



RESEARCH LETTER

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Key Points:

- Non-Gaussian transport behavior at the MADE site is unraveled
- ADE can reproduce tracer transport in heterogeneous aquifers with no calibration
- New fractal method generates heterogeneous K fields with adequate connectivity

Supporting Information:

- Readme
- Table S1
- Figure S1
- Figure S2
- Figure S3

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Predicting flow and transport in highly heterogeneous alluvial aquifers

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Abstract Successful prediction of groundwater flow and solute transport through highly heterogeneous aquifers has remained elusive due to the limitations of methods to characterize hydraulic conductivity (K) and generate realistic stochastic fields from such data. As a result, many studies have suggested that the classical advective-dispersive equation (ADE) cannot reproduce such transport behavior. Here we demonstrate that when high-resolution K data are used with a fractal stochastic method that produces K fields with adequate connectivity, the classical ADE can accurately predict solute transport at the macrodispersion experiment site in Mississippi. This development provides great promise to accurately predict contaminant plume migration, design more effective remediation schemes, and reduce environmental risks.

1. Introduction

Predicting flow and transport in heterogeneous alluvial aquifers is one of the grand challenges facing society [Cantor, 1997]. Complex solute transport behavior in such aquifers has fueled an ongoing debate in the scientific community for decades [Dagan, 1989; Cushman, 1990; Dagan et al., 1992; Rehfeldt et al., 1992; Zheng et al., 2011] because existing modeling approaches have failed to reproduce key components of observed field tracer tests in highly heterogeneous systems.

Subsurface pollutant transport is almost always simulated using the advective-dispersive equation (ADE), which describes transport by bulk flow (advection) and spreading (dispersion). Advection is mainly controlled by K according to Darcy's law, while dispersive spreading is described as a Fickian process with the effective dispersion dependent on the fluid velocity. K is the major property that governs contaminant migration in aquifers. Accurate prediction of plume migration requires precise and high-resolution measurements of K , which may vary over many orders of magnitude within a few centimeters [Molz et al., 1986]. Because such high-resolution in situ K data cannot be measured for the entire subsurface environment, numerical facsimiles of K fields are generally developed using stochastic methods. A range of methods have provided successful simulations of contaminant transport using the ADE in mildly heterogeneous aquifers in Borden, Ontario [Freyberg, 1986; Mackay et al., 1986]; Cape Cod, MA [Garabedian et al., 1991; LeBlanc et al., 1991]; and Schoolcraft, MI [Hyndman et al., 2000; Phanikumar et al., 2005].

Modeling flow and transport in highly heterogeneous aquifers has proven to be much more challenging [Eggleston and Rojstaczer, 1998; Whittaker and Teutsch, 1999]. The macrodispersion experiment (MADE) site in Columbus, Mississippi, is one of the most studied highly heterogeneous sites in the world (with a variance of $\ln K = 6.6$) [Dogan et al., 2011; Bohling et al., 2012]. Natural gradient tracer tests at this site have fueled a spirited debate about the applicability of the classical ADE models for the simulation of contaminant transport in highly heterogeneous aquifers, because such modeling approaches could not reproduce the observed transport using only measured field properties.

Three large-scale tracer tests were performed at the MADE site. Here we focus on MADE 1, conducted from October 1986 to June 1988 [Boggs, 1991]. The experiment involved injecting a calcium bromide tracer solution into five wells, aligned approximately perpendicular to the natural groundwater flow direction. More than 11,000 water samples were collected from 258 monitoring wells (Figure 1) in eight synoptic “snapshots”

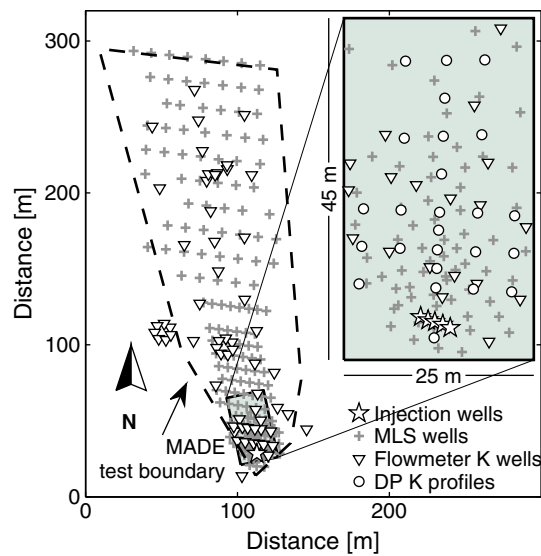


Figure 1. Map of the MADE site with the locations of tracer injection, multilevel sampler, flowmeter K wells, and DP K profiles. The MADE 1 test boundary is indicated by the dashed line; the rectangle shows the model domain used for simulations. Locations of DP K profiles outside the model domain have been excluded for clarity.

center of mass location in map view. In addition, some of these methods are difficult to use in a predictive sense, as they are not easily parameterized based solely on field data. A recent stochastic multi-indicator model was developed with summary statistics of the direct-push (DP) K data set [Bohling *et al.*, 2012] used here to simulate transport at the MADE site. The resulting one-dimensional (1-D) evaluations of solute mass reproduced some aspects of the longitudinal distribution of solute mass at the MADE site [Fiori *et al.*, 2013]. The simulated K field in that study was not conditioned on the K measurements; in this paper we condition on the DP K measurements for the presented three-dimensional (3-D) simulations. Here we present the first successful 3-D deterministic transport model for this site using only the classical ADE without calibration of K values.

2. Methods

The DP K data were collected using a recently developed tool [Liu *et al.*, 2009] that can collect a 10 m long profile in approximately 2 h, providing K estimates across the site at an unprecedented speed, with a high 1.5 cm vertical resolution [Liu *et al.*, 2009; Bohling *et al.*, 2012]. This resolution is approximately an order of magnitude finer than previous studies at the site (~15 cm intervals in the flowmeter data) and orders of magnitude finer than is common for characterizing contaminated sites. This data set allowed us to analyze the statistical properties in detail. DP K profiles were used to study the autocorrelation functions for defining the average fractal dimension and correlation lengths [Meerschaert *et al.*, 2013]. We utilized this information with a multiscaling fractal lognormal hydraulic conductivity field generator [Benson *et al.*, 2013] to populate high-resolution 3-D K fields for flow and transport simulations (Figure 2). Each equally likely realization of the stochastic K field was conditioned on either the measured DP K or lower-resolution flowmeter K data [Meerschaert *et al.*, 2013]; the realizations share the fractal dimension and correlation lengths from the DP data because the density of the flowmeter data was insufficient for accurate estimation of those quantities. This was done to assess the importance of the conditioning data; the flowmeter data set includes information from locations that could not be accessed during the DP work because the injection site was disturbed during the MADE 3 tracer test. In addition, the DP tool used here had an upper K limit of 60 m/d; the tool will produce inaccurate K values above that limit. Conditioning on the flowmeter data set enables us to exploit the high-resolution correlation information from the DP logging while avoiding the high K limits of the DP K tool used at this site. The mean and variance of the observed $\ln K$ data are -1.6 and 8.5 for DP K and 0.74 and 2.9 flowmeter $\ln K$ data, respectively (see Table S1 in the supporting information for additional details and comparisons with stochastic fields). We followed the field data collection procedure to sample our simulated fields along vertical profiles with comparable resolution to the data sets.

spanning 20 months and analyzed for tracer concentrations. The resulting data showed a highly asymmetric tracer plume in which most of the mass stayed near the injection area, but a portion of the plume was rapidly transported far downgradient.

Several modeling approaches have been proposed to simulate the observed plume behavior at the site including preferential flow paths [Zheng and Gorelick, 2003], placement of hypothetical high and low K zones with optimized parameters [Barlebo *et al.*, 2004], overlapping mobile and immobile transport domains [Harvey and Gorelick, 2000; Schumer *et al.*, 2003], and replacing the Fick's law dispersive second derivative term in the ADE by a nonlocal fractional derivative [Benson *et al.*, 2001; Zhang and Benson, 2008].

Although some of these approaches reasonably represented the average, or upscaled, plume behavior, they did not accurately replicate important plume characteristics, including spatial extent and

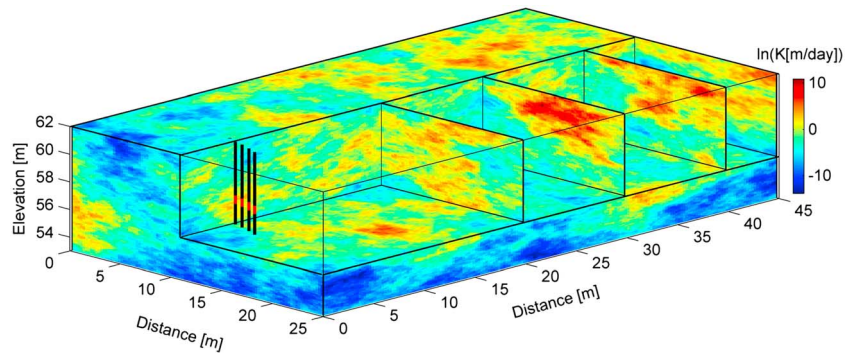


Figure 2. A sample 3-D K field created using DP K data. This volume corresponds to the saturated portion of the model domain that was used for flow and transport simulations. The red rectangles marked on black lines show the tracer injection interval for the MADE 1 and 2 tests.

The flow and transport model domain encompasses a region with dense DP K data near the MADE 1 tracer injection area (Figure 1), which held more than 93% of the recovered mass at the 503 day snapshot. The model grid was defined with over 3.2 million $0.25 \times 0.25 \times 0.05$ m (length \times width \times height) cells oriented with the long axis parallel to the average downgradient direction of the observed MADE tracer plumes. This 25×45 m domain is smaller than the horizontal extent of the entire tracer experiment (Figure 1) but is sufficiently large to capture the asymmetric plume behavior with peak concentrations near the injection point and strong downgradient tailing. The east and west edges of the model are approximately along the average flow direction and thus assumed to be no-flow boundaries. The northern and southern edges of the model were assumed to be constant head boundaries and were assigned using an interpolated plane of the average measured heads collected during the MADE 1 test. The maximum measured water level increase (0.64 m) across the five injection wells [Rehfeldt et al., 1992] was used to test the suitability of the simulated K fields. Injection rates were calculated to be proportional to the K values in each cell, and water level increases from steady state flow simulations were downscaled to each borehole using the Thiem equation. Of the 55 generated stochastic K fields, the nine that provided the maximum simulated head increases across the five injection wells within 25% of the measured maximum (0.64 ± 0.16 m) were selected for transport simulations.

Transport simulations based on the classical ADE were conducted using MT3D [Zheng and Wang, 1999] employing a third-order total variation diminishing scheme, which is mass conservative with minimal numerical dispersion. Necessary parameters were defined as follows: longitudinal dispersivity of 0.05 m [Gelhar, 1993], transverse horizontal and vertical dispersivity ratios of 0.1 and 0.01, effective molecular diffusion coefficient of 10^{-6} cm²/s, isotropic K within each cell, and porosity of 0.35 [Adams and Gelhar, 1992].

The 503 day snapshot was chosen for the comparison of simulated and observed plumes, as it is the most commonly presented in publications with MADE 1 simulations. Mass distributions for experimental data and

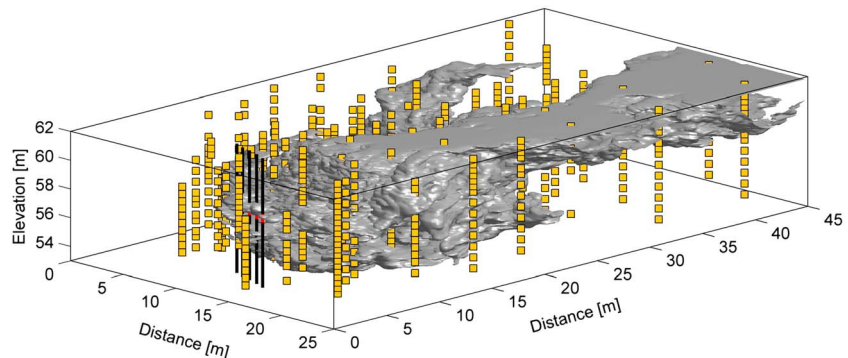


Figure 3. Snapshot from a sample transport simulation at $t = 503$ days. The 3-D volume shows the boundary of measurable concentration (0.01 mg/L for the snapshot). The yellow squares indicate the locations of multilevel samplers that were used to sample the plume.

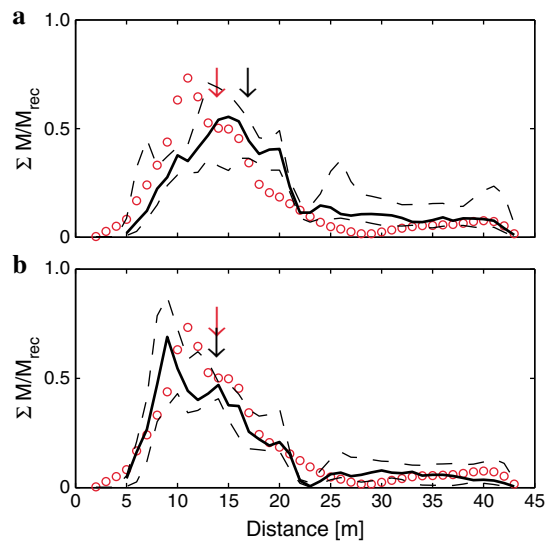


Figure 4. One-dimensional relative mass distribution profiles 503 days after the start of injection with center of mass locations marked with red (observed plume) and black (simulated plume) arrows. (a) The median of nine DP *K*-conditioned simulations (black line), with 25% and 75% quartiles given by dashed lines. The red circles show the profile for the MADE 1 observations. (b) The median of nine flowmeter *K*-conditioned simulations.

the simulations were normalized with the total recovered mass for each case. The simulated and measured concentrations were integrated vertically and then interpolated in 2-D using a 1×1 m (dx, dy) grid and then summed horizontally (along y axis) to create 1-D relative mass distribution profiles, as is commonly done for this site [Adams and Gelhar, 1992; Zheng et al., 2011; Benson et al., 2013; Fiori et al., 2013]. To replicate the experimental procedure, we sampled the simulations at the locations and depths of every multilevel sampler (MLS) used in the tracer test (Figures 1 and 3) and considered only those concentrations larger than the instrument detection limits (0.01 mg/L).

3. Results

The median of the simulated mass profiles is very similar to the experimental data for models based on both the DP *K* and the flowmeter *K* data (Figure 4 and Figure S1 in the supporting information), with similar plume locations (Table 1), extent of peaks, and

tailing behavior. Simulated results would likely be improved by calibrating porosity, but this would reduce the predictive power of the approach. The simulated plume based on the flowmeter *K*-conditioned stochastic fields was closer to the observed plume in terms of the 1-D relative mass profiles (Figure 4), the locations of center of mass (Figure 5, Table 1, and Figures S2 and S3 in the supporting information), and the horizontal and vertical displacement of the center of mass (Table 1). This is likely due to the availability of flowmeter *K* data very close to the injection wells (not possible for the DP *K* measurements as mentioned earlier) and the inaccurate *K* values produced by the DP *K* tool above its upper limit of 60 m/d.

Figure 5a shows a map of vertically integrated relative mass for the experimental data, in which the majority of mass stayed near the injection area. We provide the median of nine simulations conditioned on DP *K* data in Figure 5b and the flowmeter data in Figure 5c for comparison. The simulated plume extent for DP *K* (18×15 m, Figure 5b) and flowmeter *K* (17×15 m, Figure 5c) are both similar to what was measured (22×17 m, Figure 5a). The measured location of the maximum mass is also similar to that of the simulated plumes. Individual simulated tracer plumes are shown in Figures S2 and S3 in the supporting information.

The agreement between these plume characteristics in multiple dimensions provides strong evidence that the classical ADE can predict complex pollution plume behavior, even in highly heterogeneous aquifers, when high-resolution *K* data are collected and stochastic methods are used that provide adequate connectivity in *K* fields.

Table 1. Center of Mass Coordinates and Displacement of This Center of Mass for the Observed and the Median of the Nine Simulated Tracer Plumes Conditioned on DP *K* and Flowmeter *K* Data, Respectively

	Center of Mass (m)			Displacement (m)	
	x	y	z	Horizontal	Vertical
Observed	10.8	12.9	59.0	7.99	+1.22
DP <i>K</i> conditioning	14.5	15.9	58.1	11.23	-0.31
Flowmeter <i>K</i> conditioning	13.6	12.8	59.5	8.02	+1.73

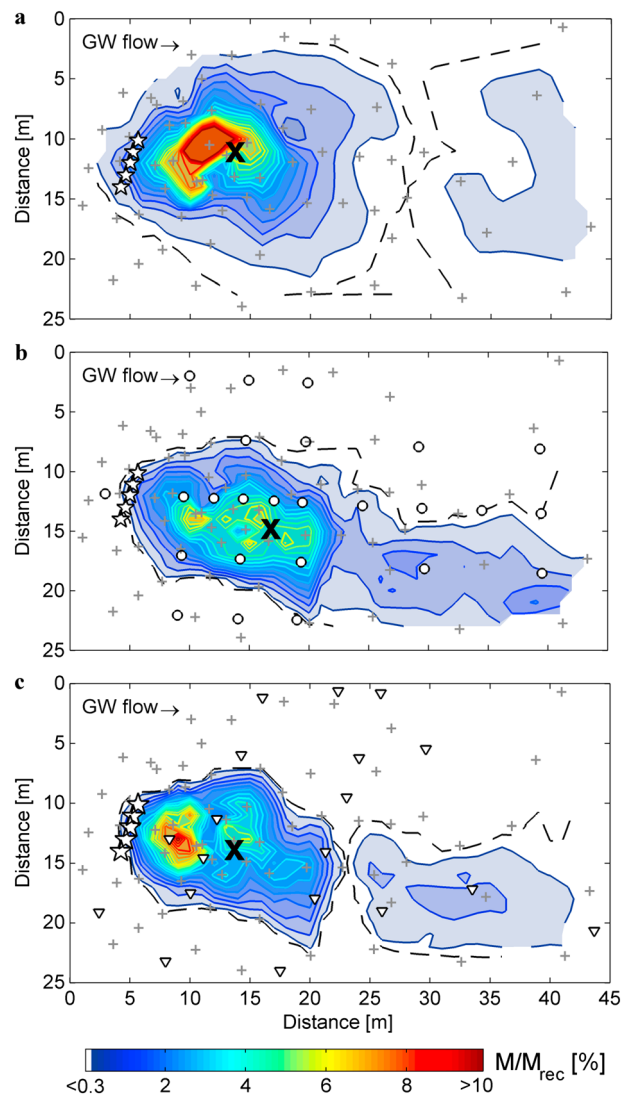


Figure 5. Vertically integrated contour maps of relative mass 503 days after the start of injection with center of mass locations marked with a black cross. (a) MADE 1. (b) The median of the nine DP K data-conditioned simulations that met the head change criterion. The injection locations are shown as open stars; the DP K locations are shown as open circles. (c) The median of nine flowmeter K -conditioned simulations that met the head change criterion. The flowmeter K locations are shown as open triangles. All contour maps were created using the interpolation procedure discussed in the text (the black dashed line shows the 0.1% contour level, and gray plus signs indicate the MLS borehole locations).

In this paper, we demonstrate that the classic ADE can reasonably reproduce observed plume behavior at the MADE site. Our analysis also demonstrates that modeling approaches are commonly limited by inadequate aquifer characterization data. Our approach uses data from novel DP tools and previously performed flowmeter logging, coupled with a fractal interpolation method conditioned to the measured K data, to overcome this long-standing limitation. This study demonstrates the value of high-resolution K data for solute transport simulations. Although our assessment focused on a portion of the MADE site, the implications of our results extend far beyond that region to heterogeneous aquifer sites around the world. The transport models successfully reproduced nearly all aspects of the observed tracer plume without calibration of K values. This includes the heavy tails, where most of the mass is near the injection point but some moves far downgradient; features that have previously only been reproduced using transport theories such as dual domain mass transport or those that superimposed high K networks on simulated K fields. Predictive solute transport based solely on field measurements of K data, such as done here, has immense value for improving the ability to design more effective remediation schemes.

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