orientation. Specifically, the orientation deviated significantly from the regional orientation for those data triplets for which the magnitude was significantly greater than the regional magnitude. By isolating those particular data triplets (deviation in orientation combined with high magnitude) and plotting them on Fig. 1, it was observed that these

FIG. 1. Photograph of Constructed Aquifer Used in This Study. Overall Dimensions of Medium Are 0.92 m (Horizontal) by 0.45 (Vertical) by 0.106 m

FIG. 2. Location of 25 Piezometers Relative to Coordinates within Flow Cell

triplets coincided with regions of low permeability where flow was moving under high gradient from one high permeability region to a second high permeability region. Hence, it is concluded that triplets that show both large deviations in orientation and high magnitude of the gradient are indicators of regions within the aquifer where flow is crossing low permeability lenses from one region of high permeability to a second region of high permeability.

In this case study, therefore, the three-point estimator provided not only high-quality estimates of the regional gradient, but also evidence of local variations in the flow field related specifically to regions of low permeability.

DOUGLAS ROAD LANDFILL

The Douglas Road Landfill (in north-central Indiana) is a hazardous waste landfill currently under study for remediation by the U.S. Environmental Protection Agency. The landfill is located in a glacial outwash aquifer consisting primarily of coarse sands and gravel with occasional silt and clay lenses. The aquifer is underlain by a discontinuous clay layer, which itself is underlain by additional sand. The local hydrology has

been studied through installation of 22 wells at three elevations within the upper and lower sands. For the present discussion, consideration is limited to 10 wells located at consistent depths between 5 and 8 m (Fig. 4). Similar results are obtained for 10 wells located at consistent depths between 17 and 20 m (also in the upper sand). However, insufficient data are available (two wells) to interpret the data from the lower sand.

In Fig. 4, the x-axis shown is parallel the east-west compass direction. Douglas Road passes south of the site. There are no significant topological or structural features in this area of the site. The landfill runs parallel to the north-south axis between approximately 670 and 760 m east on this plot. The landfill extends from approximately 600 to approximately 1200 m on the north-south axis.

The analysis presented here involves comparison of data collected during two sampling periods (May 1994 versus August 1994). As shown in Figs. 5(a) (May) and 6(a) (August), both sampling periods provide convergence of the magnitude to approximately 0.003 (± 0.0005).

As shown in Figs. 5(b) (May) and 6(b) (August), the orientation also converges for large enclosed areas for all the points from the May sampling period and the majority of the points for the August sampling period. This orientation, approximately 40° relative to the positive x-axis (the x-axis here is oriented in the east-west direction, with east being positive x) leads to the conclusion that flow is generally towards the southwest, in agreement with the contour analysis presented in the consulting report for this site ("Focused" 1995) and observed motion of a TCE plume. However, closer comparison of Figs. 5(b) and 6(b) shows that, for the August data, there is a second orientation of approximately 10 – 20° less than zero relative to the x-axis, which exists at relatively large enclosed areas. By identifying the well triplets that contribute to the points at this orientation [those triplets below the line in Fig. 6(b)], it is observed that a single well (well 8 in Fig. 4) contributes to the majority of these points. A minimum of two hypotheses may be advanced to explain these observations.

FIG. 3. Results of Application of Three-Point Method to Data Collected from Piezometers in Constructed Aquifer

First, the data from this well may be in error (there is no evidence supporting this hypothesis). Second, there may be a local influence that causes a northerly component to the ground-water flow in this region of the site.

As discussed in "Focused" (1995), there is evidence of a secondary plume to the northwest of this site. The possibility of variation in the orientation of the gradient (towards the

FIG. 4. Distribution of Wells at Douglas Road Site (Relative to Local Coordinate Axes)

FIG. 5. Three-Point Analysis Results for May Data from Douglas Road Site. Orientation is Reported in Degrees Relative to Positive x-Axis

northwest) is also suggested in the report. It is suggested here that the water level data are consistent with the interpretation that the orientation of the gradient, at least along the northern portion of the site, varies from southwest to west-northwest

FIG. 6. Three-Point Analysis Results for August Data from Douglas Road Site. Line on Orientation Plot Separates Points with Similar Orientation to May Results from Those Points Indicating Different Orientation to Gradient

during this sampling period. Significantly, the three-point estimator provides direct evidence of which portion of the site might be influenced by this variation in gradient. As such, it represents a powerful tool for monitoring variations, over time, in the hydraulic gradient.

NUMERICAL RESULTS

Both of the data sets discussed indicate that the three-point estimator may provide a means of deriving unambiguous estimates of the regional magnitude and orientation of the gradient. However, our efforts with this estimator indicate that the plots of magnitude versus area and the orientation versus area may also be useful in identifying the magnitude of the sampling scale (sample support) relative to the integral scale of the hydraulic conductivity field. In an effort to further understand this relationship, a random field was simulated and sampled at five different sample supports using the same number and pattern of piezometers. The simulation was based on the MODFLOW program (McDonald and Harbaugh 1988) using 160×160 elements, constant head boundaries on the left and right ends of the grid, and no flow boundaries along the upper and lower edges of the grid. The simulation was run at steady state.

Fig. 7 shows the random field along with the distribution

of points for the largest sampling scale. In all cases, the pattern involved nine wells in the pattern shown. The scale of measurement for the five scales in addition to the largest sampling scale were $1/2$, $1/4$, $1/8$, $1/16$, and $1/32$ of the largest scale. These translate into the sampling networks covering from approximately 25 integral scales (the largest network), 12 integral scales, 6 integral scales, 3 integral scales, less than 2 integral scales, and less than 1 integral (the smallest network). In every case, the sampling pattern was centered on the center point of the numerical grid (i.e., the center of the modeled aquifer).

In Fig. 7, the hydraulic conductivity of the light regions was 0.114 times the conductivity of the dark regions (the same ratio as used in the constructed aquifer). The locations of the wells are shown as white rectangles within the diagram. For graphical reasons, the size of the white rectangles has been exaggerated by a factor of 25 relative to the size of the numerical elements actually containing the wells. The sampling pattern geometry was maintained constant for all networks. Only the distance between monitoring locations was altered. All networks contained the center point of the aquifer as the center point of the network. The lower left quarter of this random field was the random field used in the experimental work (Fig. 1).

In Fig. 8(a), the smallest sampling network, the maximum distance between wells is equal to approximately 1.6 integral scales of the correlation structure of the hydraulic conductivity. In Fig. 8(b), the maximum distance between wells is approximately 3.1 integral scales; in Fig. 8(c), it is approximately

FIG. 7. Random Field and Largest Sampling Network for Numerical Simulation Results

FIG. 8. Variation of Estimated Magnitude versus Enclosed Area for Numerical Results and Different Areas of Coverage of Sampling Network

6.25 integral scales; in Fig. 8(d), it is approximately 12.5 integral scales; and in Fig. 8(e), it is approximately 25 integral scales.

As shown in Fig. 8, the variation of the magnitude of the gradient with the enclosed area was a strong function of the total sampling area. When samples were collected at the local scale [Fig. 8(a)], very little variation in the estimate was observed. By moving this sized sampling network relative to the numerical grid, however, it was observed that the magnitude estimate was very sensitive to the location of the sampling network within the aquifer. Such an estimate might be of significant use in local monitoring of the movement of a plume, but is an unreliable estimator of the gradient for purposes of predicting regional gradients or long term transport of a contaminant plume. As the area covered by the sampling network increases relative to the integral scale of the conductivity field [Figs. 8(b)–8(d)], the variability of the magnitude estimate increases (apparently related to the amount of heterogeneity enclosed within the sample network). With further increase in the length scales of the sampling network, however, the variability in the magnitude estimate once again decreases, approaching zero as the sampling network extends over a large number of conductivity integral scales (approximately 25 in this case). It is noted that the convergence to a large-scale "homogeneous" behavior is consistent with conclusions drawn from the literature on stochastic hydrology [e.g., Gelhar and Axness (1983); Dagan (1986)].

Similar results are seen in Fig. 9 for the estimate of the orientation of the gradient (relative to the mean flow direction). When the sampling network covers only a small area, there is little variation in the estimate of the orientation of the gradient. However, there is a significant probability in this case that the estimate will be biased. As the area covered by the network increases, noise is introduced to the system due to the sampling of different hydraulic heterogeneities. Hence, there is significantly more variability in the orientation estimate for the midsized sampling networks. Finally, as the area covered by the sampling network becomes large relative to the correlation integral scale, little noise is observed for the larger enclosed areas and a high precision estimate of the regional orientation is obtained.

Returning to Figs. 3(a) and 3(b), it might be argued that the behavior of the gradient estimates for the constructed aquifer is similar, for the magnitude estimate, to the noise observed in Figs. 8(d) and 8(e). Further, the plot of orientation versus

FIG. 9. Variation of Estimated Orientation versus Enclosed Area for Numerical Results and Different Areas of Coverage of Sampling Network. Order and Integral Scales for Plots Are Same as in Fig. 8

enclosed area contains variability which reduces with enclosed area in a fashion similar to those shown in Figs. 9(d) and 9(e). This comparison with the numerical results implies an integral scale in this medium on the order of 1/12 to 1/6 of the maximum separation among the wells. In this case, this prediction would provide an integral scale in the range of approximately 6.7 to 13 cm. This is relatively consistent with the design correlation structure of approximately 6 cm parallel to the principal flow direction.

Returning to Figs. 5(a) and 6(a), these results imply that the integral scale at the Douglas Road site is on a scale significantly less than the maximum distance between the wells [e.g., the data shown in Figs. 5(a) and 6(a) show a larger degree of variation at all length scales than observed in Figs. 8(a,b) and 9(a,b)]. Additionally, the presence of noise at the smaller enclosed areas indicates that this system is not to the stage represented by Figs. 8(f) and 9(f) (low noise at all length scales for large sampling network). One might therefore conclude that the integral scale of the conductivity distribution (if such a length scale can be defined) in this region of the aquifer is on the order of approximately 100–300 ft (i.e., between six and 13 integral scales included within the area covered by the wells). Unfortunately, at the present time, there is no independent support for this estimate.

ANALYSIS

Within this paper, it has been shown that the three-point estimator of the gradient represents a valuable tool for monitoring the hydraulic gradient at a field site. Specifically, for sampling networks that include more than 10 integral scales between the most distant wells, this method provides good estimates of the magnitude and orientation of the gradient. In addition, this method provides for a measure of the local variability in both the magnitude and orientation of the gradient. In terms of monitoring variations in the gradient over time, this method has proved quite useful in identifying relatively subtle, local variations in the gradient. Finally, for sampling networks for which the underlying integral scale of the conductivity structure is not known, this method provides insight into the length scale of heterogeneity in the conductivity field.

In terms of monitoring, this method provides significant insight in addition to those insights that would normally be gained from contouring of hydraulic head data. First, this method provides a rapid method of identifying well locations for which the local data are inconsistent with the majority of the field data. Such inconsistency may be related to data collection error or anomalous hydraulic behavior in the vicinity of these well locations. As suggested for the Douglas Road data sets, such anomalies may provide key insight into local, transient variation in the orientation or magnitude of the hydraulic gradient. Second, this method provides a means of estimating the local variation in the gradient. Such information may be valuable in designing a monitoring network with sufficient coverage to capture the potential variability in the geometry of a plume. Once again referring to the Douglas Road site, the information gained from the three-point analysis would be valuable in designing a monitoring network for study of the secondary plume northwest of the site (specifically to determine whether the secondary plume is emanating from the Douglas Road site).

In closing, it is stressed that this method is not designed to replace either contouring or more advanced analysis methods such as inverse modeling. Rather, this three-point estimator method has proved useful at a number of field sites for identification of flow characteristics. As such, it is a valuable addition to contouring (requiring no additional data collection) and may represent a useful precursor to more advanced study utilizing techniques such as inverse modeling. Additional

study to further prove this technique is required, particularly with respect to an analyses of the robustness of the gradient estimates to errors in the measured hydraulic head, extension to three dimensions, extension to flow in converging/diverging flow fields, and extension to more complex geologic settings than described here.

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Observations of measured hydraulic conductivity in two artificial, confined aquifers with boundaries

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Abstract. Recent theoretical efforts have provided strong evidence that the effective conductivity varies with distance between monitoring wells and a point source. In the present study, two artificial confined aquifers were subjected to steady state pumping tests. Analysis of these experimental results indicates dependence of the estimated hydraulic conductivity on distance from the pumping well and the form of the physical heterogeneity present. As the precision of the measured hydraulic heads within the experimental apparatus was limited to ~ 0.5 mm, the experimental results were reproduced numerically. Although some minor discrepancies were noted between the numerical and experimental results, the primary observations were consistent. In all cases the Thiem equation was used in combination with image well theory (to account for the boundary conditions) to analyze the response at 25 observation points (87 observation points used in the numerical experiments), providing a number of estimates of the hydraulic conductivity. Within each experiment it was observed that the variance of the conductivity estimate decreased with the distance between the two observation wells used in the calculation. Further, the mean conductivity varied with distance from the pumping well. In an experiment in which the medium was constructed to mimic a second-order stationary random field the mean conductivity approached, at large distance from the pumping well, the conductivity for a random, anisotropic medium under mean uniform flow. In an experiment in which the medium was constructed to mimic a structured (scaled) medium the mean conductivity decreased, at large distance from the pumping well, below the geometric mean and approached a value between the geometric and the harmonic means of the conductivity field. While the boundary conditions make it difficult to fully analyze the large-dimension behavior of these two media, it is clear from the results that the structured medium performs quite differently than the random field at large-measurement scale.

1. Introduction

A number of authors have considered the influence of hydraulic heterogeneity on the flow and distribution of hydraulic head in the vicinity of a point sink. Specifically, there have been several recent studies of the relationship between the distribution of hydraulic head and the water flux moving toward a point sink. *Gomez-Hernandez and Gorelick* [1989] considered effective model parameters in two dimensions in the presence of point sinks. Among the conclusions reached by these authors was that the effective conductivity (as applied to flow to a point sink in isotropic correlation structures) evaluated at distance from the point sink was approximately equal to the geometric mean of the conductivity distribution. As such, the large-scale effective conductivity was consistent with the result for uniform flow as discussed by other authors [e.g., *Dagan*, 1979; *Gelhar and Axness*, 1983]. These authors suggested that the effective conductivity varied with distance from the pumping well but was consistently between the harmonic and geometric means of the conductivity distribution.

Naff [1991] completed a perturbation solution for flow to a well in three dimensions. Among the conclusions presented in this work were that when the solution was reduced to two dimensions with an isotropic covariance structure, the effective

conductivity in the vicinity of the well asymptotically approached the arithmetic mean of the conductivity distribution. As distance from the well increased, the effective conductivity approached the harmonic mean.

Desbarats [1992] utilized a numerical-empirical approach to study how best to assign transmissivities to elements within numerical models in the presence of hydraulic heterogeneity and point sinks. Although much of the work described by *Desbarats* was focused on selection of parameters for numerical modeling, the author observed that the ensemble mean block transmissivity decreased from the arithmetic ensemble mean for small field sizes to the geometric ensemble mean for large field sizes. As with the earlier work, the transmissivity field was modeled as statistically isotropic.

Neuman and Orr [1993] also studied flow toward a point sink as a portion of their study of conditional moments. Utilizing high-resolution Monte Carlo analysis, these authors concluded that the effective conductivity at distance from the pumping well approached the geometric mean (for a conductivity field which is statistically isotropic), while near the well the effective conductivity approached the harmonic mean. Specifically, these authors noted that while their results were consistent with those of *Naff* [1991] in terms of the rate of reduction in the variance of the specific flux (with distance from the sink), the value for the large-scale effective conductivity obtained was inconsistent with the results reported by *Naff*.

As is apparent from this review and by the discussion pre-

sented by Indelman and Abromovich [1994], the exact nature of the effective conductivity remains a topic of investigation in the groundwater literature. One point which is consistent among these earlier papers is that the effective conductivity will vary with distance from a point source/sink.

In the present study, pump tests were conducted within artificial aquifers. The philosophy of this effort was to illustrate two examples of how the estimated hydraulic conductivity might vary both with distribution of monitoring wells and with average distance of the monitoring wells from the pumping well. In particular, interest is focused on how the measured conductivity might vary if pump tests in heterogeneous media are analyzed on the basis of the assumption of a homogeneous conductivity (the typical field test). Two media are constructed and analyzed on the basis of simulating a confined aquifer with spatially varying hydraulic conductivity. In the first experiment the spatial distribution of the hydraulic conductivity is based on a second-order stationary random field (termed the "random medium" for the remainder of the paper). In the second the spatial distribution is based on a simplified version of a Sierpinski carpet [Mandelbrot, 1983] (termed the "structured medium" for the remainder of the paper). As shown below, the results are strongly influenced by the distribution of the conductivity. Because of limitations in the precision with which hydraulic head can be measured in the laboratory, these two experiments were reproduced numerically to ensure that the observations made on these two experimental results are reproducible (and not simply a function of experimental error).

2. Experimental Design

Both experiments were conducted in a flow cell as illustrated in Figure 1. The cell allows experimental media to be constructed with dimensions of $\sim 0.89 \times 0.45 \times 0.106$ m. For experimental convenience the flow cell was run with the plane of analysis (0.89×0.45 m) oriented vertically. The upper and lower boundaries on the tank were maintained as no-flow boundaries. The left reservoir (reservoir 1) consists of an open reservoir separated from the porous medium by a stainless steel screen on a square lattice support. Tests on this screen/support system indicate that there is negligible head loss across this screen at the flow rates utilized in these experiments. Hence this edge of the flow cell is best represented as a constant head boundary. The reservoir at the right end of the flow cell (reservoir 2) was minimized in terms of volume to assist in chemical tracer tests. As a result, this reservoir could be modified to mimic either a no-flow or constant head boundary condition. In the first set of experiments discussed below this boundary was used as a constant head boundary. Because of concern that there might be measurable head loss across this boundary, this boundary was changed to a no-flow boundary for the second set of experiments.

Two fully penetrating wells and 25 partially penetrating piezometers were installed as shown in Figure 1b. Individual pump tests were run from both of the fully penetrating wells. The piezometers were connected to individual manometer tubes on a manometer board, thus allowing monitoring of hydraulic head. On the basis of working with these manometers it is estimated that the error in reading an individual manometer was slightly <0.5 mm. Manometer tubes were changed on a regular basis to avoid both contamination and microbial growth within the tubes which could negatively impact the precision of the readings. As discussed below, this

precision led to uncertainty in the reliability of the experimental results and thereby motivated the use of a numerical model in confirming the overall conclusions of this study.

Each experiment was conducted through establishing a constant head in reservoir 1 and either a constant head (experiment set 1) or no-flow (experiment set 2) in reservoir 2. Influx to the flow cell was established through injection into the well. The manometers were then monitored to determine when steady state had been achieved. Steady state was determined by monitoring the difference in hydraulic head between piezometers (as required for the Thiem analysis discussed below).

3. Analysis of Experimental Data

All analyses were performed assuming two-dimensional flow in a confined aquifer with the primary axis of flow parallel to the vertical orientation (the 0.89×0.45 m face) of the flow cell. As such, the pumping wells were fully penetrating (perpendicular to the plane of flow), and the piezometers were distributed spatially within the plane of flow. As mentioned in section 1, the analysis utilized for this work was based on the assumption (as commonly used in the analysis of field data) that a homogeneous solution is sufficient for analysis of a pump test. In this case the additional requirement of steady state flow was also assumed. Steady state analysis was required to deal with experimental limitations in the laboratory as control and measurement of the compressibility of the experimental device (necessary to allow running of a transient analysis) was beyond the capability of the current equipment.

Following completion of each experiment, the head measurements were utilized to generate conductivity estimates based on the Thiem equation (applied in the vertical cross section of the flow cell with dimensions 89×45 cm) in combination with image well theory and every combination of two piezometers. The Thiem equation [e.g., Fetter, 1995] provides an estimate of the transmissivity T through measurement of the pumping rate Q and the steady state hydraulic head $h(r)$ at two piezometers which are at distances r_1 and r_2 from the pumping well. The Thiem equation, when applied for $r_1 < r_2 < R$, provides an estimate of the transmissivity as

$$T = Q / \{ (2\pi) [h(r_1) - h(r_2)] \} \ln (r_2/r_1)$$

where R is the effective radius of influence of the well and Q is considered positive for pumping into the aquifer. For the present study it is assumed that R is large compared to the dimension of the flow cell.

The boundary conditions present in the experiment were met through application of image wells (similar to the shape factors utilized by Desbarats [1992]). In theory the boundary conditions given here require an infinite number of image wells distributed around the region of the flow cell. For the present application, only a finite number of image wells were utilized. Convergence to the final solution was based on adding image wells in layers (Figure 2) to the solution until the change in the conductivity estimate was negligible. In this way it was determined that ~ 20 layers of image wells were sufficient to ensure convergence of the conductivity estimates within the precision required for the present study. For all results presented below a total of 30 layers of image wells was used. The final conductivity estimate was therefore estimated from the equation

$$K = Q / \{ (2\pi B) [h(r_1) - h(r_2)] \} \sum I_i \ln (r_{i2}/r_{i1})$$

where B is the depth of the flow cell (10.6 cm in this case), r_{i1} is the distance from the first piezometer to the image well i ,

Figure 1. Schematic of flow cell used in both experiments. (a) Internal dimensions of region containing porous media is $89 \times 45 \times 10.6$ cm. (b) The flow cell including 25 piezometers and two fully penetrating wells as shown (open circles are piezometers and asterisks are pumping wells).

and r_{i2} is the distance from the second piezometer to the image well i . The parameter I_i accounts for the difference between injection and withdrawal at well i . In particular, $I_i = +1$ if the well is an injection well, and $I_i = -1$ if the well is a withdrawal well. The series goes from $i = 0$ through $i = N$, where N is the total number of image wells used. The first term ($i = 0$) covers the real pumping well, while the remainder of the terms represent the N image wells. Analysis of this estimator on a numerical solution of the flow equation for a homogeneous medium demonstrated that when using 30 layers of image wells, the error in the conductivity was $<1\%$ over all pairs of sampling locations used in this study.

Utilizing every possible pair of piezometers from the collection of 25 piezometers in the physical model, there was the

potential to generate ~ 300 conductivity estimates for each experiment. As discussed below, the potential pairs increased to >3000 when 87 observation points were used in the numerical results. Because of heterogeneity and measurement error, however, not every pair provided a positive conductivity estimate. It was determined (through the numerical analysis described below) that the number of pairs for which $h(r_1) < h(r_2)$ because of heterogeneity was $< \sim 7\%$ of all data pairs for all laboratory experiments conducted. Further, the points at which $h(r_1) < h(r_2)$ occurred were primarily for locations where the piezometers were close together and the distance from the pumping well to the piezometers was small (the same regions where experimental error is expected to be significant). Hence it was assumed that all pairs for which $h(r_1) < h(r_2)$

Figure 2. Locations of image wells (open circles) relative to the pumping well (asterisk) and boundaries of the flow cell. The number of layers represented in these diagrams are 1, 3, 5, and 10.

could be omitted from the analysis with negligible impact on the asymptotic means and variances estimated from the laboratory data.

4. Two Experimental Media

Two media were investigated for this study. In both cases the media were constructed with two differently sized sands (mesh sizes of 12–20 for the coarse sand and 40–50 for the fine sand; these give grain size ranges of ~ 0.85 – 1.7 and 0.30 – 0.43 mm, respectively). The hydraulic conductivity of the coarse sand was measured as ~ 0.5 cm s $^{-1}$, while the conductivity of the fine sand was measured as ~ 0.057 cm s $^{-1}$. The sands were not mixed such that both experimental media consisted of two distinct conductivities (with the potential for a thin layer of mixing at the contact between the two grain sizes). All portions of both media were saturated.

The media were packed in 1 cm layers utilizing plastic dividers (with thickness < 1 mm) to establish sharp contacts between regions of different conductivity. Packing was performed under saturated conditions with the sand being poured through ~ 1 cm of water. The flow cell was repeatedly vibrated to maintain consolidation, and the plastic dividers were removed as packing progressed.

A schematic representation and photograph of the first packing are shown in Plates 1a and 1b where the darker regions represent the high-conductivity sand and the light regions represent the low-conductivity sand. This pattern is a simplified version of the Sierpinski carpet [Mandelbrot, 1983]. The present structure is not a true fractal as the structure is limited to three scales of high-permeability inclusions. The significant aspects of this particular packing for the present discussion are that this packing has a structure which does not fit the classical concept of a random distribution of conductivity and that the high hydraulic conductivity regions are poorly connected. This latter observation refers specifically to the fact that a flow line will necessarily pass through a significant length

of the low-permeability sand as it moves from the well to the constant head boundary. The horizontal to vertical scale of the high-conductivity zones was maintained at 2:1 in order to mimic the anisotropy present in the second packing.

The second packing is shown in Plates 1c and 1d, with the light material once again being the low-conductivity sand. This pattern was generated as a second-order stationary random field on the basis of the discrete analysis algorithm [Silliman and Wright, 1988]. The correlation structure was approximately exponential with integral scales of 6 cm in the long (89 cm) direction of the flow cell and 3 cm in the short (45 cm) direction of the flow cell. The medium was uniform over the 10.6 cm width of the flow cell.

5. Results: Estimated Conductivity Versus Distance Between Piezometers

Two length scales were investigated in analyzing the data generated in this study. As shown in Figure 3, the first length scale is the distance between the two piezometers used in applying the Thiem equation. The second length scale, also shown in Figure 3, is the distance from the pumping well to the midpoint between the two piezometers used in the analysis.

Plate 1. (opposite) The two media used in these experiments. The dark regions represent coarse sand with a hydraulic conductivity of ~ 0.50 cm s $^{-1}$. The light regions represent a finer sand with a hydraulic conductivity of ~ 0.057 cm s $^{-1}$. (a) Distribution of conductivity for the structured field. The location of the pumping wells is denoted by the asterisks. (b) Photograph of the structured medium with a dye tracer being transported under mean uniform flow from left to right. (c) Distribution of conductivity for the random medium. The location of the two pumping wells is denoted by the asterisks. (d) Photograph of the random medium with a dye tracer being transported under mean uniform flow from left to right.

Figure 3. The two length scales used to measure the support of a measurement: A, the distance between the piezometers used in the Thiem analysis; and B, the distance between the pumping well and the midpoint between the piezometers used in the Thiem analysis.

While it is difficult to justify these particular measures from a theoretical basis, these two measures provided significant insight into the data collected and represent measures easily determined in the field. Hence the current discussion will be limited to use of these two measures of length scale.

The first analysis performed for both media regarded the variation of the hydraulic conductivity estimate with the distance between the two piezometers used in deriving the estimate. Figure 4a shows results for the structured medium with the well in the center of the flow cell (open circles) and the well in the corner of the flow cell (asterisks). Results for the random field are shown in Figure 4b. These results indicate that the variance of the estimate of the conductivity decreases with increasing distance between piezometers. Both media produce similar results in this regard with the variance becoming quite small when the piezometers are separated by the equivalent of 5–10 integral scales (30–60 cm) within the random medium. This result is not surprising for the random medium as increasing the distance between the measurement points decreases the influence of local variations in the hydraulic head.

While the general behavior of the two media is similar in terms of a reduction in variance with an increase in the separation distance between the piezometers, they are inconsistent in terms of the asymptotic conductivity. Shown in Table 1 are the arithmetic, harmonic, and geometric means of the two conductivity distributions used in these experiments. Also included in Table 1 is the effective conductivity predicted by the equations provided by Gelhar and Axness [1983, equations (52)–(55)] for an infinite, anisotropic, random conductivity field. Figure 4a shows that the results for the structured medium fall below the geometric mean (and approach the harmonic mean for both experiments). In contrast, the results for the random field fall at or above the geometric mean. Hence there is an initial indication that these two media behave quite differently in their asymptotic behavior. As discussed below, this conclusion is further supported by the results of the analysis of variation of the estimated conductivity with distance from the pumping well.

6. Results: Estimated Conductivity Versus Distance From Pumping Well

In order to address variation of the estimated conductivity with distance from the pumping well the individual estimates shown in Figures 4a and 4b were divided into distance classes based on distance from the pumping well. This distance was measured as the length from the pumping well to the midpoint between the two piezometers used (see Figure 3). The distance classes used for experiments in which the pumping well was in the corner of the flow cell were 0–10, 10–20, 20–30, 30–50, and 50–70 cm from the pumping well. For the experiments with the well in the center of the flow cell the distance classes were modified to 0–6, 6–12, 12–18, 18–24, and 24–30 cm. All conductivities within each of these subclasses were then averaged using a geometric mean. The geometric average was used in this case as it was observed that the estimated conductivities,

Figure 4. Variation of the estimated hydraulic conductivity with distance between the piezometers used in the Thiem analysis. Each point represents a conductivity estimate derived from one pair of piezometers. (a) Results for the structured medium (open circles are results when pumping from the center well; asterisks are results when pumping from the corner well). (b) Results for the random medium (open circles are results when pumping from the center well; asterisks are results when pumping from the corner well).

at short distances from the pumping well and for small distances between piezometers, ranged over more than an order of magnitude and were highly skewed toward the higher conductivities. As a result, use of a statistic (e.g., the geometric mean) which provides a measure of central tendency in log space was considered an appropriate measure of the behavior of the conductivity estimates. Arithmetic and harmonic averages were also investigated to determine if the form of the averaging had significant impact. Although the arithmetic average showed somewhat higher variability (because of its sensitivity to high K values in a skewed distribution), all three averaging approaches led to very similar values for the mean estimated conductivity at both large distance from the pumping well and large distance between piezometers.

Figures 5a and 5b show the resulting variation in the mean of the estimated K with distance from the pumping well. The results for the structured medium (Figure 5a) show a trend toward a mean estimated conductivity <0.1 or between the geometric and harmonic mean for this medium (Table 1). For the random field (Figure 5b) the mean estimated conductivity appears to approach a value of ~ 0.24 or approximately equal to the geometric mean. In both cases the mean appears to be approaching a relatively constant value after the equivalent of 5–10 integral scales (30–60 cm) in the random medium.

7. Experimental Error and Numerical Modeling

Prior to discussing these results in detail the question of whether these experimental results are accurate must be addressed. As stated earlier in this paper, the piezometers were read with a precision of ~ 0.5 mm. On a number of occasions in the laboratory the difference in hydraulic head between two piezometers was of the order of 0.5 mm. As a result, there was concern that experimental error could significantly influence the observations and conclusions from these experiments. In order to address the question of experimental error, two additional activities were pursued. First, each experiment was repeated several times at different flow rates. In every case the results were reproducible in terms of both the magnitude of the conductivity estimates and the general relationships among the conductivity estimate, distance between piezometers, and distance from the pumping well.

Second, MODFLOW [McDonald and Harbaugh, 1988], a finite difference solution of the flow equation, was used to simulate these experiments. In order to utilize MODFLOW both conductivity fields were divided into grids of 320×320 elements. The measured flow rates from the experiments were used as the flow rate input into MODFLOW. A discretized approximation of each conductivity field was input into MODFLOW to represent the conductivity distribution. No addi-

Figure 5. Variation of the estimated hydraulic conductivity with distance between the pumping well and the midpoint between the piezometers. (a) Results for the structured medium (open circles are results when pumping from the center well; asterisks are results when pumping from the corner well). (b) Results for the random medium (open circles are results when pumping from the center well; asterisks are results when pumping from the corner well).

tional calibration was performed. Upon completion of the MODFLOW simulation the heads at the piezometers were estimated by recording the head at the center of the element containing the piezometer.

Figures 6a–6d show the quality of the match between the predicted and observed hydraulic heads for the experiments analyzed above. The worst of the fits is considered to be that shown in Figure 6b, the match for the structured medium with the well in the center of the flow field. Even in this case, the match is quite reasonable. The close agreement between the observed and the predicted heads provided significant confidence in the overall results from the experiments as described above.

The close agreement between the experiment and the model also allowed the experimental results to be “extended” through recording heads at a larger number of locations. Specifically, the numerical model was used to generate 87 heads from which to reanalyze the results obtained in the laboratory. The distribution of these 87 “piezometers” is shown in Figure 7.

Table 1. Various Mean Conductivities for the Two Experimental Media

Parameter	Structured Medium	Random Medium
Per cent of medium containing high conductivity sand	39.8	33.7
Arithmetic mean conductivity, cm s^{-1}	0.189	0.351
Harmonic mean conductivity, cm s^{-1}	0.077	0.138
Geometric mean conductivity, cm s^{-1}	0.109	0.240
Effective conductivity using Gelhar and Atness [1983]	0.123	0.270

Figure 6. Comparison of hydraulic heads predicted using MODFLOW [McDonald and Harbaugh, 1988] (open circles) and hydraulic heads observed in the laboratory (asterisks). (a) Structured medium with pumping at the corner well. (b) Structured medium with pumping at the center well. (c) Random medium with pumping at the corner well. (d) Random medium with pumping at the center well.

In repeating the Thiem analysis on these 87 points the results shown in Figure 8 for the estimated conductivity versus piezometer separation (equivalent to Figure 4) were obtained. It is noted that these plots are far more dense in terms of number of conductivity estimates (because of the increase in the number of sampling points) but that they reproduce the general structure observed in Figures 4a and 4b. Specifically, the variance in the estimate of the conductivity decreases with an increase in the distance between piezometers.

With this larger number of estimates the mean and variance of the conductivity could be estimated by distance. Figures 9a-9d show the first and second moments of estimated conductivity for all four experiments. Here it is noted that there is relatively small variation in the mean conductivity over the various distances and that the log variance decreases dramatically with distance between the piezometers. As with the experimental results, the mean conductivity for the structured medium falls at or below 0.1 whereas the mean conductivity for the random medium is ~ 0.25 for both placements of the pumping well.

These numerical data were also analyzed in terms of the variation in the mean conductivity with distance from the

pumping well. As greater distances were possible for the results with the pumping well in the corner of the flow cell, only these results are presented here. Figure 10 shows the variation in the mean estimated conductivity for the random medium (asterisks) and the structured medium (open circles). The asymptotic behavior for the random medium appears to be ap-

Figure 7. Eighty-seven sampling points used in conjunction with the numerical solution of the flow equation.

proaching a value above the geometric mean for this medium. Interestingly, the values obtained at the largest separation distances are nearly equal to the effective conductivity predicted by Gelhar and Axness [1983] for uniform flow in a stationary, random medium with an anisotropy ratio of 2:1 (as shown in Figure 10). The large distance behavior of the mean estimate of the conductivity for the structured medium falls below the geometric mean.

8. Discussion

The combination of the experimental and numerical results presented above lead to several observations. (1) The numerical efforts provide confidence that the experimental error had minimal impact on the observations derived from the laboratory. (2) The experimental and numerical efforts indicate that the best estimates of the regional effective conductivity will be derived from a pump test when the observation well(s) is (are) at several integral scales distance from the pumping well, and for situations in which more than one observation well is necessary, the observation wells are separated by several integral scales. (3) The convergence behavior of the structured medium was quite similar to the behavior of the random medium (in terms of reduction in variance), but the large-scale behavior of the mean estimated conductivity in the structured medium was substantially different than the large-scale behavior of the mean estimated conductivity in the random medium.

8.1. Experimental Error

The original focus of this effort was on the interpretation of experimentally derived conductivity estimates. As a check on these experimentally derived results, a numerical model (MODFLOW) was used to verify the general conclusions from the experimental effort. The numerical model successfully reproduced the general trends in the distribution of the hydraulic head as observed in the experiments. Further, the conductivity estimates derived from the numerical model showed behavior consistent with that observed for the experimentally derived estimates. Hence it is concluded that errors encountered in measuring the hydraulic head had only minimal impact on the observations made regarding the behavior of the estimated conductivity.

8.2. Estimating the Effective Conductivity Using Pump Tests

From the work of such authors as Naff [1991] and Neuman and Orr [1993] one can argue that the effective conductivity varies with distance from a pumping well, approaching an asymptotic limit at a large distance from the pumping well. The results presented above for both the structured and the random media are consistent with the observations of these authors in terms of the observation that the mean estimated conductivities varied with distance from the pumping well. The experimental and numerical results are also consistent with these earlier studies in that the mean of the estimated conductivity appears to be converging to an asymptotic value at distances of ~ 10 integral scales from the pumping well.

The current results also provide additional insight into the use of piezometers in conjunction with pump tests for estimating the conductivity. As noted for both the experimental and numerical results, the variance of the estimated conductivity decreases with increasing separation between the piezometers. Further, the mean conductivity appears to be approaching a constant value. This suggests that should an estimate of the

Figure 8. Hydraulic conductivity estimate as a function of the distance between piezometers for the numerical results. (a) Results for the structured medium (open circles are results when pumping from the center well; asterisks are results when pumping from the corner well). (b) Results for the random medium (open circles are results when pumping from the center well; asterisks are results when pumping from the corner well).

conductivity be derived from a field technique requiring two or more observation points (e.g., in the application, for example, of the Thiem equation), the lowest variance estimate of the large-scale effective conductivity is likely to be obtained when the distance between the observation points is large (e.g., greater than a few integral scales).

8.3. Comparison of the Results for the Structure Medium Versus the Random Medium

Figures 4a, 4b, 8a, and 8b show that the structured medium behaves similarly to the random medium in terms of the variance of the estimated conductivity. For both media the variance decreases rapidly as the separation distance between the piezometers increases. This result is also reflected in the statistics shown in Figures 9a and 9b for the numerical results.

In contrast, the large distance behavior (both distance from the well and distance between piezometers) of the mean estimated conductivity is distinctly different for these two media. As shown for the numerical results in Figure 10, the mean

Figure 9. The mean (open circles) and variance (asterisks) of the hydraulic conductivity estimate as a function of the separation distance between piezometers: (a) structured medium: center well, (b) structured medium: corner well, (c) random medium: center well, and (d) random medium, corner well.

estimated conductivity for the random medium approaches the effective conductivity predicted using the expressions presented by Gelhar and Axness [1983] for uniform flow in a medium with a 2:1 anisotropy ratio in the correlation structure (effective conductivity parallel to the direction of the largest integral scale). In the structured medium the effective conductivity drops below the geometric mean and remains between the geometric and harmonic means.

MODFLOW was used in an effort to understand why the results for the random medium approach the effective conductivity estimated for a mean linear gradient. Using the random conductivity field as input, MODFLOW was used to simulate flow through the random medium subject to constant head boundaries in the left and right reservoirs (and zero pumping). Figure 11 shows a comparison of the equipotentials and flow lines for the original simulation of the pump test and for the latter simulation using two constant head boundaries. As indicated in Figure 11, the flow field established during the pump test is essentially identical, in the left portion of the tank, to the flow field established by constant head boundaries at the left and right ends of the flow cell (for the same total flux across the left constant head boundary). As a result, the distribution of

head between piezometers at distance from the pumping well is nearly identical to the distribution of head at these same piezometers under mean uniform flow. It is therefore not surprising that the estimated conductivity approaches the effective conductivity predicted for mean uniform flow [e.g., Gelhar and Axness, 1983].

The behavior of the estimated conductivity in the structured medium is, however, more difficult to explain. One hypothesis for the low estimated conductivity in the structured medium is illustrated by viewing the distribution of a dye tracer being transported through the structured medium. Shown in Plate 1b is a photograph of the distribution of a (red dye) tracer during a tracer test in which the dye is being transported from left to right under mean uniform flow (established through injection into the left boundary and constant head overflow at the right boundary). The dramatic difference in the characteristics of this flow field as compared to the flow field in the random medium is apparent in comparing Plates 1b and 1d (an identical tracer test for the random medium). The flow paths in the structured medium follow patterns which indicate systematic velocity variations and clear symmetry within this flow field which is not present in the random medium. As such, it is not

Figure 10. Mean conductivity estimated as a function of distance from the pumping well (open circles are structured medium; asterisks are random medium). The various lines on this plot are 1, effective conductivity based on *Gelhar and Axness's* [1983] equation for random medium; 2, geometric mean conductivity for random medium; 3, harmonic mean conductivity for random medium; 4, effective conductivity based on *Gelhar and Axness's* [1983] equations for structured medium; 5, geometric mean conductivity for structured medium; and 6, harmonic mean conductivity for structured medium.

surprising that the large-scale behavior of the estimated conductivity in the structured medium does not reflect theoretical arguments for behavior in stationary, random permeability fields.

Figure 11. Flow lines and equipotentials estimated through numerical modeling for mean uniform flow (defined here as flow driven by constant head boundaries at the left and right boundaries of the numerical grid) versus flow to the pumping well, both in the random medium.

9. Conclusions

A series of experiments on two heterogeneous porous media have led to a number of observations. (1) The distribution of hydraulic head and the distribution of estimated conductivities as observed in the laboratory can be reproduced numerically, implying that the experimental error involved in the experiments is relatively insignificant. (2) For both types of media (structured and random) the results support work by other authors in illustrating that the estimated conductivity varies with distance from the well and approaches an asymptotic limit when the average distance between the piezometers and the pumping well is of the order of several integral scales. (3) The estimated conductivity for the random medium converged, at large distance from the pumping well, to the conductivity predicted from perturbation theory for mean uniform flow in a random porous medium. (4) In contrast, the estimated conductivity for the structured medium converged, at large distance from the pumping well, to a conductivity between the geometric and harmonic mean of the medium. (5) The third and fourth observations together demonstrate that the structure of the porous medium (random versus structured) had significant impact on the large-scale behavior of the estimated conductivity.

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The use of laboratory experiments for the study of conservative solute transport in heterogeneous porous media

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Abstract Laboratory experiments on heterogeneous porous media (otherwise known as intermediate scale experiments, or ISEs) have been increasingly relied upon by hydrogeologists for the study of saturated and unsaturated groundwater systems. Among the many ongoing applications of ISEs is the study of fluid flow and the transport of conservative solutes in correlated permeability fields. Recent advances in ISE design have provided the capability of creating correlated permeability fields in the laboratory. This capability is important in the application of ISEs for the assessment of recent stochastic theories. In addition, pressure-transducer technology and visualization methods have provided the potential for ISEs to be used in characterizing the spatial distributions of both hydraulic head and local water velocity within correlated permeability fields. Finally, various methods are available for characterizing temporal variations in the spatial distribution (and, thereby, the spatial moments) of solute concentrations within ISEs. It is concluded, therefore, that recent developments in experimental techniques have provided an opportunity to use ISEs as important tools in the continuing study of fluid flow and the transport of conservative solutes in heterogeneous, saturated porous media.

Résumé Les hydrogéologues se sont progressivement appuyés sur des expériences de laboratoire sur des milieux poreux hétérogènes (connus aussi par l'expression "Expériences à échelle intermédiaire", ISE) pour étudier les zones saturées et non saturées des aquifères. Parmi les nombreuses applications en cours des ISE, il faut noter l'étude de l'écoulement de fluide et le transport de solutés conservatifs dans des champs aux perméabilités corrélées. Les récents progrès du protocole des ISE ont donné la possibilité de créer des champs de perméabilités corrélées au laboratoire. Cette possibilité

est importante dans l'application des ISE pour l'évaluation des théories stochastiques récentes. En outre, la technologie des capteurs de pression et les méthodes de visualisation donnent la possibilité d'utiliser les ISE pour caractériser les distributions spatiales à la fois de la piézométrie et de la vitesse locale de l'eau dans un champs de perméabilités corrélées. Finalement, des méthodes variées peuvent être utilisées pour caractériser les variations temporelles de la distribution spatiale (et, par conséquent, les moments spatiaux) des concentrations de soluté dans les ISE. En conclusion, donc, des développements récents des techniques expérimentales ont fourni l'occasion d'utiliser les ISE comme d'importants outils d'étude en continu des écoulement de fluides et de transport de solutés conservatifs dans des milieux poreux saturés hétérogènes.

Resumen Los experimentos de laboratorio en medio poroso heterogéneo (conocidos como Experimentos a Escala Intermedia o ISE) están cada vez mejor considerados para el estudio de los sistemas saturados y no saturados. Entre las muchas aplicaciones de los ISE se encuentra el estudio del flujo y el transporte de solutos conservativos en medios con permeabilidad que presentan una cierta estructura de correlación. Avances recientes en el diseño de los ISE han proporcionado la capacidad de crear medios de este tipo en el laboratorio. Esta capacidad es importante para la aplicación de los ISE a la evaluación de las teorías estocásticas recientes. Además, la tecnología de los transductores de presión y los métodos de visualización han permitido que los ISE se usen para caracterizar la distribución espacial de niveles hidráulicos y de las velocidades locales del agua en campos de permeabilidad con determinada correlación espacial. Finalmente, existen varios métodos para caracterizar las variaciones temporales en la distribución espacial (y por tanto los momentos estadísticos espaciales) de la concentración de solutos en los ISE. Se concluye que los desarrollos recientes en las técnicas experimentales han proporcionado una oportunidad para usar los ISE como herramientas fundamentales en el estudio del flujo y transporte de solutos conservativos en medio poroso heterogéneo y saturado.

Key words laboratory experiments · tracer tests · solute transport · intermediate-scale experiments

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Introduction

Hydrologists have a long history of utilizing laboratory experiments in the study of groundwater flow and solute transport. As early as 1905, with Slichter's (1905) remarkable observations regarding solute dispersion and the impact of fluid density on solute transport, laboratory experiments were being used to illustrate flow and transport processes as they impact groundwater systems.

Heavy use of laboratory observations was made in the 1950s and 1960s in an effort to better understand the transport and dispersion of conservative solutes within homogeneous and heterogeneous porous media. These studies provided substantial insight into the process of mechanical dispersion and the relationship among the longitudinal dispersion coefficient, properties of the porous medium (such as the hydraulic conductivity, the uniformity coefficient of the grain-size distribution, or, in a fracture, the surface roughness), and/or the mean pore water velocities (e.g., Orlob and Radhakrishna 1958; Harleman et al. 1963; Rumer 1962; Pfannkuch 1963; Klotz et al. 1980). As discussed by Dagan (1989, Figs. 2.10.3 and 2.10.4, and associated discussion), and based on data summarized in a publication by Pfannkuch (1963), early experiments involving the study of the longitudinal dispersion coefficient within homogeneous porous media covered a range of velocity regimes, including: (a) strongly diffusion-dominated transport; (b) advection dominated transport; and (c) transport velocities outside the upper limit of applicability of Darcy's Law. Dagan (1989) was able to demonstrate general agreement between these early results and early theoretical arguments regarding dispersive transport within homogeneous media (e.g., Taylor 1953; Ogata and Banks 1961; Poreh 1965; Saffman 1960).

Various authors also worked with transverse dispersion within homogeneous media. Among these, Simpson (1962) studied lateral dispersion in a laboratory flow cell by constructing artificial sandstones consisting of crushed quartz cemented with an epoxy resin. The results presented by Simpson (1962) were consistent with several other studies (e.g., Blackwell 1962; Harleman and Rumer 1963; Bruch and Street 1967; Li and Lui 1966) in showing that the ratio D_T/vd (where D_T =transverse dispersion coefficient; v =mean pore-water velocity; and d =mean grain diameter) generally decreases with an increase in the mean pore-water velocity. List and Brooks (1967) summarized the experimental results reported by Simpson (1962) and several other authors, and showed that this relationship between D_T/vd and v was generally appropriate with D_T/vd approaching a constant value as v became relatively large.

Among the earliest work on chemical transport in heterogeneous porous media was the work of Hubbert (1953). In this study, Hubbert demonstrated that regions of contact within a layered medium (layers at an

angle to the mean hydraulic gradient) represented regions in which flow lines were deflected (as suggested by hydraulic theory) and, thereby, regions of convergence/divergence of a solute plume. Skibitzke and Robinson (1963) extended this early work on heterogeneities through a qualitative study of transport in porous media containing lenticular heterogeneities. They demonstrate the impact of these heterogeneities on the dispersion of a solute. Their paper contains one of the first schematics of which we are aware showing the dramatic impact of heterogeneities on the spread of a solute moving with the groundwater flow.

Skibitzke (1964) studied transport in layered media and remarked that there are probably three regions of interest in describing the dispersive process in a heterogeneous medium. These are the local scale in which local dispersion is dominant, a transition scale in which the dispersion process does not follow the available advection-dispersion model (e.g., the Fickian model of dispersive flux), and a large scale where Fickian advection-dispersion theory once again describes the average transport behavior. This early insight into the dispersion process is remarkably consistent with recently published theoretical analyses of the dispersion process. Gelhar and Axness (1983), for example, discuss asymptotic behavior that can be consistent with Fickian dispersion. Dagan (1989), summarizing much of his own earlier work on stochastic analysis of transport, illustrates that the dispersion process is evolutionary, a function of the distance travelled relative to the integral scale(s) of the hydraulic conductivity. More recently, Neuman (1990) argued for universal scaling of the dispersion process, suggesting a continual evolution of the dispersion process as the scale of transport, and thereby the scale of heterogeneity, increases.

A review of these early laboratory efforts and the associated analyses of results (e.g., Dagan 1989; Klotz et al. 1980; List and Brooks 1967) indicates that these experiments provide substantial insight into the processes influencing the transport of conservative solutes through homogeneous and heterogeneous porous media.

Based in part on these earlier successful studies and, in part, on the advancement of theories of flow, transport, and reactions in hydrologic systems, the use of laboratory experiments, herein termed intermediate-scale experiments (ISEs), for the study of flow and transport behavior in heterogeneous media has been increasing over the past 15 years. Areas of application have included the study of fluid flow in saturated and partially saturated systems (e.g., Elsner 1994; Glass et al. 1988; Reible et al. 1990), transport of conservative solutes under uniform- and variable-density flow fields (e.g., Sudicky et al. 1985; Silliman and Simpson 1987; Elsner 1994; Schincariol and Schwartz 1990; Sternberg et al. 1996; Irwin et al. 1996; Huang et al. 1995), multiphase transport (e.g., Kueper et al. 1989; Reible et al. 1990; Dullien and Dong 1996; Caruana and Dawe 1996), chemical reactions (e.g., Cobb and Bower 1991;

Starr et al. 1985), particle transport (e.g., Harvey et al. 1993; Silliman 1996), the impact of macropores (e.g., Wildenschild et al. 1994), and microbial interactions (e.g., Szecsody et al. 1994a,b; Murphy et al. 1997; Cunningham et al. 1991).

These diverse applications of ISEs have contributed to the hydrologic literature in numerous ways, and a complete review of this work is well beyond the scope of the present paper. In the specific areas of fluid flow and transport of conservative solutes through saturated, heterogeneous media, however, the scope of the literature is more focused. Furthermore, the challenges to be faced in extending the use of ISEs to these specific areas are very well defined. This paper focuses specifically on the continuing application of ISEs for the study of fluid flow and the transport of conservative solutes in heterogeneous media. Gene Simpson's historical efforts in the laboratory continue to impact the experimental design of ISEs used in these particular applications.

Recent Applications of ISEs to the Study of the Transport of Conservative Solutes

In this section, a brief review is provided of selected papers representing the range of recent applications of ISEs in the study of the transport of conservative solutes through saturated, heterogeneous porous media. One of the earliest of the recent generation of ISEs was performed under the guidance of Gene Simpson (Silliman 1981) and was published by Silliman and Simpson in 1987. In this study, numerical experiments performed by Smith and Schwartz (1980) were mimicked in a series of laboratory experiments on artificial media containing constructed heterogeneities. By analyzing depth-averaged transport in various media, Silliman and Simpson (1987) were able to demonstrate that the scale effect in dispersion (i.e., the observed increase in the dispersivity with distance from the source) could be reproduced through consideration of hydraulic heterogeneity, a conclusion consistent with numerical results reported by Smith and Schwartz (1980) and more recent theoretical arguments (e.g., Gelhar and Axness 1983; Dagan 1989; Neuman 1990). They also discuss the observation that, for selected packings, the dispersivity appeared to be approaching a constant value at larger transport distances. This result was considered to be consistent in general terms with theoretical results that were published at approximately the same time (e.g., Gelhar and Axness 1983). However, because the heterogeneities utilized were not consistent with the correlated random fields considered in the previous theoretical efforts, no effort was made to quantitatively compare the experimental results with predictions derived from stochastic theories.

Sudicky et al. (1985) and Starr et al. (1985) worked with layered porous media. Considering flow parallel to layering, these authors measured transport of both con-

servative (Sudicky et al. 1985) and reactive (Starr et al. 1985) chemical tracers. Results from these experiments led the authors to conclude that one could match observed breakthrough curves with available transport theory as long as the transport was limited to conservative solutes and the dispersion coefficients (longitudinal and transverse) could be fitted to the experimental data. In the case of reactive transport, however, the authors observed that unique parameters could not be identified and applied to experiments run at different transport velocities. The authors concluded that, at least for the reactive chemical transport, the advection-dispersion model was not correctly representing the physics and/or chemistry of the experimental system.

Huang et al. (1995) performed a similar study, working with two-dimensional, non-layered heterogeneities. These authors demonstrated the impact of spatial heterogeneities on the breakthrough curves observed at various locations within their flow field and concluded that the breakthrough curves observed within homogeneous packings were predictable using classic convection-dispersion theory, but that the breakthrough curves observed in experiments on heterogeneous media were distinctly non-sigmoidal in shape.

Silliman et al. (1987) studied the variation of the longitudinal dispersivity with variation in the mean flow direction within anisotropic media. These authors used an experimental design which allowed flow to be oriented in one of two perpendicular directions. By modifying the form of the local-scale heterogeneity, which led to the macroscale anisotropy, these authors were able to demonstrate that the ratio of the longitudinal dispersivity in the high-conductivity direction (D_{LH}) to the longitudinal dispersivity in the low-conductivity direction (D_{LL}) was strongly dependent on the mechanisms leading to macroscale anisotropy. Specifically, these authors contrasted (a) macroscale anisotropy in the hydraulic conductivity caused by layering at the microscale (leading to $D_{LH}/D_{LL} > 1.0$) with (b) macroscale anisotropy in the hydraulic conductivity caused by anisotropy in the tortuosity of flow at the microscale (leading to $D_{LH}/D_{LL} < 1.0$).

Schincariol and Schwartz (1990) built upon previous work on density-driven transport. These authors completed a series of transport experiments involving fluids with different densities in a variety of homogeneous and heterogeneous porous media. Density contrasts ranging from 1000 to 10,000 mg/L were examined. The types of heterogeneities considered include layered media and media composed of lenticular heterogeneities. These authors were able to demonstrate the significant interaction of physical heterogeneity and hydraulic effects related to density contrasts, and they suggest that density effects could be significant even at relatively low-density contrasts. These authors suggest that their results were qualitatively consistent with theoretical arguments regarding flow stability as presented by other authors (e.g., Wooding 1959).

Elsner (1994) produced one of the first ISE studies in which a random permeability field was constructed according to specified statistics in order to test stochastic theories involving both hydraulics and solute transport. Using a one-dimensional column of length 500 cm, Elsner packed a series of 5-cm layers of sand within the column (see discussion below). Upon completion of the packing of the column, Elsner was able to verify that the physical permeability distribution created in the column was consistent with the numerically simulated random field. Based on flow tests and variable-density tracer tests, Elsner demonstrated that the distribution of hydraulic head within the column (under steady flow and uniform density) was statistically consistent with the predictions derived from earlier theoretical studies (e.g., Bakr et al. 1978). Furthermore, the observed dispersive properties of both high- and low-density plumes were observed to be qualitatively consistent with the properties predicted by the theory presented by Welty and Gelhar (1991, 1992).

Irwin et al. (1996) and Sternberg et al. (1996) also performed solute-transport experiments in one-dimensional columns packed in layers. Irwin et al. (1996) studied scale-dependent dispersion in layered media characterized by periodic heterogeneities. They concluded, based on analysis of breakthrough curves monitored both internally and at the outflow from their column, that the length scale required to reach asymptotic behavior in the dispersivity was a function of the length scale of the periodicity within the column.

Sternberg et al. (1996) considered the potential for non-local dispersion in heterogeneous media. These authors demonstrated that the shape of the observed breakthrough curves at various locations within the column is dependent not only on the material present in the immediate vicinity of the measurement point, but also on the heterogeneity surrounding that measurement location. As a result, these authors concluded that the dispersion process is non-local. These authors also provide discussion regarding the design of their experiment and their efforts to limit the mechanisms which could impact their measure of macroscale dispersion.

Conwell (1997) performed experiments in a two-dimensional permeability field. This field was created to be consistent with a simulated, correlated random field (discussed in more detail below). The goal of Conwell's work was to determine whether the variogram of the pore-water velocity field could be quantitatively estimated either from measurements of hydraulic head (subsequently used to estimate the local hydraulic gradient) or from measurements of the velocity of movement of dye tracers as recorded through the side of the flow cell. Conwell had mixed success in measuring these variograms in the laboratory, depending, to large degree, on the precision of the measurement instruments used and the distribution of sample points within the flow field. This work is discussed in additional detail below.

Quantitative Analysis of Flow and Conservative Transport with ISEs

Extending the use of ISEs into the quantitative investigation of theories of fluid flow and the transport of conservative solutes through heterogeneous media requires various capabilities in the design of the ISEs. Among these are the ability to: (a) design and pack ISEs with permeability distributions consistent with pre-defined structure, such as the correlated random permeability fields; (b) characterize local variations in hydraulic head; (c) characterize the distribution of local fluid velocities; and (d) characterize spatial and temporal variations in the concentration distribution

Ability to Create Porous Media With Correlated Permeability Structures

As noted previously, one of the first examples of designing and packing a correlated, random permeability field with specified statistics is presented by Elsner (1994) and involves a one-dimensional laboratory experiment focused on the assessment of the stochastic predictions of transport behavior in the presence of significant density contrast. Elsner notes that the design of these experiments required consideration of the interaction among various length scales. These include the maximum size of the experimental system that could be housed in the laboratory, the thickness of a sand layer necessary to allow in situ measurement of permeability, the number of sand layers desired per integral scale, and the minimum number of integral scales that were considered necessary to allow comparison with the asymptotic results provided by the stochastic theory of Welty and Gelhar (1991, 1992).

Elsner (1994) constructed the random permeability field by packing layers of sand with different permeabilities within a one-dimensional column. Based on the particular experimental design used to measure permeability (hydraulic heads were measured at pairs of piezometers placed within each layer of sand), Elsner argued that a minimum depth of approximately 5 cm is required in each layer of sand. Furthermore, he argued that a minimum of four layers of sand is desired per integral scale of the permeability distribution. As a result, the integral scale of the final permeability field was set at 20 cm. By maximizing the length of column that could fit within the available experimental facility (approximately 500 cm), Elsner (1994) was able to construct a column containing approximately 25 integral scales of the permeability field. This length was deemed sufficient to achieve asymptotic behavior, as indicated by theoretical analysis (Welty and Gelhar 1991, 1992). After packing the column, Elsner measured the permeability of the individual sand layers and calculated the spatial statistics of the final permeability field. He demonstrated that the sample statistics from the column were in good agreement with the simulated statistics. Furthermore, by using 19 different mixes of sand

sizes, he was able to reproduce a permeability field that is approximately log-normally distributed.

Silliman and Frost (1996) report on experiments conducted in a two-dimensional permeability field that was constructed to be consistent with a computer-generated bimodal permeability field with an exponential covariance structure. This work has since been extended (Conwell 1997) to a second realization of this second-order stationary field and to a porous medium consistent with three levels of a scaled medium. These efforts were performed in a flow cell that had overall dimensions of approximately $1.0 \times 0.5 \times 0.15$ m (allowing a volume for the porous medium with dimensions of approximately $0.9 \times 0.45 \times 0.1$ m).

Design considerations in the development of these fields included considerations of length scales similar to those discussed by Elsner (1994). Firstly, the overall size of the flow cell was limited by the available facility. Secondly, the hypotheses to be addressed (including hypotheses related to flow behavior, chemical transport, and measurement of pore-water velocities) led to the conclusion that a minimum of 15–20 integral scales was required in each random field. Finally, similar to the arguments of Elsner (1994), it was concluded that the integral scale of the random fields should be approximately three to four times the minimum packing dimension. Based on experience in the laboratory, it was determined that the smallest sediment unit that could be reliably packed in a controlled manner was approximately 0.5–1.0 cm in the vertical direction and 1.5–2.0 cm in the horizontal direction. Hence, the minimum integral scale that could be packed in the horizontal direction was approximately 6 cm, whereas the integral scale could be reduced to 2–3 cm in the vertical direction. It was arbitrarily decided that an anisotropy ratio in the integral scale of 2:1 was to be used for these experiments. The integral scale in the horizontal direction was therefore set at 6 cm with a vertical integral scale of 3 cm, providing approximately 15×15 integral scales within the flow cell. Two realizations of these random fields as simulated on the computer and as realized in the flow cells are shown in *Figures 1a* and *1b*. A scaled pattern with three levels of heterogeneity is shown in *Figure 1c*.

Packing strategies are significant considerations when designing ISEs in more than one dimension. As an example, the following multistep process was used to pack the sediment distributions shown in *Figure 1a,b*:

1. A random field generator (Silliman and Wright 1991) was used to simulate a series of realizations of bimodal, correlated random permeability fields with specified first- and second-order moments.
2. Individual realizations of the simulated fields were assessed to determine which realizations best reproduced the specified moments. From these, one realization was randomly selected for the flow cell. A contour plot of this permeability distribution was transferred onto the outer wall of the flow cell.

3. Sands were added to the flow cell, through approximately 1 cm of standing water to ensure saturation, using the pattern drawn on the wall as a template. Separation between regions of sand with different grain sizes was maintained using thin (ca. 0.1 mm) plastic dividers. These dividers were removed as the packing proceeded, and the flow cell was vibrated to encourage uniform consolidation of sediments.

This procedure for packing the flow cells benefited significantly from earlier work by Gene Simpson, who used various techniques in packing flow cells, including producing artificial sandstones (Simpson 1962), using gelatin to “freeze” sediments in predefined shapes prior to packing, and using thin dividers to separate regions of differing grain size (Silliman 1981; Silliman and Simpson 1987). As noted, this latter technique was used in the work described here. The “freezing” approach has been used in various forms by other investigators. For example, Murphy et al. (1997) use sediments frozen in ice to aid in the placement of low-permeability inclusions.

Measurement of the Distribution of Hydraulic Head

One challenge in using ISEs in the assessment of theories involving hydraulics (e.g., Osnes 1995; Neuman and Orr 1993; Dagan 1989; Rubin and Dagan 1988) is to derive measures of hydraulic head that are of sufficient precision to allow characterization of hydraulics at appropriate length scales. To illustrate the level of precision required to characterize the spatial variation in hydraulic head within a typical flow cell, a sample problem is considered here in which flow is occurring through a homogeneous medium with a hydraulic conductivity of 10^{-3} m/s. The Darcy velocity through this medium is set equal to 10^{-5} m/s (0.86 m/d). The resulting hydraulic gradient is equal to 10^{-2} . Consider two piezometers separated by 10 mm and the magnitude of the difference in hydraulic head between these two points. If these two piezometers are aligned parallel to flow, the difference in hydraulic head between these two points is 0.1 mm. In order to measure this difference in hydraulic head with an error of less than, for example, 10% of the true difference, the pressure measurement must be completed with a precision of greater than 0.01 mm. If the piezometers are aligned in a direction at an angle to flow, the difference in head is less than 0.1 mm (approaching zero as the angle approaches 90°), thus requiring still greater precision in the pressure measurement.

References to measurement of differences in water levels using manometers of approximately 0.01 mm occur as early as the work of Meinzer and Fishel (1934) and Fishel (1935). Furthermore, recent advances in the use of vibrating crystal-type pressure transducers reportedly allow measurement of pressures with arbitrarily small precision (precision being a function of the time of integration of counts; C. Neuzil, U.S. Geological Survey pers. commun. 1997). Finally, recent efforts

Fig 1a–c Theoretical and packed permeability fields. **a,b** Images of the permeability field generated on the computer (*left*), and the packed sands in the flow cell (*right*; darker material has the higher permeability). Fields were generated as second-order stationary random fields with a horizontal integral scale equal to approximately 1/15 of the horizontal dimension of the medium and a 2:1 horizontal to vertical anisotropy in the integral scale. The correlation structure was exponential. **c** A field developed from the Sierpinski model of a fractal system (Mandelbrot 1983). Three levels of heterogeneity are included

in our laboratory have shown that averaging over multiple readings derived from high-precision, low-range, strain-gauge pressure transducers can provide pressure measurements with precision greater than 0.01 mm. Hence, it is concluded that the technology exists to provide pressure measurements with the necessary precision.

In recent work, however, Conwell (1997) observes that significant experimental and electronic difficulties need to be overcome in order to obtain head measurements from a large array of piezometers (using either manometers or pressure transducers) with precision across the measurement set of approximately 0.04 mm. Problems observed by Conwell include electronic drift, instrument noise, minute amounts of trapped gas within the connections to the piezometers, problems in establishing a level datum for reading numerous manometers, problems with changing surface tension within

manometers related to microbial growth and chemical interactions, and transients in the pressure response related to hydraulic storage within the piezometer tubes and connections. These problems were in addition to any thermal effects that may have occurred had the experiments conducted by Conwell (1997) not been performed in a constant-temperature/constant-humidity chamber. Conwell observes that the errors in the pressure readings had greatest impact on the measurement of head differences between piezometers oriented perpendicular to the mean regional gradient. As suggested by the simple example above, this observation is due to the typically small magnitude of the gradient component (and hence the need for greater precision of measurement) in this direction.

Based on these observations, it is concluded that pressure transducers are available that, with sufficient care in choosing the recording devices and sufficient av-

eraging over measurements, can provide pressure measurements with sufficient precision for application to ISEs. However, significant experimental challenges remain in actually obtaining pressure measurements with precisions of 0.01 mm or better.

In various experiments on heterogeneous media, the difference in hydraulic head between inflow and outflow reservoirs does not always match that which would be predicted from the mean gradient as measured within the sediments under study (e.g., using piezometers). Possible reasons for this mismatch include experimental difficulties (e.g., error in measurement of the hydraulic head in the reservoirs), significant head loss across the screens separating the reservoirs from the sediments, or non-local and boundary effects (e.g., Neuman and Orr 1993; Sternberg et al. 1996). We continue to study this effect.

Measurement of Local Water Velocities

An important component in many stochastic investigations of longitudinal and lateral solute dispersion is the determination of the spatial variation of the velocity field (e.g., Dagan 1989; Neuman and Zhang 1990). Two general approaches have been employed for measuring local velocities within ISEs: (a) visualization methods, whereby movement of dyes is monitored visually; and (b) estimation of local hydraulic gradients through measurement of hydraulic heads.

An early quantitative study using visualization, or visual recording of the movement of dye tracer fronts, was published by Glass et al. (1988). In the work by Glass et al. (1988), and subsequent work published by Nicholl et al. (1992, 1993), the advective velocity of an invading infiltration front was monitored by following the movement of a colored dye dissolved within the invading fluid. Using cameras and flow cells with transparent walls, these authors were able to study several aspects of fingering in infiltration through unsaturated systems. Dye tracing was also used by Schincariol and

Schwartz (1990) to monitor the movement of variable-density plumes and the formation of density fingering in heterogeneous porous media.

Rashidi et al. (1996) worked at the microscale in saturated media by following the movement of latex spheres as they were transported through a porous medium. By employing a porous medium (glass spheres) and a fluid with matched index of refraction, these authors were able to measure the three-dimensional velocity of motion of individual latex spheres as they moved through the pores of the porous medium. This allowed characterization of the details of the fluid velocity at the pore scale.

Conwell (1997) worked with both the visualization method and the estimation of local velocities through estimation of hydraulic gradients to characterize local velocities in saturated, heterogeneous media. The gradient method required that three piezometers be utilized to provide an estimate of the hydraulic gradient at a point within the ISE. Through knowledge of the permeability and porosity at that point, an estimate of the water velocity was obtained for that point. Design considerations in developing an appropriate piezometer network for this application are discussed in Conwell et al. (1997).

Conwell's (1997) study was designed to provide an assessment of the ability of an investigator to use an ISE to derive the variogram of the velocity field. Although detailed discussion of this work is beyond the scope of the present paper, Conwell produced velocity

Fig 2a,b Variograms of the local water velocities developed by measuring the spatial distribution of hydraulic head. **a** Component of the water velocity parallel to the mean gradient (V_x) evaluated for separation distances oriented perpendicular to the mean regional gradient (y). **b** Component of the water velocity perpendicular to the mean regional gradient (V_y) evaluated for separation distances in the same direction. Variogram window was 7.5° for both cases

Fig. 3a, b Variograms of the local water velocities developed by monitoring the movement of dye tracers. Conditions for **a** and **b** are the same as in *Figures 2a* and *2b*

variograms using both measurement of hydraulic head and measurement of the velocity of motion of dye tracers. These variograms were quantitatively compared with the velocity variograms predicted using a highly discretized numerical solution. *Figures 2* and *3* provide examples of the results obtained. In both cases, the variograms of local velocities are estimated for distance classes oriented along a line perpendicular to the mean direction of flow (herein termed the *y* direction) and using a 7.5° window. *Figure 2a* shows the variogram of the component of the pore-water velocity parallel to the mean gradient (herein termed the *x* direction). The agreement with the numerical result is very good. In contrast, the estimated variogram for the *y* component of the pore-water velocity (*Fig. 2b*) shows dramatic errors. As suggested above and discussed by Conwell (1997), this poor fit is a direct result of the failure to obtain sufficient precision with a pressure transducer to adequately quantify the distribution of the hydraulic head (and thereby the hydraulic gradient and the pore-water velocity) in the direction perpendicular to the mean gradient.

Figure 3 shows the variograms derived from monitoring the movement of colored dyes injected into the flow field. Both variograms in *Figure 3* provide relatively good reproductions of the numerically derived variograms. The overestimate of the variogram at short separation distances observed in *Figure 3a* was shown by Conwell (1997) to be a result of the particular choice of sample locations utilized. The underestimate of the sill (variance) in *Figure 3b* was shown by Conwell to be partially related to the design of the sampling network and partially to a limitation of the visualization method, whereby dye spots become ill defined when they cross contacts between regions of different permeability.

Measurement of Spatial Distribution of Solute Concentrations

Much of the recent theoretical development concerning the transport of conservative solutes in heterogeneous media has involved the study of the evolution of the spatial moments of a solute plume (e.g., Dagan 1989; Neuman and Zhang 1990; Naff 1994; Dagan and Fiori 1997). Estimating the evolution of these moments requires detailed spatial and temporal measurement of solute concentrations.

Three approaches have been utilized for collecting and characterizing concentrations within ISEs. The first approach is based on visualization and involves relating dye concentration to visual intensity on a recording device; the second is withdrawal of microsamples for analysis with standard analytical chemistry techniques; the third is in-situ measurement.

As noted above, other investigators have utilized visualization methods within ISEs to follow solute plumes. As discussed by Schincariol and Schwartz (1990), visualization holds significant potential for quantification of tracer concentration when lighting and color contrast conditions can be optimized. These conditions, however, may be difficult to achieve without careful design of the laboratory within which the ISE is to be housed. A review of this technology is available in Schincariol (1989). Although this approach has the potential to provide the greatest density of concentration data (both in space and time), questions remain as to the precision with which the light intensity can be related to chemical concentration, particularly in real sediments for which the background colors are not spatially uniform. As these questions are addressed, it is anticipated that visualization techniques will be increasingly relied upon to provide detailed spatial and temporal data sets on solute transport.

The second technique, sampling of pore fluid, is based on withdrawal of a fluid sample from the flow field. This approach can be difficult when applied to ISEs, however, because the small volume of water pres-

ent within a flow cell combined with the minimal volumetric fluxes moving through the flow cell make withdrawal of significant quantities of water impossible without significantly affecting the flow field. Furthermore, as discussed by Silliman (1995), the support of the sample can be strongly dependent on both the volume withdrawn and the permeability in the immediate vicinity of the point of withdrawal. Solutions to these difficulties have typically involved use of micro-sampler devices. Szecsody et al. (1994a,b), for example, provide a description of the use of microsamplers in a series of experiments on microbial degradation of Quinoline. This approach to sampling provides the significant capability of allowing complete chemical analysis (within the limits of the sample size) of the pore fluid withdrawn from the flow cell, but limits the number of samples that can be withdrawn both temporally and spatially based on minimizing the impact of the sampling on the flow field.

The third approach expands on earlier designs by Gene Simpson, his students (Silliman and Simpson 1987; Dronfield and Silliman 1993), and other investigators (e.g., Sternberg et al. 1996; Irwin et al. 1996; Huang et al. 1995), and involves use of electrical probes in measuring the concentration of an ionic tracer. Sternberg et al. (1996) and Irwin et al. (1996), for example, used Ag/AgCl electrodes to measure the in situ concentration of a KCl tracer, whereas Huang et al. (1995) used electrical conductance to measure the in situ concentration of an ionic tracer.

Similar to the work of Huang et al. (1995), our recent efforts have involved estimating the concentration of a NaCl tracer through measurement of electrical resistance at a series of twisted pairs of platinum wires. The small diameter of the wires (ca. 0.1 mm) and the ability to install these wires directly through the Plexiglass sidewall of the flow cell allowed installation of over 350 electrodes in the permeability field shown in Figure 1b. The electrodes were arranged in the pattern shown in Figure 4. Using this network of electrodes, the variation in the spatial distribution of a NaCl tracer could be followed temporally during tracer tests.

Fig. 4 Distribution of electrodes in the flow cell shown in Figure 1b

Figure 5 shows typical results obtained from the network of electrodes. In the experiment used to generate the data in Figure 5, an NaCl tracer was advanced through the medium in Figure 1b from left to right. Data obtained from the platinum wire array allowed monitoring of breakthrough curves at various points within the medium. From these records of resident concentration, the geometry of the tracer plume could be monitored over time (e.g., Figs. 5a and 5b). The quantitative nature of these data also allowed estimation of spatial moments of the tracer plume. For example, Figures 5c and 5d show the temporal variation of the first and second spatial moments of the tracer plume parallel to the mean flow direction (x direction in Fig. 4) for the depth-averaged resident concentration (averaged over the y direction in Fig. 4). These curves are truncated at the time at which a substantial portion of the tracer plume had first reached the outflow boundary. Similarly, Figures 5e and 5f show the temporal variation in the standard deviation of the vertical (y direction) variation of the resident concentration for two distances from the inflow boundary ($x=16.5$ cm and $x=56.5$ cm in Fig. 4).

Conclusion

The application of ISEs for the study of groundwater problems is very diverse and is expanding into consideration of such diverse problems as multiphase flow (including both unsaturated systems and systems involving NAPLs), transport and behavior of microbes and colloids, and co-transport of mixed systems (e.g., mixed chemical/microbial systems, or complex mixtures of reacting chemical species). In particular, ISEs will continue to be used both in the investigation of mechanisms (e.g., Kueper et al. 1989; Wildenschild et al. 1994; Szecsody et al. 1994a,b) and in the assessment of specific modeling approaches (e.g., Murphy et al. 1997).

The technology for the use of ISEs has advanced to the stage where ISEs now have the potential to be used for quantitative studies of fluid flow and the transport of conservative solutes in media with complex heterogeneity, such as correlated random structure. Due partly to the contributions of Gene Simpson, methods are available for packing complex, correlated permeability fields within these laboratory flow cells. Furthermore, advances in pressure-transducer technologies have provided the hydrologist with the capability of measuring water levels with precisions of at least 0.01 mm. This precision provides the opportunity to characterize the details of the distribution of hydraulic heads within heterogeneous media in a laboratory setting. Advances in pressure-transducer technology, combined with recent advances in the use and interpretation of visualization methods, have also provided the opportunity to utilize multiple techniques for characterizing local fluid velocities. Finally, various methods, including electrical methods pioneered in part under the direction of Gene

Fig. 5a-f Results obtained from the measurements of concentration in the random field shown in Figure 1b, using the electrode distribution shown in Figure 4. For a and b, the point measurements of concentration were assigned to the rectangular region surrounding the measurement point, thus providing the "checkerboard" look to these plots. Furthermore, electrodes that failed or were considered unreliable were assigned a value of 2.0 and were not included in calculation of any spatial or temporal moments. **a** Spatial distribution of normalized concentration (C/C_0) after approximately 0.2 pore volumes of fluid have passed through the flow cell. **b** Spatial distribution of

normalized concentration after approximately 0.85 pore volumes of fluid have passed through the flow cell. **c** First, spatial moment of the depth-averaged resident, normalized concentration (averaged over the y direction in Figure 4) as a function of transport time. **d** Second centered spatial moment of the depth-averaged resident, normalized concentration as a function of transport time. **e** Temporal variation in the variance of the resident, normalized concentration observed at $x=16.5$ cm from the inflow reservoir. **f** Temporal variation in the variance of the resident, normalized concentration observed at $x=56.5$ cm from the inflow reservoir

Simpson, have been developed for characterizing the spatial distribution of the concentration of ionic tracers. The combination of these capabilities creates the potential for ISEs to become integral tools in the assessment of theories of fluid flow and the transport of conservative solutes. It is anticipated that the use of these types of experiments will continue to expand as the technologies required for these experiments continue to improve.

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Estimating the theoretical semivariogram from finite numbers of measurements

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Abstract. We investigate from a theoretical basis the impacts of the number, location, and correlation among measurement points on the quality of an estimate of the semivariogram. The unbiased nature of the semivariogram estimator $\hat{\gamma}(\mathbf{r})$ is first established for a general random process $Z(\mathbf{x})$. The variance of $\hat{\gamma}_Z(\mathbf{r})$ is then derived as a function of the sampling parameters (the number of measurements and their locations). In applying this function to the case of estimating the semivariograms of the transmissivity and the hydraulic head field, it is shown that the estimation error depends on the number of the data pairs, the correlation among the data pairs (which, in turn, are determined by the form of the underlying semivariogram $\gamma(\mathbf{r})$), the relative locations of the data pairs, and the separation distance at which the semivariogram is to be estimated. Thus design of an optimal sampling program for semivariogram estimation should include consideration of each of these factors. Further, the function derived for the variance of $\hat{\gamma}_Z(\mathbf{r})$ is useful in determining the reliability of a semivariogram developed from a previously established sampling design.

1. Introduction

Given a trend-removed random field $Z(\mathbf{x})$, the theoretical semivariogram $\gamma_Z(\mathbf{r})$ is defined as

$$\gamma(\mathbf{r}) = \frac{1}{2} \text{Var} [Z(\mathbf{x}) - Z(\mathbf{x} + \mathbf{r})] = \frac{1}{2} \langle [Z(\mathbf{x}) - Z(\mathbf{x} + \mathbf{r})]^2 \rangle, \quad (1)$$

where $\mathbf{x}$ is location, $\mathbf{r}$ is the vector lag distance, Var stands for the variance, and the bracket designates the ensemble averaging. Although the determination of the theoretical semivariogram requires ensemble averaging, we are often limited in practice to one realization and a finite number of measurements of $Z(\mathbf{x})$. Thus the ensemble average is commonly estimated from the spatial average by assuming ergodicity. For example, given n measurements $[Z(\mathbf{x}_i), i = 1, \dots, n]$ from a single realization, the most commonly used semivariogram estimator $\hat{\gamma}(\mathbf{r})$ for a specified vectorial lag distance $\mathbf{r}$ is [Matheron, 1965]

$$\hat{\gamma}(\mathbf{r}) = \frac{1}{2N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})} [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2, \quad (2)$$

where $N(\mathbf{r})$ is the number of the data pairs, $Z(\mathbf{x}_i)$ and $Z(\mathbf{x}_i + \mathbf{r})$, available from the n measurements.

The quality of the estimate of the semivariogram for a given $\mathbf{r}$ thus depends on the bias and precision of $\hat{\gamma}(\mathbf{r})$, as an estimator for $\gamma(\mathbf{r})$. Many have used $N(\mathbf{r})$, the number of the data pairs, $Z(\mathbf{x}_i)$ and $Z(\mathbf{x}_i + \mathbf{r})$, as an index for measuring the reliability of $\hat{\gamma}(\mathbf{r})$. For example, a common "rule" applied to estimation of the semivariogram is that at least 30 pairs of

measurements, $Z(\mathbf{x}_i)$ and $Z(\mathbf{x}_i + \mathbf{r})$, are required for each lag distance $\mathbf{r}$ in order to ensure a reliable semivariogram estimate [e.g., Journel and Huijbregts, 1978]. Review of the literature indicates that this rule is derived from a simplification of earlier work by Matheron [1965]. Webster and Oliver [1992] have studied this rule through numerical sampling of a random field generated with a known semivariogram. Using a uniform, square sampling grid and comparing sample semivariograms against the known underlying semivariogram, these authors argued that at least 200–300 measurements are needed to estimate a semivariogram reliably.

This emphasis on $N(\mathbf{r})$ has also resulted in efforts to devise algorithms to maximize $N(\mathbf{r})$ by adjusting the placement of a fixed number of measurements. Russo [1984] and Warrick and Myers [1987], for example, present algorithms which optimize the location of sampling points based on a series of constraints, including constraints on the number of sample points in each lag distance. A recent paper by Conwell et al. [1997] extended this earlier work by taking into account the role of measurement instruments in the sampling design. Morris [1991] argued that accurate estimation of the semivariogram depended not only on $N(\mathbf{r})$ but also on the correlation among the measurements. He proposed an alternative index, the maximum equivalent uncorrelated pairs, as a measure of the estimation accuracy. However, this index applies only to the special case of a concave semivariogram.

Other methods for determining the accuracy of semivariogram estimates involve Monte Carlo simulation [e.g., Russo and Jury, 1987; Corsten and Stein, 1994] and the subsampling method [Chung, 1984; Shafer and Varljen, 1990]. The Monte Carlo approach obtains the confidence interval via repeated sampling from multiple realizations of a random field. The subsampling method involves subdividing a measurement set into subsamples and then estimating the semivariogram from

each subgroup of samples thus allowing characterization of the variation of parameter estimates between subsamples.

In the present paper the variance of the semivariogram estimate $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ is determined theoretically. This theoretical result is used to obtain a functional relationship between $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ and the sampling parameters (e.g., the number of the measurements and their locations). This general relationship then enables us to examine the interactions between the magnitude of $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ and its various controlling mechanisms. The interactions among the number of measurements, sampling locations, the underlying semivariogram, and measurement error are illustrated through applying this relationship to the estimation of the semivariograms of the transmissivity and the hydraulic head field. Finally, the implications of these results are discussed and extended to possible applications for sampling design and interpretation of the reliability of semivariogram estimates.

2. Theoretical Background

The bias of the semivariogram estimator $\hat{\gamma}(\mathbf{r})$ can be examined for a random field by determining the ensemble mean value of both sides of (2). This leads to

$$\begin{aligned}\langle \hat{\gamma}(\mathbf{r}) \rangle &= \left\langle \frac{1}{2N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})} [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2 \right\rangle \\ &= \frac{1}{N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})} \frac{1}{2} \langle [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2 \rangle.\end{aligned}\quad (3)$$

Applying (1) to (3),

$$\langle \hat{\gamma}(\mathbf{r}) \rangle = \frac{N(\mathbf{r})\gamma(\mathbf{r})}{N(\mathbf{r})} = \gamma(\mathbf{r}). \quad (4)$$

Thus $\hat{\gamma}(\mathbf{r})$, as given in (2), is an unbiased estimator for $\gamma(\mathbf{r})$.

Since $\hat{\gamma}(\mathbf{r})$ is an unbiased estimator, $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ can then be written as

$$\sigma_{\hat{\gamma}(\mathbf{r})}^2 = \langle [\hat{\gamma}(\mathbf{r}) - \gamma(\mathbf{r})]^2 \rangle. \quad (5)$$

Substituting (2) into (5) provides

$$\begin{aligned}\sigma_{\hat{\gamma}(\mathbf{r})}^2 &= \left\langle \left[\left[\frac{1}{2N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})} (Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r}))^2 \right] - \gamma(\mathbf{r}) \right]^2 \right\rangle \\ &= \frac{1}{N^2(\mathbf{r})} \left\langle \left[\sum_{i=1}^{N(\mathbf{r})} \left[\frac{(Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r}))^2}{2} - \gamma(\mathbf{r}) \right] \right]^2 \right\rangle\end{aligned}\quad (6)$$

To simplify the notation, we define a new random variable, $S(\mathbf{x}, \mathbf{r})$, as

$$S(\mathbf{x}, \mathbf{r}) \equiv [Z(\mathbf{x}) - Z(\mathbf{x} + \mathbf{r})]^2/2 \quad (7)$$

and obtain

$$\langle S(\mathbf{x}, \mathbf{r}) \rangle = \gamma(\mathbf{r}). \quad (8)$$

Substituting (7) and (8) into (6) results in

$$\sigma_{\hat{\gamma}(\mathbf{r})}^2 = \frac{1}{N^2(\mathbf{r})} \left\langle \left[\sum_{i=1}^{N(\mathbf{r})} [S(\mathbf{x}_i, \mathbf{r}) - \langle S(\mathbf{x}_i, \mathbf{r}) \rangle] \right]^2 \right\rangle$$

$$= \frac{1}{N^2(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})} \sigma_{S(\mathbf{x}_i, \mathbf{r})}^2 + \frac{2}{N^2(\mathbf{r})} \sum_{j>i}^{N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})-1} \text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})], \quad (9)$$

where $\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ is the covariance between $S(\mathbf{x}_i, \mathbf{r})$ and $S(\mathbf{x}_j, \mathbf{r})$ and $\sigma_{S(\mathbf{x}_i, \mathbf{r})}^2$ is the variance of $S(\mathbf{x}_i, \mathbf{r})$.

Equation (9) is essentially identical to equations provided in classical textbooks on statistics for the variance of the sample mean [e.g., *Benjamin and Cornell*, 1970, p. 378, equation 4.1.12] with the exception that here the variable is $S(\mathbf{x}_i, \mathbf{r})$ rather than $Z(\mathbf{x}_i)$. The key to understanding the importance of (9) is to determine the dependence of $\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ and $\sigma_{S(\mathbf{x}_i, \mathbf{r})}^2$ on sampling design.

As given in the appendix, for a Gaussian random variable $Z(\mathbf{x})$, we can derive

$$\begin{aligned}\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})] \\ = \frac{1}{2} [\gamma(\mathbf{r} + \mathbf{R}_{ij}) + \gamma(\mathbf{r} - \mathbf{R}_{ij}) - 2\gamma(\mathbf{R}_{ij})]^2,\end{aligned}\quad (10)$$

where $\mathbf{R}_{ij} = \mathbf{x}_j - \mathbf{x}_i$ is the separation distance between $S(\mathbf{x}_i, \mathbf{r})$ and $S(\mathbf{x}_j, \mathbf{r})$. Since $\gamma(\mathbf{r})$ is not a function of $\mathbf{x}$, $\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ as given in (10) is also independent of $\mathbf{x}$ but depends on the relative separation between $\mathbf{x}_i$ and $\mathbf{x}_j$. It is noted further that $\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ is anisotropic even if $\gamma(\mathbf{r})$ is isotropic. When $\mathbf{R}_{ij} = 0$ in (10), S has variance of

$$\sigma_{S(\mathbf{x}, \mathbf{r})}^2 = \sigma_{S(\mathbf{r})}^2 = 2\gamma^2(\mathbf{r}). \quad (11)$$

Substituting (11) into (9), the expression for $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ may now be written as

$$\sigma_{\hat{\gamma}(\mathbf{r})}^2 = \frac{2\gamma^2(\mathbf{r})}{N(\mathbf{r})} + \frac{2}{N^2(\mathbf{r})} \sum_{j>i}^{N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})-1} \text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})], \quad (12)$$

where the expression for $\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ is given in (10). A similar expression for $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ is used by *Cressie* [1993] in the context of performing weighted least squares fitting.

To facilitate the comparison of various $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ at different lag distances, we divide $\sigma_{\hat{\gamma}(\mathbf{r})}^2$ by $\gamma(\mathbf{r})$ to obtain the coefficient of variation $\rho_{\hat{\gamma}(\mathbf{r})}$:

$$\rho_{\hat{\gamma}(\mathbf{r})} = \left[\frac{2}{N(\mathbf{r})} + \frac{4}{N^2(\mathbf{r})} \sum_{j>i}^{N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})-1} \text{coeff} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})] \right]^{1/2}, \quad (13)$$

where

$$\begin{aligned}\text{coeff} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})] &= \frac{\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]}{2\gamma^2(\mathbf{r})} \\ &= \frac{\text{Cov} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]}{\sigma_{S(\mathbf{x}_i, \mathbf{r})}^2}.\end{aligned}\quad (14)$$

Hence $\rho_{\hat{\gamma}(\mathbf{r})}$ consists of two parts. The first part is completely determined by $N(\mathbf{r})$, the number of data pairs. The second part is a function of both the number of pairs and the correlation among the $S(\mathbf{x}_i, \mathbf{r})$.

In two special cases the expression for $\rho_{\hat{\gamma}(\mathbf{r})}$ can be simplified. First, when the $S(\mathbf{x}_i, \mathbf{r})$ are independent, all $\text{coeff} [S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ are equal to zero and

$$\rho_{\gamma(r)} = \sqrt{\frac{2}{N(r)}} \quad (15)$$

In this case the estimation accuracy depends only on $N(r)$. This relationship is identical to the result from classical statistics, where all the samples are taken independently. The second special case is when $S(x_i, r)$ are completely correlated with each other; all coeff $[S(x_i, r), S(x_j, r)]$ are then equal to one. Thus $\rho_{\gamma(r)}$ reduces to a constant as follows:

$$\rho_{\gamma(r)} = \left\{ \frac{2}{N(r)} + \frac{4}{N^2(r)} \left[\frac{N(r)^2 - N(r)}{2} \right] \right\}^{1/2} = \sqrt{2}. \quad (16)$$

That is, the estimation accuracy becomes independent of sampling. An example of such extreme cases can be found in a random field whose semivariogram grows quadratically with the increasing separation distance.

Other than in these special cases, the estimation precision of $\gamma(r)$ (defined here as the magnitude of $\rho_{\gamma(r)}$) is determined not only by $N(r)$ but also by the summation of the coeff $[S(x_i, r), S(x_j, r)]$, the correlation coefficients among the squared increments, $[Z(x_i) - Z(x_i + r)]^2$. This correlation coefficient, according to (10), is determined by the separation distance between x_i and x_j ($i, j = 1, \dots, N(r)$), the lag distance r , at which the semivariogram is to be estimated, and the underlying semivariogram $\gamma(r)$. Thus (13) gives an explicit expression describing the interactions among the variance of the estimate of the semivariogram, the number of data, and the sampling design. It is a general relationship that holds in any random field $Z(x)$, provided that the increments of $Z(x)$ are normally distributed.

3. Illustration Using Hydraulic Head and Transmissivity Fields

In order to illustrate the impact of (13) on problems of interest to hydrogeologist, a comparison is made between estimating the semivariogram of the transmissivity (T) and estimating the semivariogram of the head residual (h , the hydraulic head minus the mean trend in the head). These particular parameters were chosen as they are two of the most frequently studied spatial processes in groundwater hydrology.

For the following illustration it is assumed that T is log normally distributed. A new, normally distributed random variable Y is therefore defined as $Y = \ln(T)$. It is further assumed that Y is second-order stationary, exists within an infinite spatial flow domain, and is characterized by an isotropic, exponential covariance function

$$\gamma_Y(r) = \sigma_Y^2 [1 - \exp(-r')], \quad (17)$$

where σ_Y^2 is the variance of Y and $r' = |r|/\lambda$ is the magnitude of the separation vector r normalized by the integral scale λ of Y .

Dagan [1985] has presented the first and second moments of the distribution of the head residual under mean uniform flow within the type of transmissivity field described above. Of particular interest here is Dagan's expression for the semivariogram of the head residual:

$$\gamma_h(r) = \frac{J^2 \lambda^2 \sigma_Y^2}{2} \left\{ [2(\cos \psi)^2 - 1] \cdot \left[\frac{\exp(-r')(r'^2 + 3r' + 3) - 3}{r'^2} \right] + [(\cos \psi)^2 - \frac{1}{2}] \right\} \quad (18)$$

Figure 1. Comparison of the head residual semivariogram with the log transmissivity semivariogram.

where J is the magnitude of the mean regional head gradient J , ψ is the direction of r relative to the orientation of J , Ei is the exponential integral function, and E is Euler's constant (equal to 0.5772).

Figure 1 shows plots of $\gamma_Y(r)$ (normalized by σ_Y^2) and $\gamma_h(r)$ (normalized by $J^2 \lambda^2 \sigma_Y^2 / 2$) versus the normalized separation distance r' . The anisotropic $\gamma_h(r)$ is plotted for two different directions, one being parallel with J , that is, $\psi = 0$, and the other perpendicular to J , that is, $\psi = \pi/2$. As shown in Figure 1, $\gamma_Y(r)$ asymptotically approaches a sill as the separation distance increases, whereas $\gamma_h(r)$ grows logarithmically with the increasing separation distance. Here $\gamma_Y(r)$ also has a finite integral scale λ , while the hydraulic head field is correlated over a much longer distance, and no finite integral scale can be defined. Hence these two variables have fundamentally different structure in their semivariograms.

This difference in structure has dramatic impact on the potential to estimate these semivariograms using data collected from a single realization. In order to simplify a comparison of the estimation of the semivariogram for the Y and h fields, the semivariograms are evaluated only for the cases in which the orientation of the lag r is parallel to the direction of the mean gradient J . (It is straightforward to extend the analysis and its results to other directions if necessary.) Under this condition and for fixed r the expressions for the theoretical semivariograms ((17) and (18), respectively) can be substituted into (14) and (10) to obtain coeff $[S(x_i, r), S(x_j, r)]$ as a function of R_{ij} . Figures 2a and 2b illustrate the correlation functions for $S_Y(x, r)$ and $S_h(x, r)$. As R_{ij} is a two-dimensional vector, these are three-dimensional plots with the horizontal axes defining the directional components of R_{ij} and the vertical axis providing the magnitude of the correlation coefficient. In Figures 2a and 2b, r has a magnitude of 10 units and is oriented parallel to J . Both R_{ij} and r are normalized by λ . Figures 2a and 2b show that the correlation structures for both $S_Y(x, r)$ and $S_h(x, r)$ are anisotropic. Further, a local maximum in correlation exists at $R_{ij} = r$. Finally, Figures 2a and 2b show that $S_h(x, r)$ is correlated over longer distances than is $S_Y(x, r)$.

Figure 2. Distribution of the correlation coefficient as a function of the separation vector $\mathbf{R}_{ij}$, with $r = 10$ along the direction of $\mathbf{J}$ (a) for the transmissivity and (b) for the head residual.

The contribution of the correlation between sample pairs can be illustrated only for a given sampling pattern. Hence a uniform square grid sampling network is utilized here to illustrate the cumulative contribution of coeff $[S(x_i, r), S(x_j, r)]$ to the magnitude of $\rho_{\gamma(r)}$. This sampling network is set to be aligned with the direction of $\mathbf{J}$, contains n sample points in a square array (where n is the number of points at which T and h are measured), and has minimum spacing d between sample points (see Figure 3 for an example in which $n = 36$ and $d = 1.0$). For the discussion below the minimum separation distance between the $S(x_i, r)$, m , was set to be equal to d . Thus specific to this sampling scheme, it is straightforward to show that a general relationship between the number of $S(x_i, r)$ (i.e., the number of measurement pairs for a particular r) and the number of sample points n exists as

$$N(r) = n - \frac{r\sqrt{n}}{d} \quad (19)$$

Using the sampling scheme as defined above, it is possible to calculate $\rho_{\gamma(r)}$ according to (13) for a specific r . By varying n , it is possible to adjust $N(r)$ and evaluate $\rho_{\gamma(r)}$ as a function of $N(r)$. Figure 4 shows $\rho_{\gamma(r)}$ and $\rho_{\gamma_h(r)}$ versus $N(r)$ when $r = 10$ along the direction of $\mathbf{J}$. This plot also includes the curve for $\rho_{\gamma(r)} = \sqrt{2/(N(r))}$, the result obtained by assuming zero correlation among the $S(x_i, r)$ (see (15)). From this plot it is apparent that the correlation among $S(x_i, r)$ strongly influences the rate of reduction of $\rho_{\gamma(r)}$ with increasing $N(r)$. In order to achieve, for example, a value for $\rho_{\gamma(r)}$ of 0.8, $N(r)$ need only be around 5 for both uncorrelated data and the transmissivity field. For the head residual this number increases to approximately 100. Hence nearly 20 times the data pairs are required to achieve the same coefficient of variation for semivariogram of the head residuals as would be required for an uncorrelated variable or the random fields with short correlation range.

One interesting dependence which was further investigated was the relationship between the coefficients of variation, $\rho_{\hat{\gamma}_Y(r)}$ and $\rho_{\hat{\gamma}_h(r)}$, and r . On the basis of the same sampling scheme with $N(r) = 64$, $\rho_{\hat{\gamma}_Y(r)}$ and $\rho_{\hat{\gamma}_h(r)}$ vary with r as shown in Figure 5. Once again, $\rho_{\hat{\gamma}_h(r)}$ is consistently larger than $\rho_{\hat{\gamma}_Y(r)}$ for all r , a result of the head pairs being correlated over longer distances than transmissivity pairs. Further, increasing r appears to have a greater adverse impact on the head residuals than on the transmissivity (i.e., $\rho_{\hat{\gamma}_h(r)}$ appears to grow with r , whereas $\rho_{\hat{\gamma}_Y(r)}$ appears to be relatively insensitive to r). These results imply that not only will the separation distance among sample points need to be modified for estimating the semivariogram of Y versus the semivariogram of h , the basic design of the measurement locations must be modified as well (with approximately uniform distribution of data pairs among lag classes for Y and increasing number of data pairs with increasing lag distance required for h).

4. Discussion and Conclusions

The theoretical analysis of the sample semivariogram shows that the semivariogram estimator as given in (2) is unbiased but that the coefficient of variation of the estimator $\rho_{\hat{\gamma}(r)}$ depends not only on the number of the data pairs $N(r)$ but also on the correlation among the data pairs. This correlation is, in turn, related to the form of the underlying semivariogram $\gamma(r)$, the relative locations of the data pairs, and the lag distance r , at which the semivariogram is to be estimated. When the increment, $S(x, r)$, is Gaussian, knowledge of $\gamma(r)$ is sufficient to define the correlation structure among the squared increments according to (10).

Equation (10) leads to at least three significant observations. First, the reliability of a semivariogram estimate derived from measured data is dependent not only on the number of data points collected but also on the parameter being measured (through the semivariogram of that parameter). Second, random variables exhibiting correlation over large distances are very likely to have squared increments which are highly correlated. Thus the sample semivariogram estimate for a random

Figure 3. An example of a uniform grid sampling network with an example in which $n = 36$ and $d = m = 1.0$.

Figure 4. Plots showing $\rho_{\hat{\gamma}_Y(r)}$ and $\rho_{\hat{\gamma}_h(r)}$ versus $N(r)$ with $r = 10$ along the direction of J . Figure 4 also shows $\rho_{\hat{\gamma}(r)}$ versus $N(r)$ for uncorrelated samples.

variable which is correlated at large distances will tend to be unreliable, and caution should be used in interpreting sample semivariograms exhibiting a long-range correlation structure (e.g., a power law semivariogram). Should a field data set imply such a long-range correlation structure, (10), or a modification thereof for non-Gaussian random variables, should be used to determine an estimate of the coefficient of variation of the sample semivariogram at each lag distance thus providing a measure of confidence on the structure observed in the sample data. Third, the optimal distribution of data pairs over the different lag distances at which the sample semivariogram is to be estimated is also a function of the underlying semivariogram. As was shown, a reliable estimate of the semivariogram at various lags for the transmissivity (subject to the constraints outlined above) could be accomplished with relatively uniform numbers of data pairs in each lag class. In contrast, estimating the semivariogram of the head residual, with equal coefficient of variation in each lag class, would require increasing numbers of data pairs as the magnitude of the lag distance is increased.

Figure 5. The $\rho_{\hat{\gamma}(r)}$ versus r for $\hat{\gamma}_Y(r)$ and $\hat{\gamma}_h(r)$.

Appendix: Detailed Derivation for Equation (10)

Given the definition of $S(\mathbf{x}_i, \mathbf{r})$ as in (7) and its ensemble mean (8), an expression for $\text{cov}[S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})]$ can be written as

$$\text{cov}[S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})] = \frac{1}{4} \langle [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2 \cdot [Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]^2 \rangle - \gamma(\mathbf{r})^2 \quad (20)$$

Using the joint moment generating function, Papoulis [example 7-6, page 158, 1984] shows that if two random variables, X_1 and X_2 , are jointly normal with zero mean, the following relationship holds:

$$\langle X_1^2 X_2^2 \rangle = \langle X_1^2 \rangle \langle X_2^2 \rangle + 2 \langle X_1 X_2 \rangle^2 \quad (21)$$

Assuming that the increment $Z(\mathbf{x}) - Z(\mathbf{x} + \mathbf{r})$ is jointly normally distributed (which holds, at least, for the case in which $Z(\mathbf{x})$ is jointly normal), the application of (21) to (20) leads to

$$\begin{aligned} \text{Cov}[S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})] &= \frac{1}{4} \langle [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2 [Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]^2 \rangle - \gamma(\mathbf{r})^2 \\ &= \frac{1}{4} \langle [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2 \rangle \langle [Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]^2 \rangle \\ &\quad + \frac{1}{2} \langle [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})][Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]^2 \rangle - \gamma(\mathbf{r})^2 \\ &= \frac{1}{2} \langle [Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})][Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]^2 \rangle \quad (22) \end{aligned}$$

Further expanding $[Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})][Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]$ into four terms results in

$$\begin{aligned} \text{cov}[S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})] &= \frac{1}{2} \left\langle \frac{[Z(\mathbf{x}_i) - Z(\mathbf{x}_i + \mathbf{r})]^2}{2} + \frac{[Z(\mathbf{x}_j) - Z(\mathbf{x}_j + \mathbf{r})]^2}{2} \right. \\ &\quad \left. - \frac{[Z(\mathbf{x}_i) - Z(\mathbf{x}_j)]^2}{2} - \frac{[Z(\mathbf{x}_i + \mathbf{r}) - Z(\mathbf{x}_j + \mathbf{r})]^2}{2} \right\rangle^2 \\ &= \frac{1}{2} [\gamma(\mathbf{r} + \mathbf{x}_j - \mathbf{x}_i) + \gamma(\mathbf{r} - \mathbf{x}_j + \mathbf{x}_i) - 2\gamma(\mathbf{x}_j - \mathbf{x}_i)]^2 \\ &= \frac{1}{2} [\gamma(\mathbf{r} + \mathbf{R}_{ij}) + \gamma(\mathbf{r} - \mathbf{R}_{ij}) + \gamma(\mathbf{r} - \mathbf{R}_{ij}) - 2\gamma(\mathbf{R}_{ij})]^2, \quad (23) \end{aligned}$$

where $\mathbf{R}_{ij} = \mathbf{x}_j - \mathbf{x}_i$ is the separation distance between $S(\mathbf{x}_i, \mathbf{r})$ and $S(\mathbf{x}_j, \mathbf{r})$.

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Anomalous transport in laboratory-scale, heterogeneous porous media

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Abstract. We analyze measured breakthrough curves in a laboratory model which consists of a uniformly heterogeneous porous medium; these curves were previously shown to be indicative of scale-dependent transport and therefore inconsistent with the (macroscopic) advection-dispersion equation [Silliman and Simpson, 1987]. Our analysis is based on an analytical expression for the first-passage time distribution (FPTD) of migrating contaminants in random media, developed with the use of a continuous time random walk (CTRW) formalism. The general CTRW has been shown to be effective in quantifying anomalous transport patterns frequently observed in fractured and strongly heterogeneous porous media [Berkowitz and Scher, 1997, 1998]. We calculate a family of FPTD curves, usually referred to as “breakthrough curves,” which are a function of an exponent β ; this exponent is related to the low-velocity tail of the velocity distribution. The FPTD curves fit well the measured data, with a single value of the β exponent over the spatial/temporal scale of the experiment. This is in contrast to previous analyses using solutions of the Gaussian-based advection-dispersion equation with time-independent parameters in a uniform flow field. We conclude that the CTRW may allow analysis of transport in porous media subject to complex heterogeneities at large scale, which may not be amenable to analysis using classical advection-dispersion theory. Hence the CTRW represents a potentially valuable tool in the assessment of dispersive processes in heterogeneous porous media.

1. Introduction

The issue of how to quantify the migration of contaminants in groundwater systems has received overwhelming attention during the last 3 decades, and a vast literature dealing with the subject has developed (see, e.g., the recent reviews in the book by Dagan and Neuman [1997]). The basic conceptualization is that of contaminant “particles” moving continuously through space with variable velocities in a heterogeneous porous medium. One can view this phenomenon in a variety of ways. The initial, widely applied model emphasizing the fluid-like continuous spread of a conservative, neutrally buoyant chemical plume was the advection-dispersion equation (ADE). Laboratory-scale and field-scale application of this equation is based on the assumption that dispersion behaves macroscopically as a Fickian diffusive process, with the dispersivity being assumed constant in space and time. Following the development of the classical ADE, it was observed through field, laboratory, and Monte Carlo analyses that the dispersivity is not constant but is, rather, dependent on the time and/or length scale of measurement, leading to what has been referred to as the “scale effect”; we describe this as “anomalous transport.” The spatial dependence of transport coefficients measured in the field is usually attributed to the existence of hydraulic conductivity fields with coherence lengths varying over many scales. We

observe that chemical transport at the field scale is now often recognized to be non-Fickian, so that one can argue that the “anomalous” (non-Fickian) transport discussed here is actually the norm rather than the exception.

This dependence on space (or, equivalently in some cases, time) has been the principal focus of many stochastic theories, and the ADE approach has been generalized by substituting a more detailed stochastic process for the Fickian assumption. Stochastic theories have examined, for example, the temporal variation of the first and second moments of the spatial distribution of a chemical plume, with specific interest in defining conditions under which macrodispersivities are obtained. Such analyses have provided substantial insight into the dispersion process and have been shown, through application to well-documented field experiments, to provide accurate prediction of the temporal variation in the first- and second-order moments of tracer plumes for a number of geological formations characterized by relatively small degrees of heterogeneity (e.g., the Cape Cod site [Garabedian *et al.*, 1991]). These theories have been much less successful in providing reliable prediction of the temporal variation in the first two concentration moments for field experiments carried out in highly heterogeneous aquifers, such as the detailed tracer tests at the Columbus Air Force Base [Adams and Gelhar, 1992]. Further, Fitts [1996] has recently argued that successful reproduction of the first two moments is not equivalent to successfully reproducing the details of the distribution of the concentration in space. Fitts [1996] demonstrated substantial deviation between predicted and observed concentrations in the Borden and Cape

Cod field sites despite close agreement between the predicted and observed spatial moments.

Of particular interest for the present discussion are recent stochastic theories based on Lagrangian formulations, such as those developed by Dagan [1989] and others (see also the review by Rubin [1997]). One of the applications of the Lagrangian approach is the estimation of effective dispersion coefficients at all scales of transport. The general formulation, based on analysis of particle trajectories and transition probabilities in random velocity fields, is developed in terms of a general probability density function of the velocity field without limitations on the types of velocity fields which can be considered. In practice, however, the majority of the theoretical results which have been obtained are based on the assumption that the velocity distributions can be completely characterized in terms of the first two moments. Many of these results are also limited to porous media in which the variance of the log hydraulic conductivity is small (less than unity). The reader is referred to Dagan and Fiori [1997] for a brief discussion of the justification given for the use of these assumptions.

If the variation in the velocity field is sufficiently large, a different picture of the particle motion can be developed, a highly discrete one. The formalism described below is particularly appropriate for this picture. The particle motion is approximated as a series of discrete steps, each having a different velocity. The particle's position is determined at the points of transition between these steps. A continuous time random walk (CTRW) can then be introduced [Berkowitz and Scher, 1995, 1997, 1998] to account naturally for the cumulative effects of a sequence of transitions. The transitions are characterized by $\psi(s, t)$, the probability rate for a displacement s with a difference of arrival times of t . In the CTRW approach one can determine the evolution of the particle distribution (plume), $P(s, t)$, for a general $\psi(s, t)$, so there is no a priori need to consider the moments of $P(s, t)$. The challenge is to map the important aspects of the physical picture of the particle motion in the medium onto a $\psi(s, t)$. A mapping which results in a $\psi(s, t)$ with an algebraic tail for large time (such as the functional behavior discussed in detail in section 2 in equation (7)) leads to the description of anomalous transport (e.g., non-Gaussian plumes).

An initial hydrological application of the CTRW was to a series of Monte Carlo transport simulations based on discrete fracture networks [Berkowitz and Scher, 1997]. In this case we used key characterizations of the particle motion, i.e., the measured velocity and flow distributions and the fracture fragment length distribution, to determine $\psi(s, t)$. The plausibility of this approach to heterogeneous porous media was then demonstrated by the interpretation of field data from the Columbus field site [Berkowitz and Scher, 1998]. The theory was able to reproduce, for the fracture simulations, the development of highly skewed particle plumes. In particular, the CTRW was able to capture the variation of both the movement of the center of mass of the particles and the standard deviation of particle location around this center of mass. For these simulations both the mean and the standard deviation of the particle position scaled as t^β , an observation which cannot be reproduced with the advection-dispersion equation. The value of β was determined from the algebraic behavior of the low-velocity tail of the distribution of velocities in the fracture segments. Hence, through the parameter β , the range of random low velocities controls the character of the transport. For the Columbus site, a highly heterogeneous aquifer, the CTRW

was capable of reproducing the observed behavior of the highly skewed plume, as well as the moments in the direction of flow, determined from field data [Adams and Gelhar, 1992]. Once again, the interpretation of the moments suggested that the mean and standard deviation of the plume position scale as t^β , with the same β as that governing the evolution of the plume.

The CTRW has been applied by other authors in the analysis of transport behaviors in porous media. Hatano and Hatano [1998], for example, examined concentration profiles measured from column studies. These authors found that the migration of sorptive ions in heterogeneous porous media could be accurately described by an anomalous transport model.

Common to any transport analysis is the question of the degree and form of heterogeneity that gives rise to non-Gaussian transport over long transport distances; this issue remains to be fully addressed. One approach to investigate this question is to compare predictions of models such as the CTRW to tracer or contaminant transport experiments conducted under controlled laboratory conditions. The present paper represents one such effort.

In section 2 we review the formalism of the CTRW and evaluate the first-passage time distribution (FPTD), commonly referred to as a "breakthrough curve." A full critical discussion of the application of the FPTD to experimental data measured in a random porous medium (described in section 3) follows in sections 4 and 5.

We show a range of FPTD curves (in section 2), each with a different value of β and degree of tailing in the early and late time concentration breakthrough. The application of the FPTD to experimental results published previously [Silliman and Simpson, 1987] involving a uniformly heterogeneous porous medium (section 3). Silliman and Simpson [1987] argued that the curves presented in that paper are indicative of the so-called "scale effect" and are not consistent with the classical ADE. We show that the shape of a FPTD curve fits well the range of measured data, with a single value of the β exponent over the spatial/temporal scale of the experiment (section 4). In sections 5 and 6 we discuss how the CTRW may allow analysis of transport in porous media subject to complex heterogeneities at large scale, which may not be amenable to analysis using classical advection-dispersion theory. We conclude that the CTRW represents a potentially valuable tool in the assessment of dispersive processes in heterogeneous porous media.

2. Continuous Time Random Walk (CTRW) Theory

2.1. CTRW Framework

We review here the basic transport equations of the CTRW formalism [Scher and Lax, 1973]. A standard random walk describes the accumulation of many random transitions between points in space. If $P_n(\ell)$ is the probability that the walker is at point ℓ after n transitions, then

$$P_{n+1}(\ell) = \sum_{\ell'} p(\ell, \ell') P_n(\ell'), \quad (1)$$

where $p(\ell, \ell')$ is the transition probability for a jump from ℓ' to ℓ , with

$$\sum_{\ell'} p(\ell, \ell') = 1. \quad (2)$$

The CTRW is a generalization of this random walk, wherein discrete time n is replaced by continuous time t :

$$R(s, t) = \sum_{s'} \int_0^t \psi(s - s', t - t') R(s', t') dt' \quad (3)$$

where $R(s, t)$ is the probability per time for a walker to just arrive at site s at time t , and $\psi(s, t)$ is the probability per time for a transition between sites separated by s with a difference of arrival times of t . The discrete transition sites s lie on a lattice, and we assume periodic boundary conditions such that $s = \sum_i s_i a_i$, with $|a_i| = a$ the lattice constant, $\{s_i\}$ integers and $s_i + j_i N \rightarrow s_i$ for arbitrary integer j , and where Na is the length of the lattice and $s_i = -(N-1)/2, \dots, (N-1)/2$ for odd N .

Analogous to $P_n(\ell)$ in (1), the probability distribution function $P(s, t)$ describing the spatial and temporal location of particles throughout the domain may be defined as

$$P(s, t) = \int_0^t \Psi(t - t') R(s, t') dt', \quad (4)$$

where

$$\Psi(t) = 1 - \int_0^t \psi(t') dt' \quad (5)$$

is the probability for a walker to remain on a site and

$$\psi(t') = \sum_s \psi(s, t'). \quad (6)$$

The probability distribution $P(s, t)$ is the normalized concentration distribution of particles as they migrate through a domain.

We stress that the random walk formalism considered here is distinct from the "random walk" models frequently invoked in the hydrology literature, wherein particles follow streamlines and are then perturbed by a random component to account for diffusion [e.g., Prickett et al., 1981; Kinzelbach, 1988; Uffink, 1995; LaBolle et al., 1998]. In particular, the advective, dispersive, and diffusive transport mechanisms are inextricably combined in (3), rather than being treated separately. Hence the transport equation (3) represents a different point of departure from the usual formulations. This CTRW formalism has been used in a wide variety of applications (compare reviews by Scher et al. [1991] and Klafter et al. [1996]).

The heart of the CTRW formulation of any transport behavior lies in the identification of $\psi(s, t)$ with the physical process of the particle motion (see, e.g., Berkowitz and Scher [1998] for determination of $\psi(s, t)$ in fracture networks). Anomalous transport arises in cases where the large time behavior of $\psi(s, t)$ is algebraic [Scher and Montroll, 1975]

$$\psi(s, t) \rightarrow t^{-1-\beta} \quad t \rightarrow \infty, \quad 0 < \beta < 1 \quad (7)$$

Generally, the form of $\psi(s, t)$ at large time determines the time dependence of the mean position $\bar{\ell}(t)$ and standard deviation $\bar{\sigma}(t)$ of $P(s, t)$. In the presence of a "bias" (such as a pressure gradient) and for the particular behavior denoted in (7), it can be shown [Shlesinger, 1974; Scher and Montroll, 1975] that

$$\bar{\ell}(t) \sim t^\beta \quad (8)$$

Figure 1. Schematic illustration of the contrast between anomalous and Gaussian transport (adapted from Scher et al. [1991]).

$$\bar{\sigma}(t) \sim t^\beta. \quad (9)$$

The unusual time dependence of $\bar{\ell}(t)$ and $\bar{\sigma}(t)$, which results from the infinite mean (over time) of $\psi(s, t)$, is the hallmark of the non-Gaussian propagation of $P(s, t)$. Note that a $\psi(s, t)$ following (7) does not fulfill the conditions of the central limit theorem. A highly pedagogical account of the content of the basic equations (7)–(9) is given by Scher et al. [1991, pp. 28–30].

This behavior is in sharp contrast to Gaussian models where $\bar{\ell}(t) \sim t$, $\bar{\sigma}(t) \sim t^{1/2}$ (as an outcome of the central limit theorem), and the position of the peak of the distribution coincides with $\bar{\ell}(t)$. Note that in Gaussian transport, $\bar{\ell}(t)/\bar{\sigma}(t) \sim t^{1/2}$; an important distinguishing feature of anomalous transport is that $\bar{\ell}(t)/\bar{\sigma}(t) \sim \text{constant}$. The contrasting behavior between Gaussian and anomalous transport is shown in Figure 1. Note that anomalous transport is characterized by a concentration peak which moves much more slowly than the Gaussian, with a longer forward advance of particles. As discussed in sections 2.2 and 5, the relative shapes of the anomalous transport curves and the rate of advance of the peak vary strongly as a function of β .

Equation (3) is a convolution in space and time which is solved by use of discrete Fourier transforms and Laplace transforms. Then from (4), using inverse transforms, the solution for $P(s, t)$ which describes the evolution of the contaminant plume can be derived. In principle, this solution can be developed analytically for arbitrary $\psi(s, t)$; but in practice, because of the difficulty of dealing with the inverse Laplace transforms, analytical solutions for $P(s, t)$ are not readily derived. Moreover, purely numerical solutions can be problematic, because of the extreme sensitivity of numerical routines that calculate inverse Laplace transforms. We have recently made use of a combined analytical-numerical solution [Berkowitz and Scher, 1997].

2.2. First-Passage Time Distributions

The probability per time to reach site s at time t for the first time is denoted by $F(s, t)$ and referred to as a first-passage time distribution (FPTD). It can be determined from the implicit relation

$$R(s, t) = \delta_{s,0} \delta(t - 0^+) + \int_0^t F(s, t') R(0, t - t') dt', \quad (10)$$

where $\delta_{i,j}$ is the Kronecker delta function and $\delta(t - t_0)$ is the Dirac delta function, indicating in (10) that the contaminant is at the origin at $t = 0$. Equation (10) is a convolution relation between $R(s, t)$ and $F(s, t)$. The meaning of (10) is that a particle might have visited s at an earlier time t' for the first time and returned an arbitrary number of times, in the remaining $t - t'$, described by $R(0, t - t')$. The breakthrough curves are based on the first-passage time distribution $F(s, t)$. Solving (10) using Laplace transforms, one obtains

$$F(s, t) = \mathcal{L}^{-1}[R^*(s, u)/R^*(0, u)] \quad s \neq 0, \quad (11)$$

where $\mathcal{L}^{-1}$ is the inverse Laplace transform and $R^*(s, u)$ is the Laplace transform ($\mathcal{L}$) of $R(s, t)$ and u is the Laplace variable.

In the CTRW one can generally evaluate $F(s, \tau)$ in an asymptotic regime, which in this case is $\tau \gg 1$, where now τ is a dimensionless time and will be defined in the discussion below. For a random fracture network [Berkowitz and Scher, 1997], $\tau = 2v_0 t/s_0$, where v_0 is a characteristic flow velocity, s_0 is a mean fracture-fragment length, and $\tau \gg 1$ simply corresponds to many (say, ≥ 10) transitions through fragments. We stress that this definition of "asymptotic" does not correspond to the limit of complete self-averaging in contrast to the usual definition of the term used in stochastic hydrology (e.g., in order to define macrodispersion coefficients). In the large τ regime the transport is dominated by the long time tail of $\psi(s, t)$. In Laplace space this is the limit $u \rightarrow 0$ [Scher et al., 1991].

The CTRW problem of the propagation of particles in a domain with a pressure gradient (i.e., a biased field), in the presence of an absorbing boundary (plane), has been considered by Montroll and Scher [1973]. This problem is very similar to the one that we want to solve. One first averages over the coordinates parallel to the plane and defines $f(L, \tau)$, the average spatial distribution of tracer per unit time, as

$$f(L, \tau) = \langle F(s, \tau) \rangle, \quad (12)$$

where L is the distance between the inlet (origin) and the outlet (x coordinate of s) planes and the angled brackets denote average. For $\psi(s, t) \rightarrow t^{-1-\beta}$ for $t \rightarrow \infty$, as discussed by Scher et al. [1991], the pertinent Laplace transform is determined by the dominant behavior as $u \rightarrow 0$ (e.g., $\psi^*(u) \rightarrow 1 - cu^\beta$, with c a constant), and in this limit, and for $0 < \beta < 1$, the evaluation of (12) reduces to

$$f(L, \tau) = \mathcal{L}^{-1}[\exp(-bu^\beta)], \quad (13)$$

where $b \equiv L/\langle \ell \rangle$ is a nondimensional measure of L and $\langle \ell \rangle$ is the mean displacement for a single transition [Scher and Montroll, 1975]. The parameter b is a large number if there is a separation between source and collection (measurement) plane greater than an intrinsic length scale of the heterogeneous medium (which pertains to all of the cases we shall consider).

The $\mathcal{L}^{-1}$ expression in (13) can be evaluated exactly in terms of Fox's H function [Metzler et al., 1994, 1998], but for simplicity we use an infinite series for the case $bu^\beta < 1$, which corresponds to late times (i.e., when the peak of the FPTD has already reached the measurement plane). The function

$\exp(-bu^\beta)$ is expanded in a power series and integrated term by term to derive

$$f(L, \tau) = \tau^{-1} \sum_{j=0}^{\infty} \frac{(-b/\tau^\beta)^j}{\Gamma(j+1)\Gamma(-j\beta)}, \quad (14)$$

where $\Gamma(x)$ is the gamma function and $1/\Gamma(-j\beta) = 0$ for integer values of $j\beta$. The series in (14) represents an entire function (similar to the exponential function) and can be shown to be expressible in terms of known analytic functions for rational values of β . For early times ($bu^\beta > 1$) one can evaluate the integral as an asymptotic series in

$$\omega \equiv (\tau^\beta/\beta b)^{1/\beta}, \quad (15)$$

with the saddle-point method [Copson, 1965]; the lowest-order term is ($\omega \ll 1$)

$$f(L, \tau) \equiv \exp[-(1-\beta)/(\beta\omega)]/[2\pi(1-\beta)\omega]^{1/2}\tau. \quad (16)$$

For $\beta = 1/2$ the asymptotic representation in (16) is the exact function (which can be obtained by summing the series in (14))

$$f(L, \tau) = \frac{b}{2\sqrt{\pi}\tau^{3/2}} \exp\left(-\frac{b^2}{4\tau}\right), \quad (17)$$

while for $\beta = 1/3$, one can obtain from (14)

$$f(L, \tau) = \tau^{-1} x \text{Ai}(x) \quad (18)$$

where $x \equiv b/(3\tau)^{1/3}$ and $\text{Ai}(x)$ is the Airy function [Abramowitz and Stegun, 1965, section 10.4]. The asymptotic limit of $\text{Ai}(x)$ for large x , inserted into (18), exactly yields (16) for $\beta = 1/3$.

The representation for early times (16), before the effective transit time (i.e., $b/\tau^\beta \approx 1.0$), is needed when it is numerically untenable to sum the series in (14). There is a range of b/τ^β where both representations of $f(L, \tau)$, (14) and (16), are in excellent agreement. We choose a value of b/τ^β in this range as a crossover point for the use of (14) or (16) to compute $f(L, \tau)$.

In Figure 2 we show semilog plots of both $b^{1/\beta}f(L, \tau)$ and

$$M(L, \tau) \equiv \int_0^\tau f(L, \tau') d\tau' \quad (19)$$

versus $\tau/b^{1/\beta}$ for a range of β . The function $M(L, \tau)$ is the accumulated mass reaching L because of a pulse of solute at the origin at $\tau = 0$ (normalized to a total mass of unity). In the case of a uniform concentration C_0 injection starting at $\tau = 0$, the concentration C reaching L is

$$C(L, \tau) \equiv C_0 \int_0^\tau f(L, \tau - \tau') d\tau'. \quad (20)$$

A change of the integration variable, $\tau - \tau' \rightarrow \tau'$, reproduces (19), that is, $C(L, \tau)/C_0 = M(L, \tau)$. Hence the curves $M(L, \tau)$ in Figure 2 are those of the typically measured normalized concentration. We note that in all curves $C/C_0 = 1/2$ corresponds to $\tau/b^{1/\beta} \approx 1.0$.

The basic shape of the breakthrough curves and their relation to $b^{1/\beta}f(L, \tau)$ (the spatial distribution of tracer due to pulse injection) changes dramatically from $\beta = 0.33$ to $\beta = 0.9$. For $\beta = 0.33$ the dispersive front occurs over about six log

Figure 2. Semilog plots showing a range of first-passage time distribution (FPTD) curves (probability density function $b^{1/\beta}f(L, \tau)$ and cumulative $M(L, \tau)$) versus $\tau/b^{1/\beta}$ for a range of β . (a) Plots of $b^{1/\beta}f(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.33, 0.50$, and 0.65 . (b) Plots of $b^{1/\beta}f(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.75, 0.80$, and 0.90 . (c) Plots of $M(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.33, 0.50$, and 0.65 . (d) Plots of $M(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.75, 0.80$, and 0.90 .

cycles in time, and the peak of $b^{1/\beta}f(L, \tau)$ occurs at $\tau/b^{1/\beta} \approx 0.02$, while for $\beta = 0.9$ the values of $\tau/b^{1/\beta}$ for the peak maximum and $C/C_0 = 1/2$ are nearly coincident. The temporal extent of the dispersion for $\beta = 0.9$ is reduced to approximately one and one half log cycles. The breakthrough curves in Figure 2 have highly variable basic features as a function of β with the common attribute of a large asymmetry between the dispersion before and after the $C/C_0 = 1/2$ "transit time" (i.e., long trailing tails). It is to be emphasized that the time dependence of $C(L, \tau)/C_0$ is determined by the limiting behavior of $\psi(s, t)$ at large t which, in turn, is controlled by the range of low velocities in the flow field. This result was shown explicitly by Berkowitz and Scher [1997, 1998]. The spatial dependence appears in the parameter $b^{1/\beta}$; a change in L rigidly shifts $C(L, \tau)$ along the x axis in the semilog plots in Figure 2.

As β increases ($0 < \beta < 1$), there is a "stiffening" of the concentration front. The plume approaches a box-like shape with a front steeper than a Gaussian yet with a trailing tail longer than a Gaussian. For $\beta > 1/2$ the peak of $P(L, t)$ is displaced from the origin [McLean and Ausman, 1977]. These shape changes with β cannot be associated simply with a change in an effective (longitudinal) dispersion, either constant or time-dependent. It is interesting to consider the exponent β as the parameter controlling the effective dispersion. We recall that β is determined by the range of random velocities. The other significant parameters are those that scale distance and time; they correspond to a characteristic velocity and intrinsic length scales of the medium. Values for these parameters are determined by the experimental conditions, for example, the characteristic fluid velocity and the size of the heterogeneities, as discussed in section 4.

Figure 3. Experimental setup showing the flow cell and the uniformly heterogeneous packing (adapted from *Silliman and Simpson* [1987]).

3. Experimental Setup and Measurements

The experiments discussed here were first presented and discussed by *Silliman* [1981] and *Silliman and Simpson* [1987]. The original intent of the experiments was to demonstrate, experimentally, the scale effect in dispersion within a variety of heterogeneous media. As such, the analysis of the curves focused on the spread of the center portion of the breakthrough curves; details of the shape of the breakthrough curves, particularly in the tails for the more heterogeneous packings, were not fully analyzed in the original effort. It is these details, along with the fact that the (macroscopic or "field scale") ADE does not properly describe the full evolution of the migrating tracer, which led to the reanalysis of these experiments which is presented here.

The experiments were conducted in a laboratory flow cell with internal dimensions of $2.44 \times 1.07 \times 0.10$ m. Flow was maintained across the medium through use of constant head reservoirs (Figure 3). The upper surface of the medium was unconfined and was penetrated by a series of dividers extending approximately 10 cm into the experimental medium. These dividers were intended to reduce the potential for surface water flow along the upper surface of the medium should the head in the inflow reservoir accidentally exceed the height of the upper surface of the medium. As the medium was unconfined, the difference in heads in the two reservoirs was kept to a minimum to reduce the variation in the velocity field (related to curvature of the water table). The maximum difference in head across the flow cell over all experiments was approximately 0.03 m (i.e., there was approximately a 3 cm change in saturated thickness).

Silliman and Simpson [1987] considered four different permeability distributions; of interest for the present discussion is the fourth packing (shown in Figure 3) which represents a "homogeneously heterogeneous" medium. Within this particular medium, blocks of fine sand (with dimensions of approximately 7.5 cm by 2.5 cm and uniform over the 0.1 m width of the flow cell) were placed within a matrix of coarser sand. The blocks occupied approximately 16% of the total volume, and the contrast in permeabilities between the coarse (high permeability) and fine (low permeability) sand was approximately a factor of 20. Owing to difficulties in packing the mean permeability in the lower portion of the flow cell appeared to be somewhat lower than the mean permeability near the upper surface of the flow cell. Hence, in addition to the heterogeneity

introduced by the presence of the low-permeability cubes (with the resulting drop in velocity within each cube), there is a lower mean permeability in the lower portion of the medium. This latter heterogeneity leads to an overall trend (although with a high degree of variability along the flow cell) in mean velocity with depth [e.g., *Silliman and Simpson*, 1987, Figure 8a].

Tracer was added to the experimental medium as a step change in concentration in the upgradient reservoir [*Silliman*, 1981]. Once introduced, the movement of the tracer was monitored at five different distances from the inflow reservoir through use of five columns of electrodes. Each column consisted of 72 metal wires at spacings slightly greater than 1 cm. Electrical resistance was measured between each pair of wires (providing 71 readings per electrode column), and the electrical resistance was calibrated to concentration in situ. For a particular measurement time, 71 readings were recorded at a single electrode column, transformed to concentrations utilizing the calibration curves, and then vertically averaged (a flow-weighted average was used as discussed by both *Silliman and Simpson* [1987] and *Silliman* [1981] to yield approximate flux-averaged concentrations). Breakthrough curves could then be plotted for each electrode column [e.g., *Silliman and Simpson*, 1987, Figure 4].

The instrumentation used to measure electrical resistance (see details given by *Silliman* [1981]) was relatively crude compared to what we currently use. As a result, it is anticipated that errors in individual measurements may have been as high as 2% of the input concentration in the tails of the curve and as high as 5–10% of the input concentration in the center of the breakthrough curve (the lower error in the tails is related to the fact that the initial and final concentrations in each experiment were used as calibration points). However, because the measurement errors are considered to be uncorrelated among the 71 readings within an electrode column, the standard deviation of error of the vertically averaged concentrations will be lower than the error in individual readings by a factor of approximately $\sqrt{71} \approx 8.4$. It is therefore concluded that the standard deviation of error in the vertically averaged, normalized concentrations is, at maximum, of the order of 0.25% in the tails of the curves and is slightly greater than 1% in the center of the breakthrough curves. It should be noted that the experiments discussed herein (the fourth packing) were the last experiments run by *Silliman* [1981] so that many of the sources of error in the electronics from the earlier experiments had been identified and corrected. This improved performance can be seen, for example, in the reduced noise in the data for the first electrode column in the homogeneously heterogeneous medium compared to the data for this electrode column in the original homogeneous medium [cf. *Silliman and Simpson*, 1987, Figures 4 and 8b].

It is unfortunate that the breakthrough curves presented for the fourth packing [*Silliman and Simpson*, 1987, Figure 8a] do not extend to concentrations higher than approximately 70–80%. The reasons for the limited data are twofold. First, as the tracer front moved through the medium, choices had to be made as to whether early time data were to be collected at electrode columns farther down gradient or whether late time data were to be collected at electrode columns located closer to the inflow reservoir (time did not allow data collection from multiple locations). A decision was made to focus on the early time data. Second, it was observed that the rate of increase in concentration at late time was quite slow (as supported by the data fits presented in section 4). As a result of this slow in-

crease in concentration, in combination with the facts that no automation was available for these experiments (all readings were collected by hand using a handheld multimeter) and the curves for relative concentrations up to approximately 70–80% were adequate to illustrate the scale effect, a decision was made not to pursue these curves to full breakthrough. In hindsight it would have been preferable to run these curves to larger times; our later studies using a more automated sampling system have produced curves including both early and late time tails [e.g., *Silliman et al.*, 1987]. As illustrated in section 4, however, our analysis using the CTRW indicates that the available breakthrough curves contain sufficient information to differentiate between behavior reflective of the advection-dispersion theory and behavior of the CTRW theory corresponding to $L/\langle\ell\rangle \gg 1$, especially for electrode columns 4 and 5.

4. Comparison of Theory and Experiments

We focus our attention on concentration measurements taken at electrode columns 3, 4, and 5, corresponding to distances from the inlet of $L = 0.91$, 1.37, and 1.83 m, respectively. We determine the FPTD curves using (14), (16), and (19) and obtain breakthrough curves such as those illustrated in Figure 2. As shown in Figure 4, the FPTD curves fit well the complete range of measured data with a single value of the β exponent over the spatial/temporal scale of the experiment. The most complete set of measurements is given in Figure 4b; a choice of $\beta \approx 0.87 \pm 0.01$, obtained by fitting the FPTD solutions to the data, captures both the early and later time arrivals as well as the intermediate breakthrough times. The agreement of the FPTD curve with the measurements is clear and permits differentiation between the FPTD and ADE solutions (discussed below). As we note in the following, the ADE solutions shown here represent “best fit” solutions wherein the dispersivity estimates vary by a factor as large as 1.7 from one location to the next. In contrast, we use the same β as for Figure 4b to predict the FPTD curves at shorter and longer distances from the inlet (shown in Figures 4a and 4c). We emphasize, in particular, the agreement of these FPTD curves with the measurements of the short time arrivals. Thus we find that a single value of β determined from, say, the breakthrough curve at electrode 4, is sufficient to predict transport behavior measured at electrodes 3 and 5.

Now as discussed in section 2, the x axis shown in Figure 2 is in dimensionless time units of $\tau/b^{1/\beta}$, with τ as the dimensionless time and $b \equiv L/\langle\ell\rangle$ (recall that $\langle\ell\rangle$ is the mean displacement for a single transition and L is the distance from the inlet to the measurement plane). As such, for comparison to the data (Figure 4), we must scale the (temporal) x axis appropriately. Our point of comparison is the time at which $C/C_0 = 0.5$ on the breakthrough curves. We recall that τ is a dimensionless time, defined by $\tau \equiv \bar{v}t/\langle\ell\rangle$, where $\bar{v}$, a characteristic flow velocity for the particle transitions, is larger than the average fluid velocity in the domain (to account for the influence of the higher-velocity transitions [Berkowitz and Scher, 1998]) and t is the (dimensional) laboratory experiment time. The transport system is essentially characterized by two features: (1) the velocity field, which determines β , and (2) the medium heterogeneities, which determine $\langle\ell\rangle$. The length $\langle\ell\rangle$ is used in both of the nondimensional ratios that determine the time (τ) and the transport distance (L). We observe that for the range of β values used here, $\tau/b^{1/\beta} \approx 0.9$ at $C/C_0 = 0.5$

Figure 4. (a–c) Comparison of measured concentrations (circles) to the FPTD theory (solid curves), with $\beta = 0.87 \pm 0.01$. Also shown in Figures 4a–4c (dashed lines) are solutions of the advection-dispersion equation using the “best” estimated value [from *Silliman and Simpson*, 1987] of dispersivity α at each distance from the inlet (note that this value varies at each location). For electrode column 3 (Figure 4a), $L = 0.91$ m and $\alpha = 0.073$ m. For electrode column 4 (Figure 4b), $L = 1.37$ m and $\alpha = 0.075$ m. For electrode column 5 (Figure 4c), $L = 1.83$ m and $\alpha = 0.127$ m.

(see section 2.2). Thus the experiment time at which $C/C_0 = 0.5$, t_r , is given by

$$t_r = \frac{0.9\langle\ell\rangle}{\bar{v}} \left(\frac{L}{\langle\ell\rangle} \right)^{1/\beta} \quad (21)$$

As described in section 3, the mean size of the low-permeability heterogeneities is ≈ 7.5 cm; it is reasonable to assume that the mean particle transition length will be somewhat smaller than the size of these heterogeneities, so we choose $\langle\ell\rangle \approx 1$ cm. With this choice and with any of the measured t_r (found from the point $C/C_0 = 0.5$ on the breakthrough curves), it follows that $\bar{v} \approx 9$ m/d. The average fluid velocity in the flow cell was about 5.7 m/d (see section 5). In agreement with these results, Berkowitz and Scher [1998] found the characteristic flow velocity for tracer particles in a heterogeneous natural aquifer to be larger than the estimated mean fluid velocity in the aquifer. It is important to note that the single values of $\langle\ell\rangle$ (1 cm) and $\bar{v}$ (9 m/d) describe all values of t_r . There is a degree of flexibility in choosing $\langle\ell\rangle$ (i.e., similar estimates of the breakthrough behavior were obtained using, for example, $\langle\ell\rangle \approx 5$ cm), since t_r is weakly dependent on $\langle\ell\rangle$ for $\beta \approx 0.87$.

Silliman and Simpson [1987] found from analysis of the measured breakthrough curves that the data are indicative of scale-dependent transport [cf. Silliman and Simpson, 1987, Figure 8b]; that is, they are not consistent with Gaussian transport (the ADE solution). Silliman and Simpson [1987] reported dispersivities that were estimated from the sample mean and variance, using their (14) and (15), which define the fit of the solution to the data through the point $C/C_0 = 0.5$. These dispersivities, which are determined individually at each electrode column, therefore represent "best fit" estimates for the data. Since the velocity is usually specified by the experimental setup, the dispersivity coefficient is the only real degree of freedom. Referring to Figure 4, even though the data are fit in the concentration range around $C/C_0 = 0.5$, deviations clearly occur outside this range. Moreover, the integrity of the fits is poor, given that the dispersivity values vary from one location to the next; for example, the value of the dispersivity increases significantly from 0.075 m to 0.127 m between Figures 4b and 4c. Imposition of a single overall dispersivity value, which is intrinsic to the theory underlying the ADE, leads to even greater discrepancy between the ADE solution and the data.

The curves shown in Figure 4 provide a contrast between the ADE (Gaussian) solutions and the CTRW (non-Gaussian) results. In Figure 4a the difference in the two fits occurs at the early times. At the larger electrode distances in Figures 4b and 4c, there is a clearer difference between the fits for the developing plume. The largest measured concentration range is in Figure 4b, and the CTRW theory accounts for both the early and later time breakthroughs. The evolving differences between the CTRW and ADE predictions become more substantial for the fully developed plume. Notwithstanding the fact that no measurements are available to fully define the very long time tails of the breakthrough curves (as discussed in section 3), we suggest that the consistency of the CTRW with the experimental results indicates the CTRW to have significant potential as a predictive tool.

5. Discussion

5.1. Experimental Considerations

To further our understanding of the above results, two aspects of the original data sets should be mentioned. As a reference system, Silliman and Simpson [1987] reported a series of initial experiments in a homogeneous medium. As shown in their Figure 4, the breakthrough curves for the homogeneous packing followed closely the shape predicted using a standard Fickian model with a constant dispersivity at all measurement columns. Moreover, Silliman *et al.* [1987] examined transport behavior in a homogeneous sand interspersed with a random packing of vertically oriented, impermeable plastic disks. While the flow field was disturbed so that fluid and tracer paths were diverted around these disks, the overall breakthrough curves again exhibited Fickian behavior. As a result, we are confident that the consistent deviation obtained for the homogeneously heterogeneous medium experiments is a measure of true transport behavior and not an artifact of the experimental procedures or measurement error. These measurements reflect the complex interplay between the high-permeability and low-permeability zones and their effect on fluid flow and tracer movement.

The second aspect of the experiments relates to an observed drift in the "average" seepage velocity reported by Silliman and Simpson [1987]. In their experiments on the fourth packing the seepage velocities calculated from the arrival of $C/C_0 = 0.5$ consistently increase from the first through the third electrode columns. The variation in this velocity was inconsistent from the third to the fourth column, but the velocity was always greatest at the fifth column. For example, in the experiments shown in Figure 4, the estimated velocities were 5.43, 5.70, and 5.79 m/d at distances of $L = 0.91$, 1.37, and 1.83 m (electrode columns 3, 4, and 5) from the source, respectively (while measurements taken at electrode columns 1 and 2, with $L = 0.15$ and 0.46 m, led to velocity estimates of 3.97 and 4.22 m/d, respectively). While these experiments were run under unconfined conditions, the head differences used in these experiments were such that the maximum change in saturated thickness across the entire cell was less than 3%; as such, the increase in velocity cannot be explained by arguments related to the change in saturated thickness.

Rather, we believe that the observed variation in measured velocities is due to a combination of non-Fickian behavior and measurement error. Note that these velocities were estimated from the breakthrough curve data, assuming the transport to be Fickian. However, as demonstrated above, the transport is, in fact, anomalous, and as a consequence, these estimated velocities can only be taken as rough values. The very minor variation in estimated velocities at electrode columns 3, 4, and 5, in particular, can easily be attributed to measurement and estimation errors. With our use of non-Gaussian solutions we require only one value of the velocity.

Also, as noted above, the process of packing the tank led to unavoidable heterogeneities, with the result that the mean permeability in the lower portion of the flow cell was evidently somewhat lower than the mean permeability near the upper surface of the flow cell. Silliman and Simpson [1987] found that the breakthrough curve at the first electrode column in the homogeneous medium was consistent with both Fickian transport and the large-scale dispersivity, so that the upgradient boundary condition is probably not the cause of the spatial variation in the velocity in the homogeneously heterogeneous

Figure 5. Schematic illustration of contaminant transport in heterogeneous porous media. As shown by the white arrows, we present a conceptual mapping of contaminant flow paths onto a mixture of discrete paths with varying lengths and ranges of transition times. The underlying velocity field map [after Grindrod and Impey, 1993] is representative of flow in heterogeneous media; lighter shading represents regions of higher velocity.

medium. Rather, it appears that the flow distance at least up to the second electrode column is not sufficiently large to adequately sample the velocity field and to yield representative behavior. As discussed in section 2, the calculation of the FPTD is valid for $\tau \gg 1$, which is the appropriate time range for $b \gg 1$, that is, $L \gg \langle \ell \rangle$. The analyses of these experiments therefore neglect measurements from the first and second electrode columns.

5.2. Additional Theoretical Considerations

The statistically complex geometry and morphology of aquifer heterogeneities give rise to a mixture of preferential pathways and stagnant regions, which can lead to non-Gaussian transport behavior. Such behavior was clearly demonstrated in the context of fracture networks by, for example, Berkowitz and Scher [1997, 1998] and was demonstrated for heterogeneous porous media by, for example, Scheibe and Cole [1994]. The CTRW framework is based on particles making discrete transitions in space with a distribution of transition times.

In a fracture network these discrete transitions are defined physically by the fracture segments (distances between fracture intersections). In a heterogeneous porous medium the definition of these discrete transitions is more subtle. Figure 5 presents a schematic of our conceptual picture of transport through heterogeneous porous media; particles move lengths controlled by the coherence properties of this velocity field and the scattering features between different velocity values. The presence of a large range of velocities is necessary but not sufficient to produce non-Fickian, anomalous transport. As discussed by Berkowitz and Scher [1995], the encounter of the particles with this range of velocities is crucial. Anomalous transport arises if the encounter-range relationship produces a wide spread of different sequences in the flow paths for the migrating particles. The arrows shown schematically in Figure 5 are an illustration of this encounter-range relationship where particles in high- and moderate-velocity paths interact sporadically with low-velocity regions. Plausibility for this conceptual picture is provided in the work of Berkowitz and Scher [1998] in

their application of the CTRW to the heterogeneous Columbus aquifer.

As noted in section 2, transport is anomalous for $0 < \beta < 1$, and the peak of the concentration distribution lags behind the mean plume position. It can be shown that for $\beta \geq 1$, the peak is more nearly coincident with the mean position [see Montroll and Scher, 1973, Figures 3 and 4]. However, because $\bar{\sigma}(t)$ does not vary as $\sqrt{t}$ [Shlesinger, 1974], the transport is therefore still "anomalous." The propagating distribution resembles a Gaussian distribution but with broader ("heavier") tails. The transport tends to a complete Gaussian form for $\beta \geq 2$ [Montroll and Scher, 1973; McLean and Ausman, 1977]. It seems reasonable to expect a relatively large value of β (with $0 < \beta < 1$) for the experimental system considered here since the heterogeneities in the porous medium are not highly contrasted (i.e., about 1 order of magnitude difference in hydraulic conductivity between the two types of sand). As such, our finding (recall Figure 4) that $\beta \approx 0.87$ is satisfying.

As we have discussed above, the value of β is dominated by the low-velocity tail of the flow field distribution; this feature was shown explicitly from numerical studies of fracture networks by Berkowitz and Scher [1998]. We stress that β is an experimental parameter which is physically identifiable from analysis of the flow field. Moreover, $\bar{v}$ is determined directly by measurements of the mean flow, while the length scale $\langle \ell \rangle$ is determined by details of the medium heterogeneities.

6. Concluding Remarks

As shown by Berkowitz and Scher [1997, 1998] and Hatano and Hatano [1998], the CTRW theory for anomalous transport qualitatively and quantitatively describes observed transport behaviors in geological media which have not been adequately explained by previous theories. The FPTD curves in the present study fit well the measured data from a "homogeneously heterogeneous" porous medium, with a single value of the β exponent. The consistency of the CTRW with the measurements over the spatial/temporal scale of the experiment

suggests that the CTRW may quantify transport in heterogeneous porous media at scales which are not amenable to analysis using classical advection-dispersion theory. Hence the CTRW represents a potentially valuable tool in the quantitative assessment of contaminant transport in such media.

Questions still remain as to the generality of the CTRW theory for both fractured and heterogeneous porous media. In particular, it remains to investigate what properties of a random system result in the behavior $\psi(s, t) \rightarrow t^{-1-\beta}$ as $t \rightarrow \infty$ (i.e., an algebraic drop-off in the low velocities), which leads to anomalous transport. These and related questions, including the use of complementary formalisms such as Lévy flights and fractional derivative transport equations, which are special cases of the more general CTRW [e.g., Hilfer and Anton, 1995; Compte, 1996; Metzler et al., 1998], are the subject of our continuing investigations.

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Correction to "Anomalous transport in laboratory-scale, heterogeneous porous media" by Brian Berkowitz, Harvey Scher, and Stephen E. Silliman

In the paper "Anomalous transport in laboratory-scale, heterogeneous porous media" by Brian Berkowitz, Harvey Scher, and Stephen E. Silliman (*Water Resources Research*, 36(1), 149–158, 2000), an error appears in Figure 2c in the curve labeled $\beta = 0.33$, which was included only for illustrative purposes. The analysis, results, and conclusions of the paper remain unchanged. The correct Figure 2 follows.

Figure 2. Semilog plots showing a range of first-passage time distribution (FPTD) curves (probability density function $b^{1/\beta}f(L, \tau)$ and cumulative $M(L, \tau)$) versus $\tau/b^{1/\beta}$ for a range of β . (a) Plots of $b^{1/\beta}f(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.33, 0.50$, and 0.65 . (b) Plots of $b^{1/\beta}f(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.75, 0.80$, and 0.90 . (c) Plots of $M(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.33, 0.50$, and 0.65 . (d) Plots of $M(L, \tau)$ versus $\tau/b^{1/\beta}$, $\beta = 0.75, 0.80$, and 0.90 .

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Estimating the variance and integral scale of the transmissivity field using head residual increments

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Abstract. A modification of previously published solutions regarding the spatial variation of hydraulic heads is discussed whereby the semivariogram of increments of head residuals (termed head residual increments HRI) are related to the variance and integral scale of the transmissivity field. A first-order solution is developed for the case of a transmissivity field which is isotropic and whose second-order behavior can be characterized by an exponential covariance structure. The estimates of the variance σ_Y^2 and the integral scale λ of the log transmissivity field are then obtained via fitting a theoretical semivariogram for the HRI to its sample semivariogram. This approach is applied to head data sampled from a series of two-dimensional, simulated aquifers with isotropic, exponential covariance structures and varying degrees of heterogeneity ($\sigma_Y^2 = 0.25, 0.5, 1.0, 2.0$, and 5.0). The results show that this method provided reliable estimates for both λ and σ_Y^2 in aquifers with the value of σ_Y^2 up to 2.0 , but the errors in those estimates were higher for σ_Y^2 equal to 5.0 . It is also demonstrated through numerical experiments and theoretical arguments that the head residual increments will provide a sample semivariogram with a lower variance than will the use of the head residuals without calculation of increments.

1. Introduction

The rapid development of stochastic groundwater theory has led to such diverse applications as the mapping of the transmissivity distribution via kriging or cokriging [e.g., *Hoeksema and Kitanidis*, 1984; *Rubin and Dagan*, 1987a], conditional simulation of transmissivities through the use of head and chemical data [e.g., *Harvey and Gorelik*, 1995], prediction of effective conductivities, transmissivities, or dispersivities [e.g., *Dagan*, 1987; *Gelhar and Axness*, 1983; *Neuman and Orr*, 1993], and prediction of uncertainties in capture zones for wellhead protection [e.g., *Cole and Silliman*, 1997]. All these applications require estimates of the variance and integral scale(s) of the conductivity field (or transmissivity field in a two-dimensional aquifer.)

A common approach to this field problem involves the direct measurement of the conductivity (or transmissivity) via field testing or collection of sediment cores for testing in the laboratory. From these field data, sample semivariograms for the conductivity (or transmissivity) can be constructed and fit using theoretical conductivity (or transmissivity) semivariograms [e.g., *Sudicky*, 1986; *Hess*, 1989; *Rehfeldt et al.*, 1992]. The advantage of this approach is its simplicity, which has resulted in its wide use [Gelhar, 1993]. The disadvantage, however, is that conductivity or transmissivity data are expensive to collect, and the estimates based on the sample moment can be unreliable when there are insufficient direct measurements available.

Another approach to parameter estimation is the maximum likelihood method, in which the corresponding variable's like-

lihood function is maximized given the available field data [Kitanidis and Vomvoris, 1983; Hoeksema and Kitanidis, 1984, 1985; Rubin and Dagan, 1987a, b; Sun and Yeh, 1992]. This approach has the advantage that it provides direct quantification of the uncertainties associated with the parameter estimates. In addition, this technique can be extended to the combined use of other types of data such as hydraulic head measurements, solute concentration, etc. [e.g., *Harvey and Gorelick*, 1995]. However, the use of this method requires an assumption that the data have a joint normal distribution. Further, the computation involved can be heavy, especially when the number of measurements considered is large.

In the current report we target the groundwater sites where a large number of hydraulic head measurements are available. We explore the possibility of basing estimates of the variance and integral scale of the transmissivity purely on semivariogram derived from head data. Examples of these sites can be found in the alluvial aquifer or the Ogallala Formation in southwestern and northwestern Kansas, where hundreds of head measurements have been collected annually through a multiyear water table monitoring program [Woods et al., 1998].

As argued in the theoretical work of a number of authors [e.g., *Mizell et al.*, 1982; *Dagan*, 1982, 1985; *Osnes*, 1995], the semivariogram of head residual (the head data with the mean trend removed) can be expressed as a function of transmissivity structure. As a result, it is theoretically plausible to estimate the variance and integral scale of the transmissivity field directly from the semivariogram derived from the head measurements. In the current report we propose the use of increments in the head residual to construct semivariogram. We illustrate the utility of head residual increment (HRI) semivariogram through numerical experiments. Further, we demonstrate the greater reliability of sample HRI semivariogram as compared

Figure 1a. The length scales involved in estimating the head residual increment (HRI) semivariogram, the separation between measurement locations used in estimating the HRI (Δr) and the separation between HRIs used in estimating the semivariogram (r).

to the head residual (HR) semivariogram through numerical and theoretical arguments.

The HRI-based approach is not proposed to replace the other existing parameter estimation methods. Rather, it will be used as an inexpensive (in terms of computation and field costs) tool for extracting information contained in a large hydraulic head data set and for quickly obtaining initial estimates of the variance and integral scale of the transmissivity field. These initial estimates could then be used to facilitate the interpretation of existing field observations and to design further transmissivity sampling strategies for the use of more sophisticated techniques to refine these estimates.

2. Methodology

2.1. Definition of Head Residual Increment (HRI)

The method discussed here is based on what is termed a head residual increment or HRI. In order to define the HRI, consider two head values, $H(x_1)$ and $H(x_2)$, measured at two wells located at the points x_1 and x_2 within an aquifer subject to a regional trend in the hydraulic head $\bar{H}(x)$. The corresponding head residuals, which represent the variation of hydraulic head about its regional trend, are

$$h(x_1) = H(x_1) - \bar{H}(x_1), \quad (1)$$

$$h(x_2) = H(x_2) - \bar{H}(x_2). \quad (2)$$

It is assumed here that these residuals are zero mean random variables. The HRI, located at the point $(x_1 + x_2)/2$, is defined as

$$\Delta h(x_1, x_2) = h(x_1) - h(x_2) = h(x_1) - h(x_1 + \Delta r), \quad (3)$$

where $\Delta r = x_1 - x_2$.

These HRIs are to be used to estimate a sample semivariogram; thus it is desirable that each of the HRIs be collected in a consistent manner. As a result, only those HRIs (constructed from point head measurements) with similar Δr are used in the analysis. This is similar to the requirement that only transmissivities measured with similar sampling supports can be included in the calculation of transmissivity semivariogram.

sivities measured with similar sampling supports can be included in the calculation of transmissivity semivariogram.

2.2. Relationship of HRIs to the Transmissivity Field

In pursuing an expression for the HRI semivariogram for a specified Δr , two distinct separation vectors are involved. As illustrated in Figure 1a, one is the separation between the head data pairs ($H(x_1)$ and $H(x_2)$) used for estimating the HRIs, Δr ; the second separation vector is the separation vector r between a pair of HRIs used in calculating the HRI semivariogram.

The ensemble semivariogram for a pair of HRIs, Δh_1 (calculated from $H(x_1)$ and $H(x_1 + \Delta r)$) and Δh_2 (calculated from $H(x_1 + r)$ and $H(x_1 + r + \Delta r)$), is

$$\gamma_{\Delta h}(\Delta h_1, \Delta h_2) = \frac{1}{2} \langle (\Delta h_1 - \Delta h_2)^2 \rangle, \quad (4)$$

where $\langle \rangle$ stands for ensemble averaging and $\gamma_{\Delta h}$ denotes the HRI semivariogram.

Substituting the definition of HRI (equation (3)) for Δh_1 and Δh_2 in (4) leads to

$$\begin{aligned} \gamma_{\Delta h}(\Delta h_1, \Delta h_2) = \frac{1}{2} \langle \{ [h(x_1) - h(x_1 + \Delta r)] - [h(x_1 + r) - h(x_1 + r + \Delta r)] \}^2 \rangle \\ = \gamma_h(h_1, h'_1) + \gamma_h(h_2, h'_2) \\ + \gamma_h(h_1, h_2) + \gamma_h(h'_1, h'_2) - \gamma_h(h_1, h'_2) - \gamma_h(h'_1, h_2), \end{aligned} \quad (5)$$

where h_1, h'_1, h_2, h'_2 are the head residuals at (x_1) , $(x_1 + \Delta r)$, $(x_1 + r)$, and $(x_1 + r + \Delta r)$, respectively, and γ_h is the semivariogram of the head residual. If the head residual semivariogram γ_h is spatially invariant, the semivariogram of the HRI $\gamma_{\Delta h}$ can be simplified as

$$\begin{aligned} \gamma_{\Delta h}(\Delta r, r) = [2\gamma_h(\Delta r) + 2\gamma_h(r) - \gamma_h(r + \Delta r) \\ - \gamma_h(r - \Delta r)]. \end{aligned} \quad (6)$$

The HRI semivariogram $\gamma_{\Delta h}$ is now tied to the underlying transmissivity distribution via the relationship between γ_h and

the structure of the transmissivity field. Analytical solutions for the head residual semivariogram have been presented by a number of authors, including Mizell *et al.* [1982], Dagan [1982, 1985], and Osnes [1995]. In the following illustration we take Dagan's [1985] analytical result to demonstrate the estimation of variance and integral scale.

2.3. Estimating the Variance and Integral Scale

To illustrate the parameter estimation procedure, we consider a field sampling design in which the sample semivariogram can be constructed with $r \parallel \Delta r$ (i.e., the sample HRI semivariogram is calculated in the direction parallel to the line of head data pairs used to estimate the HRIs, see Figure 1b). In this case the theoretical semivariogram, (6), reduces to

$$\gamma_{\Delta h}(\Delta r, \theta, r) = [2\gamma_h(\Delta r, \theta) + 2\gamma_h(r, \theta) - \gamma_h(r + \Delta r, \theta) - \gamma_h(|r - \Delta r|, \theta)], \quad (7)$$

where r is the magnitude and θ is the orientation of the vector r or Δr .

In a mean-uniform, steady flow field and a log transmissivity field ($Y = \ln(T)$) with isotropic, exponential covariance, Dagan [1985] gave the first-order solution for the head residual semivariogram as

$$\gamma_h(r) = (J^2 \lambda^2 \sigma_Y^2) [F(r, \theta)], \quad (8)$$

where the function $F(r, \theta)$ is given as

$$F(r, \theta) = \frac{1}{2} \left\{ (2(\cos \theta)^2 - 1) \left[\frac{\exp(-d)(d^2 + 3d + 3) - 3}{d^2} \right] + \left((\cos \theta)^2 - \frac{1}{2} \right) - \text{Ei}(-d) + \ln(d) + \exp(-d) + (E - 1) \right\}. \quad (9)$$

Within this expression, λ is the integral scale and σ_Y^2 denotes the variance; J is the magnitude of mean head gradient J , whose orientation is here assumed aligned with the positive x direction; θ is the angle between r and J ; $d = r/\lambda$; Ei is the exponential integral function; and $E = 0.5772$ is the Euler's constant. Formulae (8) and (9) are for exponential covariance only. Results for other covariance are also available [e.g., Mizell *et al.*, 1982].

Substituting (8) into (7), we obtain

Figure 1b. An illustration of the case when $r \parallel \Delta r$. Here θ is the angle between r and positive x direction. Within the Figures 1a and 1b the open circles represent locations where the hydraulic head is measured, and the closed circles represent locations where the HRIs are assigned.

Figure 2. Examples of sample HRI semivariogram and its corresponding theoretical semivariogram with $\sigma_Y^2 = 1.0$, $\lambda = 10$, $\Delta r = 4.0$, $J = 0.003$, and $\theta = 0$.

$$\gamma_{\Delta h}(\Delta r, \theta, r) = J^2 \sigma_Y^2 \lambda^2 W(\Delta r, \theta, r), \quad (10)$$

where

$$W(\Delta r, \theta, r) = 2F(\Delta r, \theta) + 2F(r, \theta) - F(|r - \Delta r|, \theta) - F(r + \Delta r, \theta). \quad (11)$$

In reviewing (10) and (11), we note that λ , J , σ_Y^2 , Δr , and θ are all fixed parameters for a specific HRI semivariogram, and r is the only independent variable. Since the HRI approach is proposed as an initial screening tool for a large hydraulic head data set, we assume that J is known from the available head data via classical contouring or other simple methods [e.g., Silliman and Frost, 1998]. Further, Δr and θ are known based on the sampling design. Hence the only unknown parameters are the variance and integral scale of the log transmissivity field, σ_Y^2 and λ , respectively. Since $d = r/\lambda$, expressions (9) and (11) indicate a complex relationship between the theoretical HRI semivariogram and the value of λ . The dependence of $\gamma_{\Delta h}$ on σ_Y^2 , however, is simply linear. Taking the logarithm of both sides of (10) results in the expression

$$\log [\gamma_{\Delta h}(\Delta r, \theta, r)] = \log [J^2 \lambda^2 W(\Delta r, \theta, r)] + \log (\sigma_Y^2). \quad (12)$$

Plotting $\log [\gamma_{\Delta h}]$ versus the independent variable r , the value of σ_Y^2 determines only the vertical location of the curve and has no impact on its overall shape. The curve shape, however, is determined by the selection of λ . Thus a simple trial-and-error approach can be used to estimate λ value by matching the curve shape. Subsequently, the value of σ_Y^2 can be determined via adjusting the curve's vertical location.

3. Application to Simulated Aquifers

The proposed HRI approach is illustrated through application to a series of simulated aquifers containing log normally distributed, exponentially correlated transmissivity fields. Four values of σ_Y^2 were evaluated in the simulations: 0.25, 1.0, 2.0, or 5.0. This provides results in a range from below to significantly greater than the limit on σ_Y^2 considered appropriate for first-order theory [Dagan, 1985]. For each σ_Y^2 used, 20 realizations of the transmissivity distribution were generated using turning

Table 1. Mean and Standard Deviation (SD) of the Estimates of σ_Y^2 As Derived From the Semivariogram Fits

True σ_Y^2	$\theta = 0$		$\theta = -\pi/4$		$\theta = \pi/4$		$\theta = \pi/2$	
	Mean σ_Y^2	SD σ_Y^2	Mean σ_Y^2	SD σ_Y^2	Mean σ_Y^2	SD σ_Y^2	Mean σ_Y^2	SD σ_Y^2
0.25	0.25	0.02	0.25	0.02	0.26	0.02	0.26	0.02
1.0	1.0	0.1	1.1	0.1	1.0	0.2	1.2	0.1
2.0	2.2	0.2	2.4	0.3	2.2	0.3	3.0	0.1
5.0	6.7	1.1	7.6	1.9	7.2	1.5	10.5	1.5

bands [Tompson *et al.*, 1989]. As the head increments are independent of the mean of the log transmissivity field [Dagan, 1985], the mean of the natural logarithm of the transmissivity was arbitrarily set equal to zero. Further, the integral scale λ was arbitrarily set equal to 10. The overall dimensions of the simulated aquifers were 50λ by 50λ .

A fine level of discretization was used in the turning bands code in order to reduce artifacts in the flow solution related to discontinuity in the transmissivities. For $\sigma_Y^2 = 0.25$ and 1.0, 10 elements were employed per integral scale; for $\sigma_Y^2 = 2.0$ and 5.0, 15 elements per integral scale were utilized. In all cases the covariance of the resulting transmissivity field was calculated to verify that the input exponential covariance function was successfully reproduced.

The numerical transmissivity fields were then used in conjunction with MODFLOW [McDonald and Harbaugh, 1988] to derive the head solution. When applying MODFLOW, constant heads were assigned along the eastern and western boundaries of the transmissivity field to establish a mean regional hydraulic gradient of 0.003. The southern and northern boundaries were maintained as no-flow boundaries.

Following solution of the flow equation, heads were sampled at a subset of the numerical grids, which were at least 5λ away from all boundaries, to minimize the boundary effects [Rubin and Dagan, 1988, 1989]. The head measurements were then used to calculate the HRIs and construct the sample HRI semivariogram. The value of Δr used in all calculations was selected as 4 times the size of one grid element. This magnitude for Δr provided HRIs at a scale less than one half the integral scale of the transmissivity field thus preserving the local variability in HRI. The angle θ was varied, taking on values of $-\pi/4$, 0, $\pi/4$, and $\pi/2$, with respect to the orientation of the mean regional gradient. Hence, for each realization of the transmissivity field, four sets of HRIs were determined, and four sample HRI semivariograms were constructed (parallel to the orientation of the HRI).

Figure 2 shows a sample of the calculated HRI sample semivariogram and its corresponding theoretical semivariogram. The details of curve fittings for all 80 realizations in four

different directions are given by Zheng [1998]. A summary of the resulting parameter estimates is reproduced here in Tables 1 and 2. As shown, the parameter estimates are most exact in the direction of the mean regional gradient and decline in quality as θ varies from 0. Along the direction of mean regional flow ($\theta = 0$) the first-order theoretical HRI semivariogram is valid up to $\sigma_Y^2 = 2.0$.

4. Reliability of Sample HRI Semivariogram

As the HRI semivariogram is based on combination of semivariograms of the head residuals (HR), it is natural to ask whether it would be more convenient and of equal utility to work directly with the HR semivariogram. The answer is no, because the HRI offers a more reliable sample semivariogram than the HR does. Figure 3 illustrates sample HR (Figure 3a) and HRI (Figure 3b) semivariograms for 10 realizations of the same numerical aquifers. Both semivariograms were calculated from all available head data on the numerical grids (over 160,000) excluding the boundary strips. As shown in Figures 3a and 3b, there is substantial noise in the HR sample semivariograms, whereas the HRI sample semivariograms show significantly less variability from one realization to the next.

Further, theoretical analysis shows that the greater variability in the sample HR semivariogram is inherent because of the long range correlation existing in the head field. Zheng and Silliman [2000] have recently completed analysis of the factors impacting the variance of the sample semivariogram and demonstrated that the variance of a sample semivariogram depends on not only the number of data pairs included in the estimate but also on the correlation among the squared increments used in the summation. Quantitatively, this relationship can be summarized as [Zheng and Silliman, 2000]

$$\sigma_{\hat{\gamma}}^2 = \frac{2\gamma^2(\mathbf{r})}{N(\mathbf{r})} + \frac{2}{N^2(\mathbf{r})} \sum_{j>i}^{N(\mathbf{r})} \sum_{i=1}^{N(\mathbf{r})-1} \text{Cov}[S(\mathbf{x}_i, \mathbf{r}), S(\mathbf{x}_j, \mathbf{r})], \quad (13)$$

where $\hat{\gamma}(\mathbf{r})$ denotes the sample semivariogram of its theoretical counterpart $\gamma(\mathbf{r})$; $N(\mathbf{r})$ is the number of the data pairs for a

Table 2. Mean and Standard Deviation (SD) of the Estimates of λ As Derived From the Semivariogram Fits

σ_Y^2	$\theta = 0$		$\theta = -\pi/4$		$\theta = \pi/4$		$\theta = \pi/2$	
	Mean λ	SD λ	Mean λ	SD λ	Mean λ	SD λ	Mean λ	SD λ
0.25	10.1	0.5	10.1	1.2	9.9	1.1	10.1	0.4
1.0	9.9	0.6	9.8	0.8	9.8	0.8	9.4	0.8
2.0	9.1	0.6	8.8	0.7	9.0	0.9	8.9	0.4
5.0	8.1	0.9	8.1	0.7	7.7	0.9	6.0	0.9

The theoretical $\lambda = 10.0$ for all the realizations.

Figure 3. (a) The sample HR semivariograms from 10 different realizations with the corresponding theoretical HR semivariogram. (b) The sample HRI semivariograms from 10 different realizations with the corresponding theoretical HRI semivariogram with $\sigma_y^2 = 1.0$, $\lambda = 10$, $\Delta r = 4.0$, $J = 0.003$, and $\theta = 0$.

specified lag distance r ; $S(x, r)$ is the squared increment between a data pair located in x ; $\text{Cov}[S(x_i, r), S(x_j, r)]$ is the covariance between $S(x_i, r)$ and $S(x_j, r)$ and can be further expressed as

$$\text{Cov}[S(x_i, r), S(x_j, r)] = \frac{1}{2} [\gamma(r + x_i - x_j) + \gamma(r - x_i + x_j) - 2\gamma(x_i - x_j)]^2. \quad (14)$$

Using the same sampling schemes for both HR and HRI and substituting the previously defined theoretical HRI and HR semivariograms into (13) and (14), we calculate and plot the coefficient of variation (the standard deviation normalized by the mean) of the sample semivariograms for both HR and HRI versus the number of data pairs used in Figure 4. Also given is the coefficient of variation of the sample semivariogram for an uncorrelated process. In the case of HR, head residual is correlated over a long distance, which results in high correlation among the squared increments and thus prevents significant reduction of the variance of the sample HR semivariogram, even when a large number of data pairs are used in the calculation (e.g., Figures 4 and 3a). The head residual increment, in contrast, is correlated over a much shorter range as illustrated in Figures 2 and 3b. As a result, the variance of the sample

HRI semivariogram reduces at a much faster rate, very much resembling the uncorrelated process, as the number of data pairs used increases.

5. Discussion

Using numerical experiments and theoretical arguments, we have demonstrated that the HRI approach holds promise as an inexpensive tool for quick assessment of σ_y^2 and λ for aquifers with σ_y^2 at least up to 2.0. The primary advantage of the proposed HRI method is that it is based solely on inexpensive head measurements. Although others have devised cokriging-based methods to incorporate the hydraulic head measurements in the transmissivity parameter estimation, the heavy computational demand and the associated convergence problem has put a limit on the number of head data that may be included in the inversion [Gelhar, 1993]. The major feature of the proposed HRI approach is that the information contained in a large head data set can be summarized quickly to provide initial assessment of the transmissivity structure and thus facilitate the interpretation of existing observations and the design of future explorations. The key in the proposed HRI approach is the conversion of HRs into HRIs thus reducing the correlation among the data pairs to improve the reliability of the sample semivariogram. A choice of a sufficiently small Δr is necessary to ensure this. In practice, the appropriateness of the choice of Δr can be judged from the smoothness of the resulting HRIs, because the reduction of correlation among the HRIs will manifest as an increased irregularity in HRI data in contrast to the smoothness in the original head data.

The application of this approach in the field requires the further resolution of several other important issues, including how the number of the head data, the irregular sampling locations, the restrictive geometry in constructing HRI, the measurement errors, the cross correlation between regional gradient estimate and HRIs, and the deviations from mean linear flow (due, for example, to local pumping and uneven distribution of recharge/discharge) impact the quality of the sample HRI semivariogram and the subsequent parameter estimates. Some of these issues have been addressed by Zheng [1998]; others are still under investigation. Results from these investigations as well as the field applications of the HRI approach will be reported in a future paper.

Figure 4. The coefficient of variation of the sample semivariograms versus the number of data pairs used in calculation.

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Utility of Simple Models for Capture Zone Delineation in Heterogeneous Unconfined Aquifers

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Abstract

Monte Carlo simulation is used to examine the utility of simple (homogeneous/isotropic) models for the delineation of wellhead protection areas (WHPAs) in heterogeneous, statistically anisotropic, unconfined aquifers. Capture zone geometry is investigated under two hydrologic conditions: regional gradient negligible compared to pumping and significant regional gradient compared to the local gradient due to pumping. Variability of capture zone geometry is quantified in terms of simple measurements (maximum transport distance and maximum width of the capture zone) and the probability of inclusion in the capture zone of a series of hypothetical contaminant source locations. These results indicate that substantial uncertainty exists in the delineation of a wellhead capture zone. This uncertainty is related, principally, to flow that occurs along preferential pathways associated with zones of high hydraulic conductivity. It is suggested that these uncertainties can be accommodated in one of three fashions depending on the value of land, the benefit of wellhead protection, and the availability of funds to perform more thorough field assessment. The first option is to modify the estimated capture zone by a factor of safety. In the absence of significant regional flow, a factor of safety of approximately three applied to the calculated fixed radius estimate of the capture zone appears to be sufficient for the conditions simulated in this project. In the case of a capture zone within a significant regional gradient, uncertainty in the location of the real capture zone will require safety factors on the width of the estimated capture zone on the order of 30, with a safety factor of approximately three in the longitudinal direction. The second option reduces these factors of safety by increasing the risk that the estimated capture zone will not cover the entire area of the real capture zone. Work available in the literature provides insight into the trade-off between the size of the estimated capture zone and the risk endured. The third option is to reduce uncertainty in the vicinity of a wellhead through additional expenditure to allow the application of classic hydrologic techniques involving conditioning or inverse modeling.

Introduction

Wellhead protection applications, particularly for small municipalities, create situations in which it is often difficult to justify the costs associated with the collection of large data sets from a field site to allow advanced parameter identification (e.g., inverse modeling) and computational analysis. Further, high demand for delineation of wellhead protection areas (WHPAs) and limited personnel for performing the analysis can lead to a reliance by state water managers on simplified analytical solution techniques (Cross 1995). For example, one of the most widely used methods for designation of WHPAs involves the calculated fixed-radius (CFR) capture zone (U.S. EPA 1987).

While reliance on simple analytical models often allows delineation of a WHPA with minimal field data, there are often significant assumptions and uncertainties associated with the resulting estimates. The CFR method, for example, requires only limited data input (single values for the pumping rate, transmissivity, and porosity), but reasonable representations of the capture zones are achieved

only for negligible regional gradients in the presence of relatively homogeneous aquifer materials. One can conclude that because practical considerations may limit the complexity of models used to analyze capture zones for water supply wells, a question arises as to how to include the uncertainty associated with capture zones defined through the use of these simple models.

Previous studies have addressed various aspects of uncertainty in capture zone delineation. Two studies (Bhatt 1993; Rifai et al. 1993) identified the sensitivity in capture zone shape to variability of model input parameters (e.g., porosity, saturated thickness, transmissivity) in a homogeneous aquifer. Bair et al. (1991) used a time-of-travel model in a homogeneous aquifer with uncertain hydraulic conductivity and porosity to identify confidence intervals to quantify variation in the capture zone shape in terms of the area and the ratio of the length and the width. Additionally, Bair et al. (1991) suggest that the distribution of flow lines, as derived from Monte Carlo simulations, will be a useful indicator for placing monitoring wells. Several studies have used published spatial statistics (e.g., log conductivity variance and correlation scale) to investigate various aspects of capture zone uncertainty through Monte Carlo simulation. Cole and Silliman (1997) investigated confidence intervals for passive (low production) wells in unconfined aquifers; other authors (van Leeuwen et al. 1999; Varljen and Shafer 1991) used simulations conditioned on both the measured spatial correlation structure of the conductivity field and measured hydraulic conductivities to demonstrate that stochastically determined capture

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zones can aid in finding confidence regions that will contain the actual capture zone. Recent attempts have been made to view capture zone uncertainty in terms of transport time statistics from a contamination source to a production well (Guadagnini and Franzetti 1999; Cole 1995).

In this paper, a somewhat different approach to delineation of wellhead capture zones is presented. For the case of negligible regional flow, the potential for using safety factors in combination with the CFR method is investigated. For the case of nonnegligible regional flow, the potential for modifying simple (assuming homogeneous/isotropic conditions) solutions with safety factors is considered. The effectiveness of safety factors is considered through the application of the Monte Carlo simulation and assessment of capture zones in a series of hypothetical, unconfined aquifers with a single pumping well.

The impact of recharge on the geometry of the capture zone is not considered herein. Recharge can have significant impact on the geometry of the capture zone. For example, if recharge is spatially uniform, the capture zone will be smaller. However, uniform recharge is likely to have only a minor effect on the uncertainties associated with identification of the capture zone. In contrast, if recharge is spatially variable, particularly if poorly characterized, recharge may dramatically increase the uncertainty inherent in the identification of the capture zone. As the primary motivation of this study is to introduce safety factors as a means of incorporating uncertainty in the delineation of capture zones, these important issues related to recharge are left for future discussion.

Problem Statement

Cole and Silliman (1997) used hypothetical case studies to examine the variation of the capture zone to a passive (low production/monitoring) well. This study is based on the same physical geometry and boundary conditions used in the Cole and Silliman (1997) paper, but the pumping rate is increased to allow investigation of the variation of the capture zones under conditions of nonpassive wells. Specifically, two classes of problems are considered: In the first (termed the *zero-gradient* class), it is assumed that pumping at the well (at a rate of 0.0044 m³/s) dominates the local hydraulics. This case is modeled as an aquifer with a zero regional gradient. In the second (termed the *regional-gradient* class), it is assumed that both the pumping and the regional gradient influence the local hydraulics. This case is modeled with a regional gradient of 0.5% and a pumping rate of 0.0044 m³/s. For both classes of simulations, six individual case studies (discussed later and summarized in Table 1) were analyzed.

The mathematical representation of the flow field is shown in Figure 1. Flow is assumed to be two-dimensional and at steady state. The aquifer is square with a side dimension of 5025 m with a single well located at (3513 m, 2513 m) relative to the bottom left corner of the grid. Hydraulic conductivities are assigned by dividing the flow domain into 201 cells in both the x- and y-directions and using the random field generator described in Cole and Silliman (1997). For the zero-gradient simulations, a constant, prepumping saturated aquifer thickness of 45 m is used (i.e., h_1 and h_r in Figure 1 are both set equal to 45 m). For the regional-gradient class, the regional gradient is aligned, on average, parallel to the x-axis between constant head boundary values of $h_1 = 70$ m and $h_r = 45$ m (Figure 1).

The impervious base of this unconfined aquifer is presumed to be at constant elevation, which for steady, two-dimensional flow ren-

Figure 1. The hypothetical unconfined aquifer and its mathematical representation. Indicated is one possible capture zone for the well based on assumed parameters regarding the spatial distribution of hydraulic conductivity. The capture zone is delineated using both particle tracking endpoints (dotted line), and the flow lines with the highest and lowest ordinate value (dashed line).

ders the flow equation linear (using the base of the aquifer as datum) in terms of the square of the head (Cole 1995) as follows:

$$\frac{\partial}{\partial x} \left(K(x,y) \frac{\partial h^2}{\partial x} \right) + \frac{\partial}{\partial y} \left(K(x,y) \frac{\partial h^2}{\partial y} \right) = 2Q\delta(3513, 2513) \quad (1)$$

where Q is the magnitude of pumping (0.0044 m³/s), and δ signifies the Dirac delta function for the well location (3513, 2513). The factor of 2 appears on the right-hand side of the equation due to linearization of the flow equation in terms of the square of the hydraulic head.

Solution of the flow equation is accomplished using the method of finite differences as represented by MODFLOW (McDonald and Harbaugh 1988), with the exception that the square of the hydraulic head replaces the hydraulic head and hydraulic conductivity replaces transmissivity for the input files. These substitutions allowed use of the linear solver for the confined aquifer solution that resulted in reduced computation times. Linearization of the governing flow equation caused intercellular flows calculated by MODFLOW to be incorrect. This precluded use of MODPATH (Pollock 1989) for delineating the capture zones using particle tracking, hence, a modified reverse particle tracking algorithm was used (Cole 1995).

The Dirac delta function in Equation 1 indicates that the well is represented in the differential equation as a point in space. To mit-

Table 1
General Parametric Description of the Six Cases Used^a

Case ^b	$\mu_{\text{Log K}}$	$\sigma^2_{\text{Log K}}$	$\lambda_x/1_x^c$	$\theta_{\text{Pri. Corr.}}^d$
1	-4.0	4/9	0.01	0
2	-4.0	4/9	0.03	0
3	-4.0	4/9	0.075	0
4	-4.0	4/9	0.239	0
5	-4.0	4/9	0.03	30
6	-4.0	4/9	0.03	60

^aAll six cases were repeated for both the zero gradient and the regional gradient studies.

^bCase numbers correspond to those used in Cole and Silliman (1997).

^cRatio of the integral scale to flow domain length in the x-direction (5025 m).

^dOrientation of the primary correlation axis relative to the mean region gradient direction (the x-axis).

igate the numerical error in calculating the hydraulic head in the vicinity of the well and performing reverse particle tracking from the region surrounding the well, the cells in the 201 by 201 grid used for assigning hydraulic conductivity values were subdivided to create a finite difference grid with up to 31 by 31 cells for each original cell in the vicinity of the pumping well. This numerical resolution increase was required to adequately delineate capture zone boundaries for the simulations including regional gradients.

The capture zone for the well in each realization was delineated using the endpoints for 360 particles placed at 1° increments on a circle surrounding the pumping well. This procedure proved adequate in applications involving the zero-gradient case studies. In the presence of a significant regional gradient, these endpoints were supplemented with the entire flow lines for the two particles having the largest and smallest ordinate values (see Figure 1). Detailed discussion of the discretization method and capture zone delineation can be found in Cole (1995, Chapter 3).

Simulation Statistics

It is assumed for the present study that the primary cause of uncertainty in delineating capture zones is limited knowledge regarding aquifer heterogeneity. As a result, a series of hypothetical aquifers were generated on the computer with heterogeneous distributions of hydraulic conductivity. Through use of a random field generator (Silliman and Wright 1991), 250 aquifers for six case studies (described later) were simulated. The hydraulic conductivity was assumed to be log-normally distributed with a mean of 10^{-4} m/s and a variance of the base 10 logarithm of 4/9. The hydraulic conductivity was subject to an anisotropic correlation structure (with a ratio between the minimum and maximum integral scale of 4:1), meaning the formation was more likely to have similar hydraulic conductivity values in one direction than in the direction perpendicular to it. To consider a series of scenarios representative of various field conditions, the integral scale and orientation of the principal axis of correlation relative to the mean regional gradient were varied as outlined in Table 1.

As these series of simulations were designed to represent realistic cases in which water is actively produced from the pumping well, it was decided that those realizations in which the "well" would fall within a zone of low hydraulic conductivity should be eliminated as being unrealistic relative to the field situation. Based on this argument, the simulations were conditioned on the requirement that the value of the hydraulic conductivity in the element con-

taining the well was required to be greater than the geometric mean hydraulic conductivity. It is noted that the selection of the cut-off being equal to the geometric mean was arbitrary, and similar results could be obtained for other conductivity cut-offs. As reported previously (Cole and Silliman 1997; Cole 1995), the resulting statistics indicate that 250 realizations were sufficient to adequately define the inclusion probabilities discussed later.

As stated previously, the case studies performed are grouped into those concerning capture zones for situations in which pumping dominates capture zone hydraulics (the zero-gradient class) and those in which the regional gradient significantly impacts the shape and orientation of the capture zone (the regional-gradient class). While results from both classes are characterized in terms of capture zone geometry and inclusion of specified source locations (defined later), the actual measurements collected for the zero-gradient simulations are different from those collected for the regional-gradient simulations.

For zero-gradient conditions the analytical solution for the capture zone in a homogeneous formation is a circle with the well at its center. The geometry of the capture zone for the zero-gradient case studies was characterized in terms of the largest distance between the well and a point on the capture zone boundary. Due to statistical anisotropy, both the absolute distance and the orientation of this point were recorded. Final results are reported in terms of a histogram of the orientation on a unit circle, and statistics including the mean, standard deviation, and skew of the maximum radial distance of a point on the capture zone boundary from the well. All zero-gradient statistics were determined for a 25-year travel time to avoid boundary effects.

The behavior of the capture zone was also analyzed by determining the number of realizations for which a group of hypothetical source points (locations that might represent a known source of contamination or a potential threat to the ground water supply) were included in the simulated capture zone. For the zero gradient cases, four source points were examined. These include two points 250 m from the well and two points 500 m from the well. For each distance, one point was placed directly to the right of the pumping well, and the other point was placed directly above the pumping well. The distances from the well were selected such that the two points at 250 m from the well were located slightly inside the cap-

Figure 2. Fifty-year homogeneous/isotropic capture zone with plus signs indicating potential source location. Note that only a portion of the 60 total potential source points are shown.

Maximum Boundary Distance Orientation (degrees)

Maximum Boundary Distance Orientation (degrees)

Figure 3. Orientation angle frequency of the capture zone boundary point farthest from the well. The primary correlation direction relative to the mean regional gradient is: (a) in alignment (Case 2), and (b) rotated 60° counter-clockwise (Case 6).

ture zone boundary predicted using the geometric mean K and assuming a homogeneous aquifer (the analytical solution provided a predicted radius of approximately 270 m). The two points at 500 m from the well were expected to be outside the capture zone predicted using the geometric mean K .

To identify whether a particular location was included in the capture zone, forward particle tracking was conducted for the specified travel time starting at the potential pollution source. If the particle entered the cell containing the well during the specified time period (at most 1 m from the well itself), the source location was

Table 2
Maximum Radial Distance Statistics
for Zero-Gradient Scenario

Case	Normalized Int. Scale	Mean Max. Distance (m) ^a	σ Max. Dist.	Skew
1	0.010	503	60	0.59
2	0.030	589	100	0.99
3	0.075	490	128	1.20
4	0.239	374	92	3.18

^aNote: Radius for 25-year CFR capture zone is 273 m.

considered to be included in the capture zone.

For the regional-gradient class, the geometry of the capture zone for a particular case study was characterized in terms of two measures: (1) the maximum width of an individual simulated capture zone, and (2) the ratio of the largest width of an individual capture zone among a set of simulations to the width of the smallest region bounding all capture zones for a particular case study. Identification of this region is accomplished using the approach discussed in Varljen and Shafer (1991) and involves recording the largest distance between the well and a point on the group of capture zone boundaries (for the given case study) for a given angular increment. By finding this maximum for a number of angular increments, the region boundary can be constructed that represents the extreme of all capture zones simulated in the case study. For this study, half-degree increments were used.

For the regional-gradient class of case studies, potential source locations were used at 60 locations: 21 along the line parallel to the mean regional gradient passing through the pumping well, and 39 perpendicular to the mean regional gradient halfway between the pumping well and the left boundary (Figure 2). These points were chosen relative to the 50-year time-of-travel capture zone determined using the geometric mean conductivity and the assumption of a homogeneous aquifer (hereafter termed the *homogeneous capture zone*). The 60 potential source locations shown in Figure 2 are arranged to include points both within and exterior to the 50-year homogeneous capture zone. The 50-year travel time was chosen to allow statistical analysis with the homogeneous capture zone covering from two (Case 4) to 45 (Case 1) integral scales.

Zero-Gradient Results

Table 2 provides the statistics calculated for the maximum distance between a point on the capture zone boundary and the well for Cases 1–4, i.e., those involving variation in the integral scale. These results indicate that regardless of correlation scale, the capture zones contain on average at least one point that is located more than 30% farther from the well than the distance predicted by the CFR method using the geometric mean conductivity (273 m). The average maximum distance is largest for a normalized integral scale (integral scale divided by the width of the numerical grid) of approximately 0.03 and then decreases as the integral scale increases. The coefficient of variation (standard deviation divided by the mean) of the maximum distance increases with increasing integral scale through a normalized integral scale of 0.075 and then is approximately constant for the largest two integral scales at a value of approximately 0.25.

The skew is large for the longer integral scales. This, combined with the observation that the average maximum distance from the

Figure 4. Capture zone inclusion probability comparisons for various integral scales along lines (a) parallel, and (b) perpendicular to the mean regional gradient. Case studies indicated are: 1 - x's, 2 - o's, 3 - +s, and 4 - *s.

Figure 5. Capture zone inclusion probability comparisons for various primary correlation axis orientations along lines (a) parallel, and (b) perpendicular to the mean regional gradient. Case studies indicated are: 2 - x's, 5 - o's, and 6 - +s.

Table 3
Potential Source Location Inclusion Probabilities
for the Zero-Gradient Scenario^a

Case	(3762,2513) ^b	(4013,2513) ^c	(3513,2762) ^d	(3513,3013) ^e
1	72.0%	3.6%	42.4%	1.2%
2	77.2%	13.6%	30.4%	1.2%
3	79.2%	8.0%	39.2%	0.0%
4	83.2%	1.6%	55.2%	0.0%

^aSource location coordinates are in m as noted. Twenty-five-year travel times were used. The well is located at (3513 m, 2513 m).

^bThis point is to the right of the well and inside the homogeneous capture zone.

^cThis point is to the right of the well and outside the homogeneous capture zone.

^dThis point is above the well and inside the homogeneous capture zone.

^eThis point is above the well and outside the homogeneous capture zone.

ing well. This distance is nearly four times the distance predicted using the CFR delineation method.

Figure 3 provides histograms of the orientation (relative to the positive x-axis on the numerical grid) of the farthest point on the capture zone boundary from the well for Cases 2 and 6. These two case studies have identical parameters except for the primary orientation of the statistical anisotropy. The orientations around the well observed for Case 2 tend to cluster around 0° and 180° relative to the x-axis. In contrast, the orientation observed for Case 6 tend to cluster about orientations around the well of 60° and 240° relative to the x-axis. In both cases, these clusters of orientations coincide with the primary directions of correlation for these case studies (Table 1), indicating that the orientation of the farthest point on the capture zone boundary tends to coincide with the orientation of the largest integral scale in the underlying conductivity structure.

Inclusion probabilities for Cases 1 through 4 (Table 3) illustrate the impact of the orientation of the correlation structure of the underlying conductivity field. In each of these cases, the direction with the largest integral scale in the correlation structure is parallel with the x-axis. In a comparison of points equidistant from the pumping well, the results in Table 3 indicate higher inclusion probabilities for the points directly to the right of the well than for the

well decreases with increasing integral scale, is interpreted as indicating that, for large integral scales, a limited number of capture zones are subject to significant preferential flow, thus channeling flow from large distances to the pumping well. In reviewing the individual realizations for the normalized integral scale equal to 0.239 (Case 4), for example, it was noted that one capture zone had boundary points that were more than 1000 m away from the pump-

Table 4
Maximum Capture Zone Width Statistics
for the Regional-Gradient Scenario

Case	Mean	Stand. Dev.	Skewness	Width Ratio
1	205 m	32.5 m	0.22	0.52
2	210 m	63.8 m	0.22	0.40
3	210 m	107.0 m	0.86	0.35
4	170 m	135.0 m	1.77	0.28
5	247 m	67.9 m	0.85	0.36
6	316 m	83.9 m	0.47	0.29

two points directly above the well.

As expected, the two points within the boundary of the capture zone calculated with the geometric mean conductivity (i.e., the capture zone that would be derived from the CFR method) have higher inclusion probabilities than the points located outside this capture zone boundary. The points inside the boundary predicted from the CFR method have inclusion probabilities as low as 30%. That is, the point above the well that was predicted to be in the capture zone (based on the CFR method) was deemed to be within the simulated capture zones (for Case 2) less than one-third of the time. At maximum, the inclusion probability was less than 85% (this for the point to the right of the well in Case 2). Finally, a relatively large inclusion probability (maximum of approximately 14%) was noted for the source point located to the right of the pumping well at a distance nearly twice as far from the well as the edge of the capture zone boundary derived using the CFR method.

Regional-Gradient Results

As noted previously, two lines of source locations were investigated in the class of cases involving a non-zero regional gradient. The first line is parallel to the x-axis of the numerical grid and passes through the well. The second line is parallel to the y-axis of the grid and is upgradient from the well (Figure 2), halfway between the well and upstream boundary. For each realization, forward particle tracking was used from each of these locations to determine whether that location was within the 50-year capture zone.

As illustrated in Figure 4a, the inclusion probability decreases rapidly in the longitudinal direction for each of Cases 1 through 4. The rate of decrease in the inclusion probability near the well increases with increasing integral scale. While the homogeneous/isotropic capture zone (after a semianalytical solution from Cole 1995) extends to approximately $x = 1800$ m (or 1713 m from the well), the probability of inclusion for points significantly closer to the well (e.g., $x = 3000$ m, 513 m from the well, for Cases 2 through 4) are often less than 50%. The primary reason for these low inclusion probabilities is that heterogeneity near the well often leads to a deviation in the orientation of the capture zone in the immediate vicinity of the well, thus moving the capture zone off the line representing the mean regional gradient.

The transverse line of source locations provided observed inclusion probabilities that were approximately symmetric around the centroid of the homogeneous capture zone (Figure 4b). The width of the homogeneous/isotropic capture zone was slightly less than 80 m at this location (i.e., the capture zone extended 40 m above and below the centerline for the homogeneous capture zone). It is observed in Figure 4b that the real capture zones cover a significantly wider region, with inclusion probabilities greater than 10% extending to approximately 100 m away from the centerline and inclusion

probabilities greater than 1% extending 200 to 300 m away from the centerline. Further, the probability of inclusion of points located at distances greater than 100 m from the center line appears to increase with increasing integral scale.

Cases 1 through 4 each use a primary axis of the correlation structure parallel to the mean regional gradient. The impact of rotating this primary axis relative to the mean regional gradient is illustrated in Figure 5, which shows inclusion probabilities for Cases 2, 5, and 6. Figure 5a shows extremely rapid decline in the inclusion probability for points upgradient of the well when the primary axis of the correlation structure is rotated either 30° or 60° from the mean regional gradient direction. Within 850 m of the well (one-half of the longitudinal dimension of the homogeneous capture zone), the inclusion probability is less than 5%.

It would appear that the major effect of a rotation in the primary axis of correlation is a rotation in the orientation of the capture zone. Figure 5b shows the inclusion probabilities along the transverse line for Cases 2, 5, and 6. The maximum inclusion probability and general distribution of inclusion probabilities around the maximum does not vary dramatically for these three case studies. However, the orientation of the maximum inclusion probability does vary, increasing the deviation from the center line of the homogeneous capture zone ($y = 2513$ m) for increasing rotation of the correlation structure.

As indicated earlier, two general measures of capture zone geometry were considered. These were: (1) the maximum width of a capture zone within a given realization, and (2) the ratio of the largest individual width among a set of realizations to the width of the region required to enclose all capture zones generated for that set of realizations (termed the width ratio). Table 4 contains summary statistics appropriate to the discussion of these two measures. In this table, the mean, standard deviation and skew refer to the statistics of the maximum width of the capture zones from individual realizations within the case study.

It is noted in Table 4 that the maximum width of the capture zones is, on average, two to four times larger than the maximum width of the homogeneous capture zone (approximately 80 m). The mean maximum width also appears to increase with rotation of the primary axis of the underlying correlation structure (as shown by comparing Cases 2, 5, and 6). Further, these widths are highly variable, with coefficients of variation ranging from 0.15 to nearly 0.8, generally increasing with increasing integral scale or increasing rotation.

In reviewing the width ratios in Table 4, it becomes apparent that the maximum width from an individual capture zone explains only a relatively small portion of the total width required to enclose the entire group of simulated capture zones. The highest value (0.52) is for the case study with the smallest integral scale. Even in this case, however, only approximately 50% of the width required in the ensemble is contained in any individual realization.

Discussion and Conclusions

The results presented here provide direct evidence that use of either the CFR method for delineation of capture zones in the absence of a significant regional gradient or a homogeneous solution based on the best estimates of the geometric mean conductivity and the regional gradient are insufficient to describe the intricate capture zones that will impact the water withdrawn at a water supply well. Neither of these simple solutions adequately incorporate the uncertainty inherent in delineation of a capture zone.

Two approaches are suggested for increasing the reliability of capture zone delineation for the purpose of protecting a wellhead: (1) using the CFR or other simple solution with appropriate safety factors, or (2) identifying the appropriate information that would decrease the uncertainty of the capture zone location.

The latter approach becomes the only realistic opportunity for increasing reliability in situations where land-use practices lead to significant costs associated with increasing the area that must be protected in the vicinity of a wellhead. This situation might occur, for example, in a previously developed area where land-use patterns will need to be altered from commercial uses to residential or public uses. In this situation, several techniques are available for improving our predictive capability for a given aquifer. These include conditioning simulations on known information (e.g., measured hydraulic conductivities or hydraulic heads; Varljen and Shafer 1991 and Gorelick 1997) or the use of inverse modeling strategies (Vassolo et al. 1998; Carrera et al. 1997).

It must be recognized, however, that there is substantial cost in terms of preliminary data collection (e.g., additional boreholes and pump tests), hydrologic analysis, and continuing monitoring for this approach. Simply stated, there is a trade-off between accepting higher risks associated with uncertainty and the increased costs associated with obtaining more information. From the results previously presented and a review of the literature, it would appear that the pieces of information most critical for reducing uncertainty regarding capture zone geometry are: (1) the geometric mean K (assumed known for this study), (2) the regional gradient (orientation and magnitude), and (3) orientation of anisotropy in the correlation structure. The integral scale and variance of log K will impact this uncertainty to a lesser extent.

In many situations, particularly for small communities, the costs associated with placing additional zoning restrictions on a wellhead protection area may be small compared to the cost of a more detailed hydrologic study. In this case, placing conservative safety factors on simple solutions may provide for reliable protection of a wellhead.

As an example, we refer to the results for the zero-gradient case studies previously presented. The mean of the maximum distance of the capture zone boundary away from the well was approximately twice the radius estimated using the CFR method. In addition, water was withdrawn from within a circle with a radius nearly three times the CFR radius for approximately 95% of the realizations. In the case previously outlined, this would require protection of a circular region with a radius of approximately 820 m, compared to a homogeneous radius of approximately 270 m. In a sewered, residential area without local commercial enterprises such as gas stations or dry cleaning facilities, this increase in the protection zone might be a reasonable compromise to ensure the protection of an important water supply.

In the presence of a nonnegligible regional gradient, working solely with factors of safety becomes more problematic. Referring to Figure 5b, it is observed that in the presence of an unidentified, anisotropic correlation structure, there may be a significant probability of inclusion of source locations, which in the case discussed were as far as 500 m away from the centerline of the homogeneous capture zone (more than 10 times the expected deviation from the centerline based on the homogeneous capture zone). Further, these results are based on the assumption that the mean regional gradient is known and constant in time. As noted previously

by Cole and Silliman (1996), it is unlikely that the regional gradient will be known with certainty when based on limited field data sets. Finally, these results are based on relatively low variance in log- K and the width of the capture zones will likely increase with an increase in the variance (Cole 1995).

It is concluded from the work of Cole (1995) and the results presented here that if a factor of safety were to be placed on a homogeneous capture zone based solely on an estimate of the geometric mean K for the aquifer and a regional estimate of the hydraulic gradient, a factor of safety of approximately 30 would be required on the width of the capture zone (for a 99% probability of bounding the actual capture zone). The length of the capture zone in the direction of the mean hydraulic gradient would require a factor of safety on the order of three times the length predicted using the homogeneous capture zone.

The factor of 30 is a direct result of two mechanisms. First, uncertainty in the orientation and strength of the anisotropy in the hydraulic conductivity correlation leads to uncertainty of the capture zone orientation (e.g., Figure 5b). Without this uncertainty, the safety factor could be reduced to approximately 10. Second, limited knowledge of the local hydraulic conductivity distribution leads to uncertainty in the location and width of the capture zone. As shown in Table 4, the region required to enclose all the simulated capture zones within a given case study was at least twice as large as the largest individual capture zone width observed over all realizations for that case study. The width of this region was often eight to 15 times the width of the homogeneous capture zone.

Reduction in these safety factors can be derived from two places. The first involves a trade-off between factors of safety and risk. As noted previously (van Leeuwen et al. 1998; Cole and Silliman 1997; Bair et al. 1991; Varljen and Shafer 1991), it may be possible to place confidence bounds on the capture zone (e.g., a capture zone boundary that has a 10% or 50% probability of being violated). As shown in Cole and Silliman (1997), the width of these capture zones tends to increase rapidly as one decreases the level of acceptable risk. As a result, one can lower the safety factor required on a homogeneous capture zone by increasing the risk one is willing to accept that the capture zone will be inadequate to identify all regions contributing to the wellhead.

The second means to reduce these safety factors is, as noted previously, to increase the information available for the specific wellhead and surrounding region. As noted above, there are a number of conditioning and inverse techniques that have been developed for this purpose. Unfortunately, the number of data necessary for obtaining accurate spatial statistics can be in the thousands of boreholes (e.g., van Leeuwen et al. 1999). Further, one of the most critical parameters to determine to reduce uncertainty in the capture zone in the presence of a significant regional gradient is one of the most difficult to quantify: the correlation anisotropy of the hydraulic conductivity (both magnitude and orientation). Substantial additional effort is required to more fully address identifying and quantifying this source of uncertainty.

The results presented herein suggest that higher costs and additional effort are necessary for obtaining greater reliability associated with delineation of wellhead capture zones. These costs may take the form of additional data collection for application of classic conditioning or inverse modeling, or as suggested here by recognizing uncertainty through establishing larger capture zones via factors of safety. Additional research is required to more fully establish the trade-off between factors of safety, data availability,

and risk. Further, it is suggested that choice of the technique for reduction of risk should be placed within an economic framework including costs associated with land acquisition, rezoning, data collection, development of alternative water supplies, and remediation of contamination events.

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Comparison of Observations from a Laboratory Model with Stochastic Theory: Initial Analysis of Hydraulic and Tracer Experiments

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Abstract. Hydraulic and tracer tests were conducted in a flow cell containing a mixture of sediments designed to mimic a two-dimensional, log-normally distributed, second-order stationary, exponentially correlated random conductivity field. With 60 integral scales in the direction of mean flow and 25 integral scales perpendicular to this direction, behavior of flow and transport in the interior of the flow cell can be compared directly with stochastic solutions for flow and transport. Using 144 piezometers and 361 platinum electrodes, the distribution of hydraulic head and the concentrations of an ionic tracer could be monitored in substantial detail. The present discussion presents the details of the experimental equipment. Results and initial analysis of hydraulic measurements and characterization of a two-dimensional tracer plume are also presented. Analysis using first-order hydraulic theory shows that the flow through the medium was consistent with an effective conductivity equal to the geometric mean of the conductivity distribution. Further, the semivariogram of head increments as observed in the experimental results was consistent with the semivariogram predicted by theory. The chemical transport experiments are here compared with the early solutions presented by Dagan (1984, 1987). The observed rate of longitudinal spread of two tracer plumes was slightly less than that predicted using this theory. Further, the spread in the transverse dimension was observed to decline from the initial plume dimensions and then remain constant or increase slightly, but at a rate lower than predicted by the theory. The difference between the hydraulic and transport results is believed to be related to the fact that the hydraulic results were averaged over a very large portion of the flow cell such that ergodic conditions could be assumed. In contrast, the initial geometry of the plume covered only approximately five integral scales in the transverse direction such that the validity of the assumption of ergodic conditions must be questioned in the analysis of results for the chemical transport.

Key words: groundwater, transport, stochastic theory, experiments.

1. Introduction

Stochastic models of groundwater flow and chemical transport hold promise to be valuable tools to assist in probabilistic and deterministic prediction of flow and transport processes in heterogeneous porous media. As early as the Monte Carlo studies of Warren and Price (1965) and Freeze (1975), characterization of

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the hydraulic conductivity as a spatial random variable has been used as the basis for investigation and prediction of the distribution of hydraulic head, the transport of chemicals, and the uncertainties inherent in the prediction of these processes.

In terms of prediction of flow processes, early work by Freeze (1975) illustrated the variability in hydraulic head that can result from spatial variation in the hydraulic conductivity. This early study led to a number of additional papers including the work of Smith and Schwartz (1979) incorporating spatial correlation into two-dimensional conductivity fields, and the work by Bakr *et al.* (1981) extending into three-dimensions. Study of hydraulics in heterogeneous fields also led to the identification of effective hydraulic parameters in two- and three-dimensional flow fields subject to mean uniform flow (see, for example, the reviews of this work contained in Gelhar (1989) and Kitanidis (1997)). This work has also been extended to theoretical, numerical and experimental studies of effective parameters in the presence of active pumping from wells (e.g., Naff, 1991; Neuman and Orr, 1993; Indelman and Abromowitz, 1994; Silliman and Caswell, 1998).

The recognition of the link between the distribution of the hydraulic conductivity (or transmissivity) and the distribution of hydraulic head has also led to the development of methods for estimating the distribution of the hydraulic conductivity (or transmissivity) from head data. Calvete (1997), for example, reviewed the inverse method as applied to heterogeneous media. More recently, Zheng (1998) and Zheng and Silliman (2000) have presented a first-order methodology whereby the semivariogram of increments in the hydraulic head can be used to estimate the integral scale and variance of the transmissivity field.

Substantial progress has also been realized in terms of transport of conservative chemicals within saturated media. Much of this work is reviewed in Rubin (1997), thus the details will not be repeated herein. Among the significant early results from this literature was the development of expressions for the spatial variation and asymptotic limits for the dispersivity and discussion of conditions under which transport will approach behavior which can be modeled appropriately using the Fickian assumption. Much of this work has been focused on ergodic conditions, with somewhat less attention being paid to nonergodic conditions (e.g., Kitanidis, 1988; Dagan, 1990; Dagan, 1991; Zhang *et al.*, 1996; Zhang, 1997). Early theoretical efforts covering ergodic conditions have been the subject of a number of field investigations, including the Borden and Cape Cod sites (e.g., Freyberg, 1986; Garabedian *et al.*, 1991). Results from the Borden site indicated relatively close agreement with theoretical expressions for the growth of the second spatial moments of the tracer plume in both the longitudinal and transverse directions. The results from the Cape Cod site showed convergence to asymptotic behavior with reasonable dispersivities in the longitudinal and horizontal transverse directions and a very small transverse dispersivity in the vertical direction. In contrast, observations collected at the MADE site (e.g., Adams and Gelhar, 1992) provided field evidence of situations in which the ergodic theory was likely to be insufficient to describe the evolution of the first and second order moments of a tracer plume.

With respect to these latter results, Berkowitz and Scher (1998) and Berkowitz *et al.* (2000) have presented a non-ergodic, continuous time random walk model which provides predictions consistent with the observed variation in the spatial moments of the tracer plume at the MADE site.

A significant number of controlled laboratory experiments have been conducted on both flow and the transport of conservative chemicals in homogeneous and heterogeneous porous media (see, for example, the review in Silliman *et al.* (1998)). However, relatively few controlled laboratory efforts have appeared in the literature in which results predicted utilizing stochastic theory have been compared directly against laboratory results. Welty and Elsner (1997) report on a series of one-dimensional experiments in which a theory for the dispersive spread of a chemical plume under non-uniform density (Welty and Gelhar, 1982) was compared directly against observed transport in a one-dimensional laboratory column packed to reproduce a correlated conductivity field. The agreement was qualitatively quite reasonable. These authors also discussed observations of head variance within these one-dimensional columns and showed consistency with first-order hydraulic theory.

More recently, Silliman and Caswell (1998) report on the distribution of hydraulic head (and, through pump test analysis, the distribution of effective transmissivities) in the presence of a pumping well within a laboratory flow cell. This latter work was based on a pair of two-dimensional conductivity fields consisting of bimodal distributions of transmissivity. One conductivity field was designed to reproduce a second-order stationary random field and another was designed to reproduce a simple scaled field. In the case of the second-order stationary random field, these authors found good qualitative and quantitative agreement with published stochastic theories. When the conductivity field was designed to deviate from the assumptions of the published theory (e.g., the scaled medium), the results were observed to deviate from the predictions generated with these theories.

The present manuscript describes the experimental conditions, results, and initial analysis of a series of laboratory experiments that are to be used to examine hydraulic and transport theories as applied to heterogeneous porous media. The hydraulic results are compared with results by Zheng and Silliman (2000), Zheng (1998), and Gelhar and Axness (1983). The transport results are compared with predictions obtained from early stochastic theories describing the first and second moments of a plume of conservative tracer (e.g., Dagan, 1984, 1987; Freyberg, 1986; Sudicky, 1986). In both cases, the laboratory results allow direct comparison of predictions obtained from stochastic theory with observations obtained under controlled experimental conditions in which the underlying hydraulic distribution is well characterized (i.e., known within the limits of our ability to pack the experimental apparatus). It is recognized that the analysis to be presented, particularly in the area of transport, is preliminary with more sophisticated models likely providing more insight into the observed behaviors. This simplified analysis is accepted for the present manuscript as the goal here is to present the experimental design and

initial results and because a second series of laboratory experiments on a second realization of the heterogeneous medium is currently being completed. More advanced analysis of these results, and the second series of results, will be presented following completion of the new experiments.

2. The Experimental Apparatus

The experiments were conducted in a laboratory flow cell with dimensions for the sediment compartment of $1.6 \text{ m} \times 0.67 \text{ m} \times 0.094 \text{ m}$ (Figure 1). The apparatus was designed to allow flow to be established with a mean gradient parallel to the 1.6-m axis of the cell. The upper and lower borders of the flow cell were maintained as no flow boundaries. Three reservoirs were utilized on the inflow end of the flow cell. For hydraulic tests, these three reservoirs were hydraulically connected. For tracer tests, the central reservoir was hydraulically isolated for short periods of time to allow introduction of a tracer plume.

Hydraulic tests were conducted by injecting water at a known volumetric flux into the three, hydraulically-connected inflow reservoirs. The outflow reservoir was maintained at a constant hydraulic head. The distribution of head within the porous medium was measured through 144 piezometers installed through the wall of the flow cell and distributed as shown in Figure 2(a). The heads in the piezometers were measured one at a time through a series of valves on a manifold. Hydraulic heads were measured with both a pressure transducer and through visual measurement in a single manometer tube (one piezometer connected to the tube at a time). Due to electronic problems, the results from the pressure transducer were considered unreliable. Hence, only the results obtained using the manometer tube are analyzed

Figure 1. Schematic diagram of the flow cell used in these experiments. On inflow (left) end, the heights of the reservoirs are, from bottom to top, 0.29, 0.08, and 0.30 m.

Figure 2. Internal instrumentation within flow cell: (a) 144 piezometers, and (b) 381 platinum electrodes.

below. Error in the measurement of the hydraulic heads was estimated, based on our experience with using these manometer boards, to have a standard deviation of approximately 0.05 cm. As discussed below, this estimate of error is consistent with the results obtained.

The distribution of piezometers was based on three considerations. First, as has been discussed in the literature (e.g., Rubin and Dagan, 1989), it is necessary to avoid measurements within a few integral scales of all no flow boundary conditions (so as to allow comparison of the experimental results with those derived theoretically for infinite systems). So as to be conservative, five integral scales were maintained from both of these boundaries (in the numerical work of Rubin and Dagan (1989) it was concluded that three integral scales was likely sufficient to avoid boundary effects). Second, it was considered necessary to avoid measurement in the immediate vicinity of constant head boundary conditions (Rubin and Dagan, 1988). Hence, once again, a minimum distance from the inflow and outflow boundaries of five integral scales was chosen as a conservative minimum distance. Third, based on the work of Zheng (1998), it was concluded that the first-order theory to be applied was optimal when the piezometers were spaced at distances

of approximately one integral scale in the horizontal direction and at distances of at least four integral scales in the vertical direction. This led to distributing the piezometers in four horizontal lines, separated vertically by four integral scales and separated horizontally by a minimum distance of one integral scale.

The tracer tests discussed in this manuscript consisted of introducing a pulse of tracer through the central inflow reservoir. This pulse was introduced by first isolating the central reservoir (hydraulically) from the upper and lower reservoirs. This reservoir was then flushed, in the absence of flow through the flow cell, with tracer fluid. Fluid was then injected through all three reservoirs (the upper and lower reservoirs were hydraulically connected and driven by one pump while the central reservoir remained hydraulically isolated and was driven by a second pump), producing a finite volume of tracer (with an approximately rectangular shape) into the flow cell. Flow was once again terminated and the central reservoir was flushed with non-tracer fluid. The three reservoirs were then hydraulically connected and flow was established (using a single pump for all three reservoirs) to advance the tracer plume.

The movement of the tracer plume was monitored *in situ* through use of 381 electrodes (Figure 2(b)). Each electrode consisted of a twisted pair of insulated platinum wires with a diameter estimated at approximately 0.1 mm and was installed through the side wall of the flow cell. The tips of each pair of wires were clipped to expose the platinum to the fluid within the cell. The average separation between the tips of the pair of wires comprising a single electrode was substantially less than 1 mm. Hence, the measurements obtained were at a scale similar to the grain scale. The response at an electrode was recorded by passing an alternating current simultaneously through the electrode and a reference resistor, thus providing a measure of the voltage drop across the electrode. Each electrode was individually calibrated *in situ* by passing various concentrations of sodium chloride (used as tracer) through the flow cell. The resulting calibration curves showed a strong log-linear relationship between concentration and voltage drop across the electrode with minimal drift over the time periods used to complete these experiments. Within the range of concentrations used in these experiments (100–1000 mg/l NaCl), it was estimated that the error in readings from any given electrode was less than approximately 5 mg/l.

Due to the fact that the tracer was introduced through the central reservoir, the sampling for the tracer was concentrated in the central portion of the flow cell. The distribution of the electrodes is shown in Figure 2(b) (with the inflow reservoirs located at the left on this image). Near the inflow reservoir, the spacing of the electrodes parallel to the mean direction of the gradient was set equal to 0.5 times the anticipated integral scale of the conductivity field with spacing perpendicular to this direction set equal to 1.0 times the anticipated integral scale. Corresponding to the anticipated increase in the longitudinal dimension of the plume, the horizontal spacing of the electrodes was varied, increasing to a maximum of 2.0 integral scales at distance from the inflow reservoir. The distribution of electrodes represents a

compromise between providing a dense sampling array with which to sample the movement of the tracer plume and the limits of our ability to install instruments through the wall of the flow cell. The limit was a function of a number of factors including the strength of the plexiglass used in constructing the flow cell, the effort involved in installing these electrodes without leaks, and our ability to sample these electrodes with reasonable sampling times.

3. The Porous Medium

Two properties were of primary interest in choosing the distribution of the sands for this experiment. The first was in establishing a realization of a log-normal, exponentially-correlated, second-order stationary, random hydraulic conductivity field. This choice was based on tying conditions within our experiments to conditions assumed in the stochastic theory used in interpreting the experimental results. The second was in establishing a correlation between the hydraulic conductivity and the sorptive capacity of the medium. The latter property was important for particle and microbial transport experiments to be reported on elsewhere.

In order to produce these fields, two field sands were obtained with various grain size distributions. The first consists of Accumin sands, a commercially available sand used in our prior experimental efforts (see, for example, Silliman and Caswell, 1998). These sands are well rounded quartz sands with minimal surface coatings. The second consists of sands excavated from a research site in Oyster, Virginia. These sands include a far wider grain size distribution than the Accumin sands and are heavily coated with iron oxides. Both sands were sieved into various sizes to produce varying hydraulic conductivities. Ten final mixtures of sands were chosen for use in the flow cell. The source and hydraulic conductivity (determined in separate falling head permeameter tests on consolidated samples of sand with variation among repetitions of less than 5%) of each sand are provided in Table I.

In order to pack these sands in the flow cell, a random field with exponential correlation was simulated on the computer using the Turning Bands field generator (Tompson *et al.*, 1989). The random field generated had an isotropic integral scale of approximately 2.75 cm and a variance of $\ln K$ of slightly less than 0.5. The final distribution generated on the computer is shown in Figure 3(a). The actual distribution of sand within the flow cell, as viewed through the front surface of the flow cell, is shown in Figure 3(b). This image shows a slight blurring of the contact between regions of different grain size that is related to minor problems involved in packing within 1 mm of the wall of the flow cell. Figure 3(c), a picture taken during destructive sampling within the flow cell, shows the sharpness of the contact between regions with different grain sizes approximately 5 cm away from both walls of the flow cell (i.e., along the centerline of the flow cell). Table I provides the percentage of each sand present within the flow cell as predicted using the Turning Bands generator and as estimated from measurements of the volumes of each sand entering the flow cell.

Table 1. Sands used in packing the flow cell

Sand classification	Source of sand	Hydraulic Conductivity (cm/s)	Simulated percent of porous medium ^a	Estimate of actual per cent of medium ^b
1	Oyster	1.61	0.34	0.05
2	Oyster	0.58	1.91	0.9
3	Accumin	0.49	5.79	4.7
4	Accumin	0.224	13.78	11.0
5	Accumin	0.15	21.97	23.0
6	Accumin	0.11	24.41	26.0
7	Accumin	0.078	18.35	22.0
8	Oyster	0.041	8.98	7.8
9	Oyster	0.030	3.24	3.9
10	Oyster	0.015	1.22	0.6

^aPer cent of porous medium comprised of particular sand type as produced by random field generator on computer.

^bPercent of porous medium comprised of particular sand based on measured volumes used in filling the flow cell.

(a)

Figure 3. (a) Computer generated distribution of hydraulic conductivity subdivided into 10 classes. Gray scale based on $\ln(K)$. (b) Final distribution of sediments within the flow cell as seen through the front wall. (c) Final distribution of sediments within the flow cell as observed during the process of unpacking the flow cell. Darkest sands represent the lowest conductivities.

Figure 3. (Continued)

4. The Hydraulic Tests

4.1. PROCEDURE

The hydraulic test was run by injecting fluid into the inflow reservoirs (hydraulically connected) and holding the outflow reservoir at constant head. Flow rate was measured at the discharge and the system was allowed substantial time to come to steady state.

Using a network of valves, each of the 144 piezometers was individually opened to allow the water level in that piezometer to come to steady state; requiring between 2 and 10 min depending on the piezometer and sand grain size in the flow cell surrounding the tip of the piezometer. A number of the piezometers were inadvertently plugged by the sand grains either during packing or during the initial saturation of the piezometer tubes and were unresponsive. Measurements could not be collected from these piezometers. Of the 144 possible water level measurements, therefore, reliable measurements were collected from 114. A measurement was considered reliable if the recorded water level at the piezometer repeatedly returned to the same level following small perturbations in the water level (created by repeatedly squeezing/release of the manometer tube).

The head measurements were utilized to estimate both the effective hydraulic conductivity within the flow cell and the semivariogram of a parameter termed the head increment. The head increment, $I(x + \Delta r)$, is defined as

$$I(x + \Delta r) = H(x) - H(x + \Delta r), \quad (1)$$

where $H(x)$ is the head measured at position x and $H(x + \Delta r)$ is the head measured at position $(x + \Delta r)$. Zheng (1998) and Zheng and Silliman (2000) show that the semivariogram of the head increment, under mean uniform flow, is closely related to the variance and integral scale of the underlying conductivity distribution. Specifically, the semivariogram of the head increment, γ_I , in the presence of a log-normally distributed, exponentially correlated conductivity distribution is given by

$$\begin{aligned} \gamma_I(r, \theta, \Delta r) = & 2\gamma_h(\Delta r, \theta) + 2\gamma_h(r, \theta) - \gamma_h(|r - \Delta r|, \theta) - \\ & - \gamma_h(r + \Delta r, \theta) + 2\sigma_e^2, \end{aligned} \quad (2)$$

where γ_h is the semivariogram of the head residual ($h(x) = H(x) - \langle H(x) \rangle$; Dagan, 1985), r is the separation between two measurements of the head increment and is here taken as being parallel to Δr , θ is the orientation of r relative to the mean gradient, σ_e^2 is the variance of the measurement error.

4.2. RESULTS

Using an r oriented parallel to the mean gradient, a magnitude equal to 11.0 cm (four times the minimum piezometer spacing), and a range of possible standard deviations in the measurement error of 0.03–0.06 cm, the theoretical semivariogram

Figure 4. Sample ('o') and theoretical semivariograms for the head increments. The theoretical curves shown are for an assumed standard deviation in measurement error of 0.06 cm (upper curve) and 0.03 cm (lower curve).

of the head increment parallel to the mean gradient was determined. Figure 4 shows the theoretical semivariograms of the head increments for both the largest (0.06 cm) and smallest (0.03 cm) measurement errors considered. This figure also shows the sample semivariogram determined from measurements collected in the flow cell. It is noted that this figure was generated without calibration as all parameters required in (2) with the exception of the measurement error were known *a priori* (the integral scale and variance of $\ln K$ generated on the computer and r selected from the geometry of the piezometer locations).

The agreement between the theoretical curves and the experimental results shown in Figure 4 is considered quite strong as the shape and vertical displacement of the theoretical curve is quite sensitive to both the variance and the integral scale of the underlying conductivity field. This is illustrated in Figures 5(a) and (b). Figure 5(a) shows the theoretical curves using a measurement error of 0.05 cm, the correct variance, and assumed values of the integral scale of 0.275, 2.75 and 27.5 cm. These values for the integral scale represent an order of magnitude smaller than the true integral scale, equal to the true integral scale, and an order of magnitude larger than the true integral scale. Figure 5(b) shows the theoretical curves using a measurement error of 0.05 cm, the correct integral scale, and assumed variances of 0.1, 0.5 and 2.5. These values for the variance represent 1/5th the true value, the true value, and 5 times the true value. Based on the results shown in Figures 4, 5(a) and 5(b), it is concluded that the theory is appropriately applied to this random

Figure 5. Bounds on fit between theory and observations with an assumed measurement error of 0.05 cm: (a) Curves with design variance in the conductivity, but using integral scales of 27.5 cm (a), 2.75 cm (b), and 0.275 cm (c). (b) Curves with design integral scale, but using variances of the $\ln(\text{conductivity})$ of 2.5 (a), 0.5 (b), and 0.1 (c).

medium and that the experimental procedures were sufficiently rigorous to provide a reliable semivariogram of the head increment.

The data were also used to estimate the effective hydraulic conductivity via measurement of the volumetric flux through this medium with estimation of the large-scale mean hydraulic gradient based on measurements from the piezometers. From the piezometer data, it was estimated that the mean gradient was approximately 0.033 when the flow rate was 145 cc/min. Based upon this gradient and flux, in combination with the cross sectional area of (67×9.4 cm), the effective conductivity of this medium can be estimated to be 0.116 cm/s. However, given the number of uncertainties involved in making this estimate (the mean gradient, the exact width of the tanks, the measured flow rate), it is considered more appropriate to report the measured conductivity as being in the range of 0.11–0.12 cm/s. The effective conductivity that is predicted from the geometric mean of the conductivity field in the flow cell is 0.112 cm/s. Given the experimental errors inherent in this type of experiment, this agreement is considered quite reasonable. The agreement with the geometric mean also contrasts sharply with comparison to the calculated values for the arithmetic (0.139 cm/s) or harmonic (0.092 cm/s) means of this conductivity field.

5. Tracer Tests

5.1. PROCEDURES

Two tracer tests were performed, the first with a mean velocity approximately 22 times slower than the second. In both cases, a two-dimensional plume of sodium chloride was injected through the central reservoir on the inflow end of the flow cell. The plume was allowed to travel down gradient until the entire plume was captured within the electrode array. At this point, the distribution of concentration within the plume was measured and this distribution was used as the initial condition for the tracer experiment. The subsequent motion of the plume through the flow cell was then monitored.

Figures 6(a)–(d) show the motion of the tracer plume at several measurement times during the slower of the two tracer experiments. The temporal development of the plumes from the two experiments were characterized in terms of (i) the first spatial moment of the plume (the center of mass) in the direction of the mean gradient (herein termed the x direction), (ii) the first spatial moment of the plume in the direction parallel to the 0.67 m dimension of the flow cell (herein termed the y direction), and (iii) the second centered, spatial moments of the plume in both the x and y directions.

As noted earlier, concentrations within the flow cell were monitored through the 381 electrodes (calibrated *in situ*). These measurements required two levels of processing in order to allow determination of moments. The first level of processing involved the identification of anomalous measurements. This identification involved locating electrodes that were not functioning properly, identifying

Figure 6. Movement of the tracer plume during the experiment with the lower velocity at times (after the time recorded for the initial plume) equal to: (a) 500 min, (b) 1440 min, (c) 2580 min, and (d) 3270 min. Outer dashed contour is for $C/C_0 = 0.01$. Solid contour is for $C/C_0 = 0.1$. Inner dashed contour, where present, is for $C/C_0 = 0.2$.

electrical noise which was clearly electrical error not related to the tracer plume, and determining the minimum concentration which could be considered reliable. Identifying electrodes that were not functioning properly was straight forward as these electrodes produced signals consistent either with a broken wire or an electrical short within the electrode. The second level of processing involved estimation of spatial moments from discrete spatial measurements and is discussed further below.

Of the 381 electrodes installed within the flow cell, 21 failed to produce readings. In terms of electrical noise, the fact that some electrodes were never impacted by the tracer plume allowed for identification of and correction for slight drift in the electrical readings. As one of the experiments discussed below was conducted over a period of approximately 4 days, there was a slight drift in the electronics during this experiment that was attributed to change in temperature of the reference resistor. This drift was identified through analysis of readings at electrodes not impacted by the plume and was then corrected for at all electrodes prior to analysis of the data. In addition, the contour of the final tracer concentrations led to identification of small (normalized concentration less than 0.01) patches of measured 'concentration' at random electrodes at distance from the location of the plume. Those patches which were clearly unrelated to the movement of the plume (e.g., the reading at the electrode was inconsistent from one time to the next) were eliminated prior to estimation of the spatial statistics of the plume.

The estimated reliability of the electrodes (5 mg/l), combined with the source strengths of the tracers used (500–900 mg/l), led to the conclusion that all normalized concentrations greater than 0.01 of the source strength would be considered reliably different than zero. This cutoff at normalized concentration of 0.01 also appeared consistent with the noise present within the final data sets (e.g., the background concentration within the cell appeared to have noise ranging in the normalized concentrations from slightly less than 0.0 to as high as 0.01).

Once the reliability of the data was established and the final data set identified, the second challenge was to estimate spatial moments from data collected at discrete points in space. These moments were estimated using linear interpolation within the regions enclosed by each quadruplet of sample locations, following the discussion of Garabedian *et al.* (1991). For locations where an electrode had failed, the concentration at that electrode was first estimated via linear interpolation from the four surrounding electrodes, and the moment estimation was then based on these interpolated values.

These moments, determined from the tracer test data, were then compared with the predicted variation in these moments. In terms of the first moments, the predicted variation according to first-order stochastic theory is simply a linear increase with time of the first moment parallel to the mean gradient and a constant value for the moment perpendicular to the mean gradient. For the present manuscript, the second moments are based on the solution originally developed by Dagan (1984, 1987) and presented by several authors (e.g., Freyberg, 1986; Sudicky, 1986; Sposito and Barry, 1987; Barry *et al.*, 1988) for the spread of a plume in two dimensions under ergodic conditions. It is noted that the analysis presented herein must be considered preliminary as it assumes that the initial size of the plume in the transverse direction is large as compared to the integral scale (see, for example, the discussion of ergodicity in Dagan, 1990). Further, it does not consider the impact of the original shape of the tracer plume. It is noted, for the tracer plumes realized in these experiments, that the initial transverse dimension of the plumes was on the order of five integral scales. This transverse dimension decreased with flow distance, reducing to a minimum of approximately 2.5–3.5 integral scales for some of the measurements. Additional analysis of these data, incorporating the initial geometry of the plume, the non-ergodic nature of the plume, and consideration of the variance of the predicted versus the observed concentrations, will be presented in a later manuscript.

The expressions utilized for the longitudinal (σ_{xx}^2) and transverse (σ_{yy}^2) variances are therefore given as (Freyberg, 1986; Sudicky, 1986)

$$\sigma_{xx}^2 = 0.74(2\sigma_y^2\lambda^2) \left\{ 0.75 - 1.5E + \frac{L}{\lambda} + 1.5 \left[\text{Ei} \left(-\frac{L}{\lambda} \right) - \ln \left(\frac{L}{\lambda} \right) + \frac{\lambda}{L} \times \right. \right. \\ \left. \left. \times \exp \left(-\frac{L}{\lambda} \right) \left(\frac{1+\lambda}{L} \right) - \frac{\lambda^2}{L^2} \right] \right\},$$

$$\sigma_{yy}^2 = 0.74(2\sigma_Y^2\lambda^2) \left\{ 1.5 \left[\frac{\lambda^2}{L^2} - \frac{\lambda}{L} \exp\left(-\frac{L}{\lambda}\right) \left(\frac{1+\lambda}{L}\right) \right] - 0.5 \left[\text{Ei}\left(-\frac{L}{\lambda}\right) - \ln\left(\frac{L}{\lambda}\right) \right] - 0.75 + 0.5E \right\}$$

where $L = \text{Velocity} \times \text{time} = \text{displacement of the center of mass}$, $\lambda = \text{the integral scale of the medium}$, Ei is the exponential integral and $E = \text{Euler's constant}$. It is noted that all parameters within these equations are known *a priori* such that no calibration occurs in comparing these equations to the moments determined from the tracer test data. It is also noted that the factor, 0.74, was developed by Dagan (1987) as a means of correcting for vertical averaging of the conductivity correlation structure as an analysis is reduced from three-dimensions to two-dimensions. The magnitude of this parameter is an artifact of Dagan (1987) assuming that the vertical depth of averaging was equal to one vertical integral scale. In our situation, the covariance for the two-dimensional analysis is predetermined (assuming minimal variation in conductivity in the third direction). Hence, an argument can be made that the 0.74 should be replaced with unity. As noted below, this modification of the equation would result in further deviation between the theory and the experimental results. The theoretical results shown below are therefore based on the use of 0.74 with the recognition that a larger coefficient would lead to an increase in the estimates of both variances.

5.2. RESULTS

The two transport experiments were run with mean velocities estimated at approximately 0.015 cm/min and 0.33 cm/min. The total mass injected in the two experiments was estimated, based on the plume concentrations and geometry, to be approximately the same. Due to the manner in which the plumes were injected into the flow cell, absolute mass balances on the tracer could not be calculated (the mass of tracer could only be estimated from the measured plume concentrations). It is noted that there was significant variation in this mass (up to $\pm \sim 15\%$ of the mean estimated mass) for different measurement sets (different times) in both experiments. This variation in mass is attributed to the errors introduced by interpolating mass between measurement points in calculating the moments and measurement error (particularly at low concentrations). For further discussion of the impact of interpolation on moments, the reader is referred to Garabedian *et al.* (1991).

Figures 7(a) and 7(b) show the first moments in the x and y directions (parallel and perpendicular to the mean direction, respectively) for the two experiments performed. To allow direct comparison of these two results, the time axis for the experiment run at the higher velocity has been adjusted (multiplied) by the ratio of the volumetric fluxes used in these two experiments ($= 22.0$). As expected, the center of mass moved in the x direction approximately linearly with time (with

Figure 7. Variation of the first spatial moment with time for the longitudinal, or x , direction (upper plot) and the transverse, or y , direction (lower plot). The slow experiment is represented by 'o's and the fast experiment is represented by '*'s.

some slight variations around this mean behavior) and the center of mass in the y direction approached an approximately constant elevation after an initial period of adjustment. The fact that the two plumes were initially located at slightly different locations within the flow field is indicated by the difference in the elevation (y coordinate) of the center of mass throughout the experiment. It is interesting to note that despite the initial difference in the elevation of the center of mass, the center of mass for the two plumes converged to very similar elevations within the porous medium. It was not surprising to note that these latter elevations coincided with a region of high hydraulic conductivity.

The second spatial moments in the x and y directions are shown in Figures 8(a) and 8(b). These plots show the value of the second moments σ_{xx}^2 and σ_{yy}^2 as functions of the dimensionless quantity D/λ (dimensionless distance) such that results from the two experiments can be directly compared. For the spatial moments in the x direction, the initial value of the sample variance of each experiment has been subtracted from the calculated variances for all later measurements. Thus these results are directly comparable with the theoretical values for σ_{xx}^2 . As the second sample moments in the y direction showed an initial decline in magnitude, no correction for the initial size of the plume was pursued.

It will be noted that both experiments provide very similar behavior in terms of both the longitudinal and transverse spread with the exception of the last data points. While the deviation in the last data points is somewhat troubling, it is noted

Figure 8. Variation of the second spatial moments as functions of normalized distance of travel of the center of mass. In these plots, the symbol 'o' represents the slow experiment while the symbol '*' represents the fast experiment. The dashed line represents the theoretical prediction. The longitudinal results are shown in (a) and the transverse results are shown in (b).

that these data points are extremely sensitive to the 'cut-off' value used in analyzing the data. As noted earlier in this manuscript, it was assumed that all concentrations below $C/C_0 = 0.01$ were considered unreliable and were ignored (based on our analysis of the reliability of the equipment). In Figure 9, the development of the longitudinal second moment for the slower experiment is shown for cut-off values of $C/C_0 = 0.02, 0.01$ and 0.005 . This figure shows that while the majority of

Figure 9. Sensitivity of calculated second moment to assumed cutoff value. This plot shows calculated second spatial moment in the longitudinal direction for the slow experiment assuming cutoffs of $C/C_0 = 0.005$ ('x'), 0.01 ('o'), and 0.02 (*).

the data points are relatively robust to the choice of cutoff, the last data point is extremely sensitive to this value (the plume is relatively disperse at this point and contains large regions of low concentration). Hence, we place relatively low reliability in the last data points for both experiments (see discussion for further comment on this cutoff).

The results for the longitudinal variance (Figure 8(a)) lead to two observations. First, the rate of increase in longitudinal spread is quite similar for the two experiments. This indicates that the dominant mechanism leading to the spread of the plume is likely the same in both experiments. If any difference exists, it would appear that the experiment run at the higher velocity is showing a slightly lower rate of spread. It is noted that, at the pore scale and using the mean velocity for the flow field, the Peclet number ($ud/D_m = \text{local velocity} * \text{mean grain diameter} / \text{Coefficient of molecular diffusion}$) was on the order of 100 for the experiment run at the lower mean velocity. Hence, it is concluded that local diffusion had only minimal impact on transport in these two experiments. The resulting similarity in the longitudinal spread is therefore to be expected as mechanical dispersion is anticipated to be the dominant dispersive mechanism for both experiments.

The second observation to be derived from these results is the fact that the observed longitudinal variance of the plume, while of the correct magnitude over these travel distances, is increasing at a rate lower than the rate predicted using the solution presented above. Further, as noted above, the coefficient (0.74) used in this expression is likely leading to an underestimate in the predicted rate of spread such that the difference between theory and observation is likely larger than that shown in the figure. We anticipate that there are at least two reasons for

this discrepancy. First, the solution used does not explicitly account for the initial dimension of the plume. A number of authors, including Sposito and Dagan (1994) and Fiori (this issue) have discussed the impact of initial geometry of the plume, and the correlation between the initial plume and the velocity field, on the final stochastic description of the transport process. Second, the initial concentration field for each of the two experiments covered only approximately five integral scales in the transverse dimension and, at the first measurement locations, was likely correlated with the velocity field. Hence, there is substantial reason to question whether the initial transverse dimensions of the plumes were sufficiently large and the concentration and velocity fields sufficiently independent to support the assumption of ergodicity (see discussion, for example, in Dagan (1990) and Sposito and Dagan (1994)). Under these circumstances, there is interest in reassessing these results from the standpoint of nonergodic formulations (e.g., Zhang, 1997; Zhang *et al.*, 1996; Dagan, 1991). New experiments are currently being performed using concentration fields with larger initial transverse dimension and lower correlation with the velocity field. However, these extensions are beyond the scope of the present manuscript.

The results for the transverse dispersion (Figure 8(b)) indicate that there is minimal spreading in the transverse direction, with some reduction in spread from the initial plume to the plume observed in the central region of the flow field. From the data shown in Figure 8(b), it is apparent that σ_{yy}^2 is not increasing as fast as predicted by the theory presented above. Further, the results do not rule out the possibility of a zero (or possibly negative) rate of spread in the transverse direction as suggested in the early work of Dagan (1984).

These results lend support to the arguments made by other authors (e.g., Garabedian *et al.*, 1991) that field observations of transverse dispersivities which are larger than predicted by theory may be related to the impact of transport mechanisms that were not included in the formulation of the theory. Further, these results provide additional impetus to consider the impact of the finite size of the initial plumes and the correlation of the initial concentration field with the velocity field on the observed second moments. In particular, both plumes were significantly impacted by a large region of low permeability sands near the point of initial measurement. Had the initial transverse dimension of the plume been significantly larger (e.g., 10–20 integral scales), the impact on this local low permeability zone would have played a much smaller role in determining the sample variance in the transverse dimension. As a result, there is suspicion that the transverse behavior of this plume may be specific to this combination of realization of the conductivity field and location of the injection point. Further discussion of transverse dispersion is, therefore, delayed until completion of a second set of experiments (currently being performed) in a second realization of this conductivity field.

6. Discussion

These results represent, to our knowledge, one of the few attempts to directly compare stochastic theory with laboratory experiments. In this case, the medium was packed to mimic a log-normally distributed, second-order stationary random field with an exponential correlation structure. As such, the medium reproduced the conductivity structure included in several previously published stochastic theories, including those by Zheng (1998) and Dagan (1984).

Results from these experiments were consistent with the predicted effective hydraulic conductivity and showed excellent agreement with the solution for the semivariogram of head increments. It was observed in the tracer tests, however, that the experimental results showed great sensitivity (at late time) of the calculated second moments to the choice of the concentration cut-off. Further, significant deviations were observed in comparison of the transport theory considered here and the results obtained from the transport experiments. In particular, while the theory reproduced the correct order-of-magnitude of the variance for the plume, it over predicted the rate of increase of the longitudinal second moment. The theory also predicted an increase in the transverse second moment when the experimental results indicated that the moment was constant, or decreasing, over much of the flow domain.

The sensitivity of the estimate of spatial variance to the assumed cut-off for the concentration (Figure 9) illustrates a potentially significant difficulty that must be addressed in estimating the spatial moments of a tracer plume in the field. Specifically, as the plume is transported through a heterogeneous medium, the second spatial moments become increasingly sensitive to the regions of the aquifer occupied by low concentration of the tracer. As the concentration decreases, however, analytical detection limits will often limit our ability to quantify the concentration. The result, again as illustrated in Figure 9 for the current series of experiments, is that estimates of the second spatial moments will become increasingly uncertain as a greater portion of the plume is occupied by concentrations below our analytical detection limits. Further, this result implies that two tracers, with different detection limits (relative to their initial concentrations) may provide different estimates of the second spatial moments.

It is argued that sampling design may also help to explain the difference between the hydraulic results (in which theory matched experiment) and the transport results (in which theory failed to match experiment). The distribution of piezometers allowed relatively uniform sampling of the head and head increment over the central portion of the flow field. As a result, the data were sampled over a large number of integral scales in both the horizontal and vertical dimension, thus allowing confidence in the assumption that ergodic conditions had been achieved. In contrast, ergodic conditions were likely not obtained in the transport experiments. Dagan (1990) argues that the transverse dimension of the plume should be much greater than the integral scale in order to observe transport behavior that is consistent with predictions based on assumptions of ergodicity. The results presented here indicate

that the initial widths of these two plumes (less than five integral scales) were insufficient to achieve such ergodic conditions.

Reassessment of these transport plumes accounting specifically for both the initial geometry of the plume and non-ergodic conditions (e.g., Zhang, 1997) are topics of continuing analysis. Further, these experiments are being repeated in a new realization of the conductivity field and with large initial plume dimension (and a different method of introducing the tracer) in an effort to minimize these non-ergodic effects.

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Laboratory study of chemical transport to wells within heterogeneous porous media

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Abstract. A laboratory flow cell was used to study chemical transport to wells producing water from two realizations of a heterogeneous porous medium. The wells produced water from a confined aquifer which was otherwise governed by a mean uniform hydraulic gradient. For both realizations the aquifer was composed of a two-dimensional, lognormally distributed, second-order stationary, exponentially correlated conductivity field. For the first realization, response was monitored at a single well 45 integral scales from the inflow boundary. Four wells were used in the second realization, with locations ranging from 10 to 45 integral scales from this boundary. The focus of this work was on the nature of the dispersion observed in the chemical arrival at the pumping well, and this work builds on an earlier study in which it was shown that results under mean uniform flow were relatively consistent with stochastic theories for fluid flow and chemical transport under mean uniform flow. Consistent results were obtained across the two realizations for the wells located 45 integral scales from the boundary as the breakthrough curves demonstrated an increase in dispersive behavior with an increase in the ratio of the pumping rate to the regional flow, consistent with existing theory for finite-sized sources. In contrast, results for the well located closest to the source (10 integral scales) demonstrated variability in the timing and shape of the breakthrough curves that did not correlate with the ratio of pumping at the well to regional flow. The results indicate that the shape and timing of a breakthrough curve at a well producing within a regional flow field may be strongly dependent on the distance of the well from the source. Further, the parameters (e.g., dispersivity) obtained from analysis of the breakthrough curve are shown to be functions of the ratio of pumping rate at the well to the regional flux.

1. Introduction

Stochastic models of chemical transport have provided useful insight into contaminant transport under mean uniform flow subject to a number of assumptions regarding the statistical description of the velocity or conductivity fields. For example, first-order theory based on an assumption of mean uniform flow in an exponentially correlated conductivity field [e.g., Rubin, 1997; Dagan, 1984] has been shown to be useful in a number of field situations, including the Borden and Cape Cod sites [e.g., Garabedian *et al.*, 1991; Freyberg, 1986], and has been compared against the results of laboratory experiments performed in heterogeneous porous media [e.g., Silliman *et al.*, 1998; Silliman and Zheng, 2001]. These early theories have been extended to discussion of ergodicity [e.g., Sposito and Dagan, 1994] and to nonergodic solutions [e.g., Zhang *et al.*, 1996] that have provided significant insight into the behavior of chemical plumes that have not converged to ensemble behavior. On the basis of different assumptions regarding the statistics of the underlying velocity field, Berkowitz and Scher [1998] have suggested an alternative model, based on their continuous time random walk model in a statistically stationary velocity field, to reproduce observations from the macrodispersion experiment (MADE) field site [Adams and Gelhar, 1992] and laboratory results on a “uniformly heterogeneous” medium [Berkowitz *et al.*, 2000]. Meerschaert *et al.* [1999] have discussed

a dispersion model based on fractional dispersion. Di Federico and Neuman [1998] also considered transport (and the evolution of the dispersion coefficient) in porous media characterized by multiscale conductivity fields.

In addition to transport under mean uniform flow, there has been substantial study of the delineation and behavior of capture zones in the vicinity of pumping wells. Bair *et al.* [1991], for example, studied confidence intervals on the shape and area of capture zones through use of a time-of-travel model in a homogeneous aquifer with uncertain hydraulic conductivity and porosity. Several studies have used published spatial statistics (e.g., log conductivity variance and correlation scale) to investigate various aspects of capture zone uncertainty through Monte Carlo simulation. Cole and Silliman [1997, 2000], for example, investigated confidence intervals for wells in unconfined aquifers. Working with confined aquifers, van Leeuwen *et al.* [1998, 1999] and Varljen and Shafer [1991] demonstrated that stochastically determined capture zones can aid in delineating confidence regions that will contain the actual capture zone. Similarly, Guadagnini and Granzetti [1999] have studied capture zone uncertainty in terms of transport time statistics from a contamination source to a production well. Vassolo *et al.* [1998] and others have also looked to inverse modeling to illustrate the worth of data in delineating the capture zone of a production well. While these studies have provided substantial insight into the uncertainties involved in delineation of capture zones in heterogeneous media, they provide little insight into the dispersive behavior to be observed for a chemical being withdrawn at the pumping well.

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Figure 1. Schematic of laboratory tank used in these experiments. For the experiments discussed here the three inflow reservoirs were hydraulically connected to form a single inflow reservoir.

Toward the study of dispersive behavior at a pumping well, Silliman [1995] has suggested, for transport in a fracture, that the magnitude of dispersion observed at a well will be a function of the ratio of the pumping rate at the well to the rate of flow through the fracture. He did not, however, support these arguments through either experimental observations or a general theory. This suggestion is consistent with the spirit of work performed for nonergodic solute transport under mean uniform flow considering finite initial plume dimensions [e.g., Zhang *et al.*, 1996].

Dagan and Indelman [1999] and Indelman and Dagan [1999] have recently provided a stochastic analysis of transport under both divergent flow from a single well and flow between an injection and a withdrawal well. Neuweiler *et al.* [2001] and Attinger *et al.* [2001] have extended this work for radial flow with finite Peclet numbers. These theoretical results do not, however, cover the very difficult problem of a well producing water from a flow field otherwise described by mean uniform flow. Neuman [1993] has provided a weak approximation that can be applied to this flow condition. However, to our knowledge, no published work has provided expressions for the dispersive properties to be observed at the well.

The present study is intended to provide insight into the dispersive properties to be observed during withdrawal of a chemical at a well producing from a heterogeneous aquifer. As such, this study builds upon the literature on delineation of well head capture zones referenced above but focuses on the dispersive characteristics observed at the well rather than on delineation of the extent of the capture zone. Further, it builds on the literature covering nonergodic transport and transport in the presence of diverging flow fields. This study is based on a series of experiments, performed under laboratory conditions, in which breakthrough curves for a conservative chemical tracer were collected at a well that is pumping at steady state from a heterogeneous aquifer subject to a regional gradient. These experiments are an extension of flow and transport studies in the same media that were conducted under mean uniform flow as reported by Silliman and Zheng [2001]. As discussed in section 4, these previous studies demonstrated relatively good agreement between experimental results ob-

tained under mean uniform flow and theoretical results obtained from the literature. All work is based on two realizations of a constructed, two-dimensional aquifer that was designed to mimic a low-variance, lognormally distributed, exponentially correlated, second-order stationary random conductivity field.

2. Apparatus

The experiments were conducted in a laboratory tank with maximum dimension for the sediment compartment of $1.6 \text{ m} \times 0.67 \text{ m} \times 0.094 \text{ m}$ (Figures 1 and 2). The tank was designed to allow flow to be established with a mean gradient parallel to the 1.6 m axis of the cell. The upper and lower borders of the tank were maintained as no-flow boundaries. Mean uniform flow parallel to the long dimension of the tank was established through injection of fluid into the upstream reservoir (left reservoir in Figure 1) while maintaining a constant head in the downstream reservoir (right reservoir in Figure 1).

Five wells with locations as shown in Figure 1 were utilized in these experiments. In each case, the wells fully penetrated the 0.094 m width of the laboratory tank. The wells consisted of 0.006 m OD copper tubing which had been penetrated with a large number of drill holes along its length. The tubing passed through a special fitting on the side of the tank and could be accessed on the exterior of the tank (see Figure 2). The interior of the copper tubing (that portion within the tank) was then lined with an 80-mesh stainless steel screen, thus preventing the migration of the sediments used to fill the tank. A peristaltic pump was used to withdraw water from a well. No more than one well was active at any given time.

The critical measure of the pumping rate used in these experiments was the ratio of pumping at the well to regional flow, R , and was defined as

$$R = Q_p / Q_T, \quad (1)$$

where Q_p is the volumetric rate of discharge from the well and Q_T is the volumetric flux crossing the inflow boundary of the tank. For the experiments discussed here, R varied from ~ 0.06 to ~ 0.31 . That is, the rate of fluid withdrawal at the well varied

Figure 2. Photographs of the (a) first realization and the (b) second realization of the heterogeneous porous medium. The dark areas are the fine sediments with iron coatings. Both media mimic computer-generated realizations of a second-order stationary, exponentially correlated, conductivity field.

from ~6% of the total volumetric flux entering from the inflow reservoir to ~31% of this flux.

For the chemical tracer tests the tracer was added as a step increase in concentration within the inflow reservoir. The step

change in concentration was accomplished by stopping all fluid flow through the tank, closing valves leading from the well and outflow reservoir, and flushing the inflow reservoir with tracer fluid. The flushing occurred by injecting tracer fluid at either

the top or bottom of the reservoir (dependent on whether the tracer was of higher or lower density than the native fluid) and withdrawing fluid from the opposite end (bottom or top) of the same reservoir at an identical rate. The pumping rate was kept at a minimum to allow the vast majority of the fluid to pass through the open reservoir (rather than infiltrating into the porous medium). It was determined that ~ 3 reservoir volumes were required to bring the reservoir to full concentration of the tracer; 5 reservoir volumes were used during these experiments to provide confidence that the reservoir was at the full concentration at the beginning of the test. Following the flushing of the reservoir, flow through the porous medium was reinitiated with tracer fluid being injected at constant volumetric flux and concentration into the inflow reservoir and the valves leading from the well and outflow reservoir open. It is noted that this method produced a brief period of nonsteady flow within the porous medium. However, monitoring of the volumetric flux from the outflow reservoir indicated that the period of nonsteady flow was substantially < 1 min in all experiments and was therefore considered negligible.

The breakthrough curve of the tracer was monitored at the well by passing the discharge through a calibrated, flow-through electrical conductivity cell. Sodium chloride was used as tracer in all experiments. The conductivity cell was calibrated over the range of tracer concentrations used (calibrated from 50 to 1000 mg L⁻¹). Because of the volume of water contained in the tubing leading to the conductivity cell and the volume of the conductivity cell the time of measurement at the cell was adjusted to account for plug flow through the tubing and the cell. The cell was always oriented such that higher concentration was at the bottom of the cell, thus minimizing mixing within the cell (because of density effects the response in the cell was unstable when lighter fluid entered from the base of the conductivity cell or heavy fluid entered from the top of the cell). Independent experiments on mixing in the cell demonstrated that dispersion arising from mixing of fluids in the cell was minimal in comparison to the temporal width of the breakthrough curves observed here. All tracer experiments were run until the conductivity cell indicated that the concentration at the well was steady and equal to the inflow concentration.

3. Porous Medium

The distribution of sands comprising the heterogeneous medium within the tank was selected so as to mimic a lognormal, exponentially correlated, second-order stationary hydraulic conductivity field. The variance of this field was selected to equal ~ 0.5 . In order to produce these fields, two sources of sand were obtained with different grain-size distributions. The first source (Accumin sand) was available commercially and has been used extensively in our prior experimental efforts. This source consisted of well-rounded quartz sands with minimal iron oxides on their grain surfaces. The second source consisted of sands excavated from a research site near Oyster, Virginia. These sands included a far wider grain-size distribution than the first source, with substantial iron coatings on the fine grain-size fraction. While the Accumin sands were purchased already sieved into multiple size fractions, the sands from the field site required sieving into various grain sizes. Following sieving, the sands were remixed in various grain-size fractions (no mixing between the two sources) to provide 10 final mixtures of sands with a total range in hydraulic conduc-

tivity of 1.61 to 0.015 cm s⁻¹. The source, conductivity, and percent of the medium comprised of each grain size has previously been reported [Silliman and Zheng, 2001].

In order to define the distribution of these sands a series of two-dimensional random fields with exponential correlation were simulated on the computer using the turning bands algorithm [Tompson *et al.*, 1989]. The random fields generated had an isotropic integral scale of 2.75 cm and a variance of $\ln K$ of slightly < 0.5 . The simulated conductivities were then reduced to the 10 conductivities available in the laboratory sands. The statistics of the conductivity field (mean, variance, and correlation structure) were then recalculated on this reduced conductivity field to ensure that reducing the continuous conductivity distribution to a discrete distribution with 10 conductivities did not seriously impact the simulation statistics. Two of these realizations were selected for packing within the tank. These distributions were then printed out, to scale, on transparency sheets and attached to the wall of the tank to be used as templates.

The sands were then added to the tank following the template and using narrow plastic dividers to separate different regions in the tank. In the second realization, for example, the entire tank was subdivided into regions of size 6.9 mm $\times$ 6.9 mm $\times$ 94 mm (94 mm is the width of the tank). Sand was added to each of these regions and the tank was regularly vibrated in order to ensure proper compaction (the plastic dividers were raised as the tank was filled with sand). The actual distributions of sand, as viewed through the front surface of the tank, are shown in Figure 2. As discussed by Silliman and Zheng [2001], the distribution at the wall as shown in Figure 2 includes slightly blurred contacts between different regions (due to leakage around the plastic divider at the wall surface), and the contact between different sands in the interior of the tank is much sharper than observed at the wall.

As discussed by Silliman and Zheng [2001], correspondence between the computer-simulated random field and the random field actually packed in the tank was checked for the first realization both visually and through comparison with hydraulic measurements (i.e., the effective hydraulic conductivity and the second-order moment of the hydraulic gradient). It was concluded that the distribution of sands obtained in the laboratory was quite close to the simulated distribution. Further, the agreement between chemical tests run in the two realizations (see section 4) and the agreement between biological transport experiments run in both realizations (S. E. Silliman *et al.*, Bacterial transport in heterogeneous porous media: Observations from laboratory experiments, submitted to *Water Resources Research*, 2000) suggest that both packings successfully reproduced the desired random fields.

4. Summary of Results Under Mean Uniform Flow

Silliman and Zheng [2001] report on results of hydraulic and chemical tracer experiments performed under mean uniform flow in the first realization of these two media. Experiments described in that paper included analysis of both the distribution of increments in the hydraulic head and the transport of a finite sized plume of conservative chemical tracer. The hydraulic measurements described within that paper demonstrated good agreement with stochastic theory.

Results for the chemical transport experiments showed agreement with the spatial trend in longitudinal dispersion

predicted by Dagan [1984, 1987]. However, it was reported that the asymptotic longitudinal dispersivity was below that predicted by this theory. This discrepancy was explained as being due to the small initial lateral dimension of the tracer plume relative to the integral scale of the conductivity field. This explanation has since been confirmed by comparing the asymptotic dispersivity observed in those experiments with the predicted asymptotic dispersivity predicted by Dagan [1991] for an initial plume of finite width. On the basis of equation (29) in the work of Dagan [1991] it was estimated that for an initial plume of ~ 3 –5 integral scales (the minimum dimension of the observed plume) the asymptotic dispersivity will be in the range of 0.475–0.65 times $\sigma^2 I$ (where I is the integral scale (2.75 cm) and σ^2 is variance of the natural log of the hydraulic conductivity (0.49)). This provides a predicted asymptotic dispersivity in the range of 0.64–0.88 cm. On the basis of the observed rate of spread of the tracer plume it is estimated that the observed asymptotic dispersivity is in the range 0.60–0.80 cm (derived from Figure 8a of Silliman and Zheng [2001]). Hence the longitudinal transport results under mean uniform flow were consistent with stochastic theory.

In terms of transverse dispersion, Silliman and Zheng [2001] report inconsistency with the theoretical predictions obtained from the early theories of Dagan [1984, 1987]. In reviewing these results relative to nonergodic theory for finite initial dimension on the plume, however, the results reported by Silliman and Zheng (their Figure 8b) are qualitatively consistent with the small asymptotic transverse dispersivities predicted by Zhang *et al.* [1996]. Quantitative comparison has not been performed.

Overall, therefore, it was concluded from the experiments on mean uniform flow that the medium and the apparatus were providing experimental results that compared favorably with stochastic theory for mean uniform flow.

5. Experimental Results Under Flow to a Well

Wells 1 through 5 are numbered as shown in Figure 1. Only well 5 was in place for experiments in the first realization. Wells 1–4 were active for experiments in the second realization. Wells 4 (second realization) and 5 (first realization) are at the same distance from the source (45 integral scales), such that results from these two wells are directly comparable. Well 4 (rather than well 5) was used in the second realization to obtain consistent measurements along the center line of the tank.

Breakthrough curves for three tracer tests performed in the first realization using well 5 (with $R = 0.06, 0.13, 0.31$, respectively) are provided in Figure 3. The time axis on this plot has been normalized as $[t - (V_T/Q_p)](Q/V)$, where t is the time since the start of flow following addition of the tracer to the inflow reservoir, Q is the volumetric flux entering the inflow reservoir, V is the total volume of the porous medium, Q_p is the rate of pumping at the well, and V_T is the volume of the tubing between the tank and the conductivity cell plus the volume of water within the conductivity cell. The ratio, V_T/Q_p , represents a correction for the time required for a sample to travel from the well to the conductivity cell. The concentration has been normalized as percent of total change in concentration during the experiment. It is noted that the finite volume of the conductivity cell (~ 30 mL between the lower inlet to the cell and the point of discharge from the cell) causes an aver-

Figure 3. Breakthrough curves observed at well 5 in the first realization. Concentration is normalized to the total change in concentration during the experiment. Time is normalized as $[t - (V_T/Q_p)](Q/V)$ to account for variation in total flux through the medium and the time delay within the tubing and conductivity cell.

aging of the concentration within the cell. Given the pumping rates at the well (see Table 1), the maximum period of averaging was ~ 2 min (that is, at any time, the water at the outlet of the conductivity cell was withdrawn from the tank a maximum of ~ 2 min earlier than the water at the inlet to the conductivity cell). In all experiments, the width of the breakthrough curve (e.g., from $C/C_0 \sim 0.05$ to $C/C_0 \sim 0.95$) was >10 min and was often >30 min. It was therefore assumed that the time averaging which occurs within the conductivity cell was insignificant.

Using the same axes as in Figure 3, Figures 4a–4d show the results for tracer tests run at each of the four wells in the second realization, with Figures 3 and 4d representing the results for the two wells at the same distance from the source

Table 1. Description of the Various Experiments Included in Figures 3 and 4^a

Well	C_i	C_e	Q	Q_p	R
1	134	510	183	13.8	0.075
1	122	500	226	27.0	0.120
1	120	510	220	80.0	0.364
2	118	517	228	14.1	0.062
2	120	505	235	32.0	0.136
2	122	500	217	73.0	0.336
3	115	506	253	13.8	0.055
3	108	527	248	35.0	0.14
3	116	496	209	71.0	0.34
4	117	542	253	17.0	0.067
4	117	519	298	32.5	0.109
4	119	497	225	71.0	0.316
5	216	536	258	15.4	0.06
5	536	219	117	15.6	0.13
5	219	117	108	33.8	0.31

^aDefinitions are as follows: Q is volumetric flux entering the medium through the inflow reservoir (mL min^{-1}); Q_p is volumetric flux being withdrawn at the well (mL min^{-1}); R is Q_p/Q ; C_i is starting concentration of NaCl (mg L^{-1}); and C_e is ending concentration of NaCl (mg L^{-1}).

Figure 4. Breakthrough curves at wells 1-4 in the second realization: (a) results for well 1 with $R = 0.08$, 0.12 , and 0.31 , (b) results for well 2 with $R = 0.06$, 0.14 , and 0.34 , (c) results for well 3 with $R = 0.06$, 0.14 , and 0.34 , and (d) results for well 4 with $R = 0.07$, 0.11 , and 0.32 . Time and concentration are normalized as in Figure 3.

(wells 5 and 4, respectively). The flow rates, pumping rates, and starting and ending concentrations for each of the tests are provided in Table 1. Reproducibility of experimental results within a single packing was excellent. In addition, there was excellent reproducibility of results at 45 integral scales (wells 4 and 5) across the two realizations. For example, Figure 5 shows a comparison of results for one experiment conducted at well 4 in the second realization (with $R = 0.32$) as compared to an experiment conducted at well 5 in the first realization (with $R = 0.31$). Because of the fact that only one well was available for the first

realization it was not possible to compare experimental results across realizations for the shorter flow distances.

6. Observations and Discussion

A number of observations are made regarding these results. These observations are summarized as follows: (1) The shape and timing of the breakthrough curves are functions of R . (2) At distance from the inflow boundary, there is a consistent relationship between R and the degree of dispersion observed in the breakthrough curves. (3) This relationship appears consistent across realizations. (4) This relationship breaks down at shorter flow distances.

It is apparent from review of Figures 3 and 4 that the timing and shape of the breakthrough curve varies with the rate of production at the pumping well. Viewing Figures 3 and 4d, for example, one observes that while the initial arrivals of the breakthrough curve at wells 4 and 5 are nearly identical (in terms of normalized time) for all R (and across the two realizations), the breakthrough curves reflect greater dispersion with an increase in R . Hence it is concluded that the degree of dispersion observed in the well was, for these experiments, dependent not only on the statistics of the conductivity field and transport distance between the source and the well but also on the ratio of production at the sampling point to the mean uniform flux moving through the aquifer.

Comparison of Figures 3 and 4d leads to the observation of an apparently consistent relationship between R and the degree of dispersion observed at the well, at least at distance from the inflow boundary. Specifically, as R increases, the degree of dispersion observed at the well increases. At least two mechanisms, summarized in Figure 6, can be hypothesized as leading to such a relationship.

The first mechanism (Figure 6a) is related to the degree of

Figure 5. Comparison of breakthrough curve obtained at well 5 (realization 1 - $R = 0.31$) with breakthrough curve obtained at well 4 (realization 2 - $R = 0.32$). The solid line represents the results from the second realization. Time and concentration are normalized as in Figure 3.

convergence of the mean flow in the vicinity of the well. For small R , for example, the capture zone consists of a narrow set of flow lines extending upgradient from the well. In this situation, one observes very little convergence of the mean flow field in the vicinity of the well, negligible variation in the mean transport time from the source to the well among all flow lines entering the well, and, therefore, a steep breakthrough curve (minimal dispersion at the well). As R increases, both the width of the capture zone and the degree of convergence of the mean flow field near the well will increase. The result is that mean transport time from a line source (perpendicular to mean flow) to the well increases with transverse distance away from the center line of the capture zone, resulting in the breakthrough curve observed at the well reflecting a greater degree of dispersion.

The magnitude of observed dispersion at the well caused purely by convergence in the mean flow field can be examined either numerically or through an analytical solution provided by Bear and Jacobs [1965]. The results shown here are based on the analytical solution but have also been reproduced numerically using combinations of MODFLOW [McDonald and Harbaugh, 1988] and MODPATH [Pollock, 1989], and also MODFLOW and MT3D [Zheng [1992], setting the longitudinal dispersion coefficient to zero). The solution of Bear and Jacobs, based on the assumption of homogeneity in the conductivity field, provides a measure of the time of travel from a specified point within the aquifer to the well and is given by

$$t' = x' + \ln \left(\frac{\sin \Theta}{\sin (y' + \Theta)} \right), \quad (2)$$

where

$$\Theta = \arctan \left(\frac{y'}{x'} \right),$$

$$x' = \frac{2\pi q_0}{Q} x,$$

$$y' = \frac{2\pi q_0}{Q} y,$$

$$t' = \frac{2\pi q_0^2}{nQ} t,$$

where

- q_0 Darcy velocity of the mean uniform flow per unit thickness;
- Q rate of production at the well per unit thickness;
- x longitudinal distance from the well to the chemical source;
- y transverse distance from the well to the chemical source;
- t time.

Assuming that the well is sufficiently distant from the inflow boundary to ensure that there is mean horizontal flow at the boundary (this has been confirmed to be true for wells 4 and 5 through numerical modeling), the mean width of the capture zone at the inflow boundary ($2L$) is given by

$$2L = (Q/Q_T) D,$$

where Q_T is equal to the measured flow through the entire tank and D is the saturated thickness of the experimental medium along the vertical axis of the tank (0.67 m). Setting $x = 1.19$ m (the distance from the inflow boundary to wells 4 and

Figure 6. Schematic illustration of two mechanisms leading to the dispersion of the breakthrough curve observed at the pumping well: (a) increased dispersion of the breakthrough curve related to the widening of the capture zone with an increase in R ($R1 < R2 < R3$) and (b) increased dispersion of the breakthrough curve with an increase in R ($R1 < R2$), related to greater variability in fluid velocities along any transverse cross section through the capture zone.

5) and letting y vary in the range of $-L$ to $+L$, one can generate the predicted breakthrough curve at the well solely because of the convergence of the flow field in the vicinity of the well (i.e., zero dispersion due to local diffusion, local dispersion, or heterogeneity). Figure 7a shows the predicted breakthrough curves for the three R values used in these experiments and illustrates that the observed dispersion in the well does increase with increase in R . Figure 7b, however, shows the comparison of this analytical solution against the observed breakthrough curves for two of the experiments at well 5, illustrating that there is substantially more dispersion observed at the well than predicted from consideration of advection via the mean flow field. Hence it is apparent that while an increase in pumping rate will result in increased dispersion of the breakthrough curve observed at the well, this increase is insufficient to explain the observed dispersion at the well in these experiments.

The second mechanism is directly related to the presence of heterogeneity within the porous medium. Figure 6b shows hypothetical capture zones at the same well for two pumping rates in a heterogeneous aquifer. Note that the capture zone at the higher pumping ratio ($R2$) will necessarily enclose the capture zone at the lower pumping ratio ($R1$) for the same orientation of regional flow. If we limit our attention to the portion of the capture zone that is far enough upgradient of the well such that flow is approximately mean uniform, we can

Figure 7. Comparison of predicted breakthrough curves (normalized time and concentration), assuming that advection in the converging flow field is the only mechanism of dispersion: (a) comparison of predicted curves at $R = 0.06$, 0.13 , and 0.31 and (b) comparison of pure advection model with experimental results at well 5 for both $R = 0.06$ and $R = 0.31$.

compare the expected behavior here to the nonergodic transport discussed by Dagan [1991] and Zhang *et al.* [1996]. In the case considered here, a high R translates into a wider initial (and mean) width of the tracer front that will eventually enter the well and, thereby, a wider sampling of velocities along any transverse cross section through the capture zone. As shown by Dagan [1991] and Zhang *et al.* [1996], this increase in the initial width of the tracer front is predicted (at least under mean uniform flow) to translate into an increase in the effective longitudinal dispersivity. It is conjectured that similar arguments of nonergodicity will apply in the presence of the well and lead to an effective longitudinal dispersivity increasing with increasing mean width of the capture zone and, therefore, increasing with R .

In order to determine whether the second mechanism had a significant impact on the observed results, the breakthrough curves shown in Figure 3 were simulated using a combination of MODFLOW [McDonald and Harbaugh, 1988] and MT3D [Zheng, 1992]. It is important to note here that we are not

making the argument that we expect Fickian behavior in this medium or that MT3D should reproduce all details of these breakthrough curves. Rather, MT3D is used here as a tool to provide a simple measure of the relative dispersion among the different experimental results (as represented by the fitted dispersivity). In completing these simulations it was assumed that the medium was (1) homogeneous with a hydraulic conductivity equal to the calculated effective conductivity of the medium [Silliman and Zheng, 2001], (2) at steady state with respect to flow, (3) subject to pumping at a rate and at a location identical to those used in the experiment, (4) subject to boundary heads for the left and right reservoirs selected to provide the observed volumetric flux across the inflow boundary, and (5) subject to the input of the tracer as a step function at the inflow boundary. The assumption of homogeneity (and, thereby, the use of effective parameters) was justified in this case by the observation that the breakthrough curves were nearly identical at 45 integral scales from the source in the two realizations, implying a lack of dependence of the large-scale transport process on the details of the local conductivity distribution. Simulations were run at a variety of discretizations (from element dimensions of 0.4 cm on a side to element dimensions of 1.0 cm on a side) and with a variety of transport solution schemes (method of characteristics (MOC), modified MOC (MMOC), and the hybrid scheme provided in MT3D) in order to ensure that the solutions had converged and were not subject to excessive numerical dispersion. Further, the numerical solution was compared with the analytical solution described above for the case of zero dispersion with excellent reproduction of the analytical result. The solutions presented in Figures 8a–8c were performed with element dimensions of 1.0 cm $\times$ 1.0 cm, the hybrid MOC/MMOC package, a Courant number of 0.9 , and the fourth-order Runge-Kutta algorithm for particle tracking. For these simulations it was assumed that the transverse dispersivity was equal to one tenth the longitudinal dispersivity.

Figure 8 shows selected (typical) fits between the observed breakthrough curves at well 5 and the predicted breakthrough at this well for different pumping ratios, R . The parameters used in these simulations are listed in Table 2, as are the range of parameters considered to provide reasonable fits to the data. The theoretical curves tend to fit the observed curves relatively well through the heart of the breakthrough curve but not in the early or late time tails. The observed behavior shows an overly steep front portion to the breakthrough curve and substantial tailing. As a result of the relatively weak fit of the model to the tails of the observed breakthrough curves, combined with the fact that this paper is focused on the overall behavior of transport to the wells rather than parameter estimation, no formal algorithm was used to optimize the fit between the theoretical results and the observed breakthrough curves. That the fitted dispersivities are distinctly different at the three pumping ratios, R , is illustrated in Figure 8d, which shows the lack of fit of MT3D to the data for $R = 0.31$ using the parameters listed in Table 2 for the experiments at $R = 0.06$ and $R = 0.13$.

Review of the parameter estimates obtained (Table 2) shows that there is a consistent change in parameters with R . Specifically, the longitudinal dispersivity and the porosity were observed to increase with increasing R . As expected from the similarity of the breakthrough curves for well 4 (second realization), the same behavior of the dispersivity and porosity was observed for well 4. It is noted that while the observed variation in the effective porosity is considered quite significant, an

Figure 8. Fit to the data observed at well 5 in the first realization using MODFLOW and MT3D: (a) $R = 0.06$, (b) $R = 0.13$, (c) $R = 0.31$, and (d) the fit to the data for $R = 0.31$ using the parameters estimated for the experiments with $R = 0.06$ and $R = 0.13$. Solid lines represent the fitted curves and the dotted lines represent the data.

adequate explanation for this behavior is currently not known to this author. Hence discussion of the fitted porosity is left for future consideration.

It is illustrative to compare the dispersivities obtained with MT3D against the asymptotic dispersivities predicted for mean uniform flow with a finite initial width of the source. As above, this prediction is based on equation (29) of Dagan [1991], assuming mean uniform flow and, in this case, an initial source width equal to the mean width of the capture zone at the inflow boundary. For the three experiments represented in Figure 8 the initial widths were set equal to 1.46, 3.17, and 7.55 integral scales based on $R = 0.06$, $R = 0.13$, and $R = 0.31$, respectively, and an integral scale of 2.75 cm. The resulting predicted longitudinal dispersivities are listed in Table 2. It is observed that the magnitude of the dispersivities and trend in dispersivity with R are quite consistent between these laboratory results and the results predicted using the theory for mean uniform flow. That is, the large-scale behavior within this experimental medium in the presence of a single pumping well appears quite consistent with the large-scale, nonergodic behavior under mean uniform flow discussed by Dagan [1991] and Zhang *et al.* [1996]. Hence these numerical results lend

support to the conjecture that the observed increase in dispersivity with increasing R , as observed in the breakthrough curves at wells 4 and 5, is related primarily to the increased mean width of the capture zone at higher R .

While the dispersion results observed at wells 4 and 5 appear to be controlled by a consistent, large-scale transport behavior, it is apparent from review of Figure 4 that there is also a strong effect of scale (distance from the source to the well) on the observed breakthrough curve. At 10 integral scales from the inflow boundary (well 1), for example, the shape and timing of the breakthrough curves vary inconsistently with R (Figure 4a). The inconsistency observed at the first well is less apparent at well 2 (15 integral scales from the boundary and shown in Figure 4b), where the breakthrough curves are apparently being averaged over sufficient length scales of heterogeneity to provide some consistency in the variation with R . Specifically, the initial arrival time appears quite similar for the three R values tested. The relationship between dispersivity and R continues to become stronger through well 3 and, finally, through wells 4 and 5. This result is, of course, quite consistent with existing transport theory for mean uniform flow.

Table 2. Parameters Used in MT3D to Produce Fits Shown in Figure 8^a

R	Range of Porosity	Range of Dispersivity, cm	Porosity Used in Figure	Dispersivity Used in Figure, cm	Dispersivity Predicted Using Dagan [1991], cm
0.06	0.285–0.290	0.2–0.4	0.288	0.2	0.21
0.13	0.290–0.295	0.3–0.7	0.295	0.4	0.50
0.31	0.330–0.335	0.7–1.5	0.335	1.2	1.0

^aDispersivity is longitudinal. Transverse dispersivity is 0.1 times the longitudinal dispersivity. Ranges on porosity and dispersivity represent the range of parameters outside of which reasonable match of theory to observation could not be obtained.

7. Conclusions

The results of this experimental effort lead to several conclusions:

1. The shape of the breakthrough curve at a well producing from a heterogeneous aquifer will be a function of R , the ratio of pumping rate at the well to the volumetric flux moving through the aquifer under the mean regional gradient.
2. At distance from the source, the degree of dispersion reflected in the breakthrough curve at the well will increase with R faster than would be expected simply through consideration of the change in the mean flow field (with R) in the vicinity of the well.
3. The heterogeneity-related dispersion reflected in the breakthrough curve appears to be a function of two length scales: (1) the distance from the source and (2) the mean width of the capture zone upgradient of the well. The dependence on the mean width of the capture zone at distance from the source is quantitatively consistent with the literature on nonergodic transport of chemical plumes of finite initial dimension being transported within mean uniform flow fields.
4. At 45 integral scales from the inflow boundary the behavior (shape, timing, and dependence on R) of the breakthrough curves was nearly identical over the two realizations tested, suggesting that transport is approaching asymptotic behavior at or near this length scale.
5. The advection-dispersion equation, applied under the assumption of homogeneity, provided reasonable reproduction of the center portion of the breakthrough curves. However, the experimentally derived curves consistently showed an overly steep early time behavior and an extended tail at late time.

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Bacterial transport in heterogeneous porous media: Observations from laboratory experiments

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Abstract. Transport of bacteria through heterogeneous porous media was investigated in small-scale columns packed with sand and in a tank designed to allow the hydraulic conductivity to vary as a two-dimensional, lognormally distributed, second-order stationary, exponentially correlated random field. The bacteria were *Pseudomonas fluorescens* R8, a strain demonstrating appreciable attachment to surfaces, and strain M1, a transposon mutant of strain R8 with reduced attachment ability. In bench top, sand-filled columns, transport was determined by measuring intensity of fluorescence of stained cells in the effluent or by measuring radiolabeled cells that were retained in the sand columns. Results demonstrated that strain M1 was transported more efficiently than strain R8 through columns packed with either a homogeneous silica sand or a more heterogeneous sand with iron oxide coatings. Two experiments conducted in the tank involved monitoring transport of bacteria to wells via sampling from wells and sample ports in the tank. Bacterial numbers were determined by direct plate count. At the end of the first experiment, the distribution of the bacteria in the sediment was determined by destructive sampling and plating. The two experiments produced bacterial breakthrough curves that were quite similar even though the similarity between the two porous media was limited to first- and second-order statistical moments. This result appears consistent with the concept of large-scale, average behavior such as has been observed for the transport of conservative chemical tracers. The transported bacteria arrived simultaneously with a conservative chemical tracer (although at significantly lower normalized concentration than the tracer). However, the bacterial breakthrough curves showed significant late time tailing. The concentrations of bacteria attached to the sediment surfaces showed considerably more spatial variation than did the concentrations of bacteria in the fluid phase. This contrast between behavior in the fluid phase and on the solids is consistent with field observations by other authors and initial modeling of these heterogeneous media.

1. Introduction

Interest in the deposition and transport of bacteria in groundwater systems ranges from the study of subsurface ecosystems, to analysis of bioremediation efforts, to the study of the movement of pathogens. A number of field and laboratory investigations have addressed the mechanisms controlling microbial transport. Among the best documented field studies are those at the field site maintained by the U.S. Geological Survey on Cape Cod [e.g., Harvey *et al.*, 1989; Harvey and Garabedian,

1991; LeBlanc *et al.*, 1991; Garabedian *et al.*, 1991; Bales *et al.*, 1995]. From the diverse range of experiments conducted at several locations within the Cape Cod site, these authors have demonstrated that microbial transport is strongly influenced by subsurface geochemistry (in particular, pH), physical heterogeneity, and the flow path taken by the bacteria. Harvey *et al.* [1993] also demonstrated that tests performed in the field are often governed by different mechanisms than those in repacked columns in the laboratory.

Laboratory studies have demonstrated that even in homogeneous media, transport is influenced by complex factors such as microbial activity and behavior. Clement *et al.* [1997], for example, used both laboratory studies and a numerical model to study the growth and transport of bacteria under denitrifying conditions and found that a first-order attachment-detachment model could describe the transport observed in the column studies. These authors suggest, from analysis of

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their results, that the rate of detachment may increase with growth rate. *Johnson et al.* [1995] combined laboratory experiments with a series of detachment models for bacteria to argue that only detachment functions that allow an initial period of rapid release combined with a longer term release mechanism were successful in reproducing observed breakthrough curves. They also argued that for their experiments a number of preferential (irreversible) sorption sites must exist within their columns and that bacteria must sorb to these sites prior to the onset of breakthrough. *Hendry et al.* [1999] demonstrated the impact of mean pore water velocity on the sorption (attachment/detachment) behavior of bacteria in homogeneous columns.

A number of these studies have focused on the influence of geochemical properties on microbial attachment/detachment. *Knapp et al.* [1998] performed a series of column experiments demonstrating that the iron oxyhydroxide coatings on mineral surfaces of a porous medium can lead to a substantial decrease in the percentage of bacteria remaining in the fluid phase and observed in the column effluent. Detachment can vary considerably in space and time for media in which the pH and ionic strength vary significantly [*Scholl and Harvey*, 1992]. Also, attachment-detachment behavior can change from first-order to second-order processes depending on the ionic strength of the fluid phase [*Deshpande and Shonnard*, 1999]. Clearly, environmental factors can have a significant influence on transport through homogeneous media.

Bacterial transport processes become even more complex in heterogeneous media. *Teutsch et al.* [1991] constructed several experimental devices (1 m columns and an 8 m long flume) for the study of bacterial transport in various sediments. These authors found that the breakthrough curves for bacteria and viruses were highly variable and strongly dependent on the composition of the medium. *Fontes et al.* [1991] found that the ability of a bacterium to travel through a laboratory column was a function of the composition of the artificial groundwater, the mean grain size of the porous medium, and the length of the flow path through the column. They also found that heterogeneity can lead to multi-peaked breakthrough curves because of strong variation in mean transport time along different flow paths through the column. *Morley et al.* [1998] have studied transverse mixing of bacteria between regions of different hydraulic conductivity. On the basis of observations from a laboratory column containing a vein of coarse sand within an otherwise homogeneous column of fine sand, they argued that transverse mixing is not significant in aquifers.

Microbial transport also has been investigated in intact sediment cores [*Bolster et al.*, 1999], with destructive sampling revealing substantial (two orders of magnitude) spatial variation in microbial deposition. The authors concluded that this spatial variability was due, in part, to heterogeneity but also to multiple attachment behaviors among the source bacteria. They argued further that destructive sampling of cores is necessary to fully understand the bacterial deposition occurring within such laboratory experiments. *Alfeider et al.* [1997] have made similar arguments regarding the need for sediment cores based on comparison of fluid samples and cores samples collected in the field.

The present laboratory study builds on these previous reports by investigating microbial transport through two heterogeneous media for which the local hydraulic conductivity varied as a stationary, correlated random field. Further, the conductivity was inversely correlated with the coating of sand

grains with iron oxyhydroxide. Although the flow field was mean uniform, it was fully two-dimensional and subject to local pumping at a well. Bacterial breakthrough curves were determined by analysis of water collected at the well. In addition, fluid samples were collected at several points in space and at several times during the transport experiment. Finally, bacterial abundance on the solid phase was determined, for one of the experiments, by destructive sampling. These experiments represent an extension of the flow and transport studies reported by *Silliman and Zheng* [2001] and *Silliman* [2001].

2. Methods

2.1. Laboratory Tank

The experiments were conducted in a laboratory tank with a sediment compartment of 1.6 m $\times$ 0.67 m $\times$ 0.094 m (Figures 1 and 2). The design allowed flow to be established with a mean gradient parallel to the 1.6 m axis of the tank and with no-flow boundaries along the upper and lower borders of the tank. Tests were conducted by injecting water at a known volumetric flux into the inflow reservoir (Figure 1) while constant head was maintained at the outflow reservoir. Water was withdrawn at well 5 (the first test) or well 4 (the second test) with the ratio of the pumping rate at the well to the volumetric flux across the inflow boundary between 0.24 and 0.25.

Chemical and microbial tracers were added concurrently to artificial groundwater, AGW (140 μ M NaHCO₃; 140 μ M MgSO₄ $\times$ 7H₂O; 80 μ M NaCl; 70 μ M CaSO₄ $\times$ 2H₂O; 15 μ M KNO₃). The chemical tracer was sodium chloride at concentrations of 100 mg L⁻¹ in the first experiment and 200 mg L⁻¹ in the second experiment. The microbial tracers are described below. Both tracers were added to the inflow reservoir through a flushing process used previously [e.g., *Silliman*, 2001] to provide sharp front and back faces to tracer pulses. The pulse volumes were $\sim$ 8.0 L for the first experiment and 6.6 L for the second experiment. Tracer-free AGW was used to chase the pulses through the tank for $\sim$ 9 pore volumes in the first experiment and 4 pore volumes in the second experiment.

Chemical breakthrough was monitored at the well using a calibrated conductivity electrode. Microbial concentrations were determined by plating (see below) samples from (1) the discharge at the well, (2) fluid samples collected from sampling ports during the course of the experiment, and (3) samples collected destructively after termination of the experiment and opening of the tank (first experiment only: the second tank remains sealed for additional experiments).

Mixtures of sands from two sources were utilized to create the porous media for all experiments completed. The first, Accumin Sand (AS) (Unimin, Le Sueur, Minnesota), was available commercially and has been used extensively in our previous experiments. AS is comprised of well-rounded quartz sands with relatively minor surface coatings. The second type of sand was excavated from a U.S. Department of Energy research site in Oyster, Virginia [*DeFlaun et al.*, 1999] and is referred to here as Oyster Sand (OS). This sand includes a far wider distribution of grain size than AS, with substantial iron coatings on the fine-grain size fraction. All sands were sieved into multiple grain size categories to produce varying hydraulic conductivities. Ten final mixtures of sands were chosen for use in packing the flow cell, providing a total range in hydraulic conductivity of 1.61–0.015 cm s⁻¹ (Table 1 provides a summary of these 10 mixtures; conductivities were determined using

Figure 1. Schematic diagram of laboratory tank. Well 5 was active for the first realization. Well 4 was active for the second realization. For the experiments discussed here the three inflow reservoirs were hydraulically connected to form a single inflow reservoir.

constant flux permeameters packed with homogeneous samples of each sand). The fines containing the bulk of the iron-oxide coatings were limited predominantly to the three finest grain size mixtures (i.e., those with the three lowest hydraulic conductivities), thus establishing a negative correlation between coating and conductivity.

The spatial distribution of the sands within the tank was selected so as to mimic a lognormal, exponentially correlated, second-order stationary hydraulic conductivity field. The variance of this field was selected to equal ~ 0.5 , and the intergral scale was 2.75 cm. The process of selecting an appropriate random field and transferring this computer-generated random field to the tank and the sand packing process are described elsewhere [Silliman, 2001]. Figures 2a and 2b show the two final realizations of the random field. The dark material represents the three categories of sand with the lowest hydraulic conductivity and highest degree of iron coatings.

2.2. Microbes

Two strains of *Pseudomonas fluorescens*, R8 and M1, were utilized in the first experiment, and only *P. fluorescens* M1 was used in the second experiment. Strain M1 is a transposon mutant of strain R8, selected for its reduced attachment ability, as previously described [Williams and Fletcher, 1996]. *P. fluorescens* R8 is a spontaneous rifampin-resistant mutant of *P. fluorescens* B0267 (U.S. Department of Energy (DOE) Subsurface Microbial Culture Collection, Florida State University), a strain collected from an uncontaminated subsurface site at the U.S. DOE Savannah River Site [Balkvill et al., 1989].

Transport of *P. fluorescens* R8 and M1 were compared in small-scale sand columns (methods described by Williams and Fletcher [1996]), constructed from 20 cc syringes with silanized, glass wool plugs used as frits. These were packed with the following sands: (a) a previously studied AS sand size fraction quite similar to one of the size fractions used in the large tank (sand 6; Table 1), (b) unfractionated OS, and (c) the 10 sand size fractions used in the tank (Table 1). All of the sands were washed with distilled water followed by deionized water to remove smaller sediment particles. The wet sands were steril-

ized by autoclaving for 90 min and then baked overnight at 250°C to dry.

Bacteria were grown to early stationary phase in peptone and yeast extract broth (PYE: 0.1% weight/volume peptone and 0.07% weight/volume yeast extract) with shaking (125 rpm) at 23°C. The cells were subsequently harvested by centrifugation, washed, and resuspended in AGW to a density of 2.2×10^9 cells mL⁻¹. To equilibrate the columns, AGW was passed through each column at 1 mL min⁻¹ for 20 min. One milliliter of cell suspension was then loaded onto each column, at the rate of 1 mL min⁻¹, just as the last of the AGW passed into the sand. As the bacterial suspension passed into the sand, an additional 20 mL of AGW was passed through the column at 1 mL min⁻¹.

To determine bacterial numbers eluted from the column, cells were stained with the fluorochrome Syto 11 (molecular probes). Syto 11 was added to a final concentration of 2 μ M in 2 mL of effluent and incubated for 30 min. Fluorescence was measured with excitation at 509 nm, and emission was measured at 530 nm and compared to a calibration curve prepared from known densities of stained cell suspensions.

In separate experiments, retention of strains R8 and M1 in the sand columns was determined by radiolabeling the cells and then measuring the amount of radiolabel retained on sand samples. The bacteria were grown to stationary phase, harvested by centrifugation, and washed and resuspended in AGW (to a final density of 0.6–0.8 optical density (OD) units at A_{590}). Suspensions of strain R8 or M1 were labeled by adding ³H-leucine (1 μ Ci mL cells) or ¹⁴C-leucine (0.5 μ Ci mL cells), respectively, along with 10 μ L mL⁻¹ of PYE followed by incubation for 2 hours at 23°C with shaking (200 rpm). Subsequently, the cells were harvested by centrifugation, washed and resuspended in an equal volume of AGW, and then incubated without nutrient amendment at 23°C for a further 24 hours. The cells were again harvested by centrifugation and washed three times in AGW before being applied to the columns. After passing the bacteria through the columns as described above, the columns were frozen at -20°C for 24 hours and then cut into ~ 1 g transverse sections. Each section

Figure 2. Photographs of the experimental tank containing: (a) the first realization and (b) the second realization of the heterogeneous porous medium. The dark areas are the fine sediments that are coated with iron.

was added to 5 mL of Optiphase Hisafe 3 scintillation cocktail (Wallac), and bacteria were removed from sand particles by vigorous shaking (400 rpm) for 3 hours. Each sample was counted in a scintillation counter (Packard Tri-Carb 1600 TR), quench corrections were made, and cell density was determined. Cell numbers were determined from counts per minute using a previously prepared calibration curve.

2.3. Sampling in the Tank

Two experiments were run with two separate packings of the tank. For the first experiment, well 5, located 45 integral scales from the inflow reservoir (Figure 1), was used as the pumping

well and location for collection of outflow samples. Approximately 8×10^{12} cells of each strain (M1 and R8) were passed into the porous medium, concurrently with the chemical tracer, through the upgradient reservoir over a period of 28 min and with an injected volume of 7980 mL. In addition to samples taken at the well, samples were collected from the 23 septa at approximately 1.5, 7.5, and 11.5 hours after the start of the tracer pulse. Following termination of the experiment, the tank was rotated so that the $1.6 \text{ m} \times 0.67 \text{ m}$ face of the tank was horizontal. The front wall of the tank was removed, and samples were collected via vertical coring (i.e., parallel to the 0.094 m dimension of the tank) at 80 locations. All destructive sam-

Table 1. Sands Used in Packing the Flow Cell

Sand	Source of Sand	Hydraulic Conductivity, cm s^{-1}	Approximate Range of Grain Size, mm	Significant Surface Coating?	Percent of Medium
0	Oyster	1.61	2.4 ^a –4.8	no	0.05
1	Oyster	0.58	0.9 ^a –2.4	no	0.9
2	Accumin ^b	0.49	0.85–1.58	no	4.7
3	Accumin ^c	0.224	0.425–1.58	no	11.0
4	Accumin ^b	0.15	0.425–0.600	no	23.0
5	Accumin ^d	0.11	0.300–0.600	no	26.0
6	Accumin ^b	0.078	0.300–0.425	no	22.0
7	Oyster	0.041	0.425 ^a –0.600	yes	7.8
8	Oyster	0.030	0.300 ^a –0.425	yes	3.9
9	Oyster	0.015	<0.300	yes	0.6

^aAll sieving on the Oyster sediments was performed dry. As a result, fines with a smaller grain diameter than the minimum reported grain size are likely to be attached to grains retained on the sieves.

^bAs provided by manufacturer without further sieving.

^cMixture of sands 2 and 4.

^dMixture of sands 4 and 6.

ples contained ~1.5 cc of sediment and the accompanying fluid. These samples were homogenized prior to separation of the bacteria (described below).

For the second experiment, well 4, also located 45 integral scales from the inflow reservoir (Figure 1), was used as the pumping well and location for collection of outflow samples. For this experiment, $\sim 2.1 \times 10^{12}$ cells of strain M1 were passed into the porous medium, concurrent with the chemical tracer, through the upgradient reservoir over a period of 24 min, and with an injected volume of 6620 mL. Only strain M1 was used in this second experiment because of the very limited transport and enumerating difficulties encountered with strain R8 in the first experiment. In addition to samples at the well, samples were collected from 20 septa (the three remaining septa locations had been changed to wells prior to the second experiment). Samples were collected from these septa at times (relative to the start of the tracer pulse) of approximately 0.5, 1.1, 3.0, and 5.0 hours. No destructive samples were collected from this packing as additional experiments were in progress.

Strains R8 and M1 were enumerated in samples taken from the tank by differential plating. Serial dilutions of liquid samples were plated in triplicate on PYE-agar plates (15 g agar per 11 PYE) with either kanamycin (5 mg mL⁻¹) or rifampin (100 mg mL⁻¹). The combined density of strains R8 and M1 in each sample was determined by direct count of colonies on PYE-rifampin plates (both strains showing resistance to rifampin). The density of strain M1 in each sample was determined by direct count using PYE-kanamycin plates (strain R8 is not resistant to kanamycin). Finally, the density of strain R8 in each sample was determined by subtracting the density of strain M1 from the combined density of strains R8 and M1.

Bacteria attached to sand removed during destructive sampling following the first experiment were assessed by dislodging the bacteria from the sand and subsequent differential plating. The samples were placed in 10 mL of extraction buffer (0.1% sodium pyrophosphate, 0.1% triton X-100) and vigorously shaken for 3 hours (300 rpm). Aliquots of the extraction buffer were then used for differential plating. Extraction efficiencies determined independently for each of the 10 sands were used to adjust all cell counts obtained from the destructive samples.

It is noted here that determination of recovery percentages of bacteria for the tank experiments was found to be unreliable and is not reported here. The reasons are twofold. First, it is

recognized that no extraction method for removing live bacteria from a mineral surface is 100% efficient. Thus it is difficult to assess the exact numbers of bacteria that remain attached to the media (the methods used here were, however, based on optimal removal of cells and were consistent across replicate experiments). Second, the heterogeneity and limited sampling performed in the large tank made spatial estimation of the numbers of bacteria attached to the sands quite subjective.

3. Experimental Results

3.1. Small-Scale Sand Columns

It was observed that the transport of strain M1 was generally greater than or equal to the transport of strain R8 in all sands (Figure 3). The greatest transport differences between strains R8 and M1 were with AS (sands 2–6), with strain M1 being transported with a significantly higher efficiency than strain R8. No transport of strain M1 was detected with the fine OS size classes (sands 7–9); however, transport of strain R8 within these size classes also was severely limited. Both strains were transported at relatively high efficiencies through the coarse OS size classes (sands 0 and 1).

The results for the destructive samples obtained from the small columns filled with OS demonstrated that both strains R8 and M1 were retained in greater numbers near the top of the column (Figure 4). Very low numbers of strain R8 were observed in the lower half of the column. Numbers for strain M1 also decreased markedly, but to a lesser degree than observed for strain R8. For the AS, both strain R8 and strain M1 were distributed throughout the column with a slight decline in numbers with distance into the column. For the AS the numbers observed for strain R8 were generally greater than or equal to the numbers for strain M1. The distribution and number of cells were similar whether the cells were initially added as pure cultures or as a mixture of the two strains. Therefore the presence of the second strain did not appear to enhance or interfere with the attachment of the first.

3.2. Experiments in the Heterogeneous Media

Figure 5 shows breakthrough curves for sodium chloride and strain M1 from the first tank experiment. Strain R8 was either not detected in samples, or their numbers were low and incon-

Figure 3. Percentage of applied cells of (a) strain R8 and (b) strain M1 eluted from the different classes of sand as listed in Table 1. "SGW" refers to a control without sediment and containing only the silanized glass wool used to support sediments in the experimental columns. "Oyster Sand" refers to unsieved sand from the Oyster site. "Accu Sand" refers to Accumin sand used in prior biological transport studies with a grain size distribution similar to sand 6. The error bars represent ± 1 standard deviation as determined from triplicate experiments for each sand.

sistent among replicates, thus leading us to conclude that the data for strain R8 were unsuitable for analysis. The concentrations given in Figure 5 have been normalized to the total mass injected. The shape and timing of the sodium chloride curve is quite consistent with previous results [Silliman, 2001], where a series of chemical breakthrough curves at wells 4 and 5 were compared and analyzed. The curves in Figure 5 also are consistent with the shape and timing of the strain M1 and sodium chloride breakthrough curves for the second tank experiment.

Three significant features of the chemical and bacteria curves (Figure 5) are noted. First, the timing of the initial peak of the bacteria breakthrough was the same as that of the sodium chloride. Second, the bacteria breakthrough curve showed a significant tail with a near-uniform concentration for times >200 min after injection. Third, the peak normalized concentration of the bacterial breakthrough was an order of magnitude lower than the peak normalized concentration of the sodium chloride.

A comparison of the breakthrough curves for strain M1 in the two experiments demonstrates several notable features (Figure 6). First, the timing and (normalized) peak concentration of the initial portion of the breakthrough were approximately the same for the two experiments. Second, both experiments showed significant tailing at later times. Third, the

Figure 4. Representative retention of strains R8 and M1 in columns containing OS and AS sands. Counts are in bacteria per gram of dry sediment. Distance is measured from the inflow end of the column: (a) retention on AS and (b) retention on OS. Triangles represent strain R8; circles represent strain M1.

width of the peak was somewhat narrower, and the concentration in the tail portion was lower for the second experiment.

The concentration of strain M1 in fluid samples from different septa changed with time, illustrating movement of bacteria with respect to the inflow boundaries. As shown in Figure 7, for the second experiment, the spatial distribution of bacteria settled into a consistent pattern for the samples collected >2 hours after the injection of the bacteria. Figure 8 shows, for the first experiment, the distribution of strain M1 on the solids as a function of distance from the inflow boundary. The numbers of bacteria on the solids generally showed high spatial variability, with the numbers of bacteria being particularly high in the

Figure 5. Breakthrough curves at well 5 for NaCl (solid line) and strain M1 (circles) for the first realization. The concentrations have been normalized to the total mass injected.

Figure 6. Comparison of the breakthrough curves for strain M1 in the two realizations. The concentrations have been normalized to the total mass injected. First experiment is shown by circles, and second experiment is shown by triangles.

fine-grained sediments: 2–5 orders of magnitude above the concentrations observed in the remainder of the tank.

4. Interpretation of Results

The bacteria breakthrough curves obtained in the two tank experiments are quite similar to those obtained from laboratory experiments with homogeneous media that have been reported in the literature [e.g., Johnson *et al.*, 1995; Hendry *et al.*, 1999]. Specifically, these curves show initial breakthrough of the bacteria timed quite similarly to that of the conservative chemical tracer (compare, for example, with Figures 1 and 2 of Hendry *et al.*, [1999]). Further, these breakthrough curves show a nearly constant bacterial concentration in the late time portion of the breakthrough, consistent with the observations of both Hendry *et al.* [1999] and Johnson *et al.* [1995] (who term

Figure 8. Distribution of the concentration of strain M1 cells on the solid phase (cells per gram) for the first realization. The horizontal axis represents the horizontal (1.6 m) axis in the tank. The samples collected in sands 7–9 are denoted by circles, and the remainder of the samples are denoted by triangles.

this late time concentration of bacteria an “elution plateau”). This demonstrates that the transport process at large scale (relative to the integral scale) in these heterogeneous media showed fundamental characteristics similar to those of a homogeneous medium.

These results also are consistent with some, but certainly not all, field observations. The field results of Harvey *et al.* [1993], for example, show very similar breakthrough behavior for their bacteria (relative to the conservative chemical tracer) at two of the depths sampled at their field site (9.0 and 9.5 m). Their results for a third sampling depth (8.8 m), however, provided bacteria breakthrough times quite inconsistent with the observed arrival of the bromide tracer. Similar comparisons with our results can be made with other field studies, such as the bacteriophage experiments reported by Ryan *et al.* [1999]. Hence, while the present results may provide insight into certain aspects of the field transport problem, these experiments clearly do not represent the range of behaviors one is likely to observe in the field.

The bacteria breakthrough curves for the two realizations showed good reproducibility. Specifically, the peak concentrations in these two experiments were quite similar, the timing of the initial peak was the same, and despite a significant post-peak decline in the second realization, the concentrations at the well were similar at late time. This result is quite significant as it provides initial evidence that bacterial transport, at least under select conditions, may show consistent large-scale behavior that may be a function of the statistical description of the conductivity/mineralogy, but that is not a function of the details of each realization of these fields. Although conclusions regarding large-scale behavior would be premature at this point, these results suggest that large-scale, mean behavior may exist in these media (as suggested by Rehmann *et al.* [1999] on the basis of stochastic theory for viral transport).

There remains, however, some distinct differences between the two results shown in Figure 6. Although the peak bacterial concentration for the second realization was quite similar to that of the first, the width of the bacterial peak for the second curve was narrower than that of the first. Further, the decline in bacterial concentration following the peak was substantially greater for the second realization than for the first. Finally, the

Figure 7. Distribution of concentration of strain M1 cells in the fluid phase (cells per mL) for the second realization. Three sampling periods after the start of injection of the cells are shown: 85 min (circles), 185 min (triangles), and 305 min (diamonds). The horizontal axis represents the horizontal (1.6 m) axis in the tank. One point at 11 cm from the inflow boundary sampled at 85 min was truncated from this plot (due to a concentration ~ 3 orders of magnitude higher than the remainder of the data).

second realization showed a delayed secondary arrival of bacteria in the form of a slow increase in the bacterial concentration in the outflow fluid at late time.

While the reduced width of the breakthrough curve for the second experiment (relative to the width for the first experiment) may be related to the reduced width of the input pulse of bacteria in the second experiment, the remaining differences in postpeak behavior of the two curves are not as easily explained. It is possible that the presence of strain R8 in the first experiment had some influence on transport of strain M1. However, as mentioned above, we concluded from the column experiments that the presence of strain R8 did not impact the transport behavior of strain M1. Hence we suggest that any interference effect between strains R8 and M1 is unlikely to be the primary mechanism leading to the differences in these two results. Further, the lack of nutrients and short duration of the experiments make it unlikely that growth or death of bacteria played a significant role in either of these experiments. We conclude therefore that the observed differences in the strain M1 breakthrough curves in the two tank experiments must be related to the interaction among the flow field, the bacteria, and the spatial distribution of the sands. Hence, while it may be suggested that certain characteristics of the bacterial transport may be approaching a large-scale, averaged behavior (as indicated in the previous paragraph), the portion of the bacterial breakthrough curve beyond the initial peak continues to show significant sensitivity to the details of the spatial distribution of the sands.

A second set of observations relate to the numbers of bacteria in the fluid samples and those remaining on the solids at the termination of the first experiment. As indicated by the data shown in Figure 7, the concentration of bacteria in the fluid phase approached an approximate steady state (the later time concentrations declined at a very slow rate). Further, and again at late time, the concentration showed a consistent spatial pattern with slightly higher concentrations near the inflow boundary and generally decreasing or constant concentrations with increasing distance from this boundary. The concentration in the fluid phase near the well was also consistent with the concentration observed in the well. This general spatial pattern of nearly uniform concentration at late time (total variability in concentration was ~ 1 order of magnitude) and consistency between the fluid samples and the concentration observed at the well were observed in both experiments.

In contrast to the minimal variation in concentration in the fluid phase, the destructive samples collected after completion of the first experiment showed >5 orders of magnitude variation in the concentration of strain M1 remaining on the solids: the highest concentrations were associated with the low permeability/high iron sediments (Figure 8). This result is generally consistent with those of the small column experiments (Figures 3 and 4) in which it was demonstrated that strain M1 attached in higher numbers (near the column inflow) and had substantially reduced ability to transport in the OS as compared to the AS.

The contrast between the concentration in the fluid phase and the concentration on the solid phase is consistent with the discussion by *Alfreider et al.* [1997], in which they infer from field samples that relatively uniform concentrations of bacteria in fluid samples may coexist with tremendous spatial variation in concentrations in the attached microbial populations. This distribution of bacteria on the solids and in the fluid is also consistent with initial numerical simulations of our experi-

ments using a kinetic sorption-desorption model (L. Zheng, Kansas Geological Survey, Personal Communication, 1999).

The different attachment behaviors of strains M1 and R8 in the small column experiments on these two different sediment types also illustrated the significance of substratum chemistry in assessing comparative bacterial attachment characteristics [*Wiencek and Fletcher*, 1995]. Strain R8 demonstrated a higher attachment ability than M1 with the AS sediment, while both strains attached readily to the high-iron OS sediments. It is premature to speculate on the mechanisms for these observed differences in attachment to the two sediment types, but they are presumably related to the compatibility in chemistries on the particle and bacterial surfaces [*Fletcher*, 1996].

These results suggest two implications of the difference between sampling from the solid phase and sampling from the fluid phase. First, as noted by *Alfreider et al.* [1997], sampling from the fluid phase will provide a measure of the spatial variation in bacterial concentrations that (1) represents an average of the microbial behavior in the porous medium over a scale which may be quite large relative to the local heterogeneities and (2) does not reflect the variation observed on the solids. Second, it is likely that following injection of a pulse of bacteria into a heterogeneous medium, the bacteria on the solids will represent a continuing source of bacteria for the fluid phase, such that bacteria are likely to be found in fluid samples (albeit at low concentrations) well after the initial breakthrough and subsequent decline in concentration has occurred. In our case, we suspect that the late time tail was due primarily to release from the lower conductivity zones. This appears consistent with the preliminary modeling performed relative to these experiments (L. Zheng, Kansas Geological Survey, personal communication, 1999). In the field it is likely that the concentration in the fluid phase will be a function both of release and growth of the bacteria remaining in the system. Both of these mechanisms would be tempered by the rate of death of the bacteria.

5. Conclusions

A pair of biological transport experiments have been conducted in two realizations of a porous medium consisting of exponentially correlated hydraulic conductivities. The results of these experiments provided several significant observations, including the following.

1. The two experiments produced bacterial breakthrough curves that were quite similar, even though the similarity between the two porous media was limited to first- and second-order statistical moments. This result appears consistent with the concept of large-scale average behavior previously observed for the transport of conservative chemicals.

2. The transported bacteria arrived at the outflow port simultaneously with the conservative chemical tracer (although at significantly lower concentration).

3. The bacteria breakthrough curves showed significant late time tailing. This tailing was probably related to high concentrations of bacteria remaining in regions characterized by small grain size, low hydraulic conductivity, and high degree of iron-oxide coating.

4. The behavior of the bacteria breakthrough, compared to the breakthrough of the conservative chemical tracer, was consistent with observations made in the laboratory on homogeneous sediments and with select field observations.

5. The concentrations of bacteria attached to the sediment

surfaces showed considerably more spatial variation than the concentrations of bacteria in the fluid phase did.

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