Discussion of "DNAPL migration through a fractured perching layer" by Daniel B. Stephens, James A. Kelsey, Mark A. Prieksat, Mel G. Piepho, Chao Shan, and Mark D. Ankeny, July-August 1998 issue of Ground Water, v. 36, no. 4: 605-610 (U)

David M. Tuck

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Westinghouse Savannah River Company Savannah River Site Aiken, SC 29808 "DNAPL migration through a fractured perching layer" by Daniel B. Stephens, James A. Kelsey, Mark A. Prieksat, Mel G. Piepho, Chao Shan, and Mark D. Ankeny, July-August 1998 issue, v. 36, no. 4: 605-610.

Discussion by D. M. Tuck, Senior Scientist, Westinghouse Savannah River Co., Savannah River Technology Center, Bldg. 773-42A, Aiken, SC 29808.

The visualization study of 1,1,1-trichloroethane (TCA) infiltration described by Stephens et al. (1998) was an elegant design, clearly demonstrating DNAPL behavior encountering a fractured perched layer. I question, however, their interpretation, stated in both abstract and conclusions, that rapid DNAPL penetration of the fracture was "in contrast to existing mathematical solutions of hydrostatic initial conditions and full saturation below the fracture." I also have a comment regarding the experimental conditions.

Essential to all visualization experiments is the need to stain one of the fluid phases (usually the organic phase) to observe its paths through the medium (Schwille 1988; Kueper et al. 1989; Poulsen and Keuper 1992; Powers et al. 1992; Kueper et al. 1993; Brewster et al. 1995; Pennell et al. 1996). By their nature, dye chromophores are likely to cause surface active behavior. Dye type and concentration are therefore important experimental conditions which should be reported. The authors reported the dye used (D & C Red 17); they did not, however, report the dye concentration.

D & C Red 17 (also known as Sudan III, among other trade names; (Lide and Milne 1995) belongs to an oil soluble azo, class of dyes (Catino and Farris 1978). Every visualization study I've found uses dyes of the same basic structure with minor variations (see Table 1). Tuck and co-workers (Tuck et al. 1996; Tuck et al. 1997; Tuck et al. 1998) have been studying the surface chemistry effects of Sudan IV in the PCE-water-glass system (see Table 2). D & C Red 17 has two fewer methyl groups than Sudan IV on its "polar" tail, increasing its polar character. It is thus likely to be more surface active than Sudan IV, causing greater interfacial tension decreases. Stephens et al. (1998) calculated a 34 cm entry pressure depth for TCA to penetrate the fracture. Assuming D & C Red 17 decreases TCA-water interfacial tension by one fifth (1/5), the entry pressure required to penetrate the fracture decreases to approximately 23 cm (Kueper and McWhorter 1991).

I have purposely used the term entry pressure rather than pool thickness. I believe the latter term is deceptive; it seems to imply that a DNAPL must always pond or pool above a fractured aquitard before it could enter the fracture. The deceptive aspect is that it leads us to ignore any pressure head other than what is in the "pool." The authors delivered dyed TCA at a 5 mL/min rate to the top of their upper, unsaturated aquifer, approximately 39 cm above the water table. The water table was another 75 cm above the fractured siltstone perching layer. Ignoring the head contribution from the unsaturated zone, we can estimate the TCA pressure head when it initially encountered the fracture as hydrostatic pressure (expressed as a vertical head) minus losses due to flow

$$\frac{P_{\rm D}}{\Delta \rho g} = h - (\text{flow losses})$$

(1)

where P_D is DNAPL pressure, $\Delta \rho$ is fluid density difference, g is acceleration of gravity, and h is vertical difference between the water table and siltstone (75

cm). Flow losses can be estimated from Darcy's equation modified for multiphase flow

$$q = \left(\frac{\rho g}{\mu}\right) k_r k A\left(\frac{dh}{dl}\right)$$

(2)

where q is specific discharge (cm³/sec), μ is dynamic viscosity (g/cm-sec, Poise), k is intrinsic permeability (cm^2), k_r is relative permeability (dimensionless), A is cross-sectional flow area (cm^2), and dh/dl is head loss per unit flow length. A gross visual estimate yielded a TCA cross-sectional flow area of approximately 10% of the tank, yielding an area of 3.2 cm^2 . The intrinsic permeability was calculated to be 7.8×10^{-8} cm² based on the hydraulic conductivity provided by the authors. Dynamic viscosity for TCA is 0.86 cP, and its density is 1.341 g/cm³ (Stephens et al. 1998). Assuming k_r is 0.5 and a 90 cm flow path (full depth of saturated upper aquifer plus half the tank width for horizontal flow), I calculate a flow head loss of approximately 40 cm. Hence, the head when TCA first reached the fracture (neglecting any unsaturated zone contribution), was approximately 35 cm, i.e., there was

sufficient head when it first encountered the fracture to enter it, even neglecting any surface active dye effects.

Regardless of the points I made above, this study demonstrates very clearly that when dealing with DNAPLs in the field, we should not assume they will pond to any great depth on an aquitard, particularly if there is significant vertical separation between the DNAPL entry point and the aquitard in question. This has, in fact, been our experience at the Savannah River Site, where the M-Area DNAPL appears to exist in relatively thin layers in the saturated zone at depths of approximately 145 feet (Jackson et al. 1996).

References

Brewster, M. L., Annan, A. P., Greenhouse, J. P., Kueper, B. H., Olhoeft, G.
R., Redman, J. D. and Sander, K. A. (1995). "Observed migration of a controlled DNAPL Release by Geophysical Methods." <u>Ground Water</u> 33(6): 977-987.

Catino, Sigmund and Farris, Russell E. (1978). Azo dyes. *in <u>Kirk-Othmer</u>
<u>Encyclopedia of Chemical Technology</u>. vol. 3 Eds. H. F. Mark, D. F. Othmer,
C. G. Overberger and G. T. Seaborg. New York, John Wiley & Sons. pp. 387433.*

Fortin, Josée, Jury, William A. and Anderson, Michael A. (1997). "Enhanced removal of trapped non-aqueous phase liquids from saturated soil using surfactant solutions." Journal of Contaminant Hydrology **24**(3-4): 247-267.

Jackson, Dennis G., Payne, Tamra H., Looney, Brian B. and Rossabi, Joseph. (1996). Estimating the extent and thickness of DNAPL within the A/M Area of

the Savannah River Site (U). Westinghouse Savannah River Company. WSRC-RP-96-0574. 121 pp.

Kueper, Bernard H., Abbott, Wesley and Farquhar, Graham (1989)."Experimental observations of multiphase flow in heterogeneous porous media." Journal of Contaminant Hydrology 5: 83-95.

Kueper, Bernard H. and Frind, Emil O. (1991). "Two-Phase flow in heterogeneous porous media 2. Model application." <u>Water Resources</u> <u>Research</u> **27**(6): 1059-1070.

Kueper, Bernard H. and McWhorter, David B. (1991). "The behavior of dense, nonaqueous phase liquids in fractured clay and rock." <u>Ground Water</u> **29**(5): 716-728.

Kueper, Bernard H., Redman, David, Starr, Robert C., Reitsma, Stanley and Mah, May (1993). "A field experiment to study the behavior of tetrachloroethylene below the water table: Spatial distribution of residual and pooled DNAPL." <u>Ground Water</u> **31**(5): 756-766.

Lide, David R. and Milne, G. W. A., Eds. (1995). <u>Names, Synonyms and</u> <u>Structures of Organic Compounds</u>. Boca Raton, CRC Press.

Pennell, Kurt D., Pope, Gary A. and Abriola, Linda M. (1996). "Influence of viscous and buoyancy forces on the mobilization of residual tetrachloroethylene during surfactant flushing." <u>Environmental Science & Technology</u> **30**(4): 1328-1335.

Poulsen, Mette M. and Keuper, Bernard H. (1992). "A field experiment to study the behavior of tetrachloroethylene in unsaturated porous media." <u>Environmental Science & Technology</u> **26**: 889-895.

Powers, Susan E., Abriola, Linda M. and Weber, Walter J., Jr. (1992). "An experimental investigation of nonaqueous phase liquid dissolution in saturated

subsurface systems: steady state mass transfer rates." <u>Water Resources</u> <u>Research</u> **28**(10): 2691-2705.

Schroth, M. H., Istok, J. D., Ahearn, S. J. and Selker, J. S. (1995). "Geometry and position of light nonaqueous-phase liquid lenses in water-wetted porous media." Journal of Contaminant Hydrology **19**: 269-287.

Schwille, Friedrich (1988). <u>Dense Chlorinated Solvents in Porous and</u>
<u>Fractured Media: Model Experiments</u>. ed. Chelsea, MI, Lewis Publishers, Inc.
146 pp.

Stephens, Daniel B., Kelsey, James A., Prieksat, Mark A., Piepho, Mel G., Shan, Chao and Ankeny, Mark D. (1998). "DNAPL migration through a fractured perching layer." <u>Ground Water</u> **36**(4): 605-610.

Tuck, David M., Iversen, Gary M. and Pirkle, William A. (1996). "Complex Organic Co-contaminant Effects on DNAPL Mobility in Saturated Porous Media." <u>EOS</u> **77**(46): F257.

Tuck, David M., Iversen, Gary M., Pirkle, William A. and Rulison, Christopher (1998). Time-dependent interfacial property effects on DNAPL flow and distribution. *in* <u>Nonaqueous-Phase Liquids: Remediation of Chlorinated and Recalcitrant Compounds</u>. vol. C1-2 Eds. G. B. Wickramanayake and R. E. Hinchee. Columbus, OH, Battelle Press. pp. 73-78.

Tuck, David M., Iversen, Gary M., Pirkle, William A., Rulison, Christopher and Denham, Miles E. (1997). "Capillary Barriers and Time-Dependent Wetting Relationships: Effects of complex DNAPL Components." <u>EOS</u>
78(46): F289.

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Table 1. Dyes used in flow visualization studies in the ground water literature.

Dye Concentration (g/L)	Active Fraction Concentration (g/L)	γ _{NW} after 30 seconds (mN/m)
0	0	51.0
0.00508	0.0041	48.9
0.508	0.411	43.2
1.27	1.03	39.2
1.69	1.37	35.5

Table 2. Interfacial tension between water and PCE dyed with Sudan IV.