Published in final edited form as:

J Contam Hydrol. 2015 ; 0: 54–63. doi:10.1016/j.jconhyd.2015.03.002.

Using Vapor Phase Tomography to Measure the Spatial Distribution of Vapor Concentrations and Flux for Vadose-zone VOC Sources

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Abstract

A test was conducted at a chlorinated-solvent contaminated site in Tucson, AZ, to evaluate the effectiveness of vapor-phase tomography (VPT) for characterizing the distribution of volatile organic contaminants (VOC) in the vadose zone. A soil vapor extraction (SVE) system has been in operation at the site since 2007. Vapor concentration and vacuum pressure were measured at four different depths in each of four monitoring wells surrounding the extraction well. The test provided a 3D characterization of local vapor concentrations under induced-gradient conditions. Permeability data obtained from analysis of borehole logs were combined with the vapor-concentration data to determine VOC mass flux within the test domain. A region of higher mass flux was identified in the deepest interval of the S-SW section of the domain, indicating the possible location of a zone with greater contaminant mass. These results are consistent with the TCE-concentration distribution obtained from sediment coring conducted at the site. In contrast, the results of a standard soil gas survey did not indicate the presence of a zone with greater contaminant mass. These results indicate that the VPT test provided a robust characterization of VOC concentration and flux distribution at the site.

Keywords

SVE; remediation; Tomography; DNAPL; VOC

1 Introduction

It is well established that significant material-property heterogeneity and complex contaminant-source distributions are present at many hazardous waste sites. Furthermore, it

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is well documented that these conditions can greatly limit effective site remediation. The standard approach for characterizing the spatial distribution of volatile organic contaminants (VOC) in the vadose zone is to measure static vapor concentrations (the soil gas survey). However, it is recognized that this method has certain limitations, such as characterizing mass associated with poorly accessible regions (e.g., low-permeability zones) or characterizing temporal variability of concentrations (e.g., DeGroot and Lutenege, 1998; Thomson and Flynn, 2000; McAlary et al., 2009). As a result of these and other issues, the use of the soil-gas survey method for characterizing VOC sources in heterogeneous systems can be influenced by a significant degree of uncertainty. This situation has elicited a need to develop innovative methods that can better characterize such heterogeneous systems (e.g., SERDP, 2006).

Methods based on high-resolution spatial or temporal sampling of VOCs under induced-gradient conditions have been employed recently to improve characterization of vadose-zone sources, as reviewed in Brusseau et al. (2013a). For example, the vapor-phase cyclic or multi-stage contaminant mass discharge (MS-CMD) test was recently developed to measure mass discharge for vadose-zone sources under both induced-gradient and natural-gradient conditions (Brusseau et al., 2010). The multi-stage contaminant mass discharge (CCMD) test has several advantages, one primary one being that the test is based on the use of extended vapor extraction to stress the system and induce pneumatic and concentration gradients. This serves to enhance the sensitivity of the measurements by improving the ability to characterize systems influenced by relatively slow rates of mass transfer. This also reduces the time required for obtaining robust measurements, and thus increases the ability to obtain measurements in real time.

One disadvantage of the CCMD test under standard configuration is the lack of spatially discrete information provided. The location and spatial distribution of contaminant sources can be characterized to an extent by conducting a series of short-term vapor-extraction tests in different sections of the site. For example, the concentration-time signal observed for a specific extraction well will be mediated by the location of that well with respect to the source. Thus, the signals obtained from the tests can be interpreted to locate the source (taking into account initial conditions and other relevant factors). This approach was applied recently by Carroll et al. (2013) at the DOE Hanford site. Local vertical variability of permeability and contaminant concentrations can be characterized by implementing high-resolution measurement of flow and concentrations along the borehole during vapor extraction (e.g., Widdowson et al., 1997; USACE, 2002; EPA, 2003).

The vapor-phase tomography (VPT) test method was recently developed to provide three-dimensional, spatially resolved characterization of VOC concentrations and vapor flux in the vadose zone (Brusseau et al., 2011). This method involves collecting depth-specific concentration and flow-rate (or surrogate) data at multiple monitoring points during a short-term vapor extraction that is conducted at a selected location. This test can then be repeated at multiple locations if desired to potentially further enhance resolution. The premise of collecting vapor samples within a 3-D sampling domain during a vapor-extraction perturbation to characterize VOC sources was recently illustrated for a bench-scale flowcell system (Mainhagu et al., 2014).

The VPT method is analogous to the hydraulic and pneumatic tomographic tests that have been developed to characterize permeability distributions in the subsurface (e.g., Baehr and Hult, 1991; Vesselinov et al, 2001; McDermott et al. 2003; Brauchler et al., 2003; Illman and Tartakovsky, 2005; Illman et al., 2007; Yeh and Zhu, 2007; Ni and Yeh, 2008). However, those methods are based on pressure interference tests, and employ inverse modeling to determine the parameter of interest (K). In contrast, the parameter of interest, VOC concentration, is measured directly with the VPT test. Partitioning tracer tomography has been recently proposed as a means to characterize DNAPL sources (Yeh and Zhu, 2007; Illman et al., 2010). It differs from VPT by requiring the injection of a tracer suite, and is applicable only to sources for which organic liquids are present.

The objective of this study is to illustrate the application of the VPT test as a means by which to obtain high-resolution characterization of VOC distribution in the vadose zone. The study was conducted at a site contaminated by trichloroethene, and at which a soil vapor extraction system has been in operation for several years. The results of the test are compared to those obtained from a soil gas survey, both of which are benchmarked to data obtained from analysis of sediment cores collected from the site.

2 Methods

2.1 Study site

The site is part of the Tucson International Airport Area (TIAA) federal Superfund site in Tucson, Arizona. The TIAA site was placed on the National Priorities List in 1983 in response to the detection of trichloroethene (TCE) in groundwater from several potable water supply wells. A large, multiple-source plume of TCE exists in the upper portion of the regional confined aquifer, which is a source of potable water for the Tucson metropolitan area. Administratively, the TIAA site is separated into three major zones, the North, Central, and South sections. The site for the current study is located within the Central section.

The study site comprises a local saturated zone, referred to as the shallow groundwater zone, which exists as a persistent feature above the regional aquifer. The site consists of a small source area and a groundwater contaminant plume extending approximately 600 m to the west (Figure 1). Trichloroethene is the primary groundwater contaminant, and aqueous concentrations upwards of 100 mg/L and greater have been reported for samples collected from groundwater monitoring wells within the source area. The source area corresponds to a facility that was used in the past to clean aircraft parts and the external surfaces of military aircraft. Dense nonaqueous phase liquid (DNAPL) contamination was determined to be present at the site based on multiple lines of evidence (Brusseau et al., 2013b). The subsurface consists of clay and silty clay units with localized intervals/lenses of silt, sand, and gravel (see Figure 2). The clay is typically highly plastic, and is comprised primarily of muscovite, illite, and montmorillonite (Matthieu et al., 2013). The potentiometric surface for the shallow groundwater zone is approximately 28–29 m below land surface (bls) in the vicinity of the source area.

A multi-element remediation project was initiated at the study site in the fall of 2007. The effort consists of three components: soil vapor extraction (SVE) for the vadose-zone section

of the source area, hydraulic containment for the saturated-zone section of the source area, and pump and treat for the groundwater-contaminant plume. Detailed analyses of the impact of hydraulic source containment and pump and treat on contaminant removal and the groundwater contaminant plume have recently been presented (Brusseau et al., 2013b; Matthieu et al., 2014). Four extraction wells are used for the soil vapor extraction system, which has a total discharge of approximately 20,000 m³/d. The vapor extraction wells are screened at two depth intervals, approximately 9–15 and 18–23 m bls. The source-zone groundwater extraction and soil vapor extraction systems have to date removed approximately 800 and 3000 kg of TCE, respectively.

The SVE system at the TIAA site had been in operation for approximately 6 years prior to the VPT test. Concentrations of TCE in the SVE effluent are low (in the several µg/L range), and appear to have reached asymptotic values. Thus, this site represents a scenario wherein the mass of contaminant remaining in the vadose zone is likely to be relatively small, and conditions are supportive of SVE closure evaluation.

2.2 Vapor Phase Tomography test

2.2.1 Installation and preparation—CRA28 (Figure 1) was used as the vapor extraction well for the VPT test. It was designed as a vadose zone vent well, screened over a 17.5m length, spanning the entirety of the vertical domain influenced by the SVE system. The well head was connected to the SVE extraction system. The four SVE extraction wells were disconnected from the SVE system during the VPT test. Four vapor monitoring wells, CRA47, 48, 49 and 50, were installed in an approximate diamond configuration surrounding the extraction well CRA28 (Figure 1). Each well was designed similarly, in order to allow for discrete monitoring of 4 different depths: 9.9, 14.5, 19 and 25.2 meters below land surface (Figure 2). The annulus space was filled with sand for each screened interval, with bentonite placed in the intervening lengths. A double-packer system was used to isolate each screened interval during sampling.

2.2.2 Vapor-phase tomography test—The SVE system was shut down for a period of 62 days prior to the start of the VPT test. Subsequently, extraction from CRA28 was conducted for 10 days. The mean extraction flow rate was 4558 L/min, with a coefficient of variation of 1%. The pore volume affected by the extraction from CRA28 was estimated to be 1.3×10^4 m³ based on the radius of influence of CRA28 of approximately 48 m. The flow rate hence translates to a residence time of approximately 2 days. The vacuum and vapor concentrations were monitored for CRA28, as well as for all four sampling points of all four monitoring wells, described earlier (CRA47-48-49 and 50) for the duration of the VPT test. The vacuum for each sampling point was measured by an absolute pressure transducer (Omega PX429-015AI) positioned inside the monitoring well, isolated by a double-packer system positioned above and below the screened interval. A second absolute pressure transducer was positioned at the surface to provide simultaneous measures of atmospheric pressure. Preliminary testing showed that the double-packer system performed properly, and that leaks during operation would have a noticeable impact on measured pressures. Additionally, the extraction flow rate for CRA28 was monitored with a turbine flowmeter (ABB, model T3000).

Vapor samples were collected using an air-sampling pump (SP 402 SA-R-V, Schwarzer Precision) that was attached to the sampling point of the well. A two-chamber cartridge containing coconut charcoal sorbent (Orbo, Supelco) was connected to the pump. The cartridges were stored on ice for shipment to the laboratory. Upon receipt in the laboratory, both chambers of each cartridge were extracted with 5 mL of dichloromethane and then sub-sampled with a 2-mL aliquot placed into a 2-mL headspace free auto-sampler vial for analysis by gas chromatography/tandem mass spectrometry (GC/MS/MS). The quantitative detection limit for TCE was approximately 0.2 µg/L-vapor.

2.3 Mass flux calculation

The intrinsic permeability was calculated as a function of depth for each borehole by using an empirical equation (Zhang and Brusseau, 1998) to convert borehole-log texture data to permeability. The equation was developed from prior research conducted at a nearby source area of the TIAA site. A three-dimensional distribution of permeability was then developed by applying inverse-distance interpolation. Mass flux between the monitoring port and extraction well was determined as follows.

$$q_c = C_i \frac{-k(P_b - P_a)_i}{\mu L} \quad (1)$$

where q_c is the contaminant mass flux, in $\text{mg}/(\text{m}^2 \cdot \text{d})$, C is the TCE vapor concentration in mg/m^3 , k the intrinsic permeability in m^2 , μ is the viscosity, $(P_b - P_a)_i$ the difference in pressure between the extraction well and the monitoring port in Pa, and L is the distance between the extraction well and the monitoring port in m.

2.4 Ancillary Characterization Efforts

A soil-gas survey (SGS) was conducted at the study site to provide a comparative data set obtained with a standard method for vadose-zone characterization. The soil-gas survey was conducted using standard methods. The SGS was conducted 2 months prior to the VPT test. The methods used for vapor sample collection and analysis were the same as described above.

High-resolution sampling of sediment cores collected from boreholes drilled within the study site (see Figure 1) was conducted to provide benchmark data for spatial distributions of contaminant concentrations (Brusseau et al., 2013). One set of cores was collected in the spring of 2012, approximately one year prior to the VPT test. The cores were obtained using roto-sonic drilling, a useful method for collecting continuous cores. The drilling was terminated in each borehole at the base of the gravel sub-unit (37–38 m bls) to avoid penetrating the clay aquitard that overlies the regional aquifer.

The cores were sampled with high spatial resolution (every 6 or 10 cm, depending on the depth) by collecting 10-mL subsamples of sediment from the internal central axis of the core with disposable open-tip plastic syringes. Approximately 200 or more samples were collected and analyzed from each core. The subsamples were placed in 40-mL glass vials containing 20 mL of methanol (>99.9% pure), placed in a chilled container, and transported

to the laboratory. The subsamples were placed on a shaker for 24 h, after which they were centrifuged for 10 minutes. A 2-mL aliquot of the eluent was removed from the vial with a glass syringe and injected into a 2-mL autosampler vial with no headspace. The samples were analyzed by GC/MS/MS. After extraction, the sediment samples were dried and weighed. The detection limit for sediment-phase concentrations of TCE was approximately 0.03 mg/kg-dry sediment.

3. Result and Discussion

3.1 Sediment Coring and Soil Gas Survey

A 3D representation of the TCE sediment-phase concentrations for the study area is presented in Figure 3. The data reveal a non-uniform spatial distribution of concentrations, with the highest concentrations occurring at the deepest interval south of CRA28. These data indicate the presence of a persistent source of VOCs in this region, most likely associated with contaminant residing in the clay layer as well as possible off-gassing from groundwater.

The results obtained from the SGS test are presented in Table 2. The TCE concentrations are low, in the range of 3 ug/L. They are similar for all monitoring points irrespective of areal and vertical location. These results indicate a uniform distribution of TCE within the sampled domain, both areally and vertically. This is not consistent with the results reported above for the sediment data.

3.2 Vapor-phase Tomography Test

TCE concentrations in the vapor effluent of the extraction well (CRA28) decreased within approximately two days of startup to a value of ~4 µg/L, with the elution curves exhibiting asymptotic behavior thereafter (data not shown). This behavior is consistent with the SVE operations data, wherein TCE concentrations in the effluent are displaying asymptotic conditions at approximately 2 µg/L. These observations are consistent with site conditions, wherein six years of SVE operations have most likely removed the majority of contaminant mass initially present (as supported by the sediment data).

The TCE-concentration, pressure, and mass-flux data obtained for the monitoring locations are presented in Figures 4, 5, and 6, respectively. No measurable increases in concentration or flux above background levels were observed for the two shallowest sampling depths (9.9 and 14.5 m below ground surface) for any of the four wells. Concentrations average approximately 3.5 ug/L, consistent with the values measured for CRA28. The low concentrations, combined with the absence of responses above background, indicate that there are no nearby sources (Mainhagu et al., 2014), which is consistent with the sediment data.

The TCE concentrations for the 19 m bgs sampling depth exhibit nonuniform behavior amongst the four monitoring wells. Concentrations for sampling ports of both CRA49 and 50, located in the northeast quadrant (or N-E) from CRA28, exhibited no change from the low background of ~3.5 ug/L, similar to the behavior observed for the shallower two depths. Conversely, initial concentrations collected for CRA47 and CRA48 are higher than

background. However, the remaining data matched background. The concentrations obtained for the deepest (25.2 m bgs) sampling interval, closest to the water table, also exhibit non-uniform behavior amongst the four monitoring wells, but to a much greater extent than for the 19-m interval. TCE concentrations for CRA49 and 50 did not increase from the background of ~ 3.6 $\mu\text{g/L}$. The concentration for CRA47 exhibited an initial increase similar to that observed for 19 m-bgs, but to a greater extent. The concentration for CRA48 exhibited a significant, sustained increase above background.

The spatial distribution of TCE vapor concentrations (averaged over the first 3 days) is presented in Figure 5. The extraction well is located at the (0,0, -8.5 to -26) coordinates. The highest concentration values are associated with the deepest interval of the S-SW section of the domain, indicating the nearby presence of a region with a greater mass of contaminant.

Absolute pressure was measured at the sampling depth and at the surface for each TCE sampling point, providing the relative vacuum at the sampling depth. Figure 6 presents the relative vacuum, in inches of mercury ("Hg), for all sampling points. The average vacuum was -0.71, -0.90, -0.77 and -0.62 "Hg for the depths of 25.2, 19, 14.5, and 9.9 m-bgs, respectively. The spatial distribution of permeability, based on conversion of borehole log data, is presented in Figure 7. The TCE concentration, vacuum, and permeability data were used to calculate contaminant mass flux.

The TCE mass fluxes between the monitoring points and the extraction well over time are presented in Figure 8. The two shallowest depths of 9.9 and 14.5 m-bgs presented a constant mass flux of 1.8×10^{-8} $\mu\text{g/m}^2 \cdot \text{s}$ and 6.3×10^{-9} $\mu\text{g/m}^2 \cdot \text{s}$, respectively. The mass fluxes for monitoring ports of CRA47, 49 and 50, at the depth of 19 m-bgs, were constant at 3.2×10^{-8} $\mu\text{g/m}^2 \cdot \text{s}$. The mean mass flux for monitoring well CRA48 at that same depth is 1.0×10^{-6} $\mu\text{g/m}^2 \cdot \text{s}$ over the first 4 days. This value is significantly higher than those determined for the other ports at the same depth. At the 25.2 m-bgs, the mass fluxes from CRA50 and 49 remained low and constant at 9.0×10^{-8} $\mu\text{g/m}^2 \cdot \text{s}$ and 6.7×10^{-8} $\mu\text{g/m}^2 \cdot \text{s}$, respectively. The mass flux between the CRA47 monitoring port and the extraction well presented a higher initial value of 1.6×10^{-6} $\mu\text{g/m}^2 \cdot \text{s}$, and stabilized for the remainder of the extraction at 5.4×10^{-7} $\mu\text{g/m}^2 \cdot \text{s}$. Finally, mass flux between the CRA48 monitoring port and the extraction well was initially high at 1.5×10^{-6} $\mu\text{g/m}^2 \cdot \text{s}$ during the first 5 days, and decreased to 8.6×10^{-7} $\mu\text{g/m}^2 \cdot \text{s}$ during the last 5 days of the extraction test.

The spatial distribution of contaminant mass flux is presented in Figure 9. The location of the region with greatest mass flux is consistent with the results observed for TCE vapor concentrations (Figure 5). However, the specific distribution varies, particularly in the vertical dimension, due to two factors. One reflects the impact of permeability (vapor flow) on flux. The second factor relates to the use of data with different scales of resolution. Specifically, the permeability data has higher resolution in the vertical dimension compared to the concentration data.

3.3 Comparison of Characterization Methods

The performance of the VPT test, in terms of characterizing the spatial distribution of VOC concentrations and fluxes in the vadose zone and identifying the potential presence of regions with greater masses of contaminant, can be evaluated by comparing the results to the sediment data. The VPT test identified the presence of a zone of higher flux in the deepest interval of the southern section of the domain. This is consistent with the results of the sediment characterization, which shows the presence of a higher-concentration zone below ~26 m bgs in the southern section of the domain. Additionally, the results of the VPT test indicated uniform flux and the absence of a source in the interval above 25 m bgs. This is also consistent with the sediment data. These results indicate that the VPT test provided a robust characterization of the test zone.

A soil gas survey of the test zone was conducted prior to the start of the VPT test to serve as a comparative baseline. As discussed, the results of the SGS indicated a uniform distribution of VOCs in the vadose zone, with no identified source. This result contrasts with those obtained with the VPT test and the sediment characterization. The poor performance of the SGS may be related to the fact that the vast majority of the remaining TCE mass resides in the thick clay layer that encompasses the groundwater/vadose-zone interface, for which mass transfer is likely significantly constrained.

4. Conclusion

Assessing the potential for SVE system closure or transition to other remedies for a contaminated site can be supported by characterization of the nature and location of the remaining contamination. The vapor-phase tomography test was developed to offer a more effective alternative to characterize VOC concentration distributions in the vadose zone. A VPT test was conducted at a TCE-contaminated site in Tucson, AZ. A single extraction well (CRA28) was used along with four monitoring wells (CRA47, 48, 49 and 50) with four depth sampling intervals (9.9-14.5-19-25.2 m-bgs). Spatial differences in mass flux were observed, indicating the presence of a vapor source located in the S-SW quadrant of the test area, between the water table and the monitoring port screened interval. These results are consistent with those of the sediment data. The VPT test provided a more robust source characterization compared to the standard SGS method, which was unable to identify spatial variability of vapor concentrations or a potential source. These results illustrate the utility of the VPT test as a method for characterizing VOC from a source in the vadose zone.

The manner by which the permeability field was determined provided a relatively high vertical resolution. It is likely that application of pneumatic tomography employing the vacuum data could produce an even greater resolution of the 3D permeability field. In contrast, it is noted that the inherent vertical and areal resolution of the concentration data is significantly lower, given the limited number of sampling points employed. However, the effective degree of resolution may be comparable to that of permeability considering the small concentration gradients present at this site. The lower inherent resolution for concentration data may be more limiting at sites for which concentration gradients are greater. Inherent resolution can be enhanced by the use of multilevel sampling devices to increase vertical resolution, along with more sampling locations to increase areal resolution.

The use of additional sampling points of course increases costs of the test. A potential alternative approach would be to combine pneumatic tomography with VPT, with the combined tool providing an integrated characterization of permeability, concentration distribution, and vapor flux, and additionally delineation of migration pathways.

Acknowledgments

This research was supported by the U.S. Department of Defense Environmental Security Technology Certification Program (ER-201125), and the National Institute of Environmental Health Sciences Superfund Research Program (ES04940). The authors thank Fred Brinker of the Tucson Airport Authority for his support of the project and Manfred Plaschke and Peter Schwarz of CRA, Inc, for their generous assistance in implementing the test conducted at the TIAA site. We also thank Daniel Ojinaga and Celia Martinez, from CRA, Inc, and Asma El Ouni and Hazel Cox, from the UA Contaminant Transport Lab, for their assistance in conducting the test and Nohely Reyes for her assistance in the permeability calculation.

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Highlights

The VPT test provides 3D characterization of VOC concentrations and vapor flux in the vadose zone

A pilot test was conducted at the TIAA Superfund site

The results are compared to sediment and soil-gas-survey data

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Figure 1. Map of the TIAA Three Hangars TI-zone field test area, in Tucson, AZ. Insert representing the TIAA site, with TCE plume.

Figure 2. Schematic cross-section within the TIAA Three Hangars TI zone. Distances are in meters below ground surface (bgs); not to scale.

Figure 3. TCE concentration distribution associated with the sediment collected from boreholes; based on a linear interpolation of the measured data.

Figure 4.
Vapor-phase concentrations of TCE for four different depths of each monitoring well.

Figure 5.
TCE concentration distribution averaged over the first 3 days.

Figure 6.
Relative vacuum pressure for four different depths of each monitoring well.

Figure 7.
Permeability distribution.

Figure 8.
Contaminant mass flux over time for four different depths of each monitoring well.

Figure 9. Contaminant mass flux distribution averaged over the first 3 days, calculated using the data in Figures 7 and 8.

Table 1

Soil gas survey results; TCE concentrations in ug/L

Depth Interval (center, m bgs)	# Non Detects	# Detects	Mean TCE Concentration
9.9	7	2	3.3
14.5	6	4	3.8
19	9	1	3.3
25.2	8	1	3.4
27	1	3	3.4