Effect of tracer buoyancy on tracer experiments conducted in fractured crystalline bedrock

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Received 18 October 2002; revised 27 November 2002; accepted 26 December 2002; published 6 February 2003.

[1] Tracer buoyancy has been shown to influence breakthrough from two-well tracer experiments conducted in porous media. Two-well tracer experiments are presented from fractured crystalline bedrock, in which the specific gravity of the tracer injectate varied from 1.0002 to 1.0133. Under the forced hydraulic conditions imposed, no difference in breakthrough was noted for the three experiments. These results show that even relatively dense tracer injectate solutions may have an insignificant effect on breakthrough when imposed gradients are sufficiently large. *INDEX TERMS:* 1832 Hydrology: Groundwater transport; 1829 Hydrology: Groundwater hydrology; 1894 Hydrology: Instruments and techniques; 8010 Structural Geology: Fractures and faults. **Citation:** Becker, M. W., Effect of tracer buoyancy on tracer experiments conducted in fractured crystalline bedrock, *Geophys. Res. Lett.*, *30*(3), 1116, doi:10.1029/2002GL016480, 2003.

1. Introduction

[2] The tracer test is a critical tool for characterization of mass transport in fractured bedrock where hydraulic tests are typically an unreliable predictor of transport. One of the complications of performing tracer tests is that tracers are not neutrally buoyant. As the specific gravity of the tracer injectate increases, so does the possibility that tracer transport may be dominated by gravitational rather than hydraulic forces. If buoyancy effects occur over time scales comparable to that of the tracer test, then the resulting tracer breakthrough curve may be influenced by the specific gravity of the tracer. The influence of buoyancy may result in incorrect estimations of hydrodynamic dispersion, matrix diffusion, sorption, and other transport parameters related to tracer breakthrough.

[3] *Istok and Humphrey* [1995] cite a number of field cases in porous media where they believe buoyancy-induced vertical transport may have caused sinking of tracer plumes. These authors also conducted laboratory ''sand-box'' experiments that showed an effect of tracer buoyancy on breakthrough from two-well tracer tests with initial concentrations of bromide tracer as low as 50 mg/L. The primary effect that the sinking tracer plume had on the breakthrough curve was to reduce the magnitude of the peak. *Simmons et al.* [2001] performed numerical simulations of the instability and migration of relatively dense solute plumes in heterogeneous porous media, including equivalent porous media simulations of vertical fractures. The interplay between medium heterogeneity and fluid dynamics in the initiation of plume instability was complex, but the structure of the heterogeneous permeability field appeared to be the single most important factor in controlling the fate of these instabilities.

[4] *Shikaze et al.* [1998] performed numerical simulations of buoyant plumes through an orthogonal fracture network. Flow and mass-transport equations were solved by superimposing one-dimensional line elements representing horizontal and vertical fractures onto a finite difference mesh representing the porous matrix. Simulations predicted complex transport patterns even though the geometry of the network was relatively simple. Convection cells appeared within the fracture network, facilitated by the intervening porous matrix. These authors concluded that "Because of the inherent uncertainty associated with fracture delineation, and because of the irregular nature of unstable dense plumes, deterministic prediction of dense-plume migration pathways and travel times in fractured geologic media will be subject to considerable uncertainty.''

[5] In a fractured geologic medium where the matrix porosity has insignificant permeability relative to the fracture, buoyancy-driven tracer transport may be better represented by an open conduit than a dual-porosity medium. In this case, tracer buoyancy may have little or no influence on transport through a perfectly horizontal fracture, but a significant influence on tracer transport in or across non-horizontal fractures. *Ronen et al.* [1995] investigated buoyancy-driven transport for small density contrasts in open vertical conduits and found that density-driven transport was rapid and turbulent. Buoyancy-driven transport may be fundamentally different in porous and fractured media due to differences in both permeability structure and fluid behavior.

[6] Two-well forced-gradient tracer tests are considered in this article. Under forced-gradient conditions the hydraulic gradient and advective velocity vary tremendously across the flow field. One might expect that in some fractures buoyancy-driven transport will be dwarfed by advective transport, but in others may significantly impact tracer breakthrough. Thus, the influence of tracer density on breakthrough is likely to be a function of how the tracer test is designed. The impact of tracer buoyancy has been investigated for two-well tracer tests in porous media [*Istok and Humphrey*, 1995] and for fractures under uniform hydraulic gradients [*Shikaze et al.*, 1998]. The influence of tracer buoyancy on forced-gradient field tracer experiments conducted in bedrock is yet to be addressed in the published literature. Although both theoretical and experimental studies are clearly needed to understand the behavior of non-neutrally buoyant tracer in fractured geologic media, this article takes a strictly empirical approach to the topic.

2. Experimental Method

[7] Tracer experiments were conducted at the Mirror Lake Fractured Rock Research Site, in Grafton County, located in the White Mountains of New Hampshire, United

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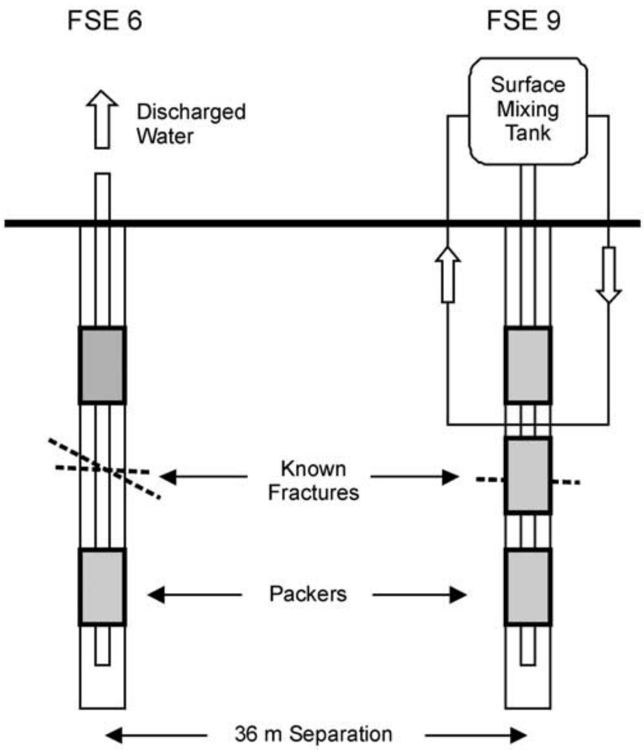

Figure 1. Diagram of the pumping (FSE6) and injection (FSE9) wells used for these experiments.

States. The Forest Service East (FSE) well field is the subject of this article, and has been the focus of many previous tracer experiments for the purpose of understanding solute [*Shapiro and Hsieh*, 1991; *Becker and Shapiro*, 2000], colloid [*Becker et al.*, 1999], and microbe [*Becker et al.*, 2003] transport in highly heterogeneous fractured geologic media. The local bedrock is typified by pelitic schist that has been metamorphosed to a sillimanite grade, and is extensively intruded by dikes, anastomosing fingers, and pods of granite [*Barton et al.*, 1997]. Bedrock in the vicinity of the well field is overlain by glacial drift that is 18 to 24 meters in depth. Ground water in many of the FSE bedrock wells tends to be reducing, possibly because of septic system discharges in the overburden. Water samples taken during pumping for the tracer tests had a pH of about 6. *Tiedeman et al.* [1998] provide further discussion of the ground-water hydrology of the Mirror Lake drainage basin, in which the FSE well field resides.

[8] Tracer experiments of focus here were conducted in August of 1999, between wells FSE6 (pumping) and FSE9 (injection). These wells are 36 m apart, 15 cm in diameter, and are cased through the glacial drift and 3 m into the bedrock. The forced gradient tracer tests discussed here are of a full-radially convergent slug-injection design [*Shapiro and Hsieh*, 1996]. For these experiments FSE6 was pumped at a rate of 10.1 liters per minute (lpm) until approximately steady-state heads were achieved. An 3 m injection interval in FSE9 was isolated by upper and lower inflatable packers while the fracture of interest was sealed by a center packer (Figure 1). Borehole fluid was circulated to a surface tank using a submersible pump. Prior to injection, a small volume of tracer-laden water was added to the surface tank and allowed to mix thoroughly with the formation fluid. Injection was accomplished by deflating the center packer and allowing injectate to enter the formation under the force of gravity. Injection ended when the fracture was resealed by the center packer, and the volume of injectate was measured by observing the volume lost from the surface tank. By sealing the fracture of interest, the amount of mass entering the formation was tightly controlled and "bleeding" of remaining tracer in the well bore was prevented.

[9] Standard geophysical logging tools, an acoustic-televiewer, and a submersible borehole television camera have been used to determine rock type and the location and orientation of fractures in the boreholes of the FSE well field [*Johnson and Dunstan*, 1998]. These data indicate that the conductive fractures isolated in pumping well (FSE6) are at a depth of 35 m below the surface, and are both steeply dipping and horizontal. The single conductive fracture isolated in the injection well (FSE9) is at a depth of 42 m below the surface and is near horizontal. Because FSE6 and FSE9 are separated by a distance of 36 m, and fractures exposed in nearby road cuts rarely exceed 10 meters in length [*Barton et al.*, 1997], it is unlikely that the two wells are connected by a single fracture. The orientation and relationship of the fractures that hydraulically connect the two wells is unknown. Based upon outcrop and borehole observations, however, it is likely that the flow path connecting FSE6 and FSE9 passes through both sub-horizontal and dipping fractures.

[10] To investigate the relationship between tracer injectate density and breakthrough, tracer tests were repeated three times under identical slug-injection conditions but using injectate fluids that varied in their specific gravity (referred to as experiments 1999A, 1999C, and 1999D). The goal was to compare the transport of nearly neutrally-buoyant tracer solution to that of negatively buoyant tracer solutions. This goal presented the difficult challenge of finding low-specific-gravity tracers that are practical for field applications. Fluorescent dyes are an attractive candidate because they can be detected at part-per-billion level concentrations so that little mass need be injected. Previous experiments in the FSE well field, however, determined that fluorescent dyes tended to be retarded in transport relative to other tracers. Benzoic acids have also been used as a tracer at Mirror Lake [*Becker and Shapiro*, 2000], but require pH neutralization by NaOH which tends to increase the specific gravity of the injectate. Consequently, we chose to use deuterated water as our best choice for nearly neutrally-buoyant tracer.

[11] Deuterated water offers a considerable advantage over ionic tracers with respect to injectate buoyancy [*Becker and Coplen*, 2001]. The density of NaBr dissolved in water is 3.7 g/cm^3 at 20°C (based upon our laboratory measurements) whereas the density of D_2O is 1.11 g/cm^3 at 20°C (based upon published values). Note that 3.7 g/cm^3 is the inverse of the volume increase (in cm^3) resulting from the addition of one gram of solid NaBr to water. As both tracers are detectable to approximately 2 mg/L, deuterated water is clearly a more neutrally buoyant tracer to use in field experiments. Deuterated water was used as a tracer in all three experiments, and sodium bromide was added to increase the specific gravity of the tracer injectate in two of the experiments. Hydrogen-isotope analyses used for the

Table 1. Density of Tracer Injectate Solutions

Experiment	Mass, g		Inj.Volume, L	Specific Gravity
	D_2O	NaBr		
1999A	76.6	0.0	38	1.0002
1999C	76.6	68.6	38	1.0015
1999D	76.6	685.4	38	1.0133

deuterated water breakthrough were conducted by the U.S. Geological Survey Isotope Fractionation Project, using a hydrogen-equilibration technique [*Becker and Coplen*, 2001]. Bromide concentrations were measured with an ion chromatograph.

[12] Solution density of the tracer injectate fluids in these tests were calculated using the formula:

$$\rho_{inj} = \frac{M_{H_2O} + \sum_j M_j}{V_{H_2O} + \sum_j V_j}, \tag{1}$$

where ρ_{inj} is the density of the injectate, M_{H2O} is the mass of water, M_j is the mass of dissolved tracer, j, V_{H2O} is the volume of water, and V_j is the volume occupied by dissolved tracer (inverse of the density in dissolved form). The use of Equation 1 requires the volume of the entire mixed injection system. This volume was found to be 63.6 liters based upon the dilution of a known mass of deuterated water in the fully mixed injection interval. The density of the tracer injectate after mixing with fluid in the borehole was confirmed with a hydrometer (Fisher Scientific ± 0.0005 SG) and by weighing a 1 L volumetric flask (±0.0002 g/cm) for experiments 1999C and 1999D. The predicted change in density due to the addition of tracer in 1999A was below the accuracy of either hydrometer or volumetric measurements. The specific gravity of the formation water was not significantly different than deionized water at room temperature (1.0000), based upon hydrometer measurements. The temperature of the circulating borehole fluid was monitored during the mixing phase to assure that the density of the borehole fluid was not reduced during the mixing phase. The borehole fluid was about 10°C during mixing, and increased less than 1°C in each experiment, implying that the temperature induced density contrast between injectate and formation fluid was less than 0.0001 g/cm^3 [*Lide*, 1995]. Injectate solution density was calculated using equation (1), and confirmed with hydrometer and volumetric measurements, for the fully mixed borehole solution. Table 1 displays the specific gravity calculated from equation (1) for all three injections (1999A, 1999C, 1999D). Results of the second experiment (1999B) are not shown because it was aborted due to a power failure and was repeated (1999C).

[13] The radially convergent slug-injection experimental design produces very repeatable results, even after equipment is removed and replaced and when experiments are conducted at different times of the year. Figure 2 compares deuterated water breakthrough from experiment 1999A conducted in August of 1999, and similar experiment conducted in May of 2000 [*Becker et al.*, 2003]. Both experiments were conducted in the slug injection mode as described above, but the pumping rate was 10.1 lpm in the 1999 experiment and 10.7 lpm in the 2000 experiment. The density of the injectate was 1.0002 g/cm^3 in the 1999 experiment (Table 1) and 1.0001 g/cm^3 in the 2000 experiment. The slight difference in pumping rate, density, and perhaps ambient flow conditions may have contributed to the minor difference in breakthrough between the 1999 and 2000 experiments shown in Figure 2.

3. Results and Interpretation

[14] Breakthrough from three tracer experiments with specific gravity varying from 1.0002 to 1.0133 are shown in Figure 3. Concentrations of tracer are normalized to the injected mass (M_0). Only the breakthrough of deuterated water is shown, but the breakthrough of bromide was identical to deuterated water in experiments 1999C and 1999D. Figure 3 clearly demonstrates that, under these experimental conditions, the specific gravity of the tracer does not affect breakthrough.

[15] It is important to note that although no relationship between the injectate density and breakthrough was noted under these experimental conditions, under the condition of denser injectate solution or slower pumping rates breakthrough may have been influenced by injectate buoyancy effects. One might conceptualize the relationship between tracer-injectate buoyancy and breakthrough as a competition between buoyancy-induced and hydraulically-induced transport. Examination of buoyancy-driven transport in stagnant vertical conduits [*Ronen et al.*, 1995] suggest that the rate of tracer plume sinking is related to $\Delta\rho^{-1/2}$, where $\Delta\rho$ is the initial density contrast of the tracer (difference between tracer density and ambient water, or 0.0002 to 0.0133 in these experiments). One might further suggest that fractures better resemble conduits than a porous media with respect to buoyancy-induced transport. These arguments suggest that the effect of increasing the initial tracer

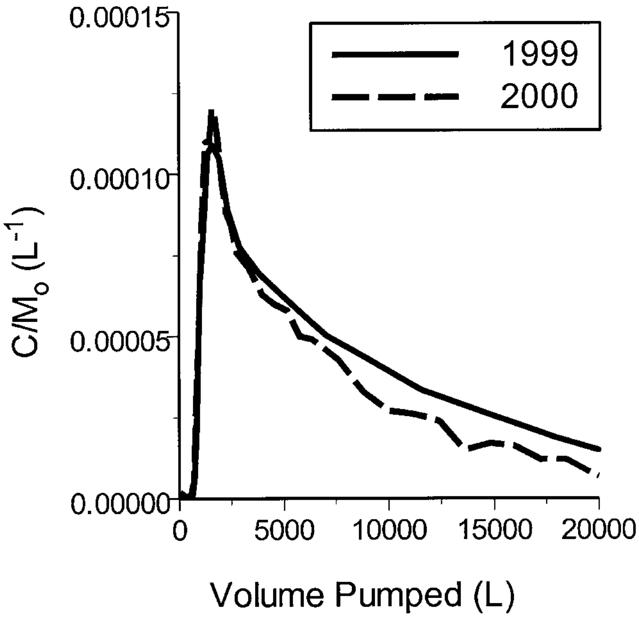

Figure 2. Deuterated water breakthrough from tracer experiments conducted between FSE6 and FSE9 using the radially convergent slug-injection design. Experiments conducted in August of 1999 May of 2000 demonstrate the repeatability of results under similar test conditions.

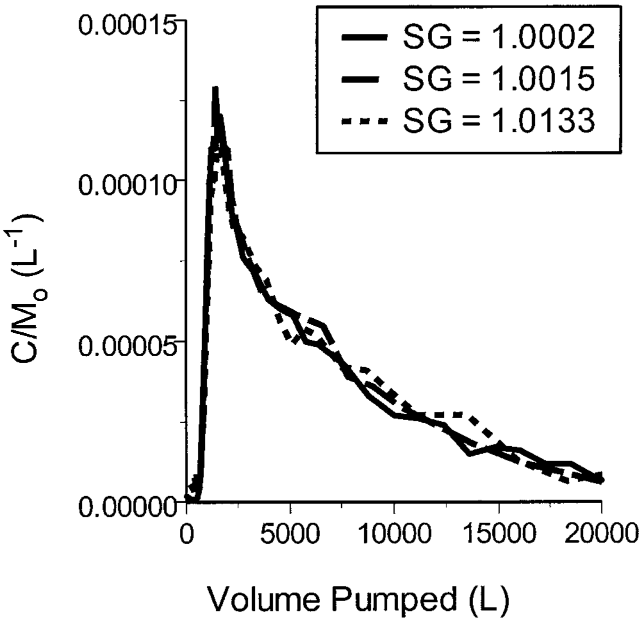

Figure 3. Breakthrough of deuterated water from a slug-injection tracer experiment, where injectate density was varied.

density contrast by a factor of 64 in these experiments would have the same general impact on breakthrough as decreasing the pumping rate by a factor of 8. In fractured rock, therefore, buoyancy-driven transport may be more sensitive to the imposed hydraulic gradient than the density of the injectate.

[16] Other two-well forced-gradient tracer experiments conducted between FSE6 and FSE9 have shown that breakthrough behavior changes with pumping rate [*Becker and Shapiro*, 2000]. Early breakthrough seems to be much more affected than later breakthrough. The results of Figure 3 indicate that breakthrough is not influenced by buoyancy-driven transport at pumping rates of about 10 lpm, but this rate is at the high end of pumping rates employed between FSE6 and FSE9. As of yet we do not have experiments comparing negatively buoyant and relatively neutrally buoyant tracer injectate at lower pumping rates used in the FSE well field (e.g. ~3 lpm).

[17] Further experiments are anticipated for the FSE well field that may add more conclusive evidence of the relationship between tracer buoyancy and breakthrough under forced gradient conditions. It would be interesting to compare the behavior of neutral and negatively buoyant tracers between another well pair to investigate the impact of specific fracture orientation on tracer breakthrough. Because the specific orientation of the fractures in the FSE well field are unknown, however, the exact interplay between fracture geometry and breakthrough would be supposition. Numerical simulations are being conducted that will better elucidate the theoretical relationship between hydraulic velocity and buoyancy-induced velocity in hypothetical fracture geometries. Clearly, the results of these experiments in the FSE well field must be considered site-specific. To rule out buoyancy effects on tracer breakthrough, investigations such as these need to be conducted for each particular tracer experiment.

[18] **Acknowledgments.** These investigations were conducted with the support of the U. S. Geological Survey Toxic Substances Hydrology Program. The author is grateful for the support provided by the Hubbard Brook Experimental Forest, which is operated and maintained by the Northeastern Forest Experiment Station, USDA Forest Service, Radnor, Pennsylvania. Field investigations and subsequent interpretation was conducted in close collaboration with Allen Shapiro of the USGS. Field assistance was provided by Samantha Collins and Michael Cummings, formerly with the University at Buffalo.

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