

EFFECTIVENESS MONITORING OF SOIL VAPOR EXTRACTION WITH REBOUND TESTING AND FORMATION OF A PATHWAY TO CLOSURE

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ABSTRACT

A pilot study was designed and installed at a small waste site in the Department of Energy's Savannah River Site to examine the effectiveness of soil vapor extraction (SVE) remediation of trichloroethylene (TCE) contamination in a layered vadose zone. The objectives of the work presented here are to report on the effectiveness monitoring of this SVE system and provide the basis for establishing that the site closure criteria have been met. The challenges encountered at this site included subsurface heterogeneity and an unknown initial contaminant distribution. These challenges were addressed by the development of a comprehensive subsurface soil gas TCE concentration rebound testing program that became the basis for site closure. Over the course of four years of SVE operation, six rebound tests have been conducted at the site. The progression of rebound tests show declining observed soil gas TCE concentration rebound at nearly all of the SVE wells. Rebound has been observed at a few SVE wells and monitoring points, but the concentrations achieved at these locations have been substantially lower than observed initial concentrations. These observations indicate that a substantial portion of the available contaminant mass has been removed and that continued SVE operation would not recover much additional contaminant mass. Additional information from mass removal and diffusion modeling support these observations and the resulting decision was to transition the SVE system to passive operation.

INTRODUCTION

Soil vapor extraction (SVE) is a common remedy for contamination caused by the release of volatile organic compounds into the subsurface. SVE studies have been reported widely in the literature. In a typical SVE installation, wells are screened in the subsurface, taking into consideration the geological features of the site and the spill distribution, and vacuum is applied to the SVE system to draw the contaminant vapors to the surface for treatment. Of the abundant discussion of SVE in the literature, few studies present a comprehensive approach to the monitoring of SVE systems and address the endpoint criteria for these systems. A problem encountered at many sites is incomplete knowledge of the spill amount and distribution. Monitoring strategies and closure criteria that overcome this limitation are needed for these sites.

Subsurface heterogeneity and spill properties such as amount, distribution and age are important parameters to consider during the design of SVE systems [1]. The presence of non-aqueous phase liquid (NAPL) is equally important. These properties will affect system behavior during the course of operation as well. The presence of low permeability materials in the subsurface increases the remediation time required because of poor advective flow in these regions as well as the slow diffusion of contaminants into and out of these materials [2, 3]. Likewise, the longer

a spill has been allowed to equilibrate in the subsurface, the more difficult it will be to remove it completely [4]. The more common site closure criteria encountered in the literature are removal of a specified fraction of the total mass present [5, 6] or termination of operation once an asymptotic limit has been reached during operation [7]. The first criterion is inappropriate for a site where the total contaminant mass is unknown. The second criterion may not reflect adequate mass removal from the subsurface, especially in the case of overly aggressive operation where substantial dilution may occur, resulting in the subsequent observation of subsurface concentration rebound and the continued need for remedial operation.

Subsurface concentration rebound after operation is a phenomenon that has been observed widely with pump and treat remediation systems [5, 8-11]. When the pump and treat system is shutdown, subsurface contaminant concentrations rise as diffusion from the less accessible pathways occurs. The nature of the remaining contaminant and its proximity to the observation point affect the speed and magnitude to which contaminant concentrations recover. Rebound may occur even in the absence of a NAPL source. Some researchers have attributed this and other observed non-equilibrium processes to the slow diffusion of contaminant molecules into and out of soil aggregates [9, 12-16]. Subsurface concentration rebound is an important phenomenon that can be harnessed for site evaluation.

The objectives of the work presented here were to follow the approach to performance tracking for an SVE system that was laid out in previous work [17] and establish that the criteria for closure had been reached.

EXPERIMENTAL

Site Background

A pilot scale soil vapor extraction and air sparging (SVE/AS) system was installed at the C-Area Burning Rubble Pit (CBRP), a small waste area within the Department of Energy's Savannah River Site. This system was designed and installed in 1999 to examine the effectiveness of those technologies in the remediation of trichloroethylene (TCE) contamination in layered soils.

The subsurface stratigraphy at CBRP is diverse, with interbedded layers ranging from clayey sands to sandy, silty clays (Fig. 1). A stiff clay layer of thickness varying up to 1.5 m (5 ft) divides the vadose zone at a depth of approximately 9 m (30 ft). The water table is present at a depth of approximately 21 m (70 ft) and is divided into two primary zones by a tan clay layer present at a depth of approximately 27 m (88 ft).

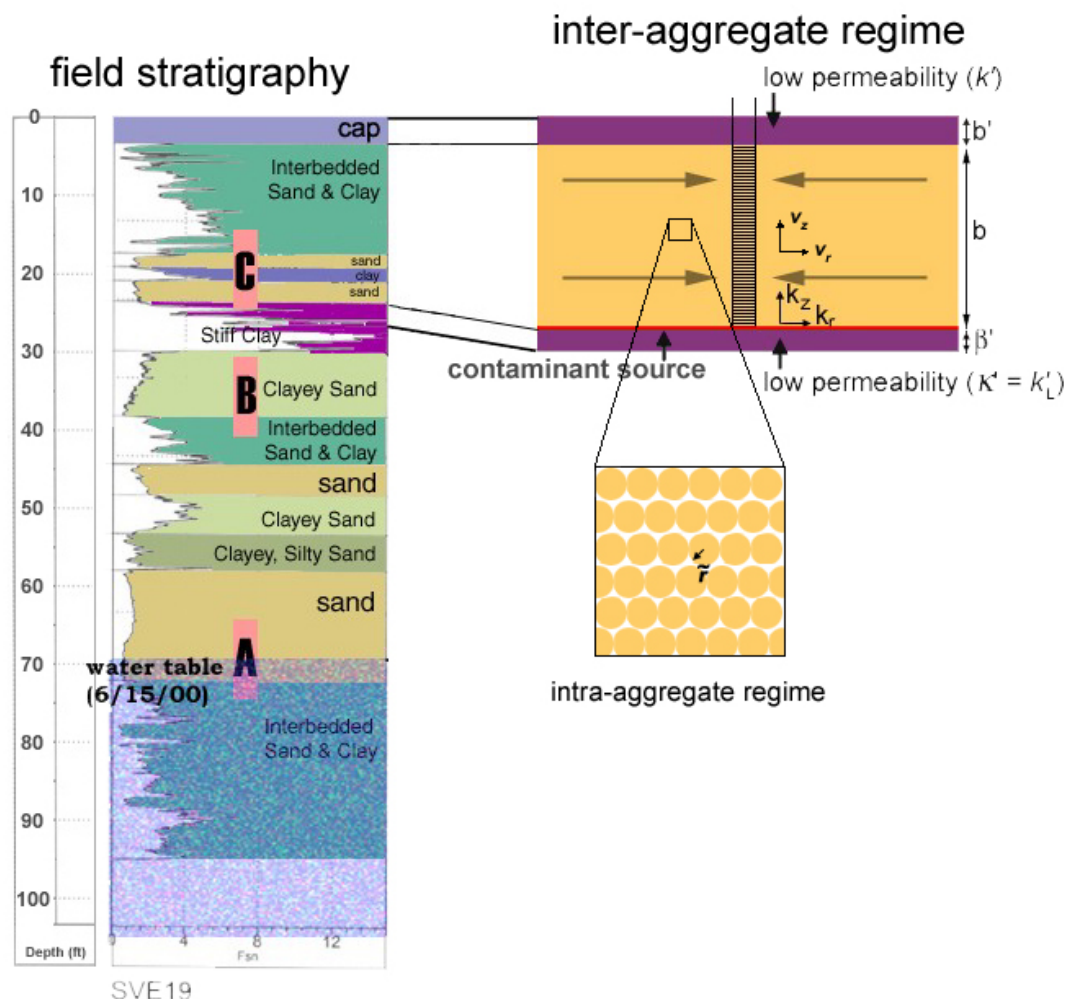


Fig. 1. Subsurface stratigraphy at CBRP is translated to the SVE model (reprinted from [18]).

In 1999, 39 SVE wells and 17 AS were installed in and around CBRP according to the subsurface stratigraphy and contaminant distribution.¹ Over the original pit, 24 of the 39 SVE wells were installed at eight locations in a clustered configuration. At each location, three wells were installed with the “A” location screened at the water table, the “B” location screened in the middle vadose zone (below the stiff clay layer) and the “C” location screened in the shallow vadose zone (above the stiff clay layer).

Three SVE locations were selected for extensive characterization (Fig. 2). These wells, SVE 18, 19 and 22, formed a triangle around the area believed to contain the most contamination based on preliminary soil gas concentration measurements. In May 1999, soil cores were collected from this area for laboratory characterization. In June 1999 and again in April 2001, a team from USEPA installed soil gas monitoring probes to track soil gas concentrations and vacuum at locations away from the SVE wells.

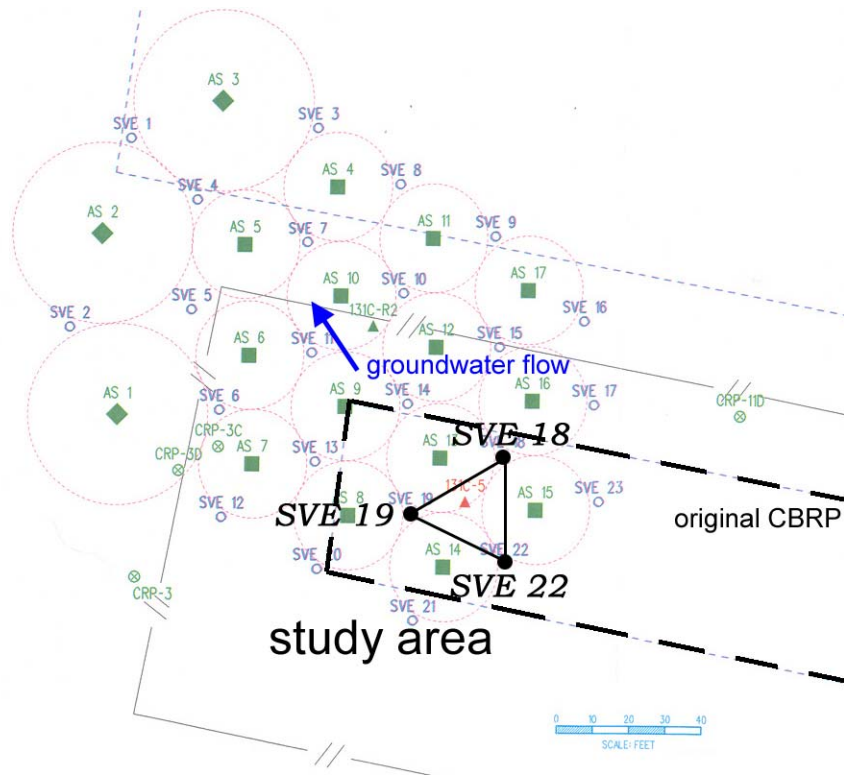


Fig. 2 Schematic diagram of the installed SVE/AS system at CBRP. The SVE 18-19-22 study area is highlighted.

Operation of the SVE system began in September 1999. The AS began operation in June 2000. The operational history at CBRP can be found in Table I. Discussion here is limited to the evaluation of the SVE system, but information about the air sparging system is included for completeness.

Table I Operational history at CBRP

Dates	Event
9/28/1999-10/13/1999	Preliminary operation and testing
10/13/1999-10/15/1999	Zone of influence testing (18BC, 19BC, 22BC)
10/18/1999-1/17/2000	Operation (18BC, 19BC, 22BC)
~12/1/1999	<i>Additional SVE wells operating</i>
1/17/2000 - 1/17/2000	System down
1/18/2000 - 1/28/2000	Operation
1/30/2000 - 2/13/2000	Rebound test #1
2/14/2000 -2/18/2000	Operation
2/19/2000 - 4/24/2000	System down for repairs
4/24/2000 - 6/30/2000	Operation
6/30/2000	Preliminary test of AS system
8/9/2000 - 10/2/2000	Zone of Influence Testing (SRTC)
10/3/2000	Operation resumed
10/16/2000	AS added to the configuration
11/6/2000 - 11/29/2000	System down for maintenance
	Rebound test #2
11/29/2000	Operation resumed
12/6/2000	AS added to the configuration
4/9/2001 - 5/9/2001	System down
	installation of new implants
	Rebound test #3
5/10/2001	SVE operation resumed
5/17/2001	AS operation resumed
11/2001	Soil coring - DNAPL detected
1/24/2002 - 2/25/2002	Rebound test #4
2/25/2002	SVE/AS resumed
9/9/2002 - 10/28/2002	Rebound test #5
10/28/2002	SVE resumed
3/24/2003 - 10/1/2003	Rebound test #6
10/1/2003	SVE resumed
1/1/2004	Transition to passive vapor extraction

Diffusion Studies

During the course of operation at CBRP, six soil gas TCE concentration rebound tests were conducted. For each test, the SVE system was shutdown for a period ranging from several weeks to several months to observe soil gas TCE concentration rebound. Often, these tests were scheduled to coincide with routine maintenance at the site. The first test was conducted for two weeks and concentration measurements were limited to the SVE 18-19-22 test area, including the monitoring probes. Subsequent tests were carried out for four weeks or longer (tests 2 – 5). The sixth and final test was carried out for six months. Soil gas concentration measurements were expanded to the entire SVE well field for the third through sixth rebound tests. All soil gas TCE concentration measurements were made with a Brüel & Kjær (B&K) portable photoacoustic detector. Periodically, soil gas samples were collected and measured by gas chromatograph mass spectrometer to verify the B&K measurements.

MODEL DEVELOPMENT

Governing Equations

A model was developed to simulate soil vapor extraction in layered materials. This model includes components that represent advection and dispersion in the bulk interstitial space and diffusion into and out of soil aggregates, as part of the overall model. The governing equations are:

$$R_{v,INTER} \frac{\partial C_v^I}{\partial t} = D_{eff} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_v^I}{\partial r} \right) + \frac{\partial^2 C_v^I}{\partial z^2} \right] - v_r \left[\frac{1}{r} \frac{\partial}{\partial r} (r C_v^I) \right] - v_z \left[\frac{\partial C_v^I}{\partial z} \right] - \sum_{k=1}^m \frac{3(1-\varepsilon)}{\tilde{R}_k} D_{INTRA,k} \frac{\partial \tilde{C}_{L,k}^{II}}{\partial \tilde{r}} \Big|_{\tilde{r}=\tilde{R}_k} \quad (\text{Eq. 1})$$

subject to the initial and boundary conditions:

$$t = 0 \quad r_w \leq r \leq R_O \quad 0 \leq z \leq b \quad C_v^I = C_0 \quad (\text{Eq.2})$$

$$t > 0 \quad r = r_w \quad 0 \leq z \leq b \quad \frac{\partial C_v^I}{\partial r} = 0 \quad (\text{Eq.3})$$

$$t > 0 \quad r = R_O \quad 0 \leq z \leq b \quad C_v^I = 0 \quad (\text{Eq.4})$$

$$t > 0 \quad r_w \leq r \leq R_O \quad z = 0 \quad \frac{\partial C_v^I}{\partial z} = 0 \quad (\text{Eq.5})$$

$$t > 0 \quad r_w \leq r \leq R_I \quad z = b \quad C_v^I = 0 \quad (\text{Eq.6a})$$

$$t > 0 \quad R_I \leq r \leq R_O \quad z = b \quad C_v^I = C_v^{sat} \quad (\text{Eq.6b})$$

after [18]. The boundary conditions are no flux at the upper boundary, zero concentration at the outer radial boundary and symmetry at the inner radial boundary (Fig. 1). The contaminant source is defined as a continuous, evaporating non-aqueous phase liquid (NAPL) pooled at the lower boundary b a distance R_I from the SVE well. NAPL volatilization is assumed instantaneous [4,13]. If the advective components in (Eq.1) are neglected, the following equation

$$R_{v,INTER} \frac{\partial C_v^I}{\partial t} = D_{eff} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_v^I}{\partial r} \right) + \frac{\partial^2 C_v^I}{\partial z^2} \right] - \sum_{k=1}^m \frac{3(1-\varepsilon)}{\tilde{R}_k} D_{INTRA,k} \frac{\partial \tilde{C}_L^{II}}{\partial \tilde{r}} \Big|_{\tilde{r}=\tilde{R}_k} \quad (\text{Eq.7})$$

represents diffusive processes in the subsurface while SVE is not operating.

Diffusion into and out of each soil aggregate regime k is represented as one dimensional in space and spherical [16]:

$$R_{INTRA} \frac{\partial \tilde{C}_{L,k}^{II}}{\partial t} = D_{INTRA,k} \left[\frac{1}{\tilde{r}^2} \frac{\partial}{\partial \tilde{r}} \left(\tilde{r}^2 \frac{\partial \tilde{C}_{L,k}^{II}}{\partial \tilde{r}} \right) \right] \quad (\text{Eq.8})$$

subject to the initial and boundary conditions:

$$t = 0 \quad 0 \leq \tilde{r} \leq \tilde{R}_k \quad \tilde{C}_{L,k}^{II} = \frac{C_0}{H} \quad (\text{Eq.9})$$

$$t > 0 \quad \tilde{r} = 0 \quad \frac{\partial \tilde{C}_{L,k}^{II}}{\partial \tilde{r}} = 0 \quad (\text{Eq.10})$$

$$t > 0 \quad \tilde{r} = \tilde{R}_k \quad \tilde{C}_{L,k}^{II} = \frac{C_v^I}{H} \quad (\text{Eq.11})$$

Model Solution

The solution to (Eq.7) with initial boundary conditions (Eq.2) – (Eq.6) was formulated in Matlab (Student Release 13, Natick, MA) as an explicit finite-difference, using central differences for space and forward differences in time. The step size in both the axial and radial directions was 7.62 cm (0.25 ft). At each temporal and spatial step in the overall solution, the aggregate equation (Eq.8) with initial and boundary conditions (Eq.9) – (Eq.11) was solved implicitly. Within each temporal and spatial step, the concentration within each aggregate domain was evaluated at five distance steps, establishing the problem as a set of five equations with five unknowns. The solution was achieved using a Gaussian elimination procedure [19, 20].

The distance from the source to the observation point was varied by evaluating selected cases between the observation point ($r = r_w$) and the outer boundary ($r = R_O$). Model concentrations for the SVE wells were determined by averaging the concentration measurements along the boundary ($0 \leq z \leq b$). Model concentrations for the monitoring probes were determined by single point concentration values at the depth of the monitoring probe.

RESULTS AND DISCUSSION

Mass Removal

Over the course of three years of operation at CBRP, approximately 820 kg of TCE has been removed from the subsurface. In the first year of operation, approximately 600 kg was removed, followed by 130 kg removed in the second year and 90 kg removed in the third year of operation. Mass removal data suggests that an asymptotic limit has been reached in terms of mass removal; however, mass removal alone is insufficient to determine the effectiveness of the SVE

installation, because a poor design would achieve this asymptotic limit sooner with insufficient mass removal from the subsurface. More information is necessary to make this evaluation.

Diffusion Studies

Six soil gas TCE concentration rebound tests were conducted during the course of four years of SVE operation at CBRP [18]. For each test, the SVE system was shutdown and the subsurface was allowed to equilibrate for several weeks. Soil gas TCE concentration measurements were made at the SVE wells and monitoring probes at several time intervals within that period. Data at SVE 18, 19 and 22 for all of the soil gas TCE concentration rebound tests can be found in Fig. 3.

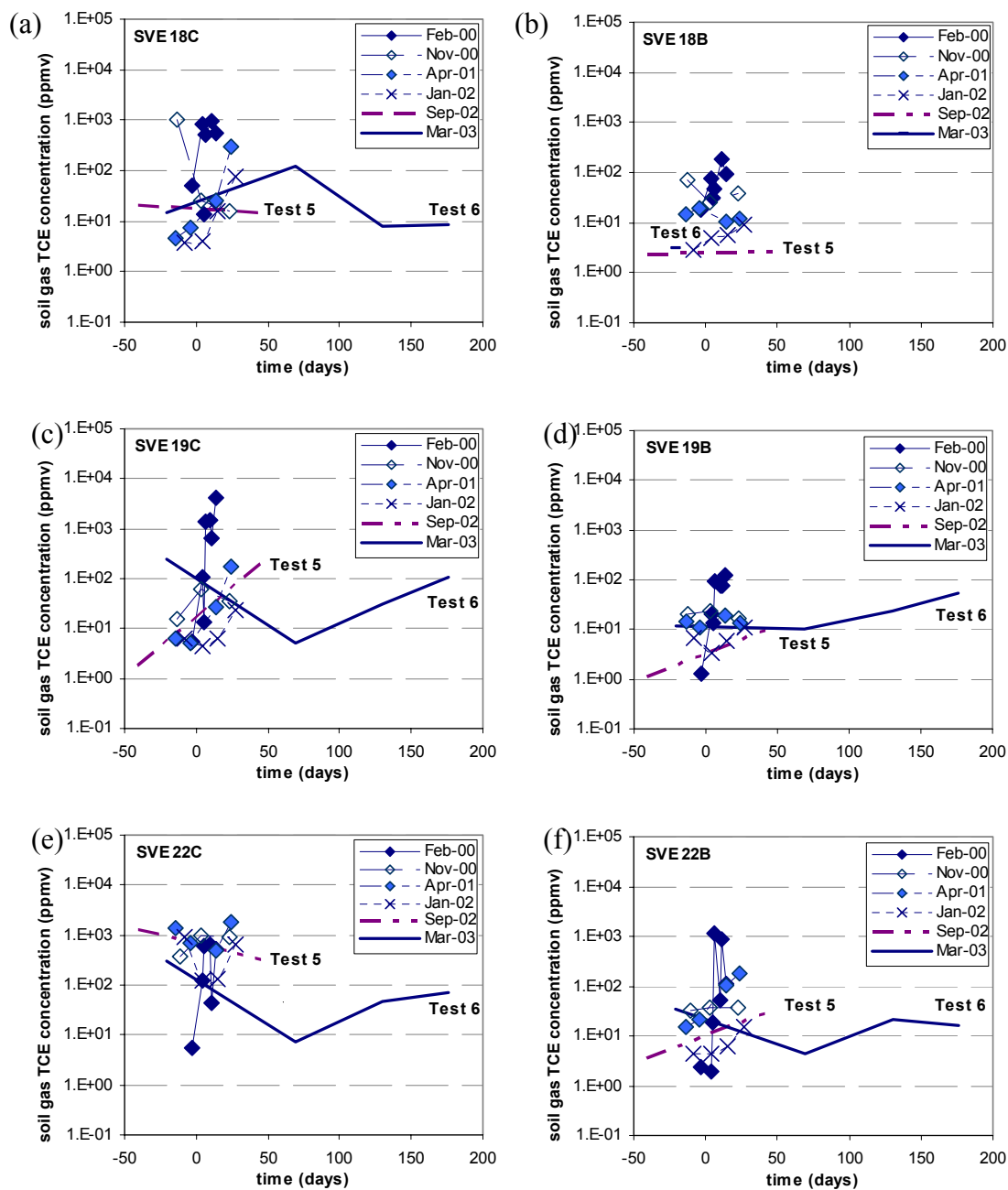


Fig. 3 Soil gas TCE concentration rebound tests conducted at (a) SVE 18C, (b) SVE 18B, (c) SVE 19C, (d) SVE 19B, (e) SVE 22C and (f) SVE 22B (reprinted from [18]).

Operation of the SVE system began in September 1999 and the first rebound test was carried out in February 2000. This test lasted two weeks. Soil gas TCE concentrations rebounded to levels at or near their starting values at many of the SVE wells. Rebound was observed at the monitoring probes as well, but in general, the concentrations achieved at these locations were lower than the starting values. The highest rebound concentrations at both SVE wells and monitoring probes correlated to the proximity of the stiff clay layer that divided the vadose zone, suggesting that the residual source had pooled above or seeped into this clay layer. TCE concentration measurements from soil cores collected in November 2001 confirmed a DNAPL source above this clay layer near the center of the SVE 18-19-22 triangle [21]

Subsequent soil gas TCE concentration rebound tests were conducted at regular intervals during the course of SVE operation. In general, these tests showed declining soil gas TCE concentration rebound at many of the SVE wells and monitoring probes. Persistent high soil gas TCE concentrations at SVE 22C and several of the monitoring probes suggested that either the SVE system was not addressing the contamination in that region or poor screen locations (wells/probes embedded in the stiff clay) were suggesting high concentrations throughout that region where in reality the high concentrations were limited to the SVE-inaccessible regions. Advective flow at SVE 22C was poor; however, advective flow at SVE 19C may have been sufficient to address the contamination in this region. For these reasons, additional monitoring probes were installed in the nearby region and that data confirmed that the second case was true. The elevated soil gas TCE concentrations were limited to the SVE-inaccessible regions.

Over the course of SVE operation, flow at SVE 22C improved. The presence of water in the subsurface has the effect of decreasing soil permeability [7, 22]. Continued SVE operation may have reduced subsurface soil moisture content thereby increasing the effective permeability of the region around SVE 22C. The improved flow and TCE mass removal at SVE 22C was followed by less observed rebound during the later tests, suggesting improved reduction of the TCE source in the immediate vicinity of this well (Fig. 3).

As the soil gas TCE concentration rebound tests showed slower, more shallow rebounds, the duration of each rebound test increased. The second through fourth tests were conducted over the course of approximately four weeks for each test. The fifth test was conducted over seven weeks. The sixth and final test extended six months to show the substantial reduction of subsurface soil gas TCE concentrations coupled with an equally substantial decline in mass removal indicated the performance limitations of the SVE process had been reached at this site.

Diffusion Model Predictions

The subsurface soil gas TCE concentration rebound tests were evaluated using a diffusion model to estimate the distance to a potential DNAPL source. Data from the first through fourth rebound tests were evaluated using the diffusion model (Eq.7). Data from the fifth test was not evaluated because of its similar behavior to that of test 4. The absence of data for many of the monitoring probes during the sixth test due to flooding prevented the analysis for this final test; however, given the observations of the fourth and fifth test, similar behavior was expected to have occurred.

The stiff clay layer in the subsurface between SVE 18-19-22 was assumed to be a continuously evaporating (but non-depleting) TCE source in the subsurface unless the distance estimates suggested otherwise. The succession of rebound tests 1-3 suggest the depletion of the DNAPL source over the course of SVE operation, with the increase in distance to the DNAPL source observed in the succession of rebound tests (Fig. 4). The location of the DNAPL that was detected by soil coring in November 2001 compares well to these predictions.

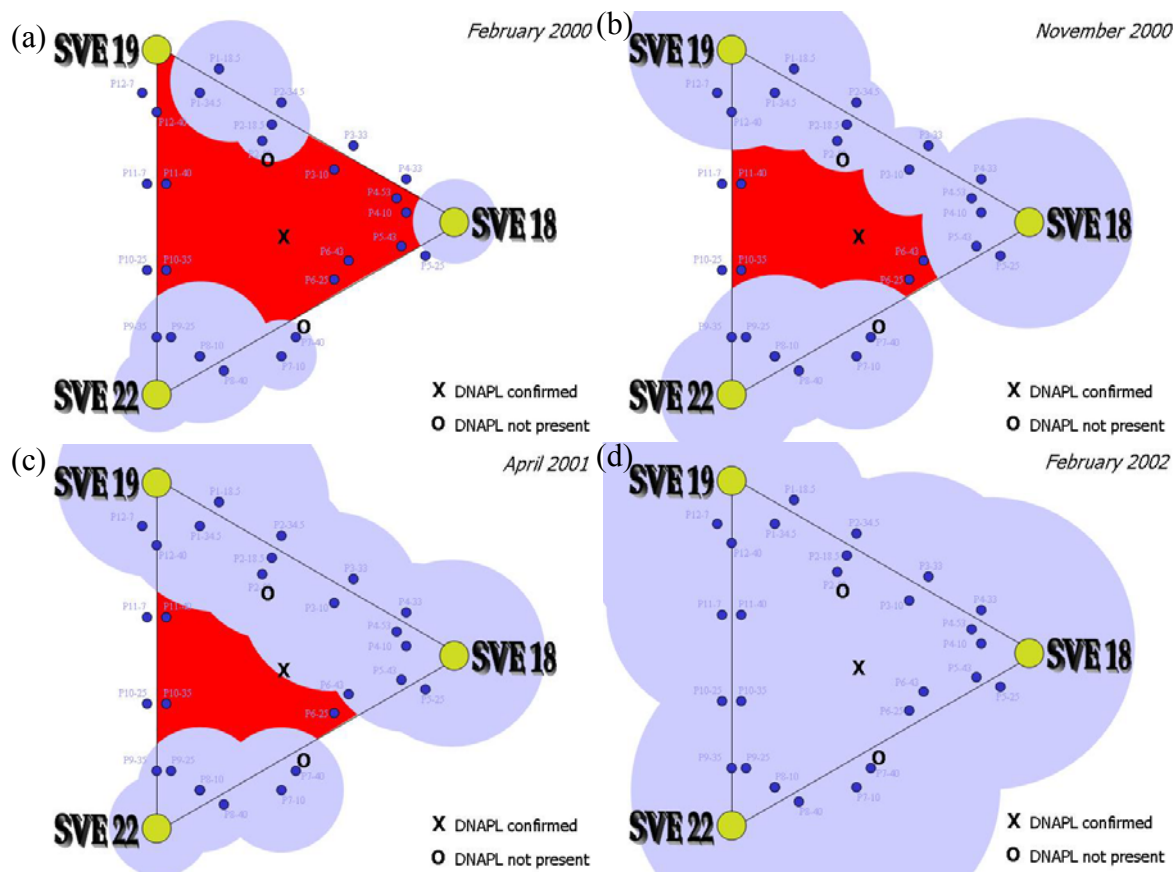


Fig. 4. Clean area at CBRP based on distance-to-source predictions from the diffusion model applied to the (a) February 2000, (b) November 2000, (c) April 2001 and (d) February 2002 rebound tests.

With the evaluation of the data from the fourth rebound test, the diffusion model predictions suggest that no DNAPL source remained at CBRP; however, factors such as subsurface heterogeneity and the formation of preferential pathways by SVE operation may result in this model evaluation being overly optimistic about the cleanup. Low concentration measurements at one location may result in a distance prediction that disguises a high concentration and therefore close source prediction. Care must be taken when evaluating this data.

Establishing a Pathway to Site Closure

The soil gas TCE concentration rebound tests were used as the basis for site closure, with supporting information from the diffusion model as well as mass removal and soil coring data.

The pathway to closure was established as two one-month rebound tests where little or no rebound was observed, followed by a six month rebound study with similar observations [17]. After the six month test, the decision between transition to passive operation versus termination of all operation would be made based on the observed rebound through the system. Based on the observed rebound data, the CBRP SVE system met the preliminary criteria and the six month rebound test was begun in March 2003. Observations at most of the SVE wells and monitoring probes showed little or no soil gas TCE concentration rebound; however, rebound was observed at several monitoring probes and SVE wells, although the concentrations achieved were substantially lower than the initial concentrations. The observation of several high concentrations (600-800 ppmv at several monitoring probes) suggested that some effort should continue at CBRP. Therefore, the decision was made to transition the SVE system to passive operation (i.e., driven by naturally-occurring barometric pressure fluctuations instead of active mechanical pumping) after January 1, 2004 [18].

CONCLUSIONS

Subsurface soil gas concentration rebound tests were used to establish the site closure criteria for a site contaminated with TCE. The declining subsurface soil gas TCE concentration rebound observations coupled with the declining TCE mass removal rate during operation suggest that a substantial portion of the available TCE mass has been recovered from the subsurface. Model predictions support these observations, showing a depleting NAPL source over the course of SVE operation that correlates well with a NAPL source that was discovered during the course of operation. Declining mass removal coupled with the recurrence of elevated soil gas TCE concentrations at a few of the SVE wells and monitoring probes as well as model observations led to the decision to terminate active SVE operation at CBRP and transition that system to passive operation in 2004.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support of the Consortium for Risk Evaluation with Stakeholder Participation through grants DE-FG26-00NT 40938 and DE-FG02-00ER63022.A000 with the U.S. Department of Energy and the U.S. Environmental Protection Agency, as well as the assistance of colleagues Jerry Nelsen (DOE), Mike Morgenstern (SRS/ER), Don Hunter (USEPA), Tim Slagle (USEPA), Mary Harris (SRTC), Greg Flach (SRTC), Joe Rossabi (SRTC), John Bradley (SRS/ER), Keith Hyde (SRTC) and Johnny Simmons (SRS/OP). This paper reflects the opinions of the authors and does not reflect the policies or practices of the DOE or USEPA. The use of trade names is for identification purposes only and does not imply endorsement by either agency.

NOMENCLATURE

t	time (s)
r	radial spatial variable (cm)
z	axial spatial variable (cm)
$R_{v,INTER}$	inter-aggregate retardation factor
C_V^I	vapor phase TCE concentration in region I (g/cm ³)

C_0	initial concentration (g/cm^3)
D_{eff}	effective diffusivity (cm^2/s)
v_r	radial velocity in the subsurface (cm/s)
v_z	axial velocity in the subsurface (cm/s)
k	aggregate regime
ε	aggregate porosity
\tilde{r}_k	aggregate radial (spherical) coordinate (cm)
\tilde{R}_k	radius of aggregate k (cm)
$D_{INTRA,k}$	intra-aggregate diffusivity (cm^2/s)
R_{INTRA}	intra-aggregate retardation factor
$\tilde{C}_{L,k}^{II}$	concentration of TCE in aggregate regime k (g/cm^3)
r_w	SVE well radius (cm)
R_I	distance from SVE well (observation point) to NAPL pool (cm)
R_O	outer boundary of NAPL pool (cm)
b	thickness of region I in the subsurface (cm)
H	Henry's Law coefficient (g/cm^3 [vapor] / g/cm^3 [liquid])

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FOOTNOTES

ⁱ The SVE/AS system was designed and installed by the contractor, Westinghouse Savannah River Company.