ANALYSIS OF TRACER MIGRATION IN A DIVERGING RADIAL FLOW FIELD

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REFERENCE: *Proceedings of the 2007 Georgia Water Resources Conference*, held March 27–29, 2007, at The University of Georgia.

Abstract. Hydrodynamic dispersion is an important factor controlling contaminant migration in the subsurface environment. However, few comprehensive data sets exist for critically evaluating the impact of travel distance and site heterogeneity on solute dispersion. Therefore, a series of field-scale experiments using tritiated water (^3H_2O), and bromide (Br^-) as tracers was conducted on the U.S. Department of Energy's Savannah River Site. For each experiment, tracer-free groundwater was injected at a fixed rate of 56.7 L min^{-1} to establish a forced radial gradient prior to the introduction of a tracer pulse. After the tracer pulse, the forced gradient was maintained throughout the experiment using non-labeled groundwater. Tracer migration was monitored using six sampling wells radially spaced at approximate distances of 2.0-, 3.0-, and 4.5-m from the injection well. Each sampling well was further divided into three discrete sampling depths that were pumped continuously throughout the course of the experiments. Longitudinal dispersivity (α_L) and travel times for ^3H_2O were estimated by fitting the field data to analytical approximations of the advection-dispersion equation (ADE) for uniform and radial flow. Dispersivity varied greatly between wells located at similar transport distances and between zones within a given well. The radial flow equation described ^3H_2O breakthrough better than the uniform flow solution, yielding lower α_L values while accounting for breakthrough tailing inherent to radial flow conditions. Temporal moment analysis confirmed the retardation of Br^-, generally considered to travel in a conservative manner, despite data truncation due to extensive tailing that biased retardation estimates when compared to ^3H_2O. Despite retardation and incomplete mass recovery, both ADE models were able to reasonably describe the Br^- data without accounting for sorption reactions, indicating that chemical interactions with the geologic matrix may be misinterpreted in terms of a physical transport process.

INTRODUCTION

A fundamental understanding of the physical processes associated with groundwater flow in the subsurface is critical for evaluating the potential hazards associated with subsurface contamination, and developing cost-effective corrective actions that reduce such risks. Groundwater tracer experiments using compounds assumed to be non-reactive with the aquifer matrix are conducted to improve our understanding of the physical processes controlling solute or contaminant migration by eliminating or reducing the impact of various chemical reactions such as precipitation and adsorption that complicate data analysis and interpretation. In such a case, the position of the tracer plume becomes a manifestation of the physical properties of the transmissive zone and not a function of a chemical interaction between the tracer and the porous media.

Recent studies, however, have suggested that anionic solutes such as bromide (Br^-) and chloride (Cl^-), often considered conservative (i.e. non-reactive) with respect to migration behavior in groundwater transport studies, may be significantly retarded (Seaman, 1998; Seaman et al., 1995; Seaman et al., 1996). In addition, researchers often incorrectly apply analytical solutions derived for uniform flow fields to systems displaying non-uniform flow (Gelhar et al., 1992; Welty and Gelhar, 1994). Therefore, the objective of the current study was to evaluate the transport behavior of ^3H_2O and Br^- in an injection well system as a function of travel distance and depth within the receiving aquifer.

EXPERIMENTAL DESIGN

The Injection Test Site (ITS), located on the U.S. Department of Energy's Savannah River Site (SRS), consists of a 15 cm ID central injection well (IW) screened throughout the water-table aquifer (≈4.6 m screened interval) and surrounded by six sampling wells screened over the same interval and radially spaced at approximate distances of 1.5 (S1 and S4), 3 (S2 and S5), and 4.5 meters (S3 and S6) from the injection well (Fig. 1). Well Obs1, located within the study site and screened within the first confined aquifer underlying the water table aquifer, was used for monitoring water depth to confirm the effectiveness of the confining layer in restricting injection of the treated groundwater to the water-table aquifer.

To avoid the introduction of colloidal artifacts (i.e. drilling mud) and the creation of preferential flow paths between closely spaced wells or sampling zones within a given well, the monitoring wells were installed using a hollow-stem auger. After augering to the desired depth (19 m), the well casing was inserted within the auger stem and the auger plug was displaced prior to withdrawing the

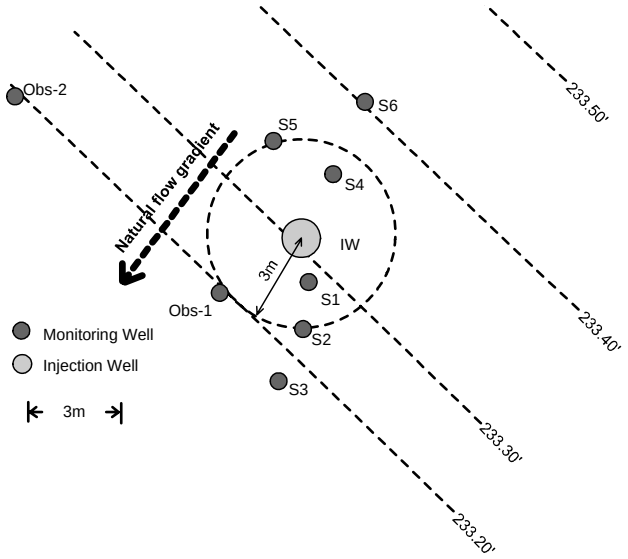

Fig. 1. Relative location of the injection well (IW) and various sampling wells (S1-S6) at the Injection Test Site (ITS).

stem from around the well casing. This allowed the formation to naturally fill in around the well casing.

The screened interval for each monitoring well was further divided into three discrete sampling zones or depths, with the deepest zone designated as Zone 1, using an inflatable packer system. Each of the three sampling zones within a well was equipped with a bladder pump that could be pumped independently or in unison with one or both of the other pumps within a well. Each tracer experiment consisted of injecting tracer-free water through the central well for approximately 24 hours at a fixed rate of 57 l min^{-1} to establish a radial gradient. Although nine tracer experiments were conducted, the current discussion will be limited to one extended breakthrough experiment consisting of 31,720 liters of a mixed tracer solution containing tritium (2,000 pCi mL^{-1}), and Br^- (3 mmol L^{-1}). After injection of the tracer pulse, injection of tracer-free water continued for approximately 7 days to maintain the radial gradient and displace the tracers past the most distant sampling wells within the test site. A more extensive discussion of the tracer results is presented in Seaman et al., (2007).

During each experiment, sampling zones within each well were pumped continuously at a minimal velocity ($\leq$ 150 mL min^{-1}) to reduce the impact of sampling on the groundwater mounding associated with injection, the volume of groundwater brought to the surface, and the generation of turbid groundwater samples associated with pumping at elevated flow rates. Groundwater samples were collected periodically from each of the 18 sampling zones for tracer analysis using standard methods.

Data Interpretation

Welty and Gelhar (1994) provided a number of approximate analytical solutions for describing tracer breakthrough under several commonly encountered non-uniform flow conditions (i.e., converging and diverging radial flow fields, and two well tracer tests) for two common inlet boundary conditions, a step input tracer test and an instantaneous Dirac pulse. In most experiments, however, the initial step change in tracer concentration is maintained for some practical duration that may be considerably less than required to observe full tracer breakthrough (i.e., $C/C_o = 1$) within even closely spaced monitoring wells at the field scale. Therefore, the analytical solution for an arbitrary pulse duration was solved through superposition using the appropriate step input approximation, assuming tracer displacement can be viewed as two distinct step inputs, the initial tracer solution (1) followed by the breakthrough of non-labeled water (2), separated by a known time interval equivalent to the pulse duration.

Under radial flow conditions hydrodynamic dispersion (D) becomes:

$$D = \alpha_L |v| + D^* \tag{1}$$

where $|v|$ is the absolute magnitude of the seepage velocity, and movement of the center of tracer mass in a radial flow field is proportional to $\sqrt{t}$. Considering only α_L in a radial system and assuming that molecular diffusion is insignificant, i.e., $D^* << \alpha_L v$, the advection dispersion equation (ADE) can be written as follows:

$$\frac{\partial C}{\partial t} = \alpha_L v \frac{\partial^2 C}{\partial r^2} - v \frac{\partial C}{\partial r} \tag{2}$$

with r representing radial distance and C representing solute concentration.

Estimates of longitudinal dispersivity (α_L) and travel time (t) for tracer breakthrough within each zone of a given sampling well were obtained using the Levenberg-Marquardt method in MATLAB (Mathworks, Inc., Natick, MA) to minimize the sum of squared residuals between the observed data and the analytical approximations, assuming the appropriate boundary conditions. For starting values, the average tracer arrival time estimate was based on the maximum tracer peak arrival time for each sampling zone, and an initial α_L value of 0.1 m.

Tritium recovery at the monitoring wells was estimated by comparing the area of the tritium breakthrough curve to that of the initial tracer pulse of a defined duration assuming ideal radial flow in a confined aquifer with a fully-penetrating injection well screened zone. Temporal moment analysis was used to compare tritium breakthrough behavior relative to that of Br^- because it is does

not require the same assumptions necessary to fit the data to a specific transport model.

RESULTS AND DISCUSSION

It is clear from the tritium breakthrough data that flow patterns within the study site were quite complex (Fig. 2), indicative of considerable three-dimensional variability in the hydraulic conductivity of the water-table aquifer at the study site. For visual comparison, the well data has been arbitrarily grouped in columns according to relative location with respect to the injection well and the natural flow gradient, with S1 through S3 located downgradient and S4 through S6 located upgradient of the injection well, IW. Data for wells located similar radial distances from IW are presented side-by-side. Note, however, that a strong radial gradient was maintained throughout each tracer experiment.

Tritium breakthrough histories indicate significant variability in terms of tracer migration velocity and maximum breakthrough concentration between wells at similar radial distances (i.e., S1 vs. S4, S2 vs. S5, etc), and even for different monitoring depths within a given well. As expected, the maximum tracer breakthrough concentration generally decreased with transport distance, a consequence of hydrodynamic dispersion, while the differences in tracer breakthrough behavior within a given well appear to increase with distance from IW.

Significant tailing is evident in many of the tritium breakthrough patterns that in some instances apparently continued after sampling for a given well had been terminated. Even greater tailing was observed for Br^- (breakthrough data not shown). Model 1, based on one-dimensional uniform flow, over-predicted initial tritium arrival, under-predicted the maximum tracer breakthrough concentration, over-predicted initial leachout, and under-predicted the long-term tailing. Model 2 based on the radial flow geometry better predicted initial tracer breakthrough and leachout tailing (Table 1).

As illustrated in the current study, discerning non-conservative tracer behavior at the field scale is often difficult due to the high degree of physical heterogeneity and the inability to thoroughly monitor displacement of the tracer plume, which limits the accuracy of mass recovery estimates and spatial moments analysis. Analytical detection limitations and logistical concerns further hamper field-scale monitoring efforts. Despite such heterogeneity, high tritium recoveries, generally greater than 90%, confirm the general radial nature of the induced flow gradient.

As indicated by r^2 values for the calibrated parameters, the radial analytical solution for solute transport was generally better than the one-dimensional uniform flow solution at describing tracer breakthrough and leachout tailing with a lower α_L (Table 1). However, the 95% confidence intervals for the estimated parameters (travel time and α_L) often overlap for the two calibrated models. Longitudinal dispersivity (α_L) values and mean arrival times differed greatly for monitoring wells placed at similar radial distances, and between sampling zones within a given well, with no clear trend in α_L observed with travel distance, inconsistent with initial expectations.

Temporal moment analysis confirmed the retardation of Br^-, with estimates of retardation ranging from 0.99, essentially conservative behavior, to a maximum value of 1.67. However, retardation was likely underestimated because of analytical limitations and data truncation resulting from extensive tailing that biased retardation estimates when compared to tritium (Table 2). Based on these results, we are developing a numerical model that accounts for the nonlinear and conditional sorption to improve the mechanistic understanding of the differences in tracer behavior. Initial results from numerical modeling are presented in Majs and Seaman (2007).

Despite tracer retardation and incomplete mass recovery, both ADE models reasonably described the anion data without accounting for anion sorption reactions or apparent multiple flow domains (modeling results not presented), indicating that chemical interactions with the geologic matrix may be interpreted in terms of a physical transport process, i.e., flow velocity, path length, pore connectivity, multiple flow domains, dispersivity, etc.

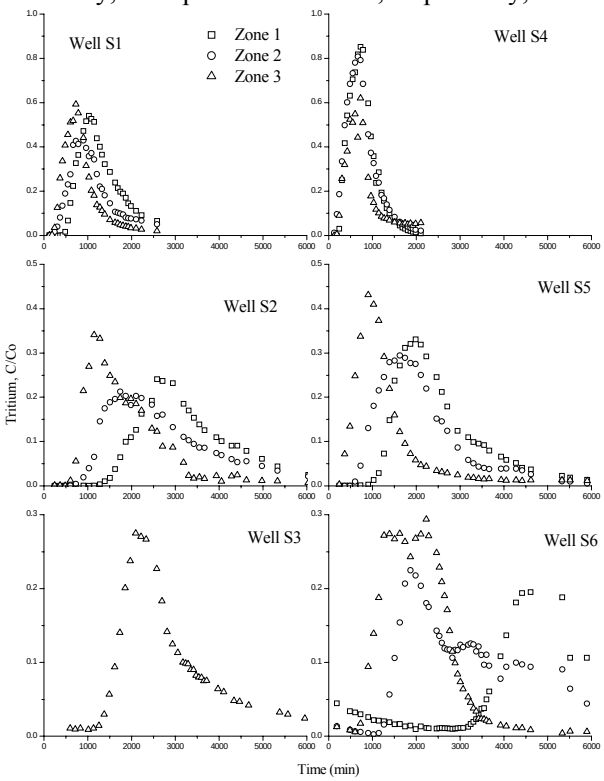

Fig. 2. Tritium breakthrough curves for each sampling zone within the six radially spaced monitoring wells.

Table 1. Summary of tritium data fitting results and 95% confidence intervals for uniform and radial flow approximate solutions.

Well no.	Radial Distance (m)	Model 1 Uniform Flow α_L (m)	Model 1 Uniform Flow Travel Time (min)	Model 1 Uniform Flow r^2	Model 2 Radial Flow α_L (m)	Model 2 Radial Flow Travel Time (min)	Model 2 Radial Flow r^2	Recovery (%)
S1-1	1.82	0.221±0.026	929±30	0.967	0.181±0.015	941±19	0.988	0.964
S1-2	1.82	0.614±0.117	878±71	0.897	0.548±0.070	898±42	0.971	0.793
S1-3	1.82	0.740±0.213	581±67	0.867	0.643±0.138	603±46	0.954	0.759
S2-1	3.19	0.232±0.028	2918±80	0.953	0.194±0.018	2966±59	0.979	0.925
S2-2	3.19	0.495±0.046	2275±65	0.967	0.464±0.039	2345±58	0.981	0.943
S2-3	3.19	0.594±0.092	1439±77	0.936	0.548±0.080	1483±70	0.962	0.891
S3-1	4.65	ND						
S3-2	4.65	ND						
S3-3	4.65	0.446±0.110	2368±146	0.739	0.381±0.081	2459±126	0.857	0.871
S4-1	2.04	0.244±0.035	422±12	0.988	0.168±0.019	420±9	0.993	1.01
S4-2	2.04	0.394±0.039	383±9	0.992	0.264±0.028	382±9	0.993	1.03
S4-3	2.04	1.53±0.63	586±118	0.793	1.23±0.45	576±84	0.902	0.673
S5-1	3.25	0.225±0.026	1694±46	0.966	0.185±0.014	1986±29	0.988	1.03
S5-2	3.25	0.417±0.030	1630±33	0.986	0.359±0.040	1679±51	0.974	0.977
S5-3	3.25	0.858±0.180	886±73	0.904	0.798±0.133	938±56	0.965	0.823
S6-1	4.41	0.111±0.023	4914±132	0.850	0.092±0.018	4943±127	0.877	0.850
S6-2	4.41	0.785±0.141	2829±213	0.738	0.691±0.128	2817±190	0.805	0.977
S6-3	4.41	0.443±0.065	1679±59	0.924	0.381±0.080	1735±90	0.888	0.945

ND = Not Determined due to analytical limitations

Table 2. Bromide retardation and mass recovery (in parentheses).

Well	S1	S4	S2	S5	S6	S3
Distance (m)	1.82	2.04	3.19	3.25	4.41	4.65
Zone 1	1.41 (0.96)	1.31 (1.02)	1.16 (0.84)	1.20 (0.94)	1.14 (0.39)	ND
2	1.67 (1.11)	1.49 (1.08)	1.24 (0.83)	1.46 (0.93)	1.02 (0.66)	ND
3	1.36 (1.09)	1.20 (0.99)	0.99 (0.87)	1.19 (0.91)	1.59 (0.92)	1.30 (0.67)
Mean	1.48 (1.06)	1.33 (1.03)	1.13 (0.85)	1.28 (0.93)	1.25 (0.66)	
SD	0.16 (0.08)	0.14 (0.04)	0.19 (0.02)	0.16 (0.01)	0.31 (0.27)	

ND = Not Determined due to analytical limitations

Acknowledgments:

The authors thank Troy Rea, Jennifer McIntosh, Angel Kelsey-Wall and Jane Logan for their assistance in the field and with tracer analyses, and Dr. L. Janecek for her assistance in creating the figures. Financial assistance was provided through Financial Assistance Award number DE-FC09-96SR18546 from the U.S. DOE to The University of Georgia Research Foundation.

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