# Partitioning Tracer Tests as a Remediation Metric: Case Study at Naval Amphibious Base Little Creek, Virginia Beach, Virginia

The partitioning tracer test (PTT) is a characterization tool that can be used to quantify the porespace saturation ( $S_{\rm N}$ ) and spatial distribution of dense nonaqueous phase liquids (DNAPLs) in the subsurface. Because the method essentially eliminates data interpolation errors by directly measuring a relatively large subsurface volume, it offers significant promise as a remediation metric for DNAPL-zone remediation efforts. This article presents, in detail, the design and results of field PTTs conducted before and after a DNAPL-zone treatment at the Naval Amphibious Base Little Creek, Virginia Beach, Virginia. The results from different tracers yield a relatively large range in  $S_N$  estimates, indicating notable uncertainty and presenting significant challenges for meaningful interpretation. Several potential interpretation methods are presented, resulting in an estimated DNAPL removal range of 15 to 109 L. While this range is large, it is consistent with the DNAPL removal (~30 L) determined from analysis of effluent concentration measurements collected during the remediation efforts. At this site, the initial and final  $S_{N}$  values are low, and the relatively inconsistent performance of the various tracers indicates that these levels are near the lower practical quantification limit for these PTTs; however, the effective lower quantification limit for these tests is unknown. Generally, an understanding of lower quantification limits is particularly important for interpretation of post-remediation PTTs because  $S_N$  values are likely to be low (due to remediation efforts) and the S $_{\scriptscriptstyle \rm N}$  estimated from the PTT may be used to predict long-term dissolved plume behavior and assess associated environmental risk. Partitioning tracer test quantification limits are test-specific, as they are dependent on a variety of factors including analytical uncertainty, tracer breakthrough characteristics, and tracer data integration techniques. The results of this case study indicate that methods for estimating lower quantification limits for field PTTs require further development. © 2004 Wiley Periodicals, Inc.

# INTRODUCTION

During the past decade, promising laboratory and field results from new remediation technologies have greatly increased interest in active treatment of subsurface dense non-aqueous phase liquid (DNAPL) zones. However, an accurate understanding of the total DNAPL mass and its spatial distribution is critical for designing effective remediation systems, estimating project costs, and assessing remedial efficiency. Unfortunately, traditional subsurface characterization methods, such as chemical analysis of drill-core and groundwater samples, are largely inadequate for quantifying total DNAPL mass and average pore-space saturations at the scale of most remediation sites. Recently, however,

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Traditional characterization methods require significant interpolation between point measurements, and extrapolation to areas in the source zone not covered by the point sampling. the partitioning tracer test (PTT) has demonstrated promise for quantifying subsurface NAPLs, and several dozen successful field-scale PTTs have been reported in the literature (e.g., Annable et al., 1998; Cain et al., 2000; Jawitz et al., 2000; Meinardus et al., 2002; Nelson & Brusseau, 1996; Young et al., 2001). Traditional characterization methods require significant interpolation between point measurements, and extrapolation to areas in the source zone not covered by the point sampling. Conversely, PTTs measure a relatively large volume of the subsurface (typically tens to hundreds of cubic meters), thereby greatly reducing the error and uncertainty associated with NAPL volume estimation in the source zone. The PTT may be a particularly useful remediation metric for NAPL-zone treatment efforts because the same subsurface volume can be directly measured in essentially the same manner before and after remediation activity.

This article presents a case study where PTTs were used to assess the remediation performance of a DNAPL-zone treatment effort at the Naval Amphibious Base Little Creek (NABLC) in Virginia Beach, Virginia. During the summer of 2002, a cyclodextrin solubility-enhancement agent was injected and extracted in a DNAPL zone in an effort to remove contaminant mass. Details of this remediation effort will be presented in a forthcoming paper. For examples of prior successful cyclodextrin-enhanced remediation operations, the reader is referred to Blanford et al. (2000), McCray and Brusseau (1998), and Tick et al. (2003). Partitioning tracer tests were conducted both before (Pre-PTT) and after (Post-PTT) the cyclodextrin application to estimate initial and final DNAPL saturation, and to identify any subzones within the treatment zone with higher DNAPL saturation. Although a number of successful field PTTs have been previously reported in the literature, most of these reports have not included detailed descriptions of the PTT design process and practical data interpretation challenges. Therefore, this case study includes a thorough description of the PTT design process, analysis and interpretation of observed field results, and related discussion of practical application and limitations of field PTTs.

# Site Description

The field site is known as "Site 11," which is located adjacent (north) to the NABLC School of Music. The primary suspected contaminant source was a small underground neutralization tank associated with a former metal plating facility that was used to store and treat plating wastes and possibly other materials during the 1960s and 1970s. Currently, a groundwater plume of several volatile organic compounds (VOCs) extends from the former tank area. Trichloroethene (TCE) was the primary contaminant of concern for the source-zone treatment efforts and was believed to represent the majority of contaminant mass at the site. Although DNAPL has not been directly observed in soil cores or monitoring wells at the site, high dissolved-phase TCE concentrations (up to 390 mg/L) measured during a previous Membrane Interface Probe (MIP) investigation (CH2M HILL, 2001) strongly suggested its presence. Several other constituents are also present in groundwater: 1,1,1-trichloroethane (1,1,-TCA); 1,1-dichloroethane (1,1-DCA); 1,1-dichloroethene (1,1-DCE); chloroform; and cis-1,2-dichloroethene (cDCE).

Hydrostratigraphic conditions at the site are relatively simple. Contaminants are present primarily in the lower 1.5 m of the Columbia Aquifer, which is locally composed of fine to medium sands (with occasional silt and clay lenses) extending from approximately 1.5 to 7.6 m below land surface (bls). The local average horizontal hydraulic conductivity in the Columbia Aquifer is approximately  $8.3 \times 10^{-4}$  cm/sec and the measured porosity is approximately 31 percent (CH2M HILL, 2001). The water table occurs at approximately 1.5 to 2.4 m bls in the Columbia Aquifer, and conditions are generally considered unconfined. Groundwater flow direction is generally consistent with the regional groundwater flow direction (approximately WSW); however, significant seasonal variations in local flow direction have been observed, which have caused significant dispersion of the dissolved plume. The Columbia Aquifer is underlain by a dense marine clay confining unit (Yorktown Confining Unit) with a vertical hydraulic conductivity of approximately 3 ×  $10^{-8}$  cm/sec (CH2M HILL, 2001). Locally, the Yorktown Confining Unit is believed to separate the surficial Columbia Aquifer from the underlying Yorktown Aquifer.

#### PTT Theory

During a PTT, a suite of conservative and partitioning tracers are injected into one or more injection wells and are subsequently recovered from one or more extraction wells. By definition, the transport of conservative tracers is unaffected by the presence of DNAPL in the tracer sweep zone. However, the partitioning tracers will temporarily partition into any accessible DNAPL, and their transport will therefore be retarded relative to the transport of the conservative tracers. The retardation factor (R) of a partitioning tracer is determined directly from the tracer breakthrough curves (BTCs) characterized from data collected at the extraction wells, and is defined relative to the transport of the conservative tracer by:

$$R = \frac{\overline{t_p}}{\overline{t_c}}$$
(1)

where  $\overline{t_p}$  and  $\overline{t_p}$  are the mean travel times for the partitioning and conservative tracers. When the tracer input is constant during the time of tracer injection  $(t_s)$ , the average tracer  $(\overline{t})$  travel time is determined from the normalized first temporal moment by:

$$\bar{t} = \frac{\int t \cdot C(t) dt}{\int C(t) dt} - \frac{t_s}{2}$$
(2)

where *t* is the measurement time and C(t) is the tracer concentration over time at the extraction well (i.e., the tracer BTC). Typically, BTCs from field PTTs exhibit significant tailing, which is primarily caused by the hydraulics of the injection/extraction system. Truncation of this tail region due to early test termination can lead to moment estimation errors; therefore, an exponential extrapolation method (see Helms, 1997) was used to extend measured elution curves beyond test termination to improve moment estimates of the BTCs.

For a fully saturated water-NAPL system (i.e., gas-phase saturation is insignificant) where tracer sorption to aquifer materials is insignificant, the pore-space DNAPL saturation  $(S_N)$  is calculated by (see Jin, 1995):

$$S_N = \frac{R-1}{R+K_{NW}-1}$$
(3a)

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practical constraints.

 $\kappa_{_{NW}} = \frac{C_{_{N}}}{C_{_{W}}}$ 

(3b) where  $K_{NW}$  is NAPL-specific the tracer partition ( $C_N$  and  $C_W$  represent the equilibrium tracer concentrations in NAPL and water). Tracer partition coefficients are typically determined in laboratory batch tests. Note that Equation 3a can be written more general-

ly so that  $S_N$  can be calculated from the travel times of two partitioning tracers:

$$S_{N} = \frac{\overline{t_{p}^{i}} - \overline{t_{p}^{j}}}{\overline{t_{p}^{j}} (K_{NW}^{i} - 1) - \overline{t_{p}^{i}} (K_{NW}^{j} - 1)}$$
(4)

where the superscripts indicate the average travel times and partition coefficients for two different partitioning tracers, *i* and *j*. The total NAPL volume measured by the test  $(V_N)$  is equal to  $S_N$  times the test sweep volume. The test sweep volume  $(V_{SW})$  is approximately calculated by:

$$V_{SW} = t_c^j \cdot Q \tag{5}$$

where Q is the fluid extraction rate.

## **Design and Field Methods**

Conceptually, PTT application is relatively simple; however, successful field implementation requires careful design to optimize test results while balancing budget, labor, and other practical constraints. For example, some of the primary test design specifications that need to be considered include:

- dimensions of the target subsurface volume;
- anticipated amount of DNAPL present;
- number and locations of injection and extraction wells;
- injection and extraction rates;
- necessity of additional hydraulic control wells;
- tracer-test duration;
- sampling frequency;
- tracer suite (i.e., selection of tracers with the appropriate partition coefficients);
- tracer injection and extraction concentrations and acceptable detection limits;
- volume of tracer pulse; and
- extraction water treatment and disposal.

Furthermore, the specifications of the various physical components required for the PTT (injection/extraction wells, pumps, storage tanks, effluent treatment system, etc.) should be consistent with the operational requirements associated with the remediation activity. For example, in this study, PTT injection/extraction wells were located and

constructed so they could be utilized for both the PTTs and the remediation activities. Because effective test design is critical for success, a detailed description of the PTT design process and test specifications is provided in the following sections.

## Target Sweep Volume, Well Locations, and Test Hydraulics

Intuitively, the target PTT sweep volume should encompass all of the DNAPL zones of interest, but not be unnecessarily large in order to reduce test time, simplify hydraulic requirements, and minimize the costs associated with purchasing tracers, chemical analysis, and extraction fluid treatment. Furthermore, as more DNAPL-free zones fall within the flow field of the PTT, the average S<sub>N</sub> over the test volume will decrease, and at some point may approach a practical lower quantification limit of the PTT method. Consequently, it is highly beneficial to accurately delineate the source zone prior to designing and conducting a PTT.

For this demonstration project, the DNAPL zone was initially delineated with data from a previous focused investigation of the suspected source zone using Geoprobe<sup>TM</sup>-driven MIPs (CH2M HILL, 2001). These data provided relatively inexpensive discrete measurements of pore-water concentrations and were used to limit the suspected DNAPL zone to a relatively small footprint area (approximately 6 m  $\times$  12 m) located immediately east and northeast of the former metal plating shop. Additionally, the MIPs data indicated that most of the contamination was located in approximately the bottom 1.5 m of the aquifer. Therefore, this specific subsurface volume was targeted as both the primary treatment/remediation zone and the target sweep volume for the PTTs.

To optimize tracer sweep efficiency through the target sweep zone, a series of PTT models was constructed using a step-wise modeling approach. Initially, an analytical model was used to provide preliminary estimates of well spacing, tracer pulse volumes, and injection/extraction rates (Exhibit 1). For this model, a series of stream tubes were constructed to account for solute transport between an injection/extraction well pair (i.e., dipole-type flowfield) and the associated hydraulic-related tracer tailing. The results of these simulations also provided a basis for the anticipated degree of tracer tailing, tracer peak concentrations, and the test duration necessary to capture a significant portion of the BTC tail region.

Initially employing analytical models in the design processes can be advantageous, as they require minimal effort to construct and can then be used as a basis for subsequent complex numerical models. As shown in Exhibit 1, various test conditions can be easily simulated and the resulting predicted tracer BTCs can be assessed to provide initial estimates of optimal test design. The results obtained from the analytical-model simulations suggested that the target zone could be swept efficiently using a series of three to six injection and extraction wells located between 1.5 to 3.0 m apart, a tracer pulse volume of 5.7 to 9.5 m<sup>3</sup> (1,500 to 2,500 gallons), and a test duration of seven to ten days.

Based on the results of the analytical models, a more complex numerical flow and transport model (TOUGH/T2VOC) (Falta et al., 1992) was then constructed to guide well-installation efforts by identifying specific optimal well configurations and locations. Initially, the DNAPL source was thought to be in the vicinity of well I1 (see Exhibit 2). The initial design was thus a traditional six-spot remediation configuration with one injection well and five extraction wells. The primary purpose of the numerical model was to

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get sweep zone, a series



**Exhibit 1.** Examples of Tracer BTCs Predicted from the Analytical Model for Various Test Conditions (Well Spacing, Pumping Rates, Tracer Slug Volume, Etc.)

design well spacing and flow rates to achieve minimal loss of injected fluids, and to ensure the tracer concentrations comprising the BTC tails were above analytical detection limits.

During drilling, it was determined that the highest contaminant concentrations were likely in the vicinity of wells E6 and E3. The well design was then altered, given that the number of wells that could be installed for this project was essentially fixed. The actual well locations are shown in Exhibit 2, and were based on both the results of the preliminary numerical modeling and field observations, including observed lithology and contaminant field-screening results.

In the final design, well I1 was designed as a PTT injection well, and wells E2, E3, and E6 were designed as PTT extraction wells. This configuration was designed to measure an entire sweep volume of approximately 60 m<sup>3</sup>, with the ability to characterize  $S_{N}$  within three subzones (I1 to E2, I1 to E6, and I1 to E3). The remaining wells (E1, E4, E5, and E7) were used to provide additional hydraulic control during the PTTs and cyclodextrin flushing. These four wells were used primarily as injection wells to optimize the groundwater flow field, thereby improving mass recovery of the tracers and cyclodextrin. Subsequent to the drilling effort, it was determined that the former tank was probably located in the vicinity of well E6. The numerical model was then refined to be consistent with as-built well locations/specifications and small-scale spatial variations in hydraulic conductivity (determined by slug testing). Various PTT simulations were conducted to identify wellinjection and -extraction rates that optimized hydraulic control, tracer mass recovery, peak concentrations, tracer pulse length, and test duration. These results were used to optimize design parameters and also provide important predictive information. For example, the anticipated arrival time of the tracer peaks was used to determine test start and stop times, injected tracer concentrations, sampling frequency, and staffing requirements.



Exhibit 2. Final Locations of PTT Injection and Extraction Wells



**Exhibit 3.** Comparison of T2VOC-Predicted BTC to the Bromide BTC Observed during the Pre-PTT for Well E3

A comparison of the T2VOC model-predicted tracer BTC to the observed bromide BTC for the Pre-PTT (well E3) is shown in Exhibit 3. There is reasonable agreement between the model and the observed concentrations for the initial breakthrough time and the peak concentrations, especially considering the model simulation was not calibrated. Initially, the actual injection and extraction rates for the Pre-PTT were consistent with the model rates. However, some of the wells were unable to sustain these initial extraction rates. Furthermore, treatment of the extracted water at these flows was less efficient than expected. Consequently, extraction rates were decreased after 1.7 days (Exhibit 4). As seen in Exhibit 3, higher-than-predicted tracer tailing was observed in the field, indicating that actual tracer mass recovery was less efficient than predicted. This is partially related to the lowering of the field extraction rates, which reduced the rate of tracer recovery (i.e., the tracer recovery rate was reduced because the total fluid recovery rate was reduced). The tailing may also be due to flow heterogeneities and mass-transfer limitations that are not simulated by the model. A summary of average well-flow rates and tracer pulse volumes is provided in Exhibit 5.

It should be noted that the change in flow rates was systemwide and occurred at a distinct time; otherwise, flows remained constant throughout the tests (see Exhibit 4). Since the modification in system hydraulics affected both the conservative and partitioning tracers equally and rate-limited processes are assumed insignificant (e.g., instantaneous tracer partitioning is assumed), the analysis of the BTCs by the method of moments remains a valid method for determining partitioning tracer retardation.



Exhibit 4. Cumulative Injection and Extraction Volumes for the PTTs

#### Tracer Suite Design

Only two tracers (typically one conservative tracer and one partitioning tracer) are necessary to estimate  $S_N$ . However, it is beneficial to use a suite of partitioning tracers with various  $K_{NW}$  values in field applications to increase the likelihood that at least one tracer pair will exhibit optimal separation. This is important because the possible range of  $S_N$  for a site is typically very large, ranging from < 0.1 percent to > 10 percent

(e.g., Young et al., 2001 and Cain et al., 2000, respectively). Additionally, use of multiple tracers can provide redundancy, which reduces uncertainty in the  $S_N$  estimate.

Based on the results of the numerical modeling, the optimal partitioning tracer retardation was estimated to be between 1.1 and 1.8 (generally, Jin [1995] suggests a practical range of 1.2 to 4.0). This range was anticipated to provide sufficient separation from the conservative tracer BTC, while also permitting reasonable tracer mass recovery over the anticipated PTT duration. Although the location of the DNAPL source zone had been previously determined with MIPs, the estimated  $S_N$  value prior to the Pre-PTT was highly uncertain. Therefore, partitioning tracers were chosen for the Pre-PTT that were optimally designed to quantify  $S_N$  values ranging from 0.1 percent to 10 percent. This is indicated by the target region in Exhibit 6, which corresponds to partitioning tracers with target  $K_{NW}$  values ranging from approximately 2 to 200.

Based on the K<sub>NW</sub> range associated with the target region shown in Exhibit 6, the partitioning tracer suite for the Pre-PTT included 2-methyl-1-butanol (2M1B), 2-ethyl-1-butanol (2E1B), 2,4-dimethyl-3-pentanol (2,4DM3P), 2-ethyl-1-hexanol

Pre-PTT Flow Rate (LPM)						
Well	0 to 1.8 days	Purpose				
I1	13.9 until 0.43 day	7.3	Tracer injection well (8.6 m <sup>3</sup> for			
	11.8 until 1.8 day		0.43 day), then clean water			
			injection for hydraulic control			
E3	9.9	8.8	Tracer extraction well			
E6	9.0	4.5	Tracer extraction well			
E2	9.0	4.4	Tracer extraction well			
E5	12.9	7.6	Hydraulic control with clean			
			water injection			

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Flow Rate (LPM)						
Well	0 to 1.9 days	1.9 to 9.2 days	Purpose			
I1	5.0	4.8	Tracer injection well (7.0 m <sup>3</sup> for			
			0.97 day), then clean water			
	injection for hydraulic control					
E3	6.7	6.7	Tracer extraction well			
E6	5.4	5.4	Tracer extraction well			
E2	4.6	4.6	Tracer extraction well			
E5	3.1	12.2	Hydraulic control and treated			
			effluent disposal			
E1	0.0	4.5 (estimated)	Hydraulic control and treated			
			effluent disposal			

Post-PTT

Notes: LPM = liters per minute

Exhibit 5. Summary of Well-Injection and -Extraction Rates for the Pre- and Post-PTTs





Р		Post-PTT			
Tracer	κ <sub>nw</sub>	C <sub>。</sub> (mg/L)	Tracer	κ <sub>nw</sub>	C <sub>。</sub> (mg/L)
bromide	0	1,012	bromide	0	923
2-methyl-1-butanol	3.71 <sup>a</sup>	300	2-methyl-1-butanol	3.38 <sup>a</sup>	1,036
2-ethyl-1-butanol	13.4ª	219	4-methyl-2-pentanol	<b>9.66</b> <sup>b</sup>	545
hexanol	18.6 <sup>c</sup>	801	2-ethyl-1-hexanol	131 <sup>a</sup>	293
2,4-dimethyl-3-pentanol	71.3ª	175	heptanol	163.1 <sup>b</sup>	346
heptanol	163.1 <sup>b</sup>	279			
2-ethyl-1-hexanol	202ª	250			
Notes:			Sources:		
Alcohol tracers were measured	<sup>a</sup> Dugan et al., 2003				
Bromide was measured with an	<sup>b</sup> Young et al., 1999				
Analytical detection limit for a	.5 mg/L <sup>c</sup> Wang et al., 19	98			
$C_{o}$ = Injected concentration, m	g/L = millig	rams per liter			

**Exhibit 7.** Tracer Suites for the Field PTTs with  $K_{NW}$  Values (Cyclodextrin-Influenced Effective  $K_{NW}$  Values for Post-PTT Partitioning Tracers Are Based on Results Presented in Dugan et al. [2003])

Several nonbranched tracers were used to improve the distribution of  $K_{\rm NW}$  values and also to provide some redundancy at relatively low cost.

(2E1H), hexanol (HEX), and heptanol (HEP). The Post-PTT partitioning tracer suite included 2M1B, 4-methyl-2-pentanol (4M2P), 2E1H, and HEP. It is possible that the site DNAPL is composed of minor fractions of other contaminants (particularly TCA) in addition to TCE. However, the reported pure TCA K<sub>NW</sub> values are similar to the TCE K<sub>NW</sub> values (relative percent differences are generally ~20 percent); therefore, K<sub>NW</sub> values based on pure TCE DNAPL were assumed. The effective K<sub>NW</sub> values and injected concentrations are summarized in Exhibit 7.

Data from several reported field PTTs indicate that some alcohol tracers exhibit significant *in situ* biodegradation, even during the typically short duration of the PTT; however, methylated and ethylated "branched" alcohols are generally more recalcitrant than straight-chain alcohols (e.g., Annable et al., 1998). Therefore, these tracers were chosen as the primary tracer for a given target  $K_{NW}$  value, although these tracers are more expensive than the nonbranched alcohols. Several nonbranched tracers were used to improve the distribution of  $K_{NW}$  values and also to provide some redundancy at relatively low cost. However, these were considered "secondary" tracers. Fewer tracers were used in the Post-PTT because the approximate breakthrough times of tracers in the suite were known.

Commonly, after flushing-based remediation of a DNAPL zone, significant concentrations of residual remediation fluid remain in the treatment zone (e.g., Battelle and Duke, 2001; Falta et al., 1999; Jawitz et al., 2000; Lee et al., 1998; McCray & Brusseau, 1998; Vane & Yeh, 2002). In some cases, the concentrations of residual remediation fluids left in situ after treatment have been reported as high as 7 percent (Jawitz et al., 1998), and these residual fluids have the potential to modify effective K<sub>NW</sub> values for Post-PTT tracers. For example, PTT estimation errors associated with the presence of residual remediation fluids have been reported for several tests (Brooks et al., 2002; Knox et al., 1999; Vane & Yeh, 2002), and Battelle and Duke (2001) determined that data from a postremediation PTT were unusable due to unanticipated sorption to residual surfactant in the subsurface. Consequently, the influence of residual cyclodextrin on  $K_{_{\rm NW}}$  values was investigated in batch partitioning tests prior to field work (the results of these experiments are presented in detail in Dugan & McCray [2003]). Generally, it was determined that cyclodextrin lowers the apparent  $K_{_{NW}}$  for some alcohol tracers. However, as observed in the data for 2-ethyl-1hexanol (Exhibit 8), there is an apparent maximum affect, and the effective K<sub>NW</sub> values for various cyclodextrin concentrations can be predicted from empirical models. During the Post-PTT, residual concentrations of cyclodextrin were as high as 2 percent; therefore, this analysis provided a basis for the effective K<sub>NW</sub> values used for the Post-PTT (Exhibit 7).

In addition to the partitioning tracers, bromide (Br) was included in the tracer suite as a conservative tracer. Although  $S_N$  can be calculated directly from the transport of two partitioning tracers by Equation 4 (i.e., a conservative tracer is unnecessary), it is generally beneficial to include a conservative tracer since it provides a direct measure of actual fluid velocity and hydraulic-related transport behavior. It can also provide information about the biodegradability of the other nonconservative tracers by providing a benchmark for tracer mass recovery. Additionally, Br is relatively inexpensive and can be easily measured with an ion selective electrode (ISE).

## Sample Collection and Analysis

Tracer samples were collected from in-line effluent sampling ports at predetermined time intervals based on the results of the numerical models. Early in the tests, samples were

collected every 30 minutes to ensure accurate characterization of the BTC front and peak, while late in the tests (when the changes in tracer concentrations were small), samples were collected every two to eight hours (sampling became progressively less frequent as the test progressed). The sampling frequency was based on the numerical-model results and was confirmed real-time in the field by using the specific conductance of extraction fluids as an initial estimate of the tracer BTC concentrations. Samples were analyzed for bromide with an ISE in the field within approximately 2 weeks of collection. Samples collected for alcohol tracers were placed in coolers and shipped to the University of Arizona for analysis by gas chromatography. To provide additional biodegradation information, dissolved oxygen was also measured in effluent samples by headspace analysis with a gas chromatograph (see Divine et al., [2003] for a similar dissolved gas analytical set-up).

## **RESULTS AND DISCUSSION**

As shown in the field BTCs for the Pre- and Post-PTTs (Exhibits 9 and 10), the transport of the partitioning tracers is generally retarded relative to the conservative tracer (Br), indicating that DNAPL was present in the sweep zone. However, the magnitude of the retardation is small, indicating that the average DNAPL saturation was relatively low both prior to and after remediation. In fact, the *maximum* observed retardation for any alcohol tracer during the Pre-PTT was 1.101, which is approximately equal to the optimal *minimal* PTT design retardation of 1.1 discussed earlier. The observed tracer retardation coefficients (relative to Br by Equation 1) and tracer mass recoveries are











Exhibit 10. Post-PTT Tracer BTCs for Extraction Wells E2 (Top), E6 (Middle), and E3 (Bottom)

		Pre-	PTT						
	Tracer Retardation								
Well	and Recovery	Br	2M1B	2E1B	2,4DM3P	2E1H			
E3	R	1.000	1.084	1.087	1.101	1.072			
	Fractional Recovery	14.8%	17.4%	17.6%	17.2%	16.0%			
E6	R	1.000	0.982	0.971	1.003	1.001			
	Fractional Recovery	36.1%	36.4%	36.6%	37.6%	36.2%			
E2	R	1.000	0.980	0.954	1.015	0.944			
	Fractional Recovery	27.4%	26.5%	26.8%	28.3%	26.9%			
	Total Recovery	78.3%	80.3%	81.0%	83.1%	79.1%			
		Post	-PTT						
	Tracer Retardation								
Well	and Recovery	Br	2M1B	4M2P	2E1H				
E3	R	1.000	1.030	1.022	1.048				
	Fractional Recovery	36.9%	33.2%	38.2%	40.3%				
E6	R	1.000	1.001	0.971	0.982				
	Fractional Recovery	30.5%	36.2%	36.6%	36.4%				
E2	R	1.000	1.058	1.177	1.101				
	Fractional Recovery	38.4%	38.2%	46.4%	49.1%				
	Total Recovery	115.7%	110.0%	127.6%	138.9%				

**Exhibit 11.** Summary of Observed Tracer R Values and Mass Recovery (Fractional Recovery Is Calculated by the Tracer Mass Recovered at a Specific Extraction Well Divided by the Total Injected Tracer Mass)

summarized in Exhibit 11. HEX and HEP are not included due to significant mass balance errors due to tracer biodegradation (discussed later).

The total tracer recoveries for the Pre-PTT ranged from 78 percent–83 percent, which is lower than the model-predicted recovery of approximately 95 percent. However, this may be explained by the lowered field extraction rates mentioned previously. During the Post-PTT, extraction fluids were reinjected into wells E5 and E1 due to an unanticipated change in regulatory requirements. Because the air-stripper treatment system was designed primarily to treat TCE and other VOCs, measurable concentrations of bromide and alcohols were present in reinjected water (up to 10 percent of the initial injection concentrations). As a result of this reinjection, a second minor tracer peak is observable in all Post-PTT BTCs. These secondary peaks were not included in the exponential tail extrapolation (as indicated by the extrapolation lines in Exhibit 10); however, the larger primary tracer peak may also mask the effects of re-injected fluids. This may explain the high tracer mass recoveries calculated for the Post-PTT (110-139 percent), even when the distinct secondary peaks are ignored. However, because the majority of the tracer response is caused by transport and partitioning processes within the target sweep zone, the analysis of Post-PTT data may still provide useful information regarding postremediation S<sub>N</sub>.

A significant reduction in dissolved oxygen occurred as water flowed between the injection well and the extraction wells during both tests, with greater depletion during the Post-PTT (Exhibit 12). Given the relatively short average residence time of



**Exhibit 12.** Dissolved Oxygen Concentrations during Pre- and Post-PTT (Samples for Injection Well I1 Were Collected from the Injection Fluid Immediately Prior to Entering the Well)

the injected water (generally less than a day or two), this suggests significant *in situ* biological activity. It has been previously documented that biodegradation of alcohol tracers can occur during PTTs (Annable et al., 1998; Cain et al., 2000), and it is likely that some biodegradation of the alcohol tracers occurred during this test. The higher microbial activity observed during the Post-PTT may have been caused by an enhancement of the *in situ* microbial population during the remediation activity due to the near-continual injection of cyclodextrin fluid (Alter et al., 2003). In particular, the propylene glycol and other byproducts that make up about 10 percent of the technical grade cyclodextrin solution may have served as a microbial food source. The magnitude of partitioning tracer biodegradation and the associated error is difficult to estimate. However, straight-chain alcohols are preferably biodegraded, and this was supported by the generally lower mass recoveries of HEX and HEP and tracer behavior in the tail regions of the BTCs. Therefore, only BTC data from methylated and ethylated alcohols were utilized for S<sub>N</sub> estimation.

Exhibit 13 summarizes the calculated raw  $S_N$  values for the various tracer pair combinations based on the observed tracer R values (using Equations 3a and 4). Ideally, all  $S_N$  estimates should agree for each tracer pair, providing redundancy and increasing

Pre-PTT				Post-PTT					
Total Sweep Volume = 62.4 m <sup>3</sup>				Total Sweep Volume = 54.6 $m^3$					
		E3				E	3		
Subzone Sweep Volume = $28.1 \text{ m}^3$				Subzone Sweep Volume = 25.0 $m^3$					
	2M1B	2,4DM3P	2E1B	2E1H		2M1B	4M2P	2E1H	
Br	2.20%	0.14%	0.65%	0.04%	Br	0.87%	0.23%	0.04%	
2M1B		0.02%	0.03%	< 0	2M1B		< 0	0.01%	
24DM3P			0.02%	< 0	4M2P			0.02%	
2E1B				< 0					
F6					E6				
Su	bzone Sv	veep Volume	$= 16.6 \text{ m}^3$		Subzone Sween Volume = $15.4 \text{ m}^3$				
	2M1R	2 / DM2D	2E1 B	2E1U		2M1R	/M2D	2E1U	
Br	< 0	0.00%		0.01%	Br		0 1/%	0.03%	
2M1R	< 0	0.00%	< 0	0.01%	2M1R	< 0	0.14%	0.03%	
24DM3P		0.05 /0	0.06%	< 0	4M2P		0.55 10	0.02%	
2E1B			0.00%	0.02%				0.02 /0	
		50				-	•		
<b>C</b>	hanna Cu	E2	170 m <sup>3</sup>		E2				
Subzone Sweep Volume = $17.8 \text{ m}^3$				Subzone Sweep Volume = 14.2 m <sup>3</sup>					
	2M1B	2,4DM3P	2E1B	2E1H		2M1B	4M2P	2E1H	
Br	< 0	0.02%	< 0	< 0	Br	1.70%	1.80%	0.08%	
2M1B		0.05%	< 0	< 0	2M1B		1.87%	0.03%	
24DM3P			0.11%	< 0	4M2P			< 0	
2E1B				< 0					

**Exhibit 13.** Summary of Raw  $S_N$  Estimates from Tracer Pairs (Equations 3 and 4) and Subsurface Sweep Volumes Determined from Br Transport (Calculated Negative  $S_N$  Values are Noted as "< 0")

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confidence in the results. Unfortunately, there is poor consistency among tracer pairs yielding positive  $S_N$  estimates, and many tracer pairs yield negative  $S_N$  values (negative values are calculated when tracer retardation values are inconsistent with the relative rank of tracer  $K_{NW}$  values). Furthermore, the general performance of specific tracer pairs is not necessarily consistent for each test. For example, the Br-2E1B pair yields an  $S_N$  estimate of 0.65 percent for the Pre-PTT I1-E3 subzone; however, this pair yields negative values for the Pre-PTT I1-E6 and I1-E2 subzones.

The high tracer mass recoveries (> 100 percent) for the Post-PTT suggest significant uncertainty is associated with the results of this test. In particular, the Post-PTT BTC for well E2 is likely the most influenced by the reinjection of tracer fluids because it was closest to the injection wells, and, therefore, these results are highly questionable. This is generally supported by the relatively high tracer mass recoveries and some anomalously high  $S_N$  estimates for this well. Since the majority of remediation efforts were focused in the subzones I1-E3 and I1-E6, it is assumed that the actual  $S_{N}$  of the I1-E2 subzone changed little between the Pre- and Post-PTTs. With this assumption, a summary review of Exhibit 13 indicates that the highest average DNAPL saturations were present in the I1-E3 subzone, with lesser saturations in the I1-E6 and I1-E2 subzones. This observation is consistent with results from field screening during well installation, background contaminant concentration measured at these wells, and probable DNAPL location based on local lithology and the geologic topography of the underlying clay unit. That is, based on well logs, DNAPL transport would be expected to be nearly vertical in the sandy, relatively homogeneous aquifer. Once DNAPL encountered the marine clay unit at the bottom of the aquifer, it may have migrated toward E3, as the clay unit dips toward E3.

The relatively large range in calculated  $S_N$  values indicates there is notable uncertainty associated with the PTT results, and poses a challenge for data interpretation. Given that no single method for analyzing the PTT data is obvious, we present the following possible interpretations and discuss relevant merits and drawbacks for the various approaches.

## Interpretation 1: Average All Positive $S_N$ Estimates

A relatively straightforward interpretation is to average all calculated  $S_N$  values for tracer pairs yielding positive values. This approach utilizes all available  $S_N$  estimates, and therefore, may reduce the influence of individual outlying high or low values. However, all values are considered equally, which assumes that all tracers have equivalent uncertainties. As noted earlier, due to the higher mass-balance errors associated with the Post-PTT BTC at well E2 and the fact that relatively little remediation effort was targeted in this area, the  $S_N$  value for the I1-E2 subzone is assumed to have not changed significantly between PTTs. Therefore, the Pre-PTT  $S_N$  results are used for the Post-PTT estimate for subzone I1-E2 instead of analyzing the Post-PTT BTC for this well (this assumption is also made for all following interpretations). By this analysis, the average sweep volume  $S_N$  estimates are 0.22 percent (Pre-PTT) and 0.16 percent (Post-PTT). However, given the large range in calculated  $S_N$  values, it is likely that there are systemic errors associated with specific tracers and/or tracer pairs. The following approaches consider only selected tracer pairs based on qualitative and quantitative criteria.

The high tracer mass recoveries (> 100 percent) for the Post-PTT suggest significant uncertainty is associated with the results of this test. Small differences in transport times may be difficult to resolve from field data due to integration errors associated with the temporal moment estimation.

## Interpretation 2: Elimination of High K<sub>NW</sub> Tracers (2E1H and 2,4DM3P)

Brooks et al. (2002) present the results of a field PTT where tracers with higher KNW values occasionally yielded inconsistent  $S_N$  estimates. This response is likely related to the high relative sensitivity of the  $S_N$  calculation for these tracers to mass-balance and temporal moment estimation errors. Therefore, results from 2E1H ( $K_{NW} = 202$ ) and 2,4DM3P ( $K_{NW} = 71.3$ ) tracer pairs are discounted. Eliminating 2E1H may be supported by the fact that most  $S_N$  estimates are negative for 2E1H pairs for the Pre-PTT. However, all 2E1H pairs yielded positive  $S_N$  estimates for the Post-PTT. By this analysis, the average sweep volume  $S_N$  estimate before remediation is 0.43 percent and after remediation is 0.32 percent. For the Pre-PTT, this interpretation eliminates all positive  $S_N$  for these subzones.

# Interpretation 3: Bromide Pairs Only

As discussed,  $S_N$  can be estimated with two partitioning tracers by Equation 4. However, the separation between partitioning tracers may be small, especially if the tracers have similar  $K_{NW}$  values. Small differences in transport times may be difficult to resolve from field data due to integration errors associated with the temporal moment estimation. Furthermore, when two partitioning tracers are used, there is uncertainty regarding the  $K_{NW}$  values for both tracers. Conversely, there is typically very little uncertainty regarding the nonpartitioning nature of the conservative tracer (especially for an anionic tracer such as Br). For this approach,  $S_N$  is estimated by considering only positive values for Br tracer pairs, resulting in average  $S_N$  values of 0.35 percent (Pre-PTT) and 0.20 percent (Post-PTT).

# Interpretation 4: Tracers with Most Consistent S<sub>N</sub> Estimates

For this approach, a single "best performer" tracer was defined for each test as the tracer yielding the most consistent results (i.e., smallest range and lowest standard deviation) for all pair combinations for each test. The average  $S_N$  was estimated by averaging positive results of all pairs for this tracer. Of course, any errors associated with a conjugate tracer will affect the average value; however, this approach may eliminate highly erroneous  $S_N$  values calculated from two "poorly performing" tracers. The tracers yielding the most consistent  $S_N$  values are 2,4DMP for the Pre-PTT and 2E1H for the Post-PTT. By this analysis, the average  $S_N$  estimates are 0.05 percent (Pre-PTT) and 0.03 percent (Post-PTT).

# Interpretation 5: Most Consistent Tracer with Br

As in the previous approach, the best performing tracer was defined as the partitioning tracer yielding the most consistent  $S_N$  estimates for all pair combinations; however,  $S_N$  was then estimated using only the Br-"best performer" pair. As pointed out in Interpretation 3, there is little uncertainty associated with the conservative behavior of Br. Consequently, this approach may identify the single most reliable  $S_N$  estimate for each BTC. The tracers yielding the most consistent  $S_N$  values are 2,4DMP for the Pre-PTT and 2E1H for the Post-PTT. The average  $S_N$  estimates by this analysis are 0.07 percent before remediation and 0.03 percent after remediation.



**Exhibit 14.** Comparison of Average Sweep Volume  $S_N$  Estimates for the Five Interpretations Approaches Described in the Text

## Interpretation Summary

The estimates from these five interpretations are summarized in Exhibit 14. These estimates indicate that the cyclodextrin flushing reduced  $S_N$ , and that the volume of DNAPL removed was between 15 to 109 L. This is generally consistent with the removal volume (~30 L) estimated by mass-balance analysis of effluent contaminant concentrations measured during the remediation. However, the various interpretations result in a large range of absolute  $S_N$  estimates, with up to an order-of-magnitude range for the estimate of final  $S_N$  (from 0.03 percent to 0.32 percent).

As noted earlier, the large range of calculated  $S_N$  values from the various tracer pairs (Exhibit 13) indicates significant uncertainty regarding the PTT results. For example, depending on the specific tracer pairs used for each BTC, the range of possible average SN estimates is greater than two orders of magnitude: between 0.02 percent (Br-2E1H pairs) and 0.99 percent (Br-2M1B pairs) for the Pre-PTT and between 0.02 percent (2M1B-2E1H pairs) and 0.52 percent (Br-2M1B pairs) for the Post-PTT. For all tracers, the observed retardation was relatively low, and at some point, the effects of tracer measurement and mass-balance errors create a lower limit for reliable  $S_N$  quantification for the PTT method. For example, the theoretical R values for partitioning tracers with  $K_{NW}$  values ranging from 2 to 20 in a case where  $S_N = 0.5$  percent are 1.01 to 1.05, which requires the ability to resolve a 1 percent to 5 percent difference in tracer travel times. For some field conditions, errors and uncertainties may not permit reliable resolution of tracer transport below these levels. Additionally, minor alcohol-tracer sorption to aquifer materials, which is typically ignored, may cause proportionally greater estimation errors at low  $S_N$  conditions (see Brooks et al., 2002; Edgar, 1997).

Generally, PTTs offer much promise for DNAPL-zone characterization, and may provide an effective metric for assessing source-zone remediation efforts for some sites. The effective lower  $S_N$  quantification limit is test-specific, as it is dependent on the specific errors and uncertainties associated with test characteristics (analytical reliability, sampling density, BTC characteristics, etc.). The calculated  $S_N$  values for several tracer pairs yield negative values (noted as "< 0" in Exhibit 13), and the range of positive SN values is relatively large, suggesting that a quantification level was approached in these tests, especially for the Post-PTT. Similarly, Brooks et al. (2002) observed significant errors for field PTTs conducted under low  $S_N$  conditions, and present additional discussion on constraints for PTT application in these applications. Clearly, further work is needed to develop methods for reliably estimating test-specific practical lower quantification levels for field PTTs.

The Post-PTT results are valuable, as they independently confirm that  $S_N$  was reduced within a probable range that is consistent with the volume of DNAPL removed based on concentration mass-balance calculations. The relative uncertainties associated with these PTT results are notably greater than uncertainties typically reported for other field PTTs; however, the estimated initial and final  $S_N$  values at this site are lower than typically reported values. These results suggest that, in general, PTT application may be less suitable when  $S_N$  is low. Furthermore, greater uncertainty should be expected for postremediation PTTs. If remediation efforts are highly effective (resulting in a low remaining  $S_N$ ), use of postremediation PTTs to quantify remedial efficiency may be of limited practicality, particularly if remedial agents remain in the subsurface during the post-PTT.

It is also valuable to note that, although Pre- and Post-PTTs can theoretically be designed to measure precisely the same target zone, duplicating exact test conditions (i.e., injection and extraction rates) for both tests can be challenging under field conditions. Deviations in both absolute and relative flow rates for injection and extraction wells will cause differences in the volume and specific dimensions of the subsurface zone measured by the PTT. For example, for these tests, the Post-PTT measured a somewhat smaller (and therefore different) subsurface target zone, introducing additional uncertainty in the DNAPL volume removal estimates.

# SUMMARY AND CONCLUSIONS

Generally, PTTs offer much promise for DNAPL-zone characterization, and may provide an effective metric for assessing source-zone remediation efforts for some sites. Specifically, PTTs can provide a direct measure of the average  $S_N$  over a sweep volume and may also provide information on DNAPL distribution. However, successful field PTT implementation is predicated on careful design. For example, preliminary PTT modeling is critical for designing an efficient hydraulic regime and anticipating tracer transport behavior. Generally, the Pre-PTT results confirm that DNAPL was present in the test zone before remediation, and the Post-PTT results indicate that  $S_N$  decreased due to remediation activities. However,  $S_N$  estimates from the various tracer pairs are relatively inconsistent, indicating significant uncertainty and suggesting that the small amount of DNAPL present was near the reliable quantification level for the tests.

Because greater uncertainty may generally be associated with postremediation PTTs, the most appropriate and cost-effective method for determining DNAPL-zone remediation efficiency may be to conduct a preremediation PTT, and then determine the remaining  $S_N$  by mass balance when recovered contaminant mass can be accounted for (i.e., effluent sampling). However, for some remedial techniques, such as chemical

oxidation, independently tracking contaminant treatment may be difficult. In these cases, performing both Pre- and Post-PTTs may be the most reliable remediation metric method. Regardless, the results of these PTTs clearly indicate that further work is needed to better understand practical limitations of the PTT method, particularly for quantifying low  $S_N$  values. In particular, it is important to understand the lower quantification limits for postremediation PTTs, because the results of these tests represent the amount of remaining DNAPL and may be used to assess long-term site risk.

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