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Changes in contaminant mass discharge from DNAPL source mass depletion: Evaluation at two field sites

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ABSTRACT

Changes in contaminant fluxes resulting from aggressive remediation of dense nonaqueous phase liquid (DNAPL) source zone were investigated at two sites, one at Hill Air Force Base (AFB), Utah, and the other at Ft. Lewis Military Reservation, Washington. Passive Flux Meters (PFM) and a variation of the Integral Pumping Test (IPT) were used to measure fluxes in ten wells installed along a transect down-gradient of the trichloroethylene (TCE) source zone, and perpendicular to the mean groundwater flow direction. At both sites, groundwater and contaminant fluxes were measured before and after the source-zone treatment. The measured contaminant fluxes (J ; $\text{ML}^{-2}\text{T}^{-1}$) were integrated across the well transect to estimate contaminant mass discharge (M_D ; MT^{-1}) from the source zone. Estimated M_D before source treatment, based on both PFM and IPT methods, were ~ 76 g/day for TCE at the Hill AFB site; and ~ 640 g/day for TCE, and ~ 206 g/day for *cis*-dichloroethylene (DCE) at the Ft. Lewis site. TCE flux measurements made 1 year after source treatment at the Hill AFB site decreased to ~ 5 g/day. On the other hand, increased fluxes of DCE, a degradation byproduct of TCE, in tests subsequent to remediation at the Hill AFB site suggest enhanced microbial degradation after surfactant flooding. At the Ft. Lewis site, TCE mass discharge rates subsequent to remediation decreased to ~ 3 g/day for TCE and ~ 3 g/day for DCE ~ 1.8 years after remediation. At both field sites, PFM and IPT approaches provided comparable results for contaminant mass discharge rates, and show significant reductions ($>90\%$) in TCE mass discharge as a result of DNAPL mass depletion from the source zone.

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1. Introduction

Multiple modeling approaches have recently been used to evaluate whether significant reduction in contaminant mass discharge (M_D ; MT^{-1}) will result from depletion of dense nonaqueous phase liquid (DNAPL) mass from source zones (Sale and McWhorter, 2001; Enfield et al., 2002; Rao et al., 2002; Rao and Jawitz, 2003; Lemke et al., 2004; Parker and

Park, 2004; Soga et al., 2004; Zhu and Sykes, 2004; Enfield et al., 2005; Jawitz et al., 2005; Wood et al., 2005; Fure et al., 2006). Changes within the dissolved plume, resulting from decreased M_D as a result of source-zone treatment, have also been examined in recent modeling analyses (e.g., Falta et al., 2005a,b). Results from these models suggest that site-specific hydrogeological conditions and spatial distribution of DNAPL within the source zone control the relationship between source mass depletion and expected reduction in M_D . Results from laboratory studies (Fure et al., 2006; Suchomel and Pennell, 2006; Totten et al., 2007) and limited field measurements in hydraulically isolated test cells (Brooks et al., 2004; Childs et al.,

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2006) suggest that reductions in contaminant mass discharge occur after removal of DNAPL source mass. Analyses of data from several field studies also provide support for the prediction of M_D reductions due to source treatment, and suggest that a linear relationship might serve as a first-order approximation (McGuire et al., 2006; Falta et al., 2005a; Brusseau et al., 2007).

Here, we present measurements of groundwater and contaminant fluxes at two DNAPL sites, one located at Hill Air Force Base (AFB), near Layton, Utah, and the other at Ft. Lewis Military Reservation, located near Tacoma, Washington. Fluxes were measured before and after aggressive DNAPL source treatment (in-situ surfactant flushing at the Hill AFB site; resistive heating at the Ft. Lewis site) for depletion of source mass. Details of the source-zone treatment at the Hill AFB site were previously reported by URS and INTERA (2003), while details of the thermal treatment at the Ft. Lewis site are presented by Beyke and Fleming (2005), TRS (2005), and Powell et al. (2007). Our focus here is on performance assessment of the DNAPL source treatment, based on M_D

estimated at a control plane just down-gradient of the source zone. Specifically, we compare M_D measurements collected at the source control plane before and after remediation to investigate changes in contaminant mass discharge resulting from DNAPL source remediation. Multiple methods were used to estimate M_D to minimize the uncertainty of any given single measurement, and while a comparison of results between methods is made, the comparison is secondary to the primary purpose of remedial performance assessment. At both sites, estimates of M_D were based on spatial integration of the contaminant fluxes measured in ten wells along a transect perpendicular to the mean groundwater flow direction; the wells were screened over the saturated zone of primary interest. Before and after source treatment, groundwater fluxes (q , LT^{-1}), and contaminant fluxes (J , $ML^{-2}T^{-1}$) were measured using two techniques: the Passive Flux Meter™ (PFM) approach (Hatfield et al., 2004; Annable et al., 2005; Basu et al., 2006), and a modified version of the Integral Pumping Test (IPT) approach, the origins for which lie in the IPT method as previously described (Bockelmann et al., 2001; Bockelmann

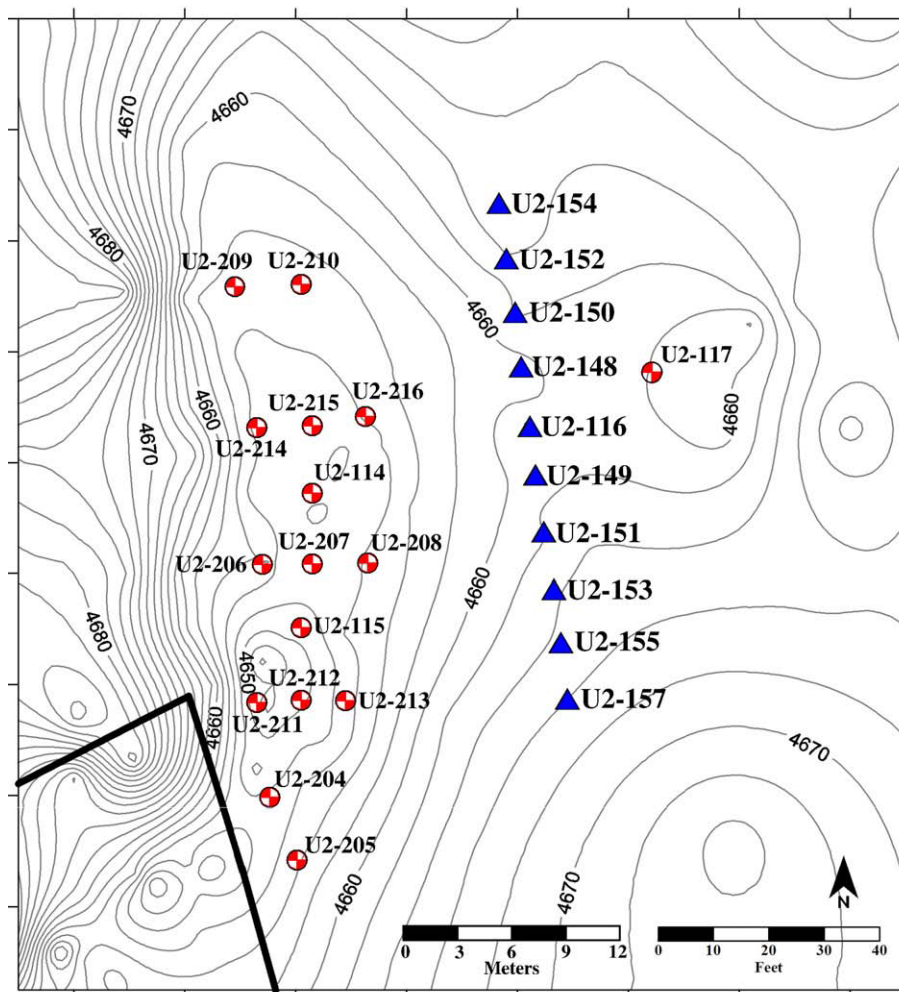


Fig. 1. Plan view of the Panel 5 area at Hill AFB. The thick black line in the lower left corner represents the containment wall installed around OU2. The triangular symbols represent wells used for mass flux measurements. The grey contour lines represent the surface of the clay unit (contours in feet) underlying the surficial aquifer.

et al., 2003; Bauer et al., 2004; Bayer-Raich et al., 2004, 2006). The PFM method enables the simultaneous characterization of the depth variations in q and J along the well screen. The IPT provides estimates of depth-averaged contaminant fluxes (\bar{J}) over the well-screen interval; the modification we introduced here to the IPT approach allows for an estimation of depth-averaged (or larger scale) groundwater flux (\bar{q}) values. Mass discharge values estimated using PFM and IPT were also compared with those estimated using a more traditional approach, the Transect Method, as described by API (2003).

2. Field sites

2.1. Hill AFB site

From 1967 to 1975, spent degreasing solvents, primarily trichloroethylene (TCE), were disposed into two unlined disposal pits dug into the sandy surface soil located along the northeast boundary of Hill AFB, near Layton, Utah. As a result, the DNAPL contaminated the underlying alluvium (the Provo Formation), predominantly as a mobile phase which pooled in topographic depressions on top of a thick clay aquiclude (the Alpine Formation), but also as an immobile residual phase above the aquiclude. A dissolved-phase contaminant plume (predominantly TCE) extends from the source area to the north north-east for a distance of ~1000 m (~3000 ft). A source recovery system (SRS) has been operating since 1993, recovering over 150 kL (37,000 gal) of DNAPL, and treating over 38 ML (9.5 million gallons) of contaminated groundwater as of May 1998 (URS Corporation and Duke Engineering Services, 2001). In 1996, a containment wall (Fig. 1) was constructed around the known source zone, and surfactant-enhanced aquifer remediation (SEAR) was conducted within the contained zone. However, during additional site characterization efforts conducted in 1997, DNAPL was discovered outside of the containment wall in a depression in the clay surface (see Fig. 1). This area, referred to as the Panel 5 area is ~430 m² in size, and is the focus of the present study.

The site overlooks the Weber Valley and is located on a terrace in an east-facing slope of an old floodplain formed by the Weber River as it carved successively younger flood plains into the Alpine Formation, a thick sequence of fine-grained deltaic sediments that the river had originally deposited. The shallow, unconfined, paleo-channel aquifer at the site consists of the heterogeneous alluvium of the Provo Formation (composed of silt, sand, and gravel) that was deposited on the eroded surface of the underlying Alpine Formation (composed of a thick layer of clay and silt). The hydrogeology and DNAPL distribution within the containment wall have been described by Meinardus et al. (2002) and Holbert et al. (2004). As the clay contours in Fig. 1 indicate, the Panel 5 DNAPL source area is located in a shallow unconfined paleo-channel aquifer that generally lies in a south to north orientation. At the northern end, the paleo-channel has an eastern outlet (oriented along a line defined by wells U2-216 and U2-117), and groundwater flow along this outlet is bounded to the north and south by the saddle-shaped clay surface (see Figs. 1 and 2).

Flux measurements were made in 10 wells along a transect down-gradient of Panel 5 before and after SEAR source treatment. Phase I groundwater and contaminant fluxes were measured prior to the surfactant flood, first using the PFM approach (May 1–8, 2002) and then the IPT approach (May 9–13, 2002). SEAR was performed using the wells to the northeast of the containment wall (see Fig. 1) during June and July 2002 (URS and INTERA, 2003), and post-SEAR (Phase II) PFM flux measurements were conducted between June 12 and 23, 2003; and the Phase II IPT flux measurements were conducted between June 24 and 27, 2003.

2.2. Ft. Lewis site

The East Gate Disposal Yard (EGDY) site is located on the Ft. Lewis Military Reservation near Tacoma, Washington, and is part of the Ft. Lewis Logistics Center Superfund site. The EGDY was used from 1946 to 1960 as a disposal area for drums of used solvents and oils that were placed in excavated

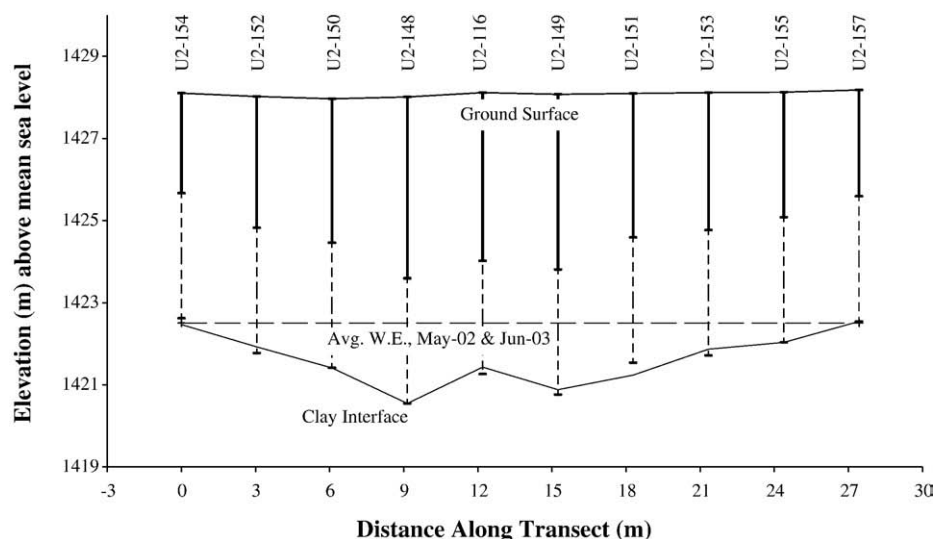


Fig. 2. Well transect cross section at Hill AFB, illustrating the well construction details and clay interface relative to the average water table elevation (Avg. W.E.) during the Phase I (pre-remediation) and Phase II (post-remediation) tests.

trenches, and is the source for a large chlorinated solvent plume (predominantly TCE) which extends to the northwest for ~4 km (~2.5 miles) towards the American Lake (USACE, 2002). The operation of a pump-and-treat system was started in 1995 for hydraulic control purposes, and drum excavation activities at the EGDY site were conducted between late 2000 and mid-2001. Site characterization work conducted in 2001 and 2002 identified three main areas of DNAPL contamination within the EGDY site, which are referred to as NAPL Areas 1, 2, and 3 (NA1, NA2, and NA3, respectively). The focus of our flux measurements was on the performance assessment associated with source treatment activities conducted in NA1, which is ~2400 m² in area.

At the EGDY site, the surficial, unconfined aquifer is composed of the Vashon Recessional Outwash/Steilacoom gravel unit (consisting of loose, well-graded sandy, cobbly gravel or gravelly sand). In the immediate vicinity of NA1, this unit is underlain by Vashon Till (consisting of loose to dense silty, sandy gravel with some clay), which is considered to be a generally continuous intermediate aquitard. This layer, in turn, is underlain by more Vashon Recessional Outwash/Steilacoom gravel or Vashon Advance Outwash (loose sandy gravel to gravelly sand with cobbles) (USACE, 2002).

Prior to thermal treatment of the NA1 source area, contaminant fluxes were measured using both PFMs and the modified IPT in ten wells along a transect down-gradient of NA1 (Fig. 3). For pre-remediation tests (Phase I), PFMs were deployed for a period of three days: October 22–25, 2003, and the IPT was conducted during November 3–7, 2003. In-situ resistive heating thermal treatment was conducted by Thermal Remediation Services (TRS) Inc. in NA1 from December 2003 until August 2004, and consisted of heating the subsurface to target temperatures of 90 °C in the unsaturated zone and 100 °C in the saturated zone to 11.6 m (38 ft) below ground surface, and recovery of contaminant

mass through a multi-phase extraction system (Beyke and Fleming, 2005; TRS, 2005; Powell et al., 2007). Flux changes associated with the NA1 source treatment (Phase II) were evaluated with PFM deployments during June 2–5, 2006 and with the IPT during June 12–17, 2006.

3. Flux measurements

3.1. Well installation and contaminants of concern

At the Hill AFB site, ten wells used in the flux studies were installed during April 9–16, 2002 using hollow-stem augers. The formation was cored to locate the clay interface and the wells were screened from above the seasonal high water table to the clay. Each well was constructed using 5.08 cm (2-in.) diameter PVC well screens, 3.05 m (10 ft) in length, which were installed with a surrounding sand filter pack. The wells were spaced ~3 m (~10 ft) apart. A cross-sectional view of the well transect is shown in Fig. 2.

During August 23–25, 2003, a roto sonic drilling method was used to install seven wells (LC-201 through LC-207; Figs. 3 and 4) down-gradient of the NA1 source zone at the Ft. Lewis site. These wells were installed with a nominal spacing of 6.1 m (20 ft), and were screened from above the water table to the top of the till unit at a depth of approximately 10.1 m (33 ft). The wells were installed in a 15.2-cm (6-in.) diameter borehole, and were constructed using 5.08-cm (2-in.) diameter stainless steel casings, 2.44 m (8 ft) in length, and 5.08-cm (2-in.) diameter stainless steel well screens, 7.62 m (25 ft) in length. A 12/20 sand filter pack was installed in the annular space around the well screens, on top of which was placed a bentonite seal, and the remaining space to land surface was filled with grout. Initial samples collected from these wells indicated a generally increasing trend in concentration from LC-201 toward LC-207 (data not shown), consequently the well transect was extended

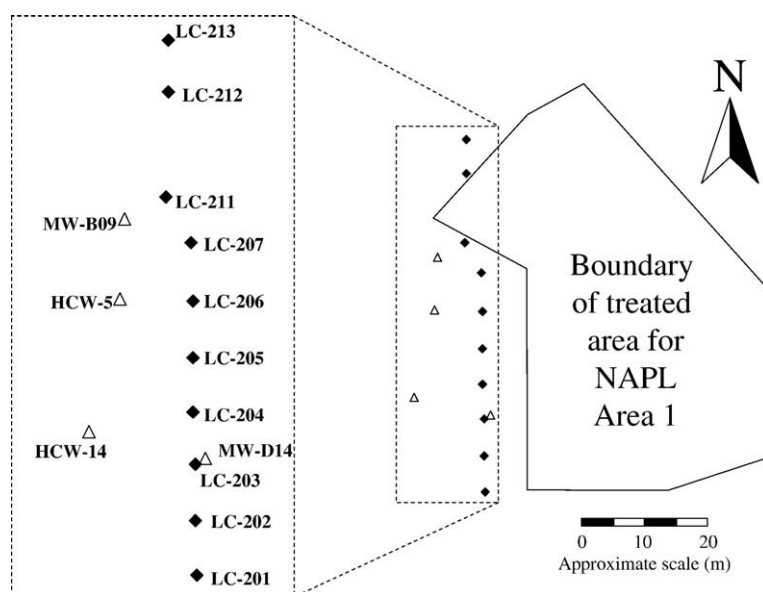


Fig. 3. Plan view of the NA1 source area at the East Gate Disposal Yard site at Fort Lewis, and the down-gradient flux well transect. The diamonds represent flux wells and the triangles represent hydraulic monitoring points.

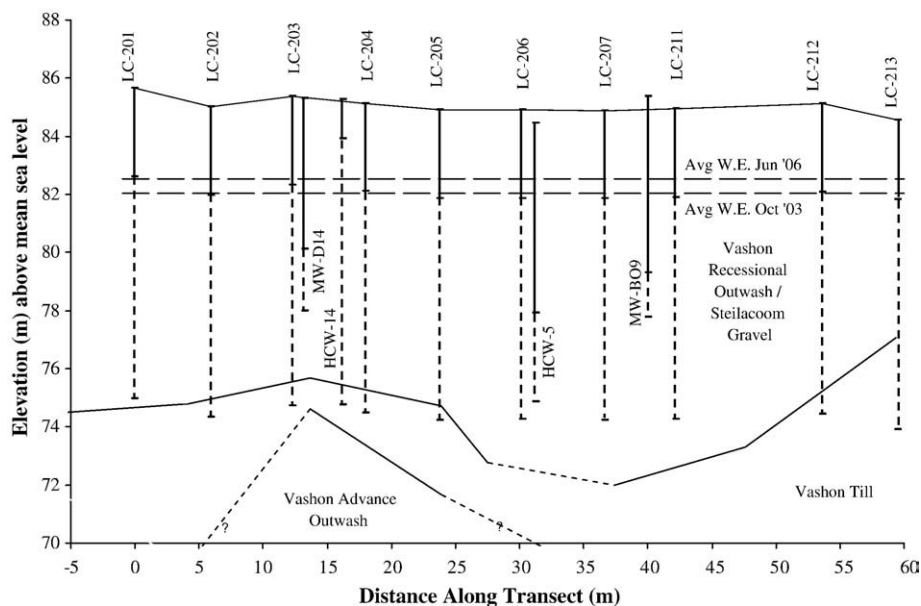


Fig. 4. Well transect cross section at NA1, Fort Lewis, illustrating the well construction details and Vashon Till interface relative to the average water table elevation (Avg. W.E.) during the pre-remedial (Oct-03) and post-remedial (Jun-06) tests.

to the north by three additional wells (LC-211, LC-212, and LC-213). These wells were installed during October 16–17, 2003 using an air rotary drilling method; each well was constructed using materials similar to those described for wells LC-201 through LC-207, with the exception that the filter pack surrounding the well screens consisted of 10/20 silica sand. A cross-sectional view of the well transect is shown in Fig. 4.

The transect is located close to the NA1 boundary, and in fact crosses a portion of the designated source zone between LC-211 and LC-212 (Fig. 3). This was necessary, however, to avoid the NA2 source area located immediately to the west of NA1. For the region where the control plane is within the source, the measured source strength may (depending on the distribution of DNAPL up-gradient of the control plane) be underestimated due to the fraction of the source down-gradient of the control plane. DNAPL immediately up-gradient of a well in this region would not affect the flux measurements any more than distant, up-gradient DNAPL would. DNAPL immediately adjacent in the lateral direction may result in mass discharge estimates larger than the true mass discharge. During the installation of well LC-211, NAPL was noted in the drill cuttings from 3.4 to 4.9 m below grade, but was not observed in the cuttings from any of the other nine flux wells.

Preliminary contaminant characterization (Meinardus et al., 2002) and initial IPT sample analysis (see Supplementary Material) at the Hill AFB site indicated that the constituent of primary concern for the Panel 5 site was TCE. For the Phase II test, *cis*-dichloroethylene (DCE) was included as a contaminant of interest because of suspected degradation resulting from the surfactant flood remedial activities. At the Fort Lewis site, the multi-component NAPL in the source zone contained other gasoline-related contaminants; but, our focus here is limited to an assessment of the changes in mass discharge rates for the chlorinated solvent components (predominantly TCE and DCE).

3.2. Passive Flux Meter (PFM) approach

General experimental methods for deployment and recovery of PFMs are described by Annable et al. (2005) and Basu et al. (2006). Briefly, the approach to the measurement of groundwater and contaminant fluxes involves deployment of a permeable, sorbent pack (i.e., PFM) in a well transect, where the wells are screened across the aquifer depth of interest. The PFM sorbent (silver-impregnated granular activated carbon [SI-GAC]) is pre-saturated with resident tracers that are desorbed from the sorbent and depleted with the groundwater flow through the PFM during the deployment period; groundwater fluxes are calculated from the tracer mass depletion. The PFM sorbent material is selected to capture the target contaminants dissolved in the groundwater flowing through the device, under natural gradient groundwater flow conditions, during the designated deployment period of exposure. The sorbed contaminant mass is used to directly estimate contaminant fluxes. Thus, flux measurements using this approach are referred to as “passive” in contrast to the IPT method (see below) that requires pumping.

The PFMs were packed on site. PFMs were constructed to match the saturated thickness in each well, and multiple PFMs (1.5 m long) were deployed as needed in wells to cover well-screen intervals larger than 1.5 m. Each PFM sock was divided into ~25-cm long segments separated using Norprene rubber washers [5.08 cm (2 in.) outside diameter with 0.635 cm (1/4 in.) holes] to prevent vertical water flow in the PFM and section the device upon retrieval. A sample of SI-GAC was collected for analysis of the initial tracer concentrations, and analytical details are provided in Annable et al. (2005). The completed PFM was then inserted into the well to the desired screen interval.

Estimations of groundwater flux based on depletion of alcohol tracers, and contaminant fluxes based on contaminant

accumulation, were completed as described by Hatfield et al. (2004) and Annable et al. (2005). Retardation factors for tracer depletion from the PFM sorbent were as reported by Annable et al. (2005). The depth profiles of contaminant fluxes from each well were averaged to determine \bar{J} for each well. The averages were calculated weighting the local contaminant flux values (J_i) by the length of the vertical interval (b_i) as follows:

$$\bar{J} = \frac{\sum_{i=1}^{n\text{-vert}} J_i b_i}{\sum_{i=1}^{n\text{-vert}} b_i} \quad (1)$$

where $n\text{-vert}$ is the number of intervals in the well screen, b_i is the length of each interval [L], and J_i is the local contaminant flux [$\text{ML}^{-2}\text{T}^{-1}$]. The J values were integrated over the width of the control plane to determine a transect-wide contaminant mass discharge, \bar{M}_D (g/day):

$$\bar{M}_D = \sum_{j=1}^{n\text{-well}} \left(\sum_{i=1}^{n\text{-vert}} (J_{ij} A_{ij}) \right) \quad (2)$$

where $n\text{-well}$ is the number of wells and A_{ij} is the area [L^2] represented by the vertical sampling interval (b_i times the horizontal spacing between wells). This calculation is predicated on the assumption that contaminant fluxes measured in the wells are representative of the entire well spacing; it is recognized that better estimates may be obtained by various interpolation techniques.

3.3. Integral Pumping Test (IPT) approach

The IPT technique to measure \bar{J} and \bar{M}_D is primarily based on the measurement of the contaminant concentration-time series, $C(t)$, in the effluent of multiple pumping wells aligned perpendicular to the prevailing direction of groundwater flow. While the use of $C(t)$ data for groundwater investigations was introduced in the early 1980s (Keely, 1982; Keely and Wolf, 1983), estimation of J and M_D using the IPT method was first described by Schwarz et al. (1998), and IPT field applications were described by Bockelmann et al. (2001, 2003) and Bauer et al. (2004). Under steady-state conditions, the mass discharge entering the capture zone of the pumping wells is equal to the product of the pumping rate and pumping well effluent concentration (Holder et al., 1998; Einarson and Mackay, 2001). However, achieving this steady-state condition is not a practical goal for short-term pumping tests. Alternatively, the transient concentration-time series from a pumping well can be used to estimate the average spatial concentration in the capture zone of the well (e.g., Bayer-Raich et al., 2004, 2006), which is combined with an estimate of the groundwater flux to give the contaminant flux.

For the IPTs conducted at Hill AFB and Fort Lewis, all wells were pumped concurrently rather than sequentially as conducted in previous deployments of the IPT method. Wells were pumped concurrently in our tests to minimize the disturbance to the contaminant distribution that would result from pumping each well sequentially, and to avoid the uncertainty associated with double counting mass located between adjacent wells. However, disadvantages to concurrent pumping are: (1) development of stagnation points

between wells, and (2) a more complex flow field, which precludes the use of the Bayer-Raich et al. (2004) analytical solution. With respect to the first disadvantage, it is nonetheless possible to sample a large fraction of the space between wells, so that the general goal of the IPT (i.e., a measured response integrated over a large area) is maintained. With respect to the second disadvantage, the average spatial concentration in the capture zone of the well was estimated using the average concentration of the concentration-time series. The mass discharge was then obtained from the product of the contaminant flux and the cross-sectional flow area associated with the wells in the transect.

The IPT method was further modified to allow the estimation of groundwater fluxes based on the hydraulic information collected during the IPT. The basis for this new approach was the assumption that the aquifer in the vicinity of the well transect could be described as one with homogeneous and isotropic hydraulic conductivity, and a uniform saturated thickness. Consequently, the hydraulic head (ϕ ; L) can be described using superposition of uniform flow and sink terms (Bear, 1979):

$$\phi(x, y) = -\frac{qB}{T}x + \frac{-Q}{4\pi T} \ln \left[(x-x_w)^2 + (y-y_w)^2 \right] + \phi_0, \quad (3)$$

where: B is the saturated thickness [L]; T is the aquifer transmissivity [L^2T^{-1}], equal to the product of hydraulic conductivity K [LT^{-1}] and B ; and Q is the pumping rate [L^3T^{-1}] for a well located at x_w, y_w . Assuming the origin of the coordinate system coincides with a pumping well of interest and that the well transect is aligned perpendicular to the groundwater flow direction, the difference in head between the pumping well and a down-gradient monitoring well is

$$\Delta\phi = -\frac{qB}{T}\Delta x + \frac{-1}{4\pi T} \sum_{i=1}^n Q_i \ln \frac{r_{\text{obs}[i]}^2}{r_{w[i]}^2}, \quad (4)$$

where the summation term accounts for concurrent pumping from n wells in the transect, and $r_{\text{obs}[i]}$ and $r_{w[i]}$ are the distances from the observation well to the i th pumping well and from the pumping well of interest to the other pumping wells, respectively. Note that $r_{w[i]}$ for the pumping well of interest itself refers to the well radius. Eq. (4) is linear with a slope of $(4\pi T)^{-1}$ and an intercept of $-qB\Delta x T^{-1}$. By measuring the difference in hydraulic head under a series of pumping rates, the hydraulic conductivity and the groundwater flux can be estimated using linear regression techniques and an *a priori* estimate of the saturated thickness. Eq. (4) is based on the assumption that the ambient flow direction is perpendicular to the well transect. If this condition is not met, Eq. (3) can be modified to account for non-perpendicular flow, resulting in:

$$\Delta\phi = -\frac{qB}{T}(\Delta x \cos \theta + \Delta y \sin \theta) + \frac{-1}{4\pi T} \sum_{i=1}^n Q_i \ln \frac{r_{\text{obs}[i]}^2}{r_{w[i]}^2}, \quad (5)$$

where θ is the angle between the flow direction and the positive x -axis (oriented perpendicular to the transect), determined by *a priori* head measurements.

Each of the IPTs was conducted immediately following the retrieval of the PFMs from the wells, thus allowing for a comparison of measured fluxes with minimum time lag. During the IPT, flow rates and water levels were measured

periodically at the wells and the combined flow was recorded as it accumulated in a temporary storage tank. Water levels were also monitored in nearby observation wells. The pumping rates were selected to bracket the calculated specific discharge rate across the control plane area, while still yielding a sufficiently large volume of pumped water (and hence sampling space surrounding the well) over the duration of the test. At the Hill AFB site, pumping rates ranged from 0.06 to 0.6 m³d⁻¹, and at the Fort Lewis site, pumping rates ranged from 2 to 40 m³d⁻¹ (see Supplementary Material for more details on the pumping rates). Moreover, at the Fort Lewis site, the transect was divided into three segments to minimize the spatial extent over which the conceptual model was applied, and a single groundwater flux was estimated for each segment by averaging results for the observation wells and the five nearest pumping wells (see Supplementary Material).

Groundwater samples from the effluent of each pumping well were collected for contaminant analysis approximately once every 3 to 6 h throughout each IPT. Once collected, the samples were immediately placed in coolers with frozen ice packs, and shipped overnight to the laboratory for refrigerated storage and analysis. Samples collected from the Hill AFB site were analyzed at the University of Florida on a Perkin-Elmer Autosystems gas chromatograph (GC) with flame ionization detector (FID) and high performance liquid chromatography with a reverse-phase column (C-18, Supelco), and samples collected from the Fort Lewis site were analyzed at Purdue University on a Shimadzu GC17A with FID detection. For the Phase II test at Fort Lewis, initial results indicated that the contaminant concentrations were below method detection limits. Consequently, aqueous samples were hexane extracted to concentrate contaminant and then re-analyzed by the Shimadzu GC17A with electron capture detection.

3.4. Transect Method

The “Transect Method” (TM), as described in API (2003), involves estimating contaminant discharge per well as the product of the average groundwater flux (\bar{q} , where the double over bar indicates a spatial average over a region larger than the well), the well cross-sectional area (A), and the flux-averaged contaminant concentration in the well (\bar{C}_f , where the subscript f indicates a flux-averaged value, and the over bar indicates a spatial scale associated with the well). In turn, \bar{q} is estimated as the product of the hydraulic gradient (\bar{i}) and saturated hydraulic conductivity (\bar{K}). Measured hydraulic gradients and previously reported values of \bar{K} were used to estimate \bar{q} whereas contaminant concentrations measured during the IPTs were used to estimate \bar{C}_f . Note that the first value of the concentration-time series measured during the IPTs most closely represents the concentration measured by traditional groundwater sampling (e.g., bailing or pumping), and was therefore used in this approach.

4. Results and discussion

4.1. Groundwater fluxes

4.1.1. Hill AFB site

Consistent spatial trends in groundwater flux across the well transect were not evident (see Supplemental Material), and

depth-averaged groundwater fluxes (\bar{q}) were estimated from PFM deployments in the wells (Table 1). Note that one well (U2-157) was dry during Phase I deployments and another well (U2-154) did not have sufficient water to use during the IPT, while two wells (U2-154, U2-157) were dry during the Phase II deployment. The transect-wide average groundwater flux (q), determined from PFM deployments, was 2.5 ± 1.8 cm/day (spatial mean and standard deviation of all PFM measurements) for Phase I, and 1.5 ± 0.7 cm/day for Phase II. Correction factors accounting for flow convergence around the well screens were estimated to range from 1.03 to 1.05 for PFM applications at Hill AFB and Ft. Lewis, and were consequently neglected (see Supplementary Material; Hatfield et al., 2004; Klammer et al., 2007). For Phase I and II IPTs, q values were 2.9 ± 1.8 cm/day, and 1.7 ± 1.0 cm/day, respectively, which are in general agreement with the PFM estimates. Based on the topography of the underlying clay surface (see Figs. 1 and 2, and Supplementary Material), groundwater flow at the Hill AFB site was directed perpendicular to the well transect; therefore, Eq. (4) was used in the IPT analysis.

The average hydraulic gradient across the transect using wells U2-216 and U2-117 based on water level measurements collected both prior to the Phase I flux measurements and prior to the Phase II flux measurements was ~0.002. Using a value of 17 m/day for the hydraulic conductivity based on site characterization data (Meinardus et al., 2002; Rao et al., 1997), the estimated average groundwater flux is 3.4 cm/day, which compares reasonably well with the Phase I PFM and IPT results (~20% difference), but less so for the Phase II PFM and IPT results (~70% difference). Moreover, these estimates do not support the PFM and IPT measurements that indicate the Phase II q was approximately 40% less than the Phase I q . One possible explanation for these differences is temporal variability of the hydraulic gradient between U2-216 and U2-117 that is not reflected in the longer term averages used in the analysis above (see Supplementary Material for an analysis of temporal variation in hydraulic gradients across the transect). Furthermore, the reduced groundwater flux estimates during Phase II based on the PFM and IPT methods may reflect a reduction in hydraulic conductivity due to biomass from microbial degradation activity (see discussion below regarding DCE mass discharge).

4.1.2. Ft. Lewis site

Strong depth patterns were absent in groundwater flux profiles within a well as determined from PFM deployments

Table 1

Summary of depth-averaged groundwater fluxes, \bar{q} (cm/day), estimated based on Phase I (pre-remediation) and Phase II (post-remediation) PFM deployments in wells along source transects at two DNAPL sites

Hill AFB			Ft. Lewis		
Well	Phase I	Phase II	Well	Phase I	Phase II
U2-154	0.5	Dry well	LC-201	38.1	28.5
U2-152	2.8	1.7	LC-202	41.0	16.2
U2-150	2.8	1.3	LC-203	21.9	17.5
U2-148	3.1	1.5	LC-204	24.1	13.5
U2-116	2.9	1.9	LC-205	24.2	8.8
U2-149	1.9	1.4	LC-206	20.4	8.3
U2-151	3.4	2.0	LC-207	20.2	13.0
U2-153	2.1	1.2	LC-211	23.1	17.8
U2-155	2.3	1.2	LC-212	20.5	11.7
U2-157	Dry well	Dry well	LC-213	39.2	32.2

(data shown in the Supplementary Material). Depth-averaged groundwater fluxes (\bar{q}) calculated for the wells are summarized in Table 1. Tracer depletion data from PFM deployments yielded a transect-wide average groundwater flux (\bar{q}) estimate of 27 ± 19 cm/day for Phase I, and 16 ± 12 cm/day (spatial mean and standard deviation of all PFM measurements) for Phase II deployments. The difference noted in q for the two deployments is consistent with the observed difference in hydraulic gradients (See Supplementary Material). The groundwater fluxes at the Ft. Lewis site are about an order of magnitude larger than those observed at the Hill AFB site, reflecting the differences in site hydrogeology.

The pumping rates used during the Phase I IPT were not large enough to induce sufficient changes in hydraulic head to apply the modified IPT method to estimate groundwater flux, an issue that was compounded by interference from a series of precipitation events that occurred prior to and during the IPT. As a result, the Phase I groundwater fluxes were estimated by scaling the Phase II IPT groundwater flux using the ratio of the hydraulic gradients for the two tests, as measured in surrounding wells (see Supplementary Material for information on measured hydraulic gradients). Furthermore, an analysis of the

hydraulic gradient in the vicinity of the well transect indicated considerable variation in the flow direction; thus, it was necessary to use Eq. (5) to explicitly account for the flow direction relative to the transect.

The estimated groundwater flux values based on IPT results during Phase II were 16 ± 13 cm/day, 18 ± 6 cm/day, and 21 ± 4 cm/day for the southern, middle, and northern portions of the transect, respectively, and \bar{q} for the entire transect was estimated at 18 ± 9 cm/day. The Phase I estimate of \bar{q} was 32 cm/day (assuming no error in the hydraulic gradient estimates, the estimated standard deviation is 16 cm/day) and the values for each segment were 28 cm/day, 31 cm/day, and 37 cm/day for the southern, middle, and northern portions of the transect, respectively. The transect-wide averages based on the IPTs are very comparable to those based on the PFMs for Phase I and II.

Based on typical values of hydraulic conductivity from pumping tests (5 m/day to 335 m/day), the groundwater flux was reported to range from approximately 2 cm/day to 460 cm/day (USACE, 2002) in the Vashon aquifer unit. An average hydraulic conductivity of 91 m/day (300 ft/day) was used for the Ft. Lewis site in a regional groundwater transport model (Truex

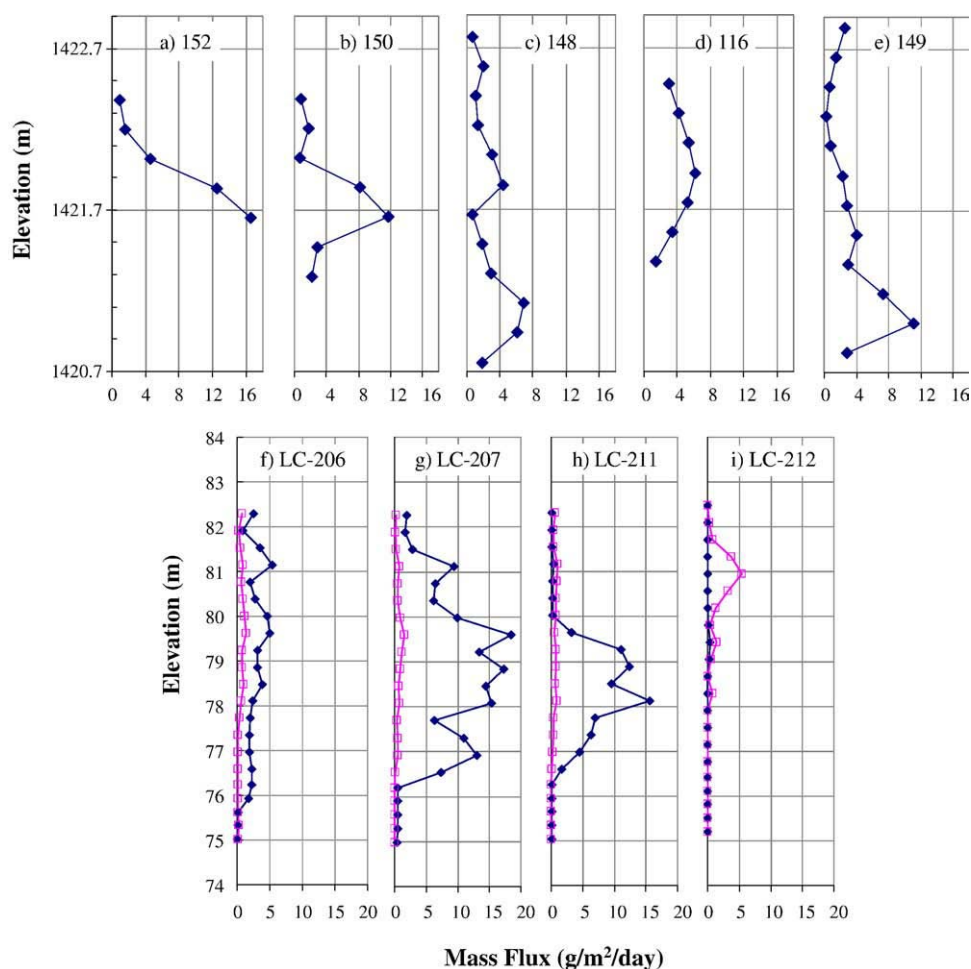


Fig. 5. Mass flux profiles measured in selected wells using the PFMs. The solid diamond represents TCE, and the open square represents DCE. Panels (a) through (e) show the wells with the largest fluxes during Phase I at Hill AFB, and panels (f) through (i) show the largest Phase I flux values at NA1, Fort Lewis. Note the change in scale on both axes to accommodate the data.

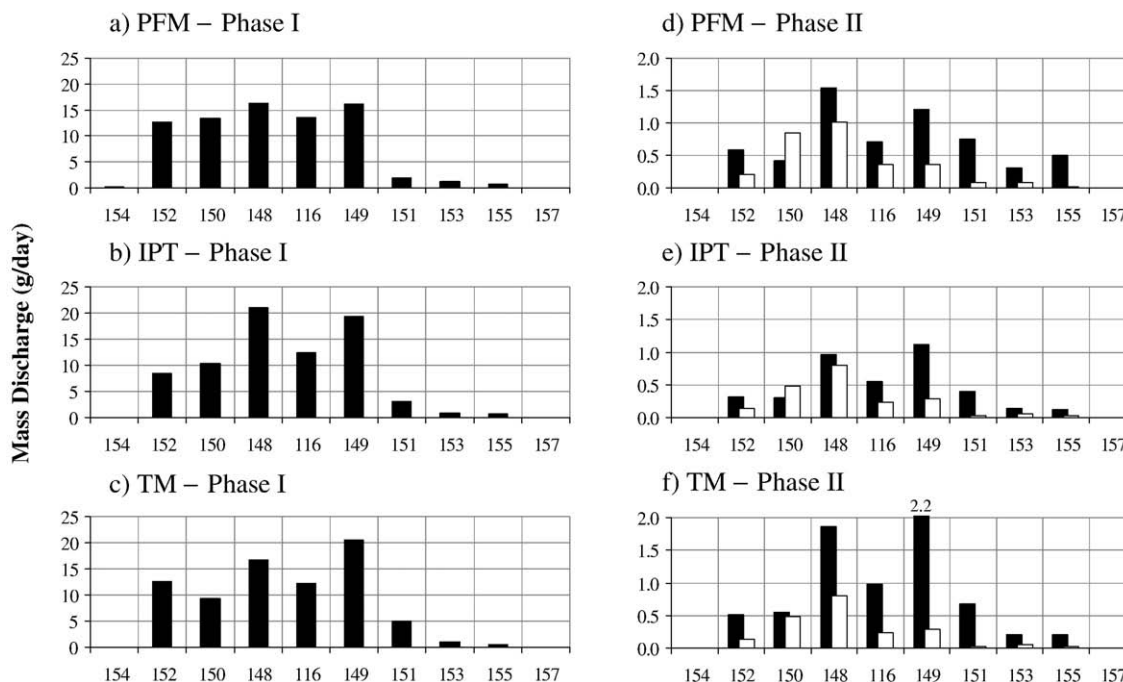


Fig. 6. Average mass discharge (g/day) for each well at Hill AFB, as measured in Phase I (May-02) by a) PFM, b) IPT, and c) TM; and as measured in Phase II (Jun-03) by d) PFM, e) IPT, and f) TM. TCE is shown in black and *cis*-DCE is shown in white. Note the change in scale on the y-axis to accommodate the reduced discharge during Phase II (Jun-03) measurements.

et al., 2004). Using this value, the groundwater flux for Phase I and II tests was 34 cm/day and 19 cm/day, respectively, using the average Phase I and II hydraulic gradients.

4.2. Contaminant fluxes and mass discharge

4.2.1. Hill AFB site

The contaminant flux profiles as measured by the PFM deployments are shown in Fig. 5 for the five wells with the largest flux values. Based on all Phase I PFM measurements, the average mass flux across the transect was ~ 3 g/m²/day, and the

maximum measured flux was ~ 17 g/m²/day in well U2-152 (Fig. 5). The PFM measured contaminant flux profiles were integrated to generate estimates of contaminant mass discharge contributed by each well. These values were then compared with equivalent estimates using the IPT method (see Fig. 6). During the IPTs, significant changes in concentrations for a given well within each test were not observed (see Supplementary Material), which suggested a uniform concentration within the capture zone of the wells (Bockelmann et al., 2001). Both methods provide comparable magnitudes and patterns of TCE mass discharge along the well transect.

Table 2

Summary of contaminant (TCE and DCE) mass discharge rates (g/day) as estimated using PFM and IPT results, and comparison with corresponding estimates based on the Transect Method (TM)

Contaminant	Method	Hill AFB		Ft. Lewis	
		Phase I (pre-remediation) May 2002	Phase II (post-remediation) June 2003	Phase I (pre-remediation) Oct 2003 ^a	Phase II (post-remediation) June 2006 ^b
TCE (g/day)	PFM	76	6.0	743 (646)	3.4 (2.3)
	IPT	76	3.9	536 (466)	2.2 (1.5)
	TM	78	7.2	688 (599)	2.8 (1.9)
DCE (g/day)	PFM	– ^c	3.0	155 (135)	5.7 (3.9)
	IPT	– ^d	2.0	257 (224)	0
	TM	– ^d	3.8	288 (251)	0
TCE+DCE (moles/day)	PFM	0.58	0.077	7.3 (6.4)	0.085 (0.059)
	IPT	0.58	0.051	6.7 (5.8)	0.017 (0.012)
	TM	0.59	0.094	8.2 (7.1)	0.021 (0.015)

^a Shown in parenthesis are flow-angle corrected values (13% less than uncorrected values).

^b Shown in parenthesis are flow-angle corrected values (31% less than uncorrected values).

^c DCE concentrations in SI-GAC extracts were below the level of quantification.

^d DCE was not included in the analysis of all samples.

Moreover, estimates based on the TM approach were also comparable to the PFM and IPT results (Table 2 and Fig. 6), and the similarity in results between the TM and IPT methods is not unexpected given the relatively uniform concentration-time series data. In Phase I, it is evident that only five wells (152, 150, 148, 116, 149) on the northern half of the well transect contributed much of the TCE mass discharge. URS and INTERA (2003) reported that during SEAR in Panel 5, the majority (78%) of the DNAPL mass was removed at the two extraction wells U2-211 and U2-207. These wells are not immediately up-gradient of wells 152, 150, 148, 116, and 149 (Fig. 1); however, this is not surprising given the complex shape of the paleo-channel aquifer, which may serve to funnel water from the source zone through a more narrow spillway created by the clay surface. TCE flux distributions in these five wells (see Fig. 5) consistently show the largest flux magnitudes towards the bottom. This is consistent with the observation that the majority of the DNAPL in Panel 5 was pooled on the clay aquitard (Meinardus et al., 2002; Holbert et al., 2004). For the Phase II test, \bar{M}_D for TCE is about an order of magnitude smaller compared to that measured during Phase I, and the relative distributions of TCE and DCE during Phase II are similar. The mass discharge distribution is still predominantly located in wells 152, 150, 148, 116, and 149; although a larger fraction of the total discharge is located in wells 151, 153, and 155.

The total TCE mass discharge for each test and the DCE mass discharge for Phase II are summarized in Table 2. Substantial reduction (>90%) in TCE mass discharge was noted from source treatment, from ~76 g/day (Phase I) to ~5 g/day (Phase II). On the other hand, the DCE mass discharge (estimated at ~0.1 g/day, see Supplementary Material) was roughly three orders of magnitude lower than the TCE mass discharge in Phase I, but

increased to ~3.0 g/day during Phase II. Since DCE was not present in significant concentrations prior to source remediation, these results suggest the surfactant used for in-situ flushing contributed to the reductive dechlorination of TCE (Holbert et al., 2004). However, the total Phase I molar discharge of TCE and DCE was ~0.6 moles/day (Table 2), and the total Phase II molar discharge was ~0.07 moles/day. Based on these measures, the total molar discharge reduction was 88%, which still suggests a significant discharge reduction even when accounting for degradation of TCE to DCE.

4.2.2. Ft. Lewis site

Contaminant mass discharge variations, as estimated by all three methods (Fig. 7), indicate that for the Phase I tests four wells (LC-205, LC-206, LC-207, LC-211) along the northern end of the well transect contributed the majority of the mass discharge (92% on average using the PFM, IPT, and TM methods). Of these four, LC-207 and LC-211 account for about 63% of the total mass discharge, and in these wells TCE flux distribution as measured by PFM deployments suggest peak values between elevations of 78 m to 80 m (~5 to 7 m below grade; Fig. 5). The largest Phase I flux measured by the PFMs was ~18 g/m²/day in well LC207, and based on all Phase I PFM measurements the average mass flux across the transect was ~2 g/m²/day. For Phase II tests, on average 89% of the mass discharge occurred in wells LC-201 through LC-205, and 57% of the TCE mass discharge occurred in wells LC-202 and LC-203. This shift in the location where the majority of the mass discharge crosses the transect in general corresponds to the change in groundwater flow direction: the estimated flow direction was at an azimuth of 294° for the Phase I tests and 219° for the Phase II tests (the azimuth of a line perpendicular to the transect is 265°). In addition, there is

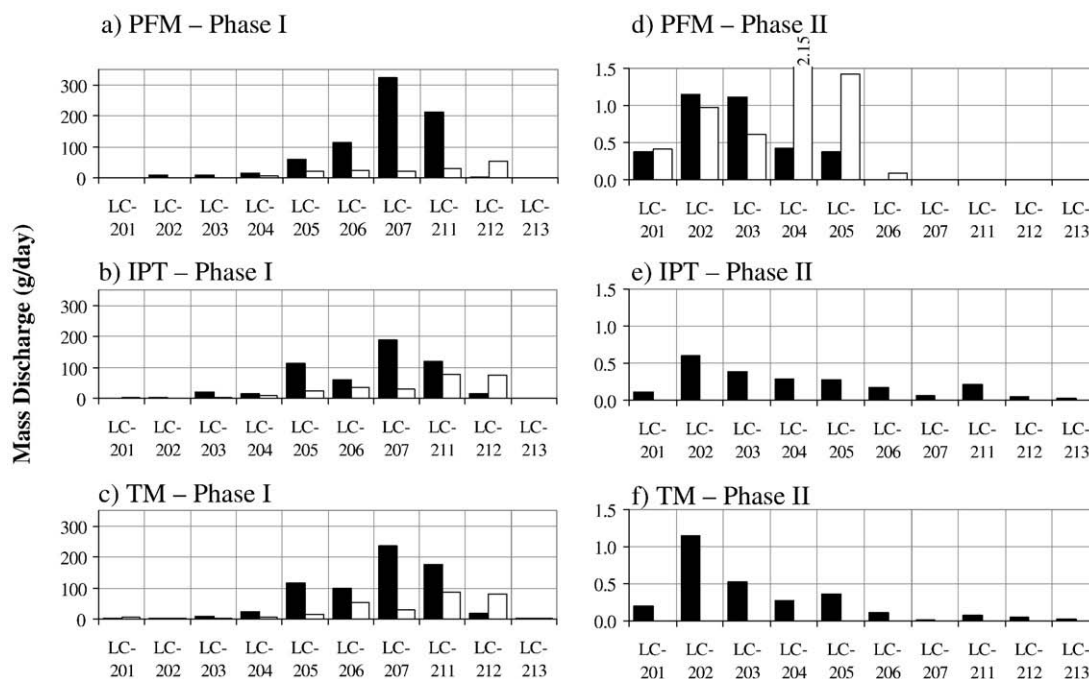


Fig. 7. Average mass discharge (g/day) for each well at NA1 Fort Lewis, as measured by a) PFMs, b) IPT, and c) TM for Phase I tests (Oct-03); and as measured by d) PFMs, e) IPT, and f) TM for Phase II tests (Jun-06). TCE is shown in black, and *cis*-DCE is shown in white. Note the change in scale on the y-axis to accommodate reduced discharge during Phase II (Jun-06) tests.

general agreement between the Phase I well locations of elevated flux, the Phase I groundwater flow direction, and the locations of elevated concentration as measured during thermal treatment (USACE, 2008).

Estimates of total contaminant mass discharge, based on the PFM, IPT, and TM approaches conducted at the NA1 source area are summarized in Table 2. TCE and DCE mass discharge for Phase I, averaged for three methods, was 655 g/day and 233 g/day, respectively. Thermal treatment of the source zone resulted in substantial decrease in mass discharge for both TCE and DCE: 2.8 g/day and 1.9 g/day, respectively. It is evident that reduction in mass discharge was >99% for both TCE and DCE using the average values of \bar{M}_D . If only the PFM data were used (since DCE was not detected during the Phase II IPT), the reduction in DCE mass discharge is >96%. These \bar{M}_D values are uncorrected for the oblique flow direction, estimated to be 29° and 46° from the transect orthogonal during the Phase I and Phase II tests, respectively (see Supplementary Material). While the oblique flow angle does not impact the estimates of mass flux by any of the techniques as applied here, they do reduce \bar{M}_D by a fraction equal to the cosine of the angle defined by the transect orthogonal and the oblique flow direction. Consequently, accounting for the oblique angle reduces \bar{M}_D by a factor of 13% for Phase I and 31% for Phase II, resulting in slightly higher mass discharge reductions for TCE and DCE compared to the uncorrected estimates.

The concentrations measured in the pumping well effluents during the IPTs at Ft. Lewis varied more compared to those measured at Hill AFB (see Supplementary Material), suggesting a heterogeneous contaminant concentration distribution around the wells (Bockelmann et al., 2001). For Phase I, the relative variation in concentration within most wells did not have a clear, monotonically increasing or decreasing pattern. During Phase II, the effluent concentration likewise indicated relatively heterogeneous spatial concentration distributions near the well, which became more uniform as the volume of extracted groundwater increased.

During Phase II, DCE was only detected by the PFM technique. The presence of DCE in the PFM data, but not the IPT data, suggests that TCE was converted to DCE in the vicinity of the well or on the PFM sorbent, and the absence of DCE from the initial sample collected during the IPT may suggest DCE was diluted below detection limits. In any case, the total molar discharge prior to thermal treatment based on the PFMs was ~7 moles/day (Table 2), and the total molar discharge after treatment was ~0.09 moles/day. The mole ratio of TCE to DCE based on the PFMs was approximately 3.6 prior to treatment, and approximately 0.4 after treatment. The higher DCE molar ratio measured during Phase II may suggest an increase in biodegradation activity during or subsequent to thermal treatment (e.g., Powell et al., 2007; Truex et al., 2007). However, even accounting for the molar sum of both TCE and DCE, the percent reduction in molar discharge was ~99% after thermal treatment, indicating that transformation of TCE to DCE was only a minor factor in the post-treatment reduction of TCE.

As noted in Section 3.1, NAPL was noted in the drill cuttings recovered from ~3 m to ~5 m below grade (80 m to 83 m in elevation, water table at ~82 m) during the installation of well LC-211, directly above the peak PFM flux values located between elevations of 78 m to 80 m (~5 to 7 m below grade; Fig. 5). While the interval over which NAPL was noted in the drill cuttings and

the PFM interval with the peak values do not coincide, their proximity does raise the question as to whether the flux measurements reflect a localized DNAPL hotspot. In which case, the Phase I mass discharge estimates for this well would overestimate the true mass discharge because the elevated flux estimate is assigned to the entire sampling space associated with the well. While not definitive, inspection of the concentration-time series measured in this well during the IPT (see Supplementary Material) generally does not support the presence of a local DNAPL hot spot. To assess the uncertainty of our conclusions, \bar{M}_D for Phase I was recalculated for each method assuming \bar{M}_b for well LC-211 during Phase I was the same as during Phase II. The average \bar{M}_D of all three methods is 486 g/day, which is 26% lower than the \bar{M}_D including results from LC-211. However, the average percent reduction is 99.4%, which is essentially the same result obtained with the inclusion of LC-211.

4.3. Mass discharge reduction and source mass depletion

The in-situ surfactant flood of the Panel 5 DNAPL source zone at Hill AFB resulted in an estimated DNAPL mass removal ranging from 1340 kg (221 gal) to 2250 kg (371 gal) (URS and INTERA, 2003), based on liquid-phase separation in the effluent treatment system and based on effluent TCE breakthrough curves from the extraction wells, respectively. A partitioning tracer test conducted prior to surfactant flooding suggested an initial TCE mass of ~2180 kg (360 gal) (URS and INTERA, 2003). These data suggest that >60% of the DNAPL mass was removed from the source zone. Free-phase DNAPL has since been detected in wells within Panel 5, indicating some DNAPL was not removed. Based on the PFM and IPT contaminant flux measurements reported here, reduction in TCE mass discharge resulting from source mass depletion ranged from 92% to 97%. Increased biodegradation of TCE after the SEAR is indicated by an increase in DCE flux.

At the Ft. Lewis site, estimates of pre-remediation TCE mass based on soil core samples range from ~3800 kg (688 gal) to 13,400 kg (2400 gal), depending on whether a single elevated NAPL saturation estimate is excluded or included, respectively, in the calculation of average saturation (USACE, 2002, 2008). If this single data point is treated as an outlier and excluded from the average saturation calculation, then the pre-remedial TCE mass estimate is ~3800 kg. An alternative estimate of pre-remedial TCE mass is obtained using the mass of TCE removed during thermal treatment added to the TCE mass estimated from post-remedial soil sampling results. A total TCE volume of 2576 kg (466 gal) was removed by the thermal treatment (Beyke and Fleming, 2005), using this and an average value of 45 kg (range was 20 to 70 kg; USACE, 2008) for the post-remedial soil coring mass provides a second estimate of pre-remediation TCE mass of 2621 kg (472 gal). This suggests a reduction in TCE source mass ranging from ~68% to ~98%. These estimates do not account for TCE mass that may have been destroyed in place by thermal treatment, which may have ranged from 8% to 70% of the mass removal estimate based on results from NA2 and NA3 (USACE, 2008). Accounting for mass destroyed if known would increase the estimate of mass reduction.

Immediately following thermal treatment, TCE concentration in groundwater, averaged over the area interior to the treated zone, was reduced by ~87% from an initial concentration

of approximately 1 mg/L) (Beyke and Fleming, 2005, USACE, 2008), and was reduced by ~99.5% 2 years after treatment (Powell et al., 2007, USACE, 2008). The additional reduction in TCE concentration two years after treatment may be explained by microbial degradation, or by a reduction in groundwater temperature following treatment, which would reduce the TCE solubility. Based on the contaminant flux measurements reported here, the reduction in TCE mass discharge was $\geq 99.5\%$. Corresponding mass discharge reductions for DCE ranged from 96.4% to $>99\%$.

Substantial DCE fluxes were detected at the Ft. Lewis transect before thermal treatment of the NA1 source area, while at the Hill AFB OU2 Panel 5 site significant DCE fluxes were measured only after SEAR of the source zone. Furthermore, the spatial distribution of the DCE mass discharge at NA1 is different from that of TCE (Fig. 7). This suggests that the spatial distribution of DCE in the NA1 source zone may have been different from that of the TCE, that degradation rates were spatially variable, or that the DCE originated from an older or further up-gradient TCE source and was converted before reaching the flux transect.

4.4. Mass discharge measurement uncertainty

Factors that may have affected \overline{M}_D for a given method vary, since each method has unique advantages and disadvantages. PFM's provided both groundwater and contaminant mass flux as a function of depth, but the lateral distance (~5 cm) interrogated was relatively small compared to the well spacing (~3 to ~5 m) at both sites (see Kübert and Finkel, 2006 and Li et al., 2007 for an analysis of errors in \overline{M}_D estimates from sampling techniques with relatively small-scale support volumes). Likewise, the sampling distance associated with the TM approach is similar to that for the PFM. In addition, TM results at the Ft. Lewis site may have been impacted by sampling bias created by long well screens (Martin-Hayden, 2000a,b). As employed here, TM results may have been impacted by uncertainty associated with homogeneity assumptions in order to use \overline{K} and \overline{i} to calculate \overline{q} . The appropriateness of using \overline{K} and \overline{i} estimates to complete the TM approach given the spatial variability of PFM results may be questioned; however, hydraulic conductivity values based on pump tests or slug tests completed at one or a few locations across a site, and site-wide estimates of hydraulic gradient are routinely used in typical site characterization approaches.

The advantage of the IPT is that it does integrate information over a much larger volume. However, it does not provide information on the vertical distribution, and as employed here, estimates of \overline{q} from the IPT data are based on assumptions of homogeneous and isotropic conditions. These assumptions, however, are not dissimilar from those underlying the theoretical basis for typical pump tests (i.e., traditional pump test conducted to measure \overline{K}), and as such results from the IPTs are viewed as an average over the volume interrogated. Furthermore, the modified IPT was employed in this study not because of insignificant heterogeneity, but rather as a way to maximize the amount of information obtained from conducting an IPT in which wells were pump concurrently.

Finally, given the vertical variability in horizontal groundwater flux based on the PFM results at both sites, uncertainty may result from vertical flow in the well (Elci et al., 2001,

2003) prior to \overline{M}_D measurements by any technique. At the Hill AFB site, the maximum thickness over which the vertical gradients may create bias was relatively small; ≤ 2 m in two wells and ≤ 1 m in the remaining wells (see Figs. 2 and S-3). At the Ft. Lewis site, vertical gradients have been measured (USACE, 2002), and relatively long wells screens were employed (saturated thickness of ~7.6 m). A critical issue with respect to \overline{M}_D measurements at Ft. Lewis then is the potential dilution of concentration below detection limits as a result of vertical flow in the well, otherwise, dilution of concentration (but still detectable) across the well depth should not significantly effect the mass discharge estimates since the depth over which the dilution occurred is explicitly included in the estimate of the control plane area. A sensitivity analysis using minimum detected concentrations during the IPTs indicates that the sensitivity of the \overline{M}_D estimates to dilution issues is approximately an order of magnitude lower than the \overline{M}_D estimates obtained during Phase I and II at both sites. Consequently, it is unlikely that dilution due to vertical flow would significantly change our conclusions about mass discharge reductions.

5. Conclusions

Both PFM and IPT flux measurement techniques provide critical site characterization data required for assessing remedial performance based on changes in contaminant flux and mass discharge from DNAPL source zones. Transect-wide average TCE mass flux prior to source treatment at the Ft. Lewis site (~2 g/m²/day) is very similar to that at the Hill AFB Panel 5 site (~3 g/m²/day). The larger mass discharge at the Ft. Lewis site (~640 g/day) relative to the Hill AFB site (~76 g/day) can be attributed to the larger cross-sectional area at Ft. Lewis (418 m²) compared to that at Hill AFB (25 m²), as well as the much larger groundwater flux at Ft. Lewis (~25 cm/day) compared to that at Hill AFB (~3 cm/day). Contaminant flux measurements made using both PFM and IPT approaches at these two DNAPL sites indicate that TCE mass depletion ($>60\%$) in the source zone through aggressive treatment (surfactant flooding; resistive heating) resulted in substantial ($>90\%$) reduction in TCE mass discharge at the source control plane. At the Hill AFB site, enhanced biodegradation in the source zone after surfactant flooding was manifested in increased DCE flux. At the Ft. Lewis site, thermal treatment of the source zone resulted in reductions of mass discharge rates for both TCE and DCE.

Contaminant flux distributions measured using PFM and IPT approaches provide data on the spatial patterns in contaminant mass discharge at the source control plane. Data for the two DNAPL sites evaluated suggest that a significant fraction of the mass discharge occurs over a small portion of the source-zone control plane. This is consistent with the field observations reported by Guilbeault et al. (2005) and Basu et al. (2007). Based on detailed groundwater profiling at three DNAPL sites, Guilbeault et al. (2005) noted that about 80% of the contaminant mass discharge occurs over 10% or less of the cross-sectional area. These data suggest that use of either PFM or IPT tests to characterize DNAPL source zone will provide critical data needed for more efficient targeting of aggressive treatment to achieve the desired reduction in source strength.

Performance monitoring at two sites, where DNAPL source-zone remediation was completed, suggest that source

mass depletion through aggressive treatment can lead to a significant reduction in source strength, as measured by the contaminant mass discharge at the source control plane. Examining the impacts of such reduction on the dissolved plume behavior was beyond the scope of this study. In any case, such changes would need to be monitored within the plume over much longer timelines. Moreover, the techniques described herein could be used to measure contaminant fluxes in wells along transects oriented either along the centerline of the plume (longitudinal transect) (see Basu et al., 2006) or oriented perpendicular to the flow direction (transverse transects) at one or more locations down-gradient of the source (see Bauer et al., 2004). Such measurements could be used to estimate the impacts of biogeochemical processes contributing to natural attenuation of the contaminants, and provide context for mass discharge reductions measured at the source control plane.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jconhyd.2008.05.008.

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