

Remediation of Chlorinated Ethenes, Ethanes, and Methanes in Groundwater Using Carbon- and Iron-Based Electron Donor

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Field-scale pilot tests were performed to evaluate enhanced reductive dechlorination (ERD) of dissolved chlorinated solvents at a former manufacturing facility located in western North Carolina (the site). Results of the site assessment indicated the presence of two separate chlorinated solvent-contaminated groundwater plumes, located in the northern and southern portions of the site. The key chlorinated solvents found at the site include 1,1,2,2-tetrachloroethane, trichloroethene, and chloroform. A special form of EHC[®] manufactured by Adventus Americas was used as an electron donor at this site. In this case, EHC is a pH-buffering electron donor containing controlled release carbon and ZV Iron MicroSphere 200, a micronscale zero-valent iron (ZVI) manufactured by BASF. Approximately 3,000 pounds of EHC were injected in two Geoprobe[®] boreholes in the saprolite zone (southern plume), and 3,500 pounds of EHC were injected at two locations in the partially weathered rock (PWR) zone (northern plume) using hydraulic fracturing techniques. Strong reducing conditions were established immediately after the EHC injection in nearby monitoring wells likely due to the reducing effects of ZV Microsphere 200. After approximately 26 months, the key chlorinated VOCs were reduced over 98 percent in one PWR well. Similarly, the key chlorinated solvent concentrations in the saprolite monitoring wells decreased 86 to 99 percent after initial increases in concentrations of the parent chlorinated solvents. The total organic carbon and metabolic acid concentrations indicated that the electron donor lasted over 26 months after injection in the saprolite aquifer. © 2009 Wiley Periodicals, Inc.

INTRODUCTION

The study site is located in Buncombe County, North Carolina (Exhibit 1). The site assessment indicated the presence of two separate groundwater plumes, located in the northern and southern portions of the site. Both plumes contained key chlorinated solvents, including 1,1,2,2-tetrachloroethane (TeCA), trichloroethene (TCE), and chloroform. In the northern plume study area, chlorinated solvent impacts occurred primarily in the PWR, while chlorinated solvent impacts in the southern plume occurred mostly in the saprolite zone.



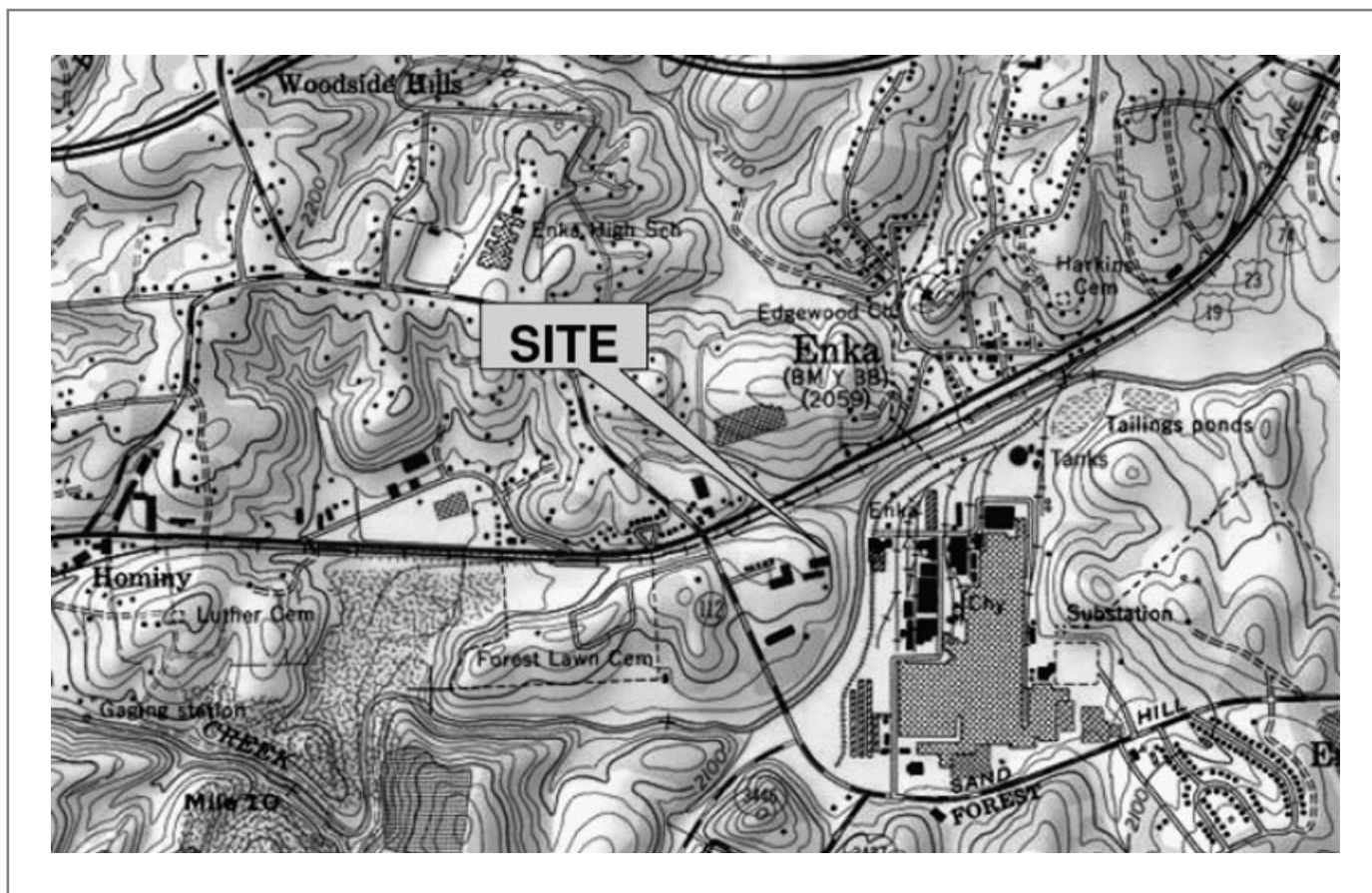


Exhibit 1. Location map, former manufacturing facility, Enka, North Carolina

A pilot study involving injection of a controlled released carbon source with BASF's micron scale zero-valent iron (ZVI) was implemented in January 2005 to enhance reductive dechlorination of chlorinated solvents in groundwater at the site. This article summarizes the pilot-study implementation and performance monitoring results.

Geologic Conditions

The site is located in the Appalachian Piedmont physiographic province (mountainous) of North Carolina and consists of silty-clay (saprolite) soils overlying a PWR zone. Fractured bedrock underlies the PWR zone. The saprolitic soils range from 40 to 55 feet thick. The PWR ranges in thickness from 5 to 25 feet and the top of bedrock ranges from 50 to 75 feet below land surface (bls).

Hydrogeologic Conditions

Data collected during the site assessment indicate that an unconfined water table resides between 8 and 30 feet bls in saprolitic soils. The groundwater flow data indicate that groundwater in shallow and deep saprolite and in partially weathered rock (PWR) generally flows to the east-northeast. Hydraulic conductivity values for saprolite wells, estimated using slug-test data during the site assessment, ranged from 2.22×10^{-3} to

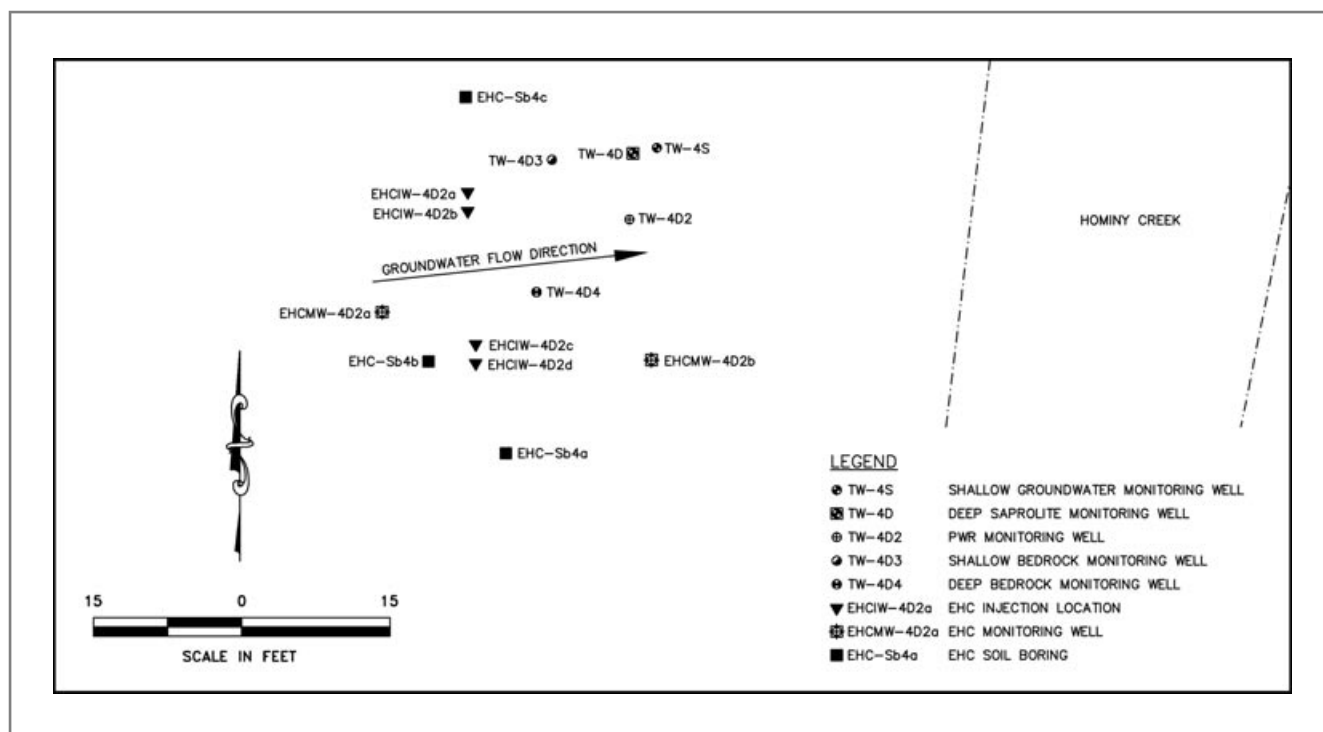


Exhibit 2. EHC injection and monitoring locations, northern plume

6.2×10^{-4} feet per minute. The estimated hydraulic conductivity value for a PWR well was 7.7×10^{-4} feet per minute.

MATERIALS AND METHODS

A special form of EHC[®], containing controlled-release carbon and BASF ZVI (ZV Iron Microsphere 200), was used as an electron donor to enhance reductive dechlorination of chlorinated solvents at the Site. The ZV Iron Microsphere 200 is spherical in shape and 3 to 5 microns in size. The advantage of ZV Iron Microsphere is that consistent reaction rates can be expected due to its narrow particle-size distribution. This iron was used in place of irregularly shaped micron-scale ZVI. The presence of low-permeability soils presented a unique challenge for placement of the EHC. Therefore, EHC was injected using hydraulic fracturing techniques. The EHC injection was performed at two locations: northern plume (see Exhibit 2) and southern plume (see Exhibit 3).

Northern Plume Injection

Two pairs of injection points (EHCIW-4D2a, EHCIW-4D2b, EHCIW-4D2c, and EHCIW-4D2d), at locations shown in Exhibit 2, were installed in the PWR zone of the aquifer by using a 4.25-inch (inner diameter) hollow-stem auger rig. Deep injection points were installed at the base of PWR on top of bedrock (34 to 36 feet bls) while the shallow PWR injection points were installed approximately 3 to 4 feet above the depth of auger refusal at or below the PWR/saprolite interface. A two-inch diameter steel casing equipped with a drive point was inserted into each of the injection points, advanced

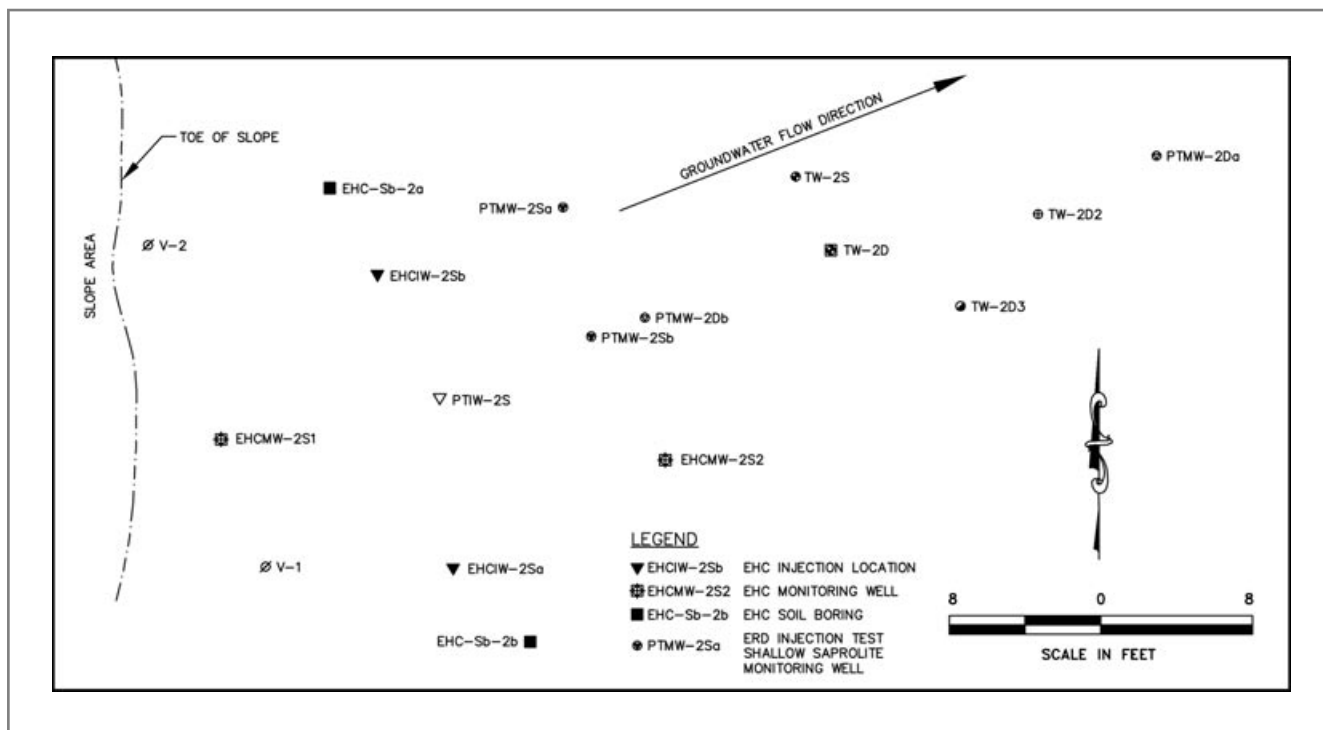


Exhibit 3. EHC injection and monitoring locations, southern plume

approximately two inches into the PWR, and grouted in place to the ground surface. Once the grout had cured for at least 24 hours, the hydraulic fracturing/injection process was initiated. A total of 1,800 lbs of EHC, 1,000 lbs of sand, and 535 gallons of guar gel and water were injected in two injection points, EHCIW-4D2a and EHCIW-4D2b. A total of 1,800 lbs of EHC, 1,000 lbs of sand, and 525 gallons of guar gel and water were injected in two injection points, EHCIW-4D2c and EHCIW-4D2d. The injection pressures ranged from 70 to 100 pounds per square inch-gauge (psig) depending upon the depth and location of the injection. Exhibit 4 presents a summary of injection pressures and EHC mixture used at each fracturing interval. Pressure transducers were placed in monitoring wells TW-4D2 and TW-4D3 to monitor the response of the water table to injection pressures.

Southern Plume Injection

Two injection points (EHCIW-2Sa and EHCIW-2Sb) were installed by FRx, Inc. (Cincinnati, Ohio) in the saprolite zone of the southern plume (see Exhibit 3) using a Geoprobe[®] rig. Initially, a high-pressure water jet was used to cut a horizontal notch in the exposed soil or strata at the injection depth. Then a mixture of EHC, sand, and guar gel slurry was injected under pressure to create fractures from the notch. A total of six fractures with approximately 3-foot vertical spacing were created in each of the injection points. Exhibit 4 presents a summary of fracture information, injection pressures, and EHC mixture. Fracture depths ranged from 8 to 25 feet bls. A total of 3,000 lbs of EHC, 3,400 lbs of sand, and 1,270 gallons of guar gel and water were injected into the six fractures at location EHCIW-2Sa. A total of 3,000 lbs of EHC, 2,900 lbs of sand, and 765

Exhibit 4. Hydraulic fracturing data, EHC injection pilot test

Injection Information			Fracture Information			Injection Information					
Injection Date	Well	Fracture No.	Injection Depth (ft)	Injection Pressure (psig)	Solids			Total		Fluid Injected	
					EHC (lbs)	Sand (lbs)	Sand Chase (lbs)	Sand and EHC (lbs)	Guar Gel (gal)		
South Plume (Saprolite)											
1/25/2005	EHC-IW-2sa	F-1	10	15-18	500	850	0	1350		200	
1/25/2005	EHC-IW-2sa	F-2	13	25-30	500	400	300	1200		225	
1/25/2005	EHC-IW-2sa	F-3	16	51	500	300	300	1100		200	
1/25/2005	EHC-IW-2sa	F-4	19	31-100	500	250	200	950		195	
1/26/2005	EHC-IW-2sa	F-5	22	45-100	500	200	200	900		225	
1/26/2005	EHC-IW-2sa	F-6	25	54-70	500	200	200	900		225	
1/26/2005	EHC-IW-2sb	F-1	8	13	500	350	500	1350		140	
1/26/2005	EHC-IW-2sb	F-2	13	25	500	300	200	1000		100	
1/26/2005	EHC-IW-2sb	F-3	16	35-88	500	200	100	800		150	
1/26/2005	EHC-IW-2sb	F-4	19	37	500	250	200	950		125	
1/26/2005	EHC-IW-2sb	F-5	22	40-60	500	200	200	900		100	
1/27/2005	EHC-IW-2sb	F-6	25	40	500	200	200	900		150	
North Plume (PWR)											
1/27/2005	EHC-IW-4D2a	F-1a	35.6	100+	900	0	500	1400		300	
1/27/2005	EHC-IW-4D2b	F-1b	32.6	100+	900	0	500	1400		235	
1/27/2005	EHC-IW-4D2c	F-1c	33.5	70-100+	900	0	500	1400		250	
1/27/2005	EHC-IW-4D2d	F-1d	30.4	NA	900	0	500	1400		275	

Notes: ft = feet; gal = gallons; lbs. = pounds; psig = pounds per square inch (guage).

gallons of guar gel and water were injected into the six fractures at location EHCIW-2Sb. The injection pressures ranged from 13 to 100 psig depending upon the depth and location of the injection (refer to Exhibit 4 for details).

Following the injection, the injection-point boreholes were abandoned by pressure grouting from the bottom to the top of the borehole. The lateral distribution of EHC and propagation of fractures were evaluated by installing soil borings around the injection points. Three soil borings were installed in the northern plume—EHC-Sb4a, EHC-Sb4b, and EHC-Sb4c (see Exhibit 2)—and two borings were installed in the southern plume—EHC-Sb-2a and EHC-Sb-2b (see Exhibit 3).

A groundwater monitoring program was implemented to evaluate the performance of the injection program. Three monitoring wells (EHCMW-4D2a, EHCMW4D2b, and TW-4D2) in the northern plume (see Exhibit 2) and three monitoring wells (EHCMW-2S1, EHCMW-2S2, and PTMW-2Sa) in the southern plume (see Exhibit 3) were sampled periodically. Prior to sampling, depth to water was gauged in the monitoring wells with an electronic water-level meter. Field measurements, consisting of dissolved oxygen (DO), oxidation-reduction potential (ORP), temperature, pH, and specific conductance, were collected during purging using a water quality meter equipped with a flow-through cell. Groundwater samples from these wells were analyzed for volatile organic compounds (VOCs) using United States Environmental Protection Agency Method 8260B. In addition, geochemical parameters (ferrous iron, total iron, dissolved and total manganese, nitrate, nitrite, sulfate, sulfide, total organic carbon [TOC], volatile fatty acids [VFAs], and dissolved gases [carbon dioxide, methane, ethene, ethane, and propane]) were also analyzed. Groundwater samples from wells EHCMW-2S2, PTMW-2Sa, EHCMW-4D2a, and EHCMW-4D2b were also analyzed for halorespiring bacteria using polymerase chain reaction (PCR) analysis. The PCR analysis was performed once, approximately five months after injection.

RESULTS AND DISCUSSIONS

Distribution of EHC Material

The observed depths and thicknesses of fractures in the soil borings are summarized in Exhibit 5. EHC was observed at several depths in soil boring EHC-Sb2a, located approximately 5 feet northwest of injection point EHCIW-2Sb (southern plume). As shown in Exhibit 5, the largest fractures were observed in the shallower locations.

The three soil borings (EHC-Sb4a, EHC-Sb4b, and EHC-Sb4c) performed in the vicinity of the PWR injection points showed very little evidence of fracturing. Only one fracture containing a small amount of EHC was observed in boring EHC-Sb4b at a depth of approximately 32.7 feet bls.

Pressure transducer data in monitoring wells TW-4D2 and TW-4D3 during injection in the northern plume are presented in Exhibit 6. Pressure response due to the injection was observed in well TW-4D3 (shallow bedrock well located approximately 9 feet from injection point EHCIW-4D2a), but no response was observed in the PWR well TW-4D2 (located approximately 16 feet from injections EHCIW-4D2a and EHCIW-4D2b).

Exhibit 5. Observed fractures in soil borings

Observed Fractures		
Boring	Depth (feet below land surface)	Thickness (inches)
South Plume		
EHC-Sb-2a	3.5	1
	6.5	1
	13.5	<0.1
	15	<0.1
	19.8	<0.1
	22.5	0.5
EHC-Sb-2b	Not Available (NA)	NA
North Plume		
EHC-Sb-4a	none	NA
EHC-Sb-4b	2.7	0.7
EHC-Sb-4c	none	NA

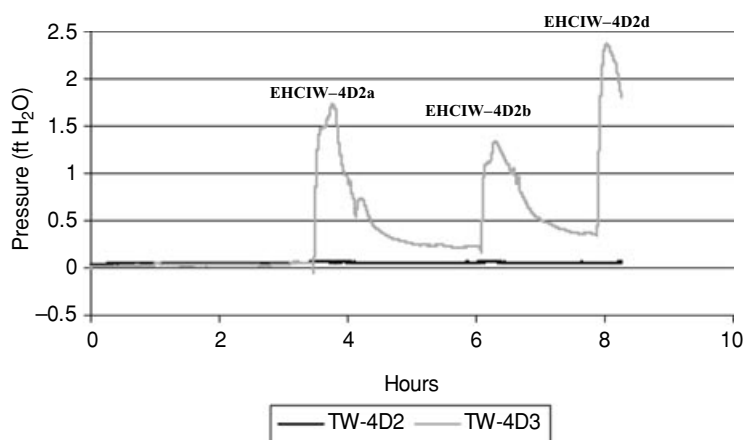


Exhibit 6. Pressure transducer data, northern plume EHC injection

Groundwater Monitoring Results

Geochemical parameters in wells EHCMW-2S2 and PTMW-2Sa in the southern plume and well EHCMW-4D2 in the northern plume indicated favorable conditions for reductive dechlorination immediately after the injection, likely due to ZVI. Moderate concentrations of TOC were found immediately following EHC injection in these wells, which gradually increased over time, indicating the influence of EHC (electron donor) injection, and later decreased, likely due to utilization of the electron donor in the biodegradation processes and dispersion/dilution due to natural movement of groundwater. As presented in Exhibit 7, well EHCMW-4D2a in the northern plume and wells EHCMW-2S2 and PTMW-2SA in the southern plume showed strong reducing conditions due to EHC influence. Strong initial reducing conditions appeared to be due to the reducing effect of the ZV MicroSphere200 used in the EHC. In addition, ferrous iron,

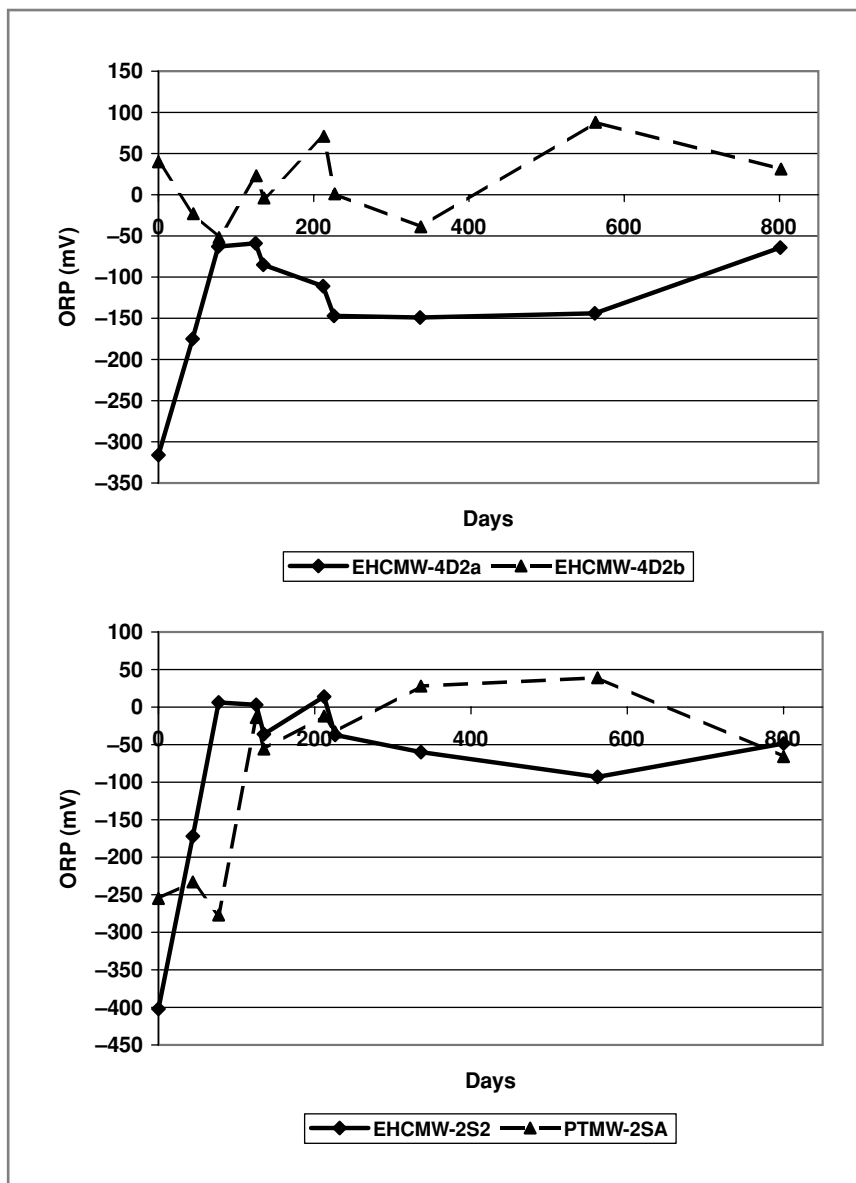


Exhibit 7. Oxidation-reduction potential in pilot monitoring wells

dissolved manganese, carbon dioxide, alkalinity, and VFA concentrations increased. The presence of degradation products (e.g., carbon dioxide, methane) and reduction in electron acceptor concentrations indicated that biodegradation of the electron donor was occurring. Exhibit 8 presents the trend in TOC concentrations in the pilot monitoring wells. The TOC and VFA data indicate that the injected electron donor lasted over 26 months in the saprolite aquifer. As presented in Exhibit 9, groundwater microbial analysis indicated the presence of dechlorinating bacteria, such as *Dehalococcoides* spp., *Dehalobacter* spp., and methanogens, at the site.

Northern Plume

The EHC injection had the most effect on well EHCMW-4D2a (located upgradient of the injection) in the northern plume, as many geochemical parameters indicated the

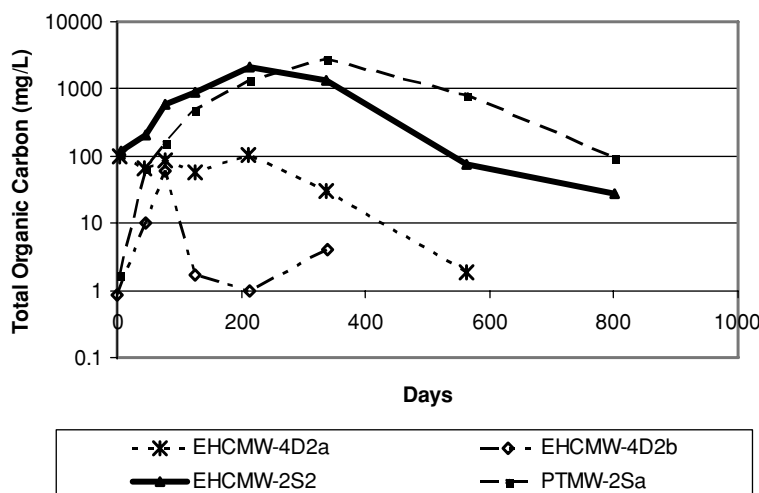


Exhibit 8. TOC in monitoring wells

generation of conditions favorable for reductive dechlorination (e.g., reduced DO, reduced ORP, and increased TOC). Similar to the southern plume (saprolite), the effect of the EHC on geochemical conditions was transient. Overall reductions in concentrations of TeCA, chloroform, and TCE (key parent chlorinated solvents) observed in well EHCMW-4D2a were approximately 98 percent, 99 percent, and 99 percent, respectively. Significant reductions in concentrations of these chlorinated solvents were observed around three to four months following injections and rebounds were observed during the later months (see Exhibit 10). Following these reductions, significant concentrations of daughter products (methylene chloride, *cis*-1,2-dichloroethylene [*cis*-1,2-DCE], and *trans*-1,2-dichloroethylene [*trans*-1,2-DCE]) were observed. Low concentrations of vinyl chloride, ethene, ethane, and methane were also observed. No significant concentrations of hydrogenolysis daughter products (1,1,2-trichloroethane [1,1,2-TCA] and 1,2-dichloroethane [1,2-DCA]) were observed. However, *cis*-1,2-DCE and *trans*-1,2-DCE concentrations significantly increased (approximately in equal quantities) during the first six months after injection and later decreased to below cleanup standards.

The EHC injection only slightly affected the geochemistry of well EHCMW-4D2b and had no noticeable effect on well TW-4D2. A slight increase in TOC concentrations was observed in well EHCMW-4d2b between 45 and 78 days of EHC injection, which later decreased to background concentrations (see Exhibit 8). No measurable influence of electron-donor injection was observed in the downgradient PWR well TW-4D2. However, these wells exhibited measurable decreases in concentrations of the primary parent compounds, which include TeCA, TCE, and chloroform, likely due to natural attenuation processes and possibly due to mass removal in the upgradient direction (e.g., well EHCMW-4D2a) from EHC injection.

Southern Plume

Wells EHCMW-2S2 and PTMW-2Sa both revealed changes in geochemical parameters favorable for reductive dechlorination following injection. Specifically, the DO and ORP readings decreased while ferrous iron, dissolved manganese, carbon dioxide, alkalinity,

Exhibit 9. Summary of polymerase chain reaction analysis

Location	Sample Date	Dehalococcoides spp.	Dehalobacter spp.	BAV1 VC R-Dase	TCE R-Dase	Methanogens
North Plume						
EHCMW-4D2a	6/16/2005	1.26E + 02	8.65E + 03	1.19E + 01	1.05E + 00	2.14E + 05
EHCMW-4D2b	6/16/2005	6.46E + 02	7.13E + 03	2.23E + 00	8.09E-01	2.68E + 05
South Plume						
EHCMW-2S2	6/16/2005	3.38E + 03	3.02E + 04	2.73E-01(J)	3.59E-01(J)	1.12E + 05
PTMW-2SA	6/16/2005	5.33E + 03	5.26E + 03	3.58E-01(J)	8.13E-01	2.06E + 05

Notes: All units are in cells per milliliter.

J = Estimated gene copies below PQL but above LQL.

NA - Not analyzed.

and VFA increased, indicating that the geochemical conditions were suitable for biodegradation to occur (refer to Exhibits 7 and 8).

In well EHCMW-2S2, the concentrations of the parent chlorinated compounds TeCA and TCE gradually increased after the electron-donor injection and later decreased. Concentrations of *cis*-1,2-DCE and *trans*-1,2-DCE, likely daughter products of TeCA and TCE, began increasing approximately six months after the injection (see Exhibit 11). No vinyl chloride was observed above the reporting limits during the first eight months after injection in this well. Vinyl chloride observed during the ninth month continued to increase and later decreased. Chloroform slightly increased during the first six months following injection but later decreased. Methylene chloride, a likely daughter product of chloroform, gradually increased and later decreased, likely indicating hydrogenolysis. Nonchlorinated daughter products (e.g., ethene, ethane, and methane) generated in this well indicate complete dechlorination of the parent chlorinated compounds. TeCA, CF, and TCE decreased over 86 percent, 88 percent, and 95 percent, respectively, after initial increases in their concentrations.

In well PTMW2Sa, the concentrations of TeCA and TCE initially increased and later decreased, similar to that observed in well EHCMW-2S2. Chloroform concentrations began declining gradually following the injection. Methylene chloride, a likely daughter product of chloroform, initially increased and later decreased. No significant production of hydrogenolysis daughter products of TeCA (i.e., 1,1,2-TCA and 1,2-DCA) and TCE (*cis*-1,2-DCE and vinyl chloride) were observed in this well. However, nonchlorinated daughter products (i.e., ethane, ethane, and methane) were observed. The overall decrease in TeCA, CF, and TCE in this well ranged from approximately 96 to 98 percent.

The influence of electron-donor injection appears to have reached well TW-2S, which indicated the presence of TOC at 335 mg/L and 52 mg/L in January 2006 and April 2007, respectively. The TOC was below the detection limit of 1 mg/L in year 2000 (before the pilot study). This information suggests that organic carbon, likely originating from EHC, is reaching well TW-2S as far away as 23 feet from the injection area. The analytical results in 2006 and 2007 indicate that key chlorinated solvents, including TeCA, chloroform, and TCE, decreased over 98 percent from those measured prior to the electron-donor injection. The presence of ethane, ethane, and methane indicates complete dechlorination of these key chlorinated solvents.

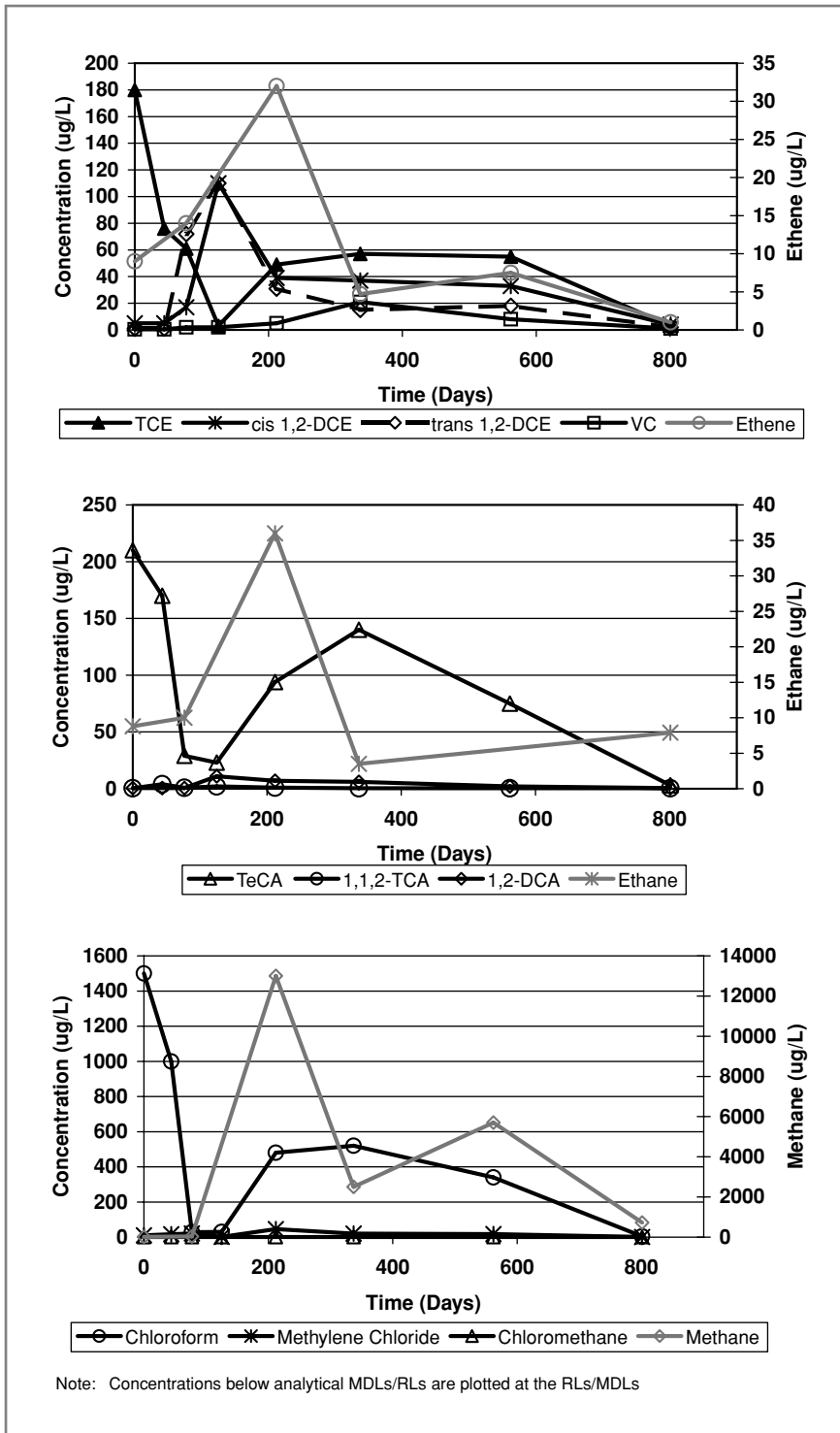


Exhibit 10. Concentrations of ethenes, ethanes, and methanes in well EHCMW-4D2a

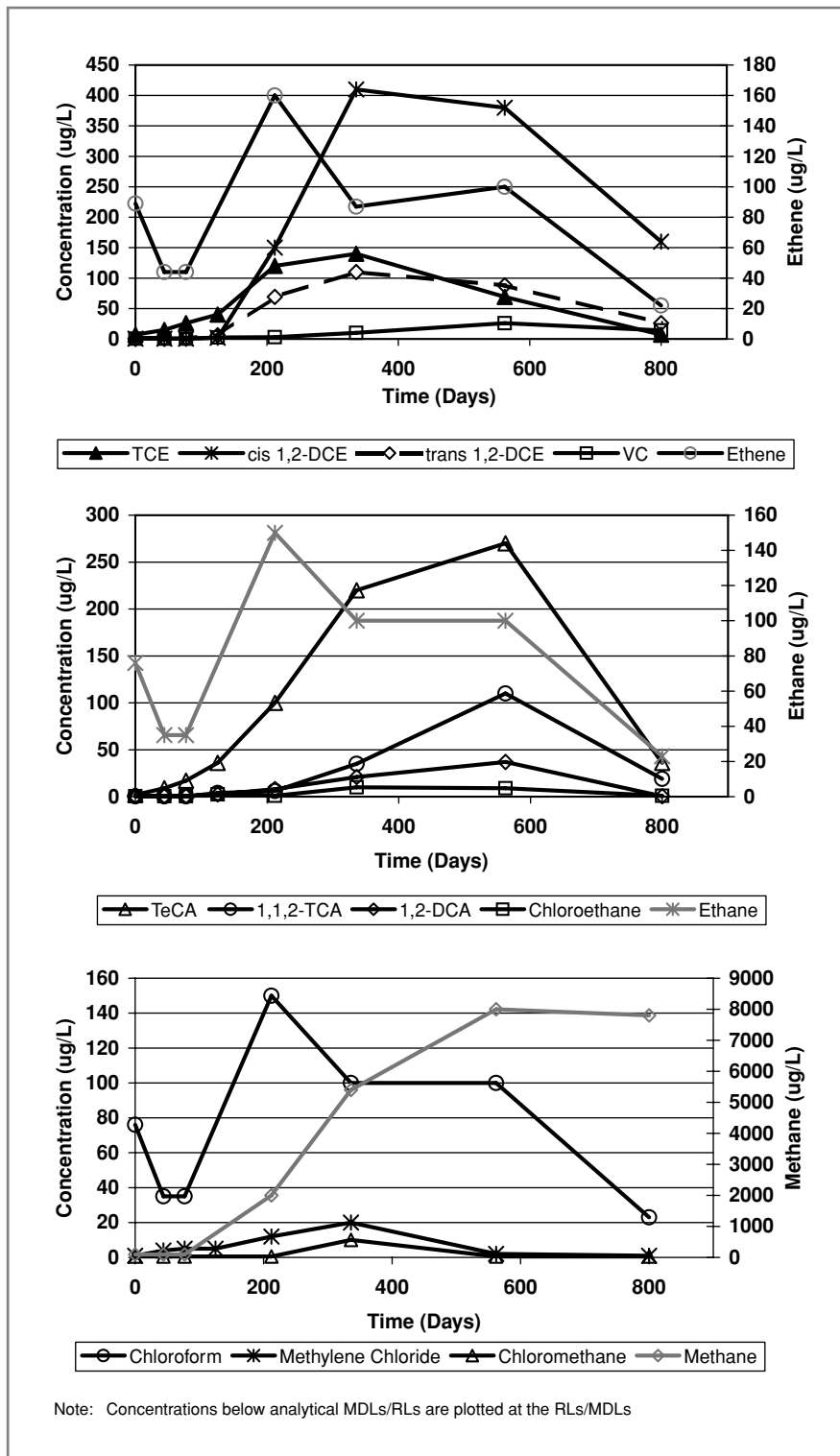


Exhibit 11. Concentrations of ethenes, ethanes, and methanes in well EHCW-2S2

TeCA Degradation Pathway

TeCA can be reductively dechlorinated to 1,1,2-TCA and subsequently to 1,2-DCA via hydrogenolysis (Lorah et al., 2003). TeCA can also be reduced to *cis*-1,2-DCE and *trans*-1,2-DCE through dichloroelimination (Arnold et al., 2002) biologically or via abiotic reduction by ZVI. The analytical data from this study indicate the accumulation of *trans*-1,2-DCE in approximately equal quantities in relation to its isomer *cis*-1,2-DCE in well EHCMW-4D2a (northern plume). Although a hydrogenolysis daughter product of TeCA, 1,1,2-TCA, was observed in well EHCMW-2S2, significant concentrations of *cis*-1,2-DCE and *trans*-1,2-DCE were also observed. Typically, *cis*-1,2-DCE is produced in greater quantities than its *trans* isomer during reductive dechlorination (hydrogenolysis) of TCE (Bouwer, 1994). This information indicates that TeCA is likely being degraded through dichloroelimination either biologically or abiotically (possible dechlorination by ZVI). Other researchers (Griffin et al., 2004; Miller et al., 2005) have recently identified TCE-degrading bacteria that produce *trans*-1,2-DCE in greater quantities than previously expected.

SUMMARY AND CONCLUSIONS

Based on the data collected, the distribution of EHC appears to be in layers or soil planes of weakness in saprolite. Hydraulic fracturing had limited effectiveness in distributing EHC in the PWR zone. However, hydraulic fracturing was successful in distributing EHC in the saprolite zone (southern plume) as it was distributed effectively and its influence reached downgradient monitoring wells. The data also suggest that the largest fractures were observed in the shallower locations, likely due to less overburden pressure.

The electron-donor injection at the site was successful in reducing concentrations of chlorinated solvents. Strong reducing conditions were established in the aquifer immediately after the EHC injection, which appears to be the reducing effect of the ZV Iron Microsphere 200. These conditions are critical for chlorinated solvents dechlorination. The degradation rates were increased as a result of the EHC injection.

Although unexpected, electron-donor influence reached the upgradient well EHCMW-4D2a in the PWR zone (northern plume), where degradation of chlorinated solvents was observed. Monitoring data in 2008 suggest that organic carbon has been depleting over time. The injection of the electron donor significantly enhanced dechlorination of chlorinated solvents in wells PTMW-2Sa and EHCMW2S2 (southern plume). Daughter products of dechlorination of the key chlorinated solvents were observed in these wells. Although TeCA and TCE concentrations initially increased in the southern plume, these VOCs were successfully dechlorinated. The initial increase was likely due to desorption of residual chlorinated solvents in soil by the electron donor (possibly cosolvent effect). The electron donor lasted over 26 months after injection in the saprolite aquifer. The overall reductions in chlorinated compounds in groundwater in the two study areas ranged from 86 percent to 99 percent.

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Matthew J. Zenker, PhD, P.E., is an environmental engineer at AECOM Environment. He received his PhD in civil engineering from North Carolina State University and is a board-certified environmental engineer in the American Academy of Environmental Engineers. His professional experience includes site investigation, natural attenuation, enhanced reductive dechlorination, *in situ* chemical oxidation, and engineering design of groundwater remediation systems.

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