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In Situ Bioremediation of Chlorinated Solvents: A Case Study

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ABSTRACT: In situ bioremediation is ongoing at a former manufacturing facility in Portland, Oregon. Shallow groundwater at the site was impacted by chlorinated solvents, primarily trichloroethene and degradation products *cis*-1,2-dichloroethene and vinyl chloride. In situ chemical oxidation treatment was completed in 2003 via the injection of sodium permanganate in source areas. Subsequent stimulation of in situ biological treatment via reductive dechlorination was evaluated and found to be viable. Emulsified vegetable oil with vitamin B_{12} and other nutrients (EOS® 598 B42) was injected in March and April 2005. Injections were completed over a depth range of 25 to 40 feet (7.6 to 12.2 m) using push probe equipment with an air diaphragm pump. Treatment costs are provided. Regular groundwater monitoring has been conducted following the initiation of the bio-remediation treatment. Nearly two years of post-injection groundwater monitoring data are presented. These data show that the emulsified oil continues to provide increased total organic carbon concentrations in the injection area wells and that concentrations of chlorinated ethenes in the treatment zone have decreased more than 99%. This success has been achieved despite some parameters (e.g., ORP) being outside of optimal ranges for reductive dechlorination. Continued groundwater sampling will be conducted to monitor the longevity of the emulsified oil product and the progress toward achieving site remediation goals.

INTRODUCTION

In situ bioremediation was completed to address chlorinated solvents at a former manufacturing facility in Portland, Oregon. The facility manufactured small appliances such as wall heaters between 1967 and 1973. The site is currently an undeveloped parcel located approximately 1.5 miles (2.4 km) southeast of the Portland International Airport Terminal. Figure 1 is a site plan showing current features, the extent of chlorinated solvents in groundwater prior to biological treatment, and the location of the emulsified oil injections. The site is bordered on the south by the Columbia

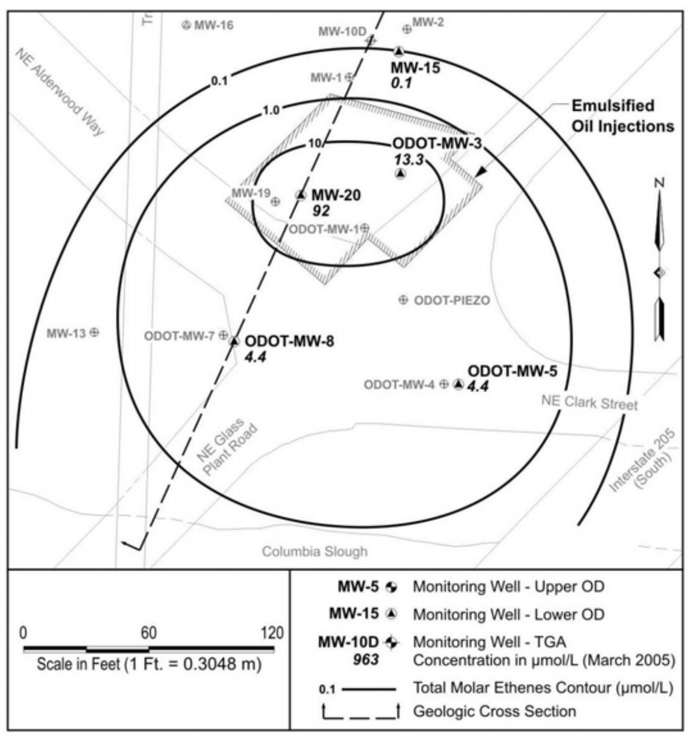

FIGURE 1. Site plan.

Slough, and the site is located within the western expansion area of the City of Portland's Columbia South Shore Wellfield. The wellfield is used as a backup municipal water supply for the Portland Metropolitan area.

BACKGROUND

Geology and Hydrogeology. Figure 2 is a cross section showing the geology in the area of concern along the predominant groundwater flow direction. There are two primary layers of concern: the Overbank Deposit (OD) and the Troutdale Gravel Aquifer (TGA). The OD is primarily composed of silt, with occurrences of laterally continuous fine sand layers (silty sand and fine sand). These layers typically range in thickness from less than 1 foot to 9 feet (0.3 to 2.7 m). The OD is present from the surface to a depth that ranges from 37 to 60 feet (11 to 18 m) across the site, and overlies the TGA. The TGA consists of unconsolidated to partially cemented gravels, with varying amounts of fines. The contact between the OD and TGA is typically marked by 2 to 8 feet (0.6 to 2.4 m) of dense silty gravel acting as a semi-confining layer.

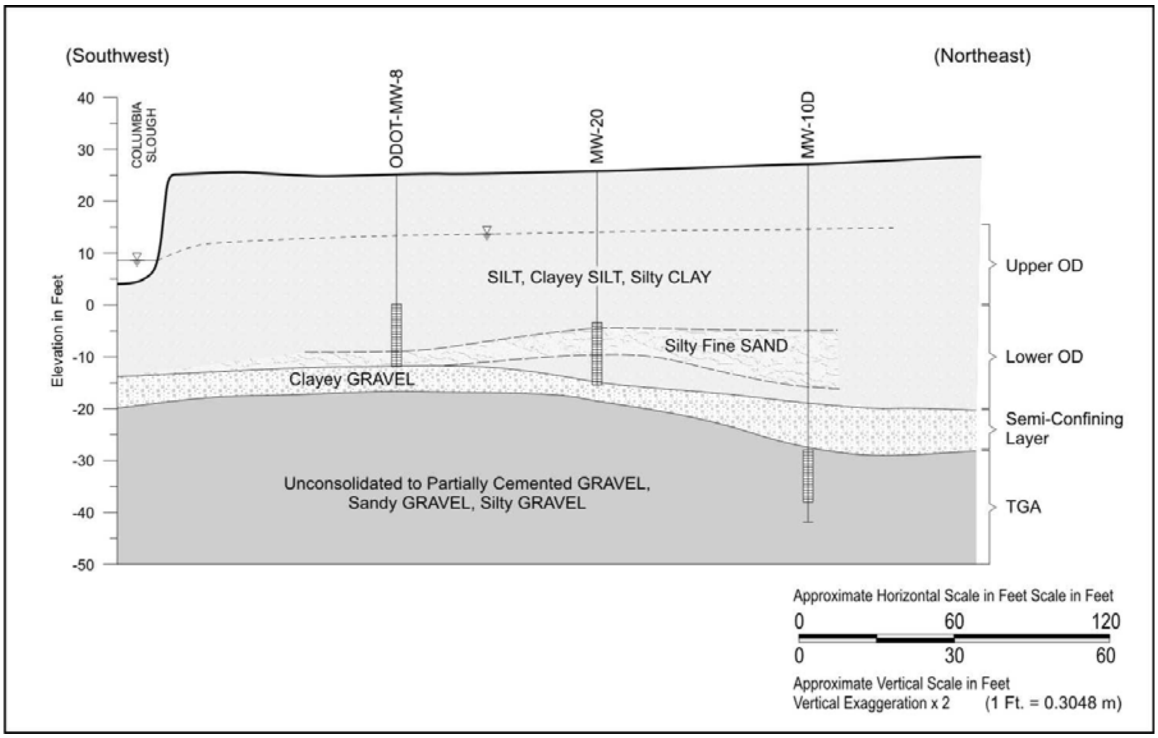

FIGURE 2. Cross section.

Groundwater flow direction in the OD is southerly (toward the Columbia Slough). Flow direction in the upper portion of the OD is locally influenced by the presence of a 72-inch (1.8 m) diameter storm water pipeline so has a westerly component. Groundwater flow in the lower OD (the primary zone of concern) is generally toward the south-southeast and the Columbia Slough. The groundwater flow in the TGA is typically toward the west-southwest.

Prior Interim Remedial Action. Initial remedial action at the site was completed in Spring 2003 and consisted of in situ chemical oxidation using sodium permanganate. The results of this initial remedial action were previously reported (Fernandes, et al., 2004). The permanganate treatment successfully addressed the upper OD, but the lower OD continued to exhibit relatively high concentrations of chlorinated compounds.

Site Investigation Results. Two categories of groundwater contaminants were present in the OD: chlorinated solvent volatile organic compounds (VOCs) (primarily tetra-chloroethene [PCE], trichloroethene [TCE], *cis*-1,2-dichloroethene [cis-DCE], and vinyl chloride [VC]), and fuel-related VOCs (primarily aromatics such as benzene, ethyl-benzene, and xylenes). The highest concentrations of both types of VOCs occurred in the same general area, near the northwestern corner of the former facility building. Chlorinated solvent VOCs have historically been detected at higher concentrations (up to 15 mg/L for *cis*-DCE) than fuel-related VOCs (up to 0.07 mg/L for ethylbenzene). Fuel-related VOC concentrations were below risk-based screening levels in most wells. Chlorinated solvent VOC concentrations were more widespread in the OD, with concentrations 10 to 100 times higher in the lower OD than in the upper OD.

Indicators of biological reductive dechlorination were present following per-manganate treatment: increased concentrations of DCE, VC, and ethene; lower oxidation-reduction potential (ORP) levels; non-detect concentrations for higher electron acceptors nitrate and sulfate; and the presence of *Dehalococcoides ethenogenes* (a species of bacteria shown to be capable of completely dechlorinating PCE to ethene). However, other indicators suggested that conditions were not optimal for complete degradation (e.g., relatively high concentrations of DCE suggested the reductive process may be stalling). Total organic carbon levels were between 5 and 10 mg/L, less than the rec-ommended minimum of 20 mg/L (U.S. Air Force, 2004), indicating that reductive dechlorination may be limited by the lack of a suitable electron donor. A detailed evaluation of the suitability of biological treatment following permanganate treatment was provided in a prior paper (Westersund et al., 2006).

BIOLOGICAL TREATMENT — EMULSIFIED OIL INJECTION

Selected Substrate. Multiple potential substrates were evaluated for use at the site. The evaluated electron donors consisted of sodium lactate, a slow-release lactic acid source, and three varieties of oil products (whole and emulsified oils). The evaluation process considered the delivery mechanisms for each substrate, the expected longevity of the substrate material, and the total installed cost of the injection program over the life of the project. The selected substrate was emulsified oil with vitamin B_{12} and other nutrients (EOS® 598 B42). The material was delivered to the site in drums as a 50 percent oil by volume emulsification.

Injections. Injections were performed in March and April 2005. 825 gallons (3,100 L) of emulsified oil substrate were diluted with water at a 9:1 ratio. Municipal water used for dilution was first dechlorinated with a common home aquarium product. Thirty-one injection locations (Figure 3) were used to cover an area of 9,000 ft^2 (800 m^2). The substrate was injected over the depth range of 25 to 40 feet (7.6 and 12.2 m) bgs.

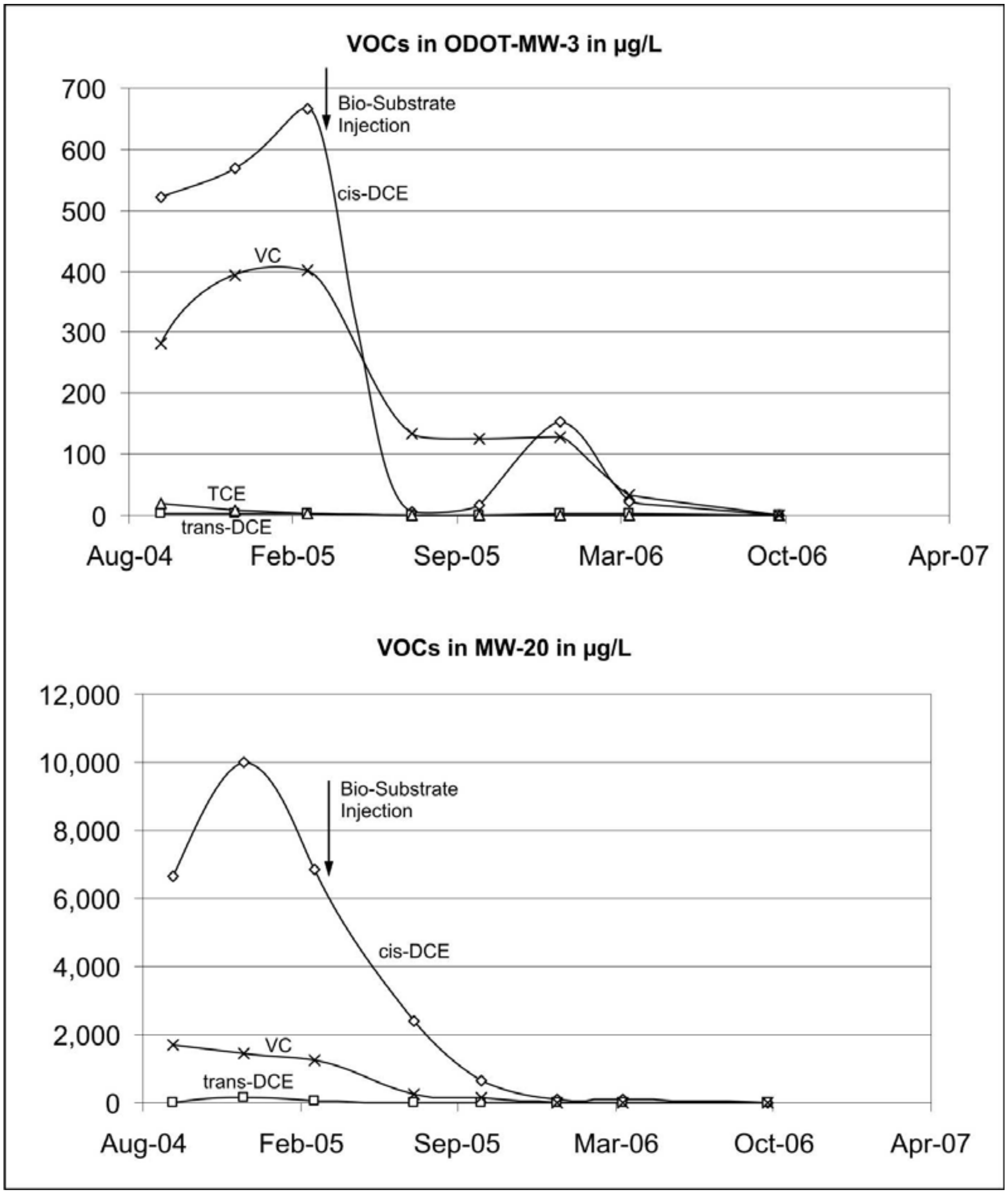

FIGURE 3. VOCs in groundwater treatment area.

Injections were completed from the bottom up, beginning at the top of the silty gravel at the lower OD/TGA contact and injecting 20 gallons (76 L) at each 1-foot (0.3 m) vertical interval. Injection flow rates varied between 1 and 7 gpm (4 and 26 L/min). Push probe equipment was used with an air diaphragm pump and injection tooling designed to provide horizontal as opposed to vertical delivery.

Costs. The costs associated with the emulsified oil injections included design ($14,000), field oversight ($10,000), drilling/injection services and supplies ($13,000), emulsified oil product ($14,000), and reporting ($8,000). The design costs were lower than typical because of the prior pilot study and injections at the site associated with the permanganate treatment.

GROUNDWATER MONITORING RESULTS

Regular groundwater monitoring has been conducted at the site. The most recent sampling event was October 2006, eighteen months after injection of the emulsified oil substrate. Tables 1 and 2 present the groundwater data for selected chlorinated VOCs and other parameters, respectively.

TABLE 1. TCE, DCE, and VC in µg/L (EPA Method 8260)

Sample Location	Sample Date	cis-1,2-DCE	trans-1,2-DCE	TCE	VC	Total Chlorinated Ethenes (µmoles/L)
MW-15	12/8/2004	2.00	<1.00	<1.00	1.44	0.04
	3/7/2005	3.80	<1.00	<1.00	3.86	0.10
	7/12/2005	9.20	<1.00	<1.00	6.12	0.19
	10/4/2005	8.99	<1.00	<1.00	6.33	0.19
	1/10/2006	175	1.33	<1.00	50.1	2.62
	4/3/2006	8.90	<1.00	<1.00	3.32	0.14
	10/2/2006	6.06	<1.00	<1.00	<1.00	0.06
MW-20	12/8/2004	10,000	128	<100	1,470	127.99
	3/7/2005	6,860	74.0	<50.0	1,270	91.85
	7/12/2005	2,410	20.2	<20.0	269	29.37
	10/4/2005	662	20.0	<5.0	148	9.40
	1/9/2006	90.9	12.8	<1.00	17.3	1.35
	4/3/2006	81.4	8.08	<1.00	10.2	1.09
	10/2/2006	2.87	2.88	<1.00	<1.00	0.06
ODOT-MW-3	12/8/2004	570	<5.00	7.40	392	12.21
	3/7/2005	666	<5.00	<5.00	401	13.29
	7/12/2005	5.65	<1.00	<1.00	133	2.19
	10/3/2005	16.0	<1.00	<1.00	125	2.17
	1/9/2006	152	4.09	<1.00	129	3.67
	4/3/2006	22.1	2.32	<1.00	34.3	0.80
	10/2/2006	<1.00	<1.00	<1.00	<1.00	ND
ODOT-MW-5	12/8/2004	154	2.23	1.75	6.66	1.73
	3/10/2005	369	5.68	63.9	3.42	4.41
	7/11/2005	357	5.50	65.0	2.64	4.28
	10/4/2005	489	6.20	23.6	5.55	5.38
	1/10/2006	399	5.05	43.6	<5.00	4.50
	4/3/2006	359	6.66	62.0	2.46	4.28
	10/2/2006	391	6.28	30.2	2.48	4.37
ODOT-MW-8	12/8/2004	291	2.56	<2.00	26.8	3.46
	3/10/2005	358	2.44	<2.00	42.8	4.40
	7/11/2005	222	<2.00	<2.00	17.0	2.56
	10/4/2005	275	2.96	<2.00	25.8	3.28
	1/10/2006	252	2.00	<2.00	33.4	3.15
	4/3/2006	237	2.28	<2.00	25.5	2.88
	10/2/2006	230	3.06	<2.00	11.9	2.59

TABLE 2. Other parameters.

Sample Number	Sample Date	Field Parameters				Alkalinity EPA 310.1 (mg/L)	Biological				Substrate		
							Dehalococcoides Ethenogenes						
		Temp (°C)	pH	Elect. Cond. (µMHOs)	Oxygen Reduction Potential (mV)		Plate Count (cells/bead or cells/mL prior to Mar 2005)	% having the BAV1 gene (VC reductase)	% having the TCE reductase gene	Total Heterotrophic Plate Count SM 9215B (CFU/mL)	Total Organic Carbon EPA 415.1 (mg/L)	Total Volatile Fatty Acids Microseeps AM23 (mg/L)	Dissolved Hydrogen AM20GAX (nM)
MW-15	2/25/2004	--	--	--	--	--	44,600	--	--	--	--	--	1.2
	6/8/2004	--	--	--	--	--	7,810	54.8%	1.2%	3,380	9.31	--	--
	9/7/2004	--	--	--	--	--	9,170	0.3%	0.0%	395	4.89	--	--
	12/8/2004	--	--	--	--	--	593	24.0%	24.0%	211	3.69	--	--
	3/7/2005	14.5	6.54	464	-86	308	2,180	17.0%	0.0%	7,210	1.53	--	--
	6/6/2005	--	--	--	--	--	4,730	3.4%	0.01%	--	--	--	--
	7/12/2005	14.8	6.36	438	-77	265	--	--	--	780	2.28	--	--
	10/4/2005	14.1	6.61	511	-8.7	328	1,660	~100%	~100%	1,650	2.53	--	--
	1/10/2006	13.5	6.66	495	-94.9	--	--	--	--	--	--	0.141	--
	4/3/2006	13.6	6.78	400	-81	--	--	--	--	--	--	--	--
	10/2/2006	15.1	7.19	425	-16	--	--	--	--	--	--	--	--
MW-20	2/25/2004	--	--	--	--	--	26,700	--	--	--	--	--	0.93
	6/8/2004	--	--	--	--	--	274,000	0.8%	3.3%	465	5.30	--	--
	9/8/2004	--	--	--	--	--	236,000	0.0%	0.0%	123	6.59	--	--
	12/8/2004	--	--	--	--	--	4,210	1.0%	1.0%	2,300	7.14	--	--
	3/7/2005	14.5	6.73	866	14.6	680	90,000	0.05%	0.06%	1,300	5.25	--	--
	6/6/2005	--	--	--	--	--	16,100	0.21%	0.16%	--	--	--	--
	7/12/2005	14.6	5.54	1,122	-40	592	--	--	--	5,300	1,000	--	--
	10/4/2005	14.4	5.83	1,232	32.6	619	8,680	6.80%	18.9%	12,500	812	--	--
	1/9/2006	13.5	5.84	1,225	-58.8	--	--	--	--	--	827	1616	--
	4/3/2006	13.7	6.01	998	-49	--	--	--	--	--	--	--	--
	10/2/2006	15.6	7.48	1,602	-121	--	--	--	--	--	--	--	--
ODOT-MW-3	2/25/2004	--	--	--	--	--	29,800	--	--	--	--	--	1.4
	3/7/2005	15.3	6.17	935	-69	648	20,500	61.0%	1.0%	2,060	9.51	--	--
	6/6/2005	--	--	--	--	--	210,000	19.4%	0.0%	--	--	--	--
	7/12/2005	14.7	6.3	1,032	-77	647	--	--	--	1,270	29.6	--	--
	10/3/2005	16.1	6.36	1,199	-36	763	49,700	23.5%	4.4%	95.0	15.1	--	--
	1/9/2006	14.0	6.42	1,085	-89.8	--	--	--	--	--	44.7	86.4	--
	4/3/2006	13.8	6.45	1,114	-77	--	--	--	--	--	--	--	--
	10/2/2006	15.6	7.66	1,456	-106	--	--	--	--	--	--	--	--
ODOT-MW-5	3/10/2005	14.4	6.43	353	-75	--	--	--	--	--	2.31	--	--
	7/11/2005	18.5	6.12	379	-64	--	--	--	--	--	2.58	--	--
	10/4/2005	14.2	6.35	349	9.1	--	--	--	--	--	5.79	--	--
	1/10/2006	13.7	6.59	350	-80.9	--	--	--	--	--	3.08	--	--
	4/3/2006	13.5	6.55	338	-69	--	--	--	--	--	--	--	--
	10/2/2006	15.6	7.27	426	-88	--	--	--	--	--	--	--	--
ODOT-MW-8	3/10/2005	14.5	6.41	488	-94	--	--	--	--	--	3.04	--	--
	7/11/2005	15.1	6.26	443	-78	--	--	--	--	--	2.57	--	--
	10/4/2005	14.2	6.42	420	-6.2	--	--	--	--	--	2.77	--	--
	1/10/2006	13.7	6.69	427	-104.0	--	--	--	--	--	3.60	0.074	--
	4/3/2006	13.9	6.68	432	-97	--	--	--	--	--	--	--	--
	10/2/2006	15.6	7.37	435	-60	--	--	--	--	--	--	--	--

TABLE 2. Other parameters continued.

Sample Number	Sample Date	Limiting Nutrients		Competing Electron Acceptors							Fermentation		End Products		
		Phosphorus EPA 365.1 (mg/L)	Ammonia Nitrogen EPA 350.1 (µg/L)	Dissolved Oxygen (mg/L)	Nitrate-Nitrogen EPA 300.0 (µg/L)	Nitrite-Nitrogen EPA 300.0 (µg/L)	Total Manganese EPA 6020 (µg/L)	Ferrous Iron SM 3500 (µg/L)	Sulfate EPA 300.0 (µg/L)	Sulfide EPA 376.2 (µg/L)	Methane GC/FID (ug/L)	Carbon Dioxide SM 4500-C (ug/L)	Chloride EPA 300.0 (ug/L)	Ethene GC/FID (ug/L)	Ethane GC/FID (ug/L)
MW-15	2/25/2004	0.342	--	--	--	--	--	--	--	--	--	--	--	--	--
	6/8/2004	0.126	--	--	--	--	--	--	--	--	2,220	--	--	<10.0	--
	9/7/2004	0.214	--	--	--	--	--	--	--	--	3,650	--	--	<10.0	--
	12/8/2004	0.301	--	--	<100	--	4,820	--	<1,000	--	2,370	--	--	<10.0	<10.0
	3/7/2005	0.201	717	0.63	<100	<100	2,740	4,000	<1,000	<200	2,590	57,200	4,740	<10.0	12.4
	6/6/2005	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	7/12/2005	0.134	596	0.53	<100	<100	2,550	3,600	<1,000	<200	1,910	36,900	<500	<10.0	11.0
	10/4/2005	<0.100	671	0.38	<100	--	--	3,600	<1,000	--	1,650	36,200	--	<10.0	<10.0
	1/10/2006	--	--	0.45	--	--	--	--	--	--	--	--	--	--	--
	4/3/2006	--	--	0.94	--	--	--	--	--	--	--	--	--	--	--
	10/2/2006	--	--	0.06	--	--	--	--	--	--	--	--	--	--	--
MW-20	2/25/2004	<0.02	--	--	--	--	--	--	--	--	--	--	--	--	--
	6/8/2004	0.0327	--	--	--	--	--	--	--	--	377	--	--	840	--
	9/8/2004	<0.0200	--	--	--	--	--	--	--	--	611	--	--	2,210	<10.0
	12/8/2004	<0.0200	--	--	<100	--	10,200	--	12,000	--	688	--	--	129	<10.0
	3/7/2005	0.0473	1,510	0.80	<100	<100	20,800	<1,000	<1,000	<200	1,150	35,200	27,500	200	<10.0
	6/6/2005	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	7/12/2005	0.398	3,600	0.13	<100	<1,000	157,000	3,200	<1,000	<400	355	382,000	22,200	2,060	76.3
	10/4/2005	0.216	3,300	0.49	<100	--	117,000	8,400	<1,000	--	421	292,000	--	2,450	119
	1/9/2006	--	--	0.61	--	--	114,000	--	--	--	9,020	--	--	2,730	227
	4/3/2006	--	--	1.78	--	--	105,000	--	--	--	--	--	--	--	--
	10/2/2006	--	--	0.04	--	--	80,200	--	--	--	--	--	--	--	--
ODOT-MW-3	2/25/2004	0.301	--	--	--	--	1,060	--	--	--	--	--	--	--	--
	3/7/2005	0.314	809	0.89	<100	<1,000	1,100	5,000	<1,000	<200	2,620	208,000	21,200	158	<10.0
	6/6/2005	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	7/12/2005	0.134	522	0.24	<100	<100	1,550	4,400	<1,000	<400	7,460	171,000	22,300	<10.0	141
	10/3/2005	0.0835	727	0.92	<100	--	1,570	5,200	<1,000	--	5,610	139,000	--	228	118
	1/9/2006	--	--	1.03	--	--	1,560	--	--	--	13,900	--	--	230	178
	4/3/2006	--	--	2.27	--	--	1,370	--	--	--	--	--	--	--	--
	10/2/2006	--	--	0.06	--	--	1,380	--	--	--	--	--	--	--	--
ODOT-MW-5	3/10/2005	--	--	0.87	--	--	987	--	--	--	--	--	--	--	--
	7/11/2005	--	--	0.72	--	--	765	--	--	--	--	--	--	--	--
	10/4/2005	--	--	0.31	--	--	664	--	--	--	--	--	--	--	--
	1/10/2006	--	--	0.43	--	--	644	--	--	--	--	--	--	--	--
	4/3/2006	--	--	0.92	--	--	612	--	--	--	--	--	--	--	--
	10/2/2006	--	--	0.08	--	--	602	--	--	--	--	--	--	--	--
ODOT-MW-8	3/10/2005	--	--	0.61	--	--	--	--	--	--	--	--	--	--	--
	7/11/2005	--	--	1.00	--	--	--	--	--	--	--	--	--	--	--
	10/4/2005	--	--	0.64	--	--	--	--	--	--	--	--	--	--	--
	1/10/2006	--	--	0.43	--	--	--	--	--	--	2,840	--	--	<10.0	<10.0
	4/3/2006	--	--	1.07	--	--	--	--	--	--	--	--	--	--	--
	10/2/2006	--	--	0.42	--	--	--	--	--	--	--	--	--	--	--

RESULTS AND DISCUSSION

VOCs Reduced More Than 99%. Figure 3 shows plots of chlorinated VOC concentrations for the past two years. Chlorinated VOC concentrations in the two lower OD groundwater monitoring wells in the source area (MW-20 and ODOT-MW-3) have decreased by more than 99% on a total molar basis since the emulsified oil injections. Concentrations of target VOCs in the area treated are approaching or below detection limits. Nearly two years after injection, the emulsified oil continues to provide a sufficient electron donor supply to support reductive dechlorination.

Reductive Dechlorination Indicator Parameters. A number of parameters have been identified as indicators for suitability of a site for enhanced reductive dechlorination (US Air Force, 2004). Table 3 lists common parameters together with optimal ranges and results from this site for the two wells in the source area. Since injection of the emulsified oil, none of the parameters stayed consistently within the optimal range for both wells. Yet the concentrations of target chlorinated VOCs have fallen to near or below detection limits. This suggests that these parameters should be used cautiously to discount reductive dechlorination for a site. Ultimately, the reduction in concentrations of target chlorinated VOCs and the generation of ethene are the best indicators of successful reductive dechlorination. Consequently, monitoring has been scaled back to focus on VOCs. Only in the event that monitoring of VOCs suggests that complete dechlorination is not occurring will additional testing be conducted in an effort to identify the cause.

TABLE 3. Comparison of indicator parameter results to optimum ranges.

Sample Location	pH	Reduction Potential (mV)	Total Organic Carbon (mg/L)	Dissolved Oxygen (mg/L)	Methane (µg/L)	Ethene (µg/L)
Optimal Range	**6 to 9**	**-130 to -300**	**> 20**	**< 1**	**< 6,000**	**> 0**
MW-20	5.5 to 7.5	33 to -121	810 to 1000	0.04 to 1.8	350 to 9020	2000 to 2730
ODOT-MW-3	6.3 to 7.7	-36 to -106	15 to 45	0.06 to 2.3	5600 to 13900	<10 to 230

Metals Not a Concern. With the creation of reducing conditions, groundwater metals concentrations were a potential concern. Groundwater monitoring identified an increase in manganese and ferrous iron concentrations in MW-20 after emulsified oil injections. Manganese concentrations are higher, probably due to the prior permanganate treatment. The elevated metals concentrations have been detected only within the middle of the treatment area (primarily MW-20), and are not migrating from the source area. The concentration of manganese has returned to baseline levels in all wells except MW-20 where the concentration of 80 mg/L detected in October 2006 is about four times the baseline level. These results support the general conclusion that secondary water quality impacts will not be a problem at reductive dechlorination treatment sites because the elevated metals concentrations will quickly return to baseline levels once the reducing conditions are eliminated (either over time as the substrate is exhausted or over distance as groundwater migrates away from the treatment zone).

SUMMARY AND CONCLUSIONS

Emulsified oil was injected into the subsurface to address chlorinated solvents by reductive dechlorination. Injections were completed in March/April 2005. Two years after the injection, the following conclusions are drawn:

- In the treated area, concentrations of target compounds (TCE, DCE, and VC) have decreased by more than 99%.
- The emulsified oil continues to support reductive dechlorination.
- Despite conditions outside recommended optimal ranges, reductive dechlorination has been very effective.
- Increased metals concentrations as a result of the induced reducing conditions are not a long-term concern.

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