Case Study of the Biotreatment of a Dilute Chlorinated Solvent Plume in an Acidic Aerobic Aquifer

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ABSTRACT: In situ bioremediation was selected in the Record of Decision as the remedial technology for a nearly 30-acre plume of dilute chlorinated solvents (principally TCE and 1,1-DCE) for a Superfund site located in central New Jersey. Implementation of the remedy at full scale began in late 2010. In situ reductive dechlorination treatment, including bioagumentation, has been conducted at the site over the last seven years. Treatment has consisted principally of injection of electron donor and basic neutralizer solution, followed by bioaugmentation solution injection after treated aquifer areas indicated anaerobic conditions. Injections were conducted in multi-level injection wells, to maintain control over the vertical intervals of amendment delivery. The areal coverage of the plume has been reduced over 80 percent and the contaminant mass has been reduced by roughly 70 percent through seven years of treatment. Lessons learned from this project include the need for bioaugment in the acidic aquifer, an efficient manner of well construction and amendment injection using multi-screen single-casing injection wells and packer systems, differences in longevity of the electron donor amendment versus the bicarbonate, and the need for varied injection techniques in limited areas of the site to attain treatment in some of the less permeable areas, using direct injection and a horizontal injection well.

INTRODUCTION

The in situ biodegradation of chlorinated solvents by enhanced reductive dechlorination (ERD) has been in practice for nearly two decades. Enhanced in situ biodegradation of chlorinated solvents is typically implemented by injecting one or more carbon sources into the impacted aquifer in some configuration to contain a plume at its downgradient end or to treat the plume body using injection points in a grid or barrier line configuration. Distribution of the injected amendment may be enhanced by using groundwater recirculation in concert with injection or by other injection point techniques such as high pressure or hydraulic fracturing injection.

This case study includes the results of six years of treatment of a large, dilute concentration plume, containing primary contaminants trichloroethylene (TCE) and 1,1-dichloroethylene (DCE). Over the six year period, several different amendment injection approaches were employed to improve amendment distribution or to inject into otherwise inaccessible plume areas, while at the same time striving for cost reduction for complete plume treatment.

BACKGROUND

Site Description. Groundwater contamination at this Superfund site originated from shallow surface or drain pipe discharges in 1980 at a building where circuit board manufacturing operations were occurring. The plume emanating from this source contains TCE and 1,1-DCE as the principal contaminants. The TCE is one of the original chemicals released at the site, while the 1,1-DCE presence is attributable to it being a breakdown product from the originally released 1,1,1-trichlorethane (TCA). At the time plume remediation began in 2010, the plume covered an area of 29 acres, and the leading edge of the plume extended 2,100 feet southeast and downgradient from the suspected point source (EPA 2005). Figure 1 depicts the TCE plume in mid-2011, just after the start of remedial activities in the fall of 2010.

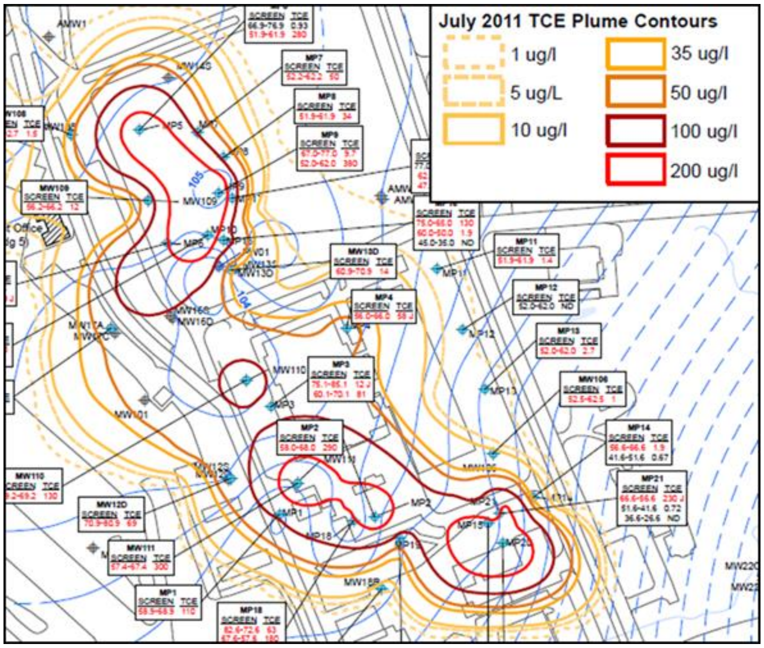

FIGURE 1. TCE plume extent at beginning of the in situ treatment process

The hydrogeology at the MDI site consists of highly stratified sands, silts, and clays known as the Cohansey (upper) and Kirkwood (lower) formations, which sits on top of the Manasquan formation, being the uppermost aquitard at the site (Ecology and Environment 2007). The Manasquan aquitard lies at a depth of 100 to 150 feet below ground surface (bgs) at the site. The occurrence of fine sands, as well as the amount of silts and clays interbedded with the sand, increases with depth in the aquifer. The depth of the water table below the ground surface varies from approximately 40 feet bgs at the northwest portion of the site, to less than 5 feet bgs in the far southeastern portion of the plume. A depiction of the plume in cross-section is presented in Figure 2.

Plume Extent and Aquifer Conditions. The plume has three distinct lobes or areas of higher concentration of TCE, as depicted in Figure 1 These are referred to as Hot Spot #1, located in the northern area of the plume, Hot Spot #2, located principally on the western side of the Zodiac Aerospace (formerly Air Cruisers) building, and Hot Spot #3 on the south and southwest side of the Zodiac Aerospace building. The maximum concentration of TCE detected prior to remediation was approximately 600 micrograms per liter (µg/L), while the highest concentration of 1,1-DCE detected prior to remediation was less than 200 µg/L. The 1,1-DCE contamination distribution spatially mimics the TCE contamination across the site.

The baseline (undisturbed) condition of the groundwater at the MDI site indicates pH levels in the range of 4 to 5 standard units (SU), aerobic dissolved oxygen (DO) levels in the range of 2 to 5 milligrams per liter (mg/L), and positive oxidation reduction potential (ORP) levels, except in deeper portions of the aquifer where slightly negative ORP levels are more commonly observed. The aquifer pH is significantly lower than the range of 6 to 8 SU considered necessary for effective ERD. Therefore, the amendment mixture chosen for injection needs to have a basic component to neutralize the pH upwards to the appropriate range. Additionally, an electron donor source added to drive the ERD process will result in lowering of the DO and ORP to respective appropriate levels (DO less than 1.0 mg/L and ORP less than -50 millivolts (mV)).

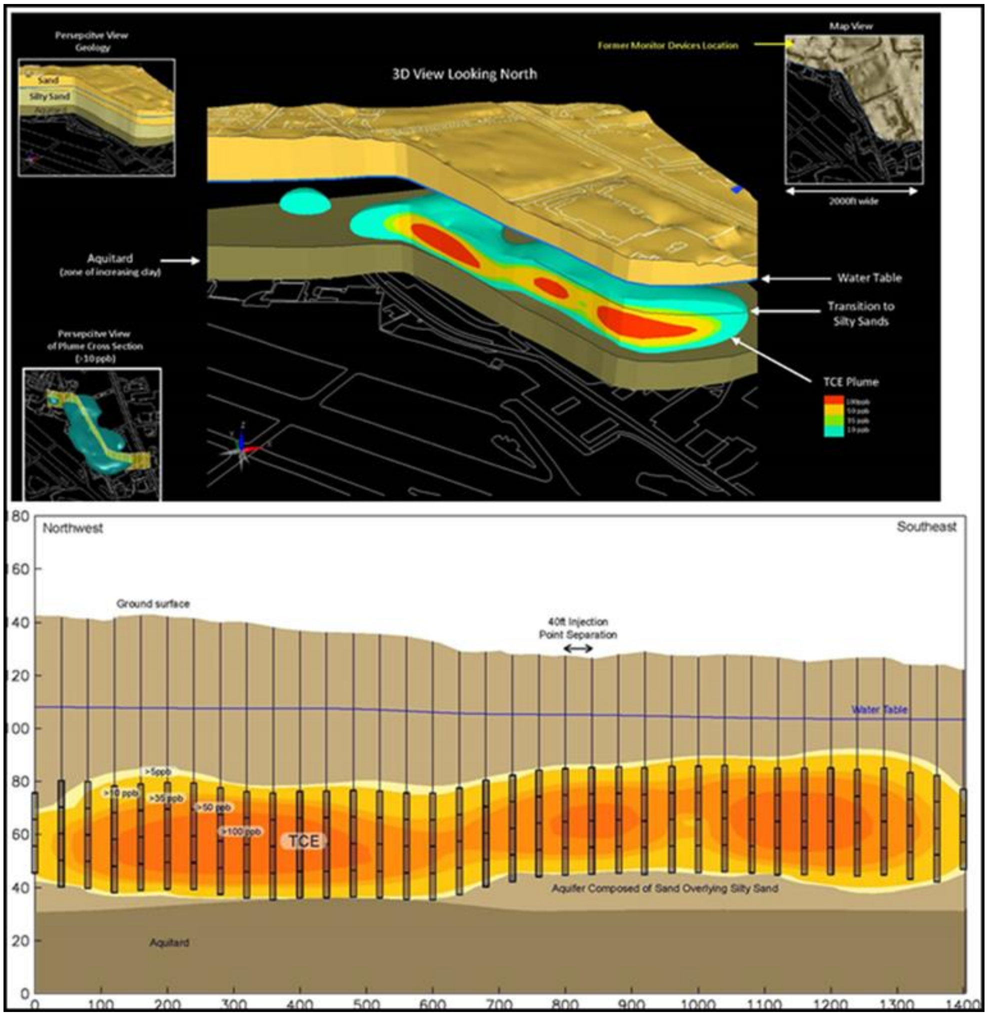

FIGURE 2. TCE plume three-dimensional and cross-section view

MATERIALS AND METHODS

The approach to total plume treatment at the MDI site was to treat high concentration areas of the plume first, with subsequent treatment effort directed at moderate to lower concentration areas. This top-down treatment approach was continued until all areas of TCE contamination of 35 µg/L or greater was treated. The first two years of treatment effort addressed those areas of the plume with concentrations greater than 100 µg/L TCE. Subsequent annual treatment campaigns targeted periphery plume areas down to 50 and 35 µg/L. After the initial amendment injection was completed in these three areas, subsequent treatment campaigns concentrated on plume areas indicating treatment resistance, contaminant rebound, or newly identified of TCE contamination discovered subsequent to the plume definition sampling effort of spring 2010. The primary new areas of TCE contamination that have been identified since the remedial program was started in 2010 include the western portion of HS #2, the southeast portion of HS #3 (immediately south of Zodiac Aerospace building), the leading edge of the HS #3 plume located east of Route 34, and the leading edge of the HS #1 plume area. Table 1 presents the history of the annual amendment injection events at the MDI site, which typically occurred each year in the late summer to mid-fall time frame. The approach employed in each injection campaign was to mix large volumes of amendment and then inject that amendment into a number of injection wells simultaneously via appropriate distribution lines. The objective was to achieve a high rate of amendment delivery and thus minimize or shorten total amendment delivery duration.

Table 1. Overview of MDI Treatment Program

Program Year	Treatment Target	Number of Injection Locations (Wells)	Volume of Amendment Injected (gallons)
1 (2010)	100 ppb TCE isocontour	50	250,000
2 (2011)	100 ppb TCE isocontour	119	2,000,000
3 (2012)	50 ppb TCE isocontour	86	1,050,000
4 (2013)	35 ppb TCE isocontour	105	1,404,000
5 (2014)	Specific targets	36	560,000
6 (2015)	Specific targets	51†	544,000
7 (2016)	Specific targets	8	101,000

†--Twenty of these locations were direction injection points instead of installed injection wells.

Amendment Injection. Emulsified vegetable oil (EVO) was chosen as the electron donor (carbon source) for implementation of ERD at the MDI site, based upon favorable microcosm test results conducted during the remedy planning stage (SAIC 2010). Additionally, the groundwater quality data indicated a requirement for addition of a basic chemical to raise the aquifer pH, since the optimal range for reductive dechlorination bacteria activity is 6 to 8 SU, and the baseline groundwater is in the range of 4 to 5 SU. Both carbonate and bicarbonate were considered for this neutralization of the groundwater pH. Bicarbonate was chosen as the most appropriate chemical for this task, after initial trials with both chemicals at the beginning of the first annual injection campaign indicated it was easier to work with. The final target concentrations of EVO and bicarbonate in the amendment injectate solution were 3.4 weight percent and 8.3 weight percent, respectively. These levels correspond to an amendment solution recipe of 4,000 lbs of bicarbonate and 200 gallons of EVO mixed with 5,800 gallons of water per each 10-foot injection well screen.

Injections of a mixed bacteria bioaugment culture containing *Dehalococcoides* (DHC) microorganisms were necessary for completion of the dehalogenation of TCE. Injections of bioaugment culture were performed separately from amendment injections, only after groundwater field parameter data indicated aquifer conditions were suitable for bioaugment survival and activity. The conditional requirements for bioaugmentation are pH above 6.0 SU, ORP less than -50 mV, and DO less than 1 mg/L.

Injection Well Design. Project personnel chose semi-permanent injection wells (IWs) as the preferred method for amendment solution delivery to the aquifer for several reasons, including the ability to administer repeat injections without having a drilling or injection rig mobilized for each event. Because of the size of the plume area requiring treatment (all areas with TCE above 35 μg/L), the use of pre-installed injection wells was favored over any direct injection technique that would require the presence of a drill rig at each injection location for some or all of the time of amendment delivery. The standard approach for locating injection wells for each year's target treatment area was placement of wells on a 40-foot square grid, with grid position adjustments made to accommodate surface, overhead electrical, road, permanent building, or underground utility obstructions. The injection locations across the entire site are depicted in Figure 3. At this spacing, the 5,800 gallons of amendment injection per 10-foot screen was estimated to represent a 25 percent pore volume replacement when injected into the aquifer. To accomplish the vertical limits of the treatment target, multiple injection well screens were used at each injection point. Over the majority of the site, the plume thickness that required treatment was determined to be 25 to 45 feet, based upon data from push-boring investigative probes. Multiple 10-foot screens stacked vertically across the complete plume thickness were used to protect against uneven vertical distribution of amendment that could occur with a single screen delivering amendment to a vertically-heterogeneous lithology. In the first several years of the injection program, this was implemented using stacked screen injection wells constructed with three separate casings installed in a single large borehole drilled by hollow-stem auger. After several years of implementing this design and encountering problems with wellhead subsidence and lower injection flow rates, a revised design was implemented using all three screens constructed situated on a single casing, and installed by the roto-sonic drilling method.

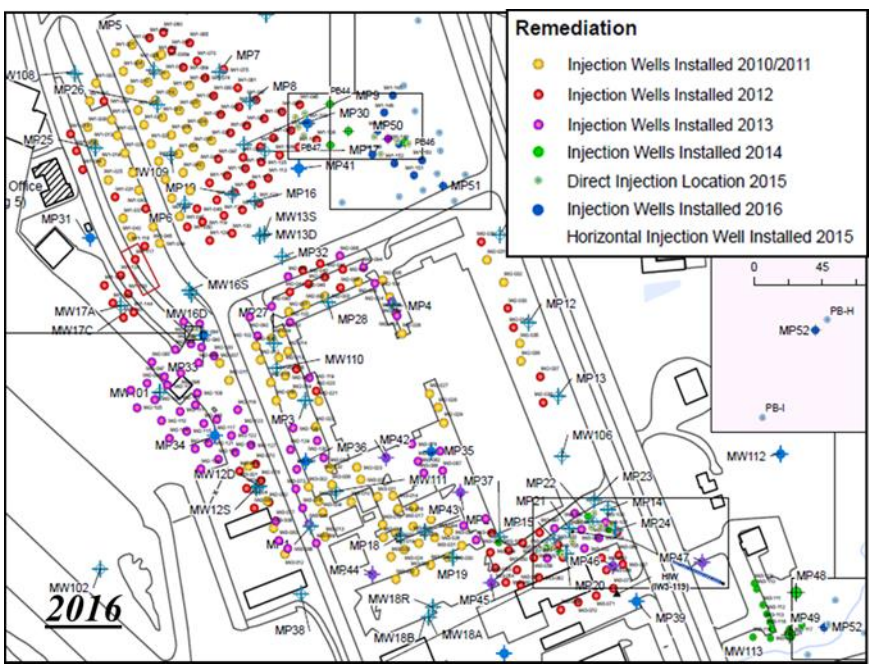

FIGURE 3. TCE plume injection well and direct inject locations through 2016

A couple of other amendment delivery approaches were utilized in special circumstances in the last couple of injection campaigns (2015 and 2016). Specifically, direct injection with limited pneumatic fracturing was used in isolated areas of HS #1 and HS #3 during the 2015 injection campaign, where significantly lower injection rates were previously indicated. Additionally, a single horizontal well with a 120-foot long screen was installed in 2015 underneath Route 34, in order to access and treat plume contamination present underneath that roadway, where vertical injection well placement was not possible.

The progress of groundwater treatment for TCE and 1,1-DCE has been assessed throughout the treatment period using a performance monitoring program consisting of groundwater sampling monitoring points (MPs) and monitoring wells and field parameter data collection ((SAIC 2011). The groundwater parameters sampled included volatile organic compounds (VOCs) and total organic carbon (TOC). The field parameters have included pH, DO, ORP, and specific conductivity (SC).

RESULTS AND DISCUSSION

Contaminant Degradation. TCE and 1,1-DCE were degraded in the plume after both the amendment (EVO and bicarbonate solution) and bioaugment injections were completed. Contaminant concentration declines were observed at a few locations prior to bioaugment injection, however they were attributed to seasonal concentration fluctuations, which were also observed in some cases prior to any in situ treatment. This information, along with the low pH and aerobic DO levels of the undisturbed aquifer, limited detections of ERD-associated daughter products cis-1,2-DCE and VC in the pre-treated plume, and the low results of DHC samples analyzed during the 2010 initial characterization, led to the conclusion that there was no substantial native population of DHC or other dehalogenating bacteria present at the site. As indicated in Table 1, amendment injections occurred at the site as annual campaigns starting in 2010 and continuing to 2016. The first bioaugment injection occurred in late 2011 in the form of a pilot test at six isolated areas, and this was followed by the first plume-wide bioaugment injections in mid-2012. Significant TCE concentration declines were commonly observed at most all monitoring locations where amendment was injected, there was evidence (altered DO, ORP, and specific conductivity) of sufficient amendment distribution from IWs to the MPs, and where bioaugment culture was subsequently injected. However, the lag time between bioaugment injection (always after amendment injection) and the observance of TCE degradation ranged from slightly less than a month to greater than six months. This variation of the lag time was believed to be due to several factors, principally the reliance on bioaugment distribution by advection, which varied due to heterogeneous lithology, and the different distances and orientations between injection wells and adjacent MPs. Figures 1 and 4 depict the TCE contaminant decline over the six years of treatment. Quantitatively, the TCE plume for the period 2010 to 2015 has been reduced 70 percent at the 35 ppb isocontour, 77% reduced at the 50 ppb isocontour, 80 percent reduced at the 100 ppb isocontour and 100 percent reduced (elimination) at the 500 ppb isocontour. Overall, the TCE plume area has been reduced by a minimum of 48 percent (10 ppb isocontour) plume-wide treatment began in 2010.

Some of the MP locations indicated rebound of TCE levels after treatment was observed. Rebound was attributed to either desorption of adsorbed contaminant mass after initial treatment of dissolved mass, or advection of higher concentrations from up gradient areas not well treated, due to inconsistent amendment distribution. Not surprisingly, the instances of highest concentration rebound occurred among wells that originally exhibited the highest TCE levels across the site, thus consistent with the explanation of desorption of adsorbed contamination, after treatment of dissolved contaminant. Many of these locations were targeted for re-treatment in 2014 and subsequent years, after all plume areas down to 35 µg/L TCE had undergone treatment for the first time in the 2013 annual campaign.

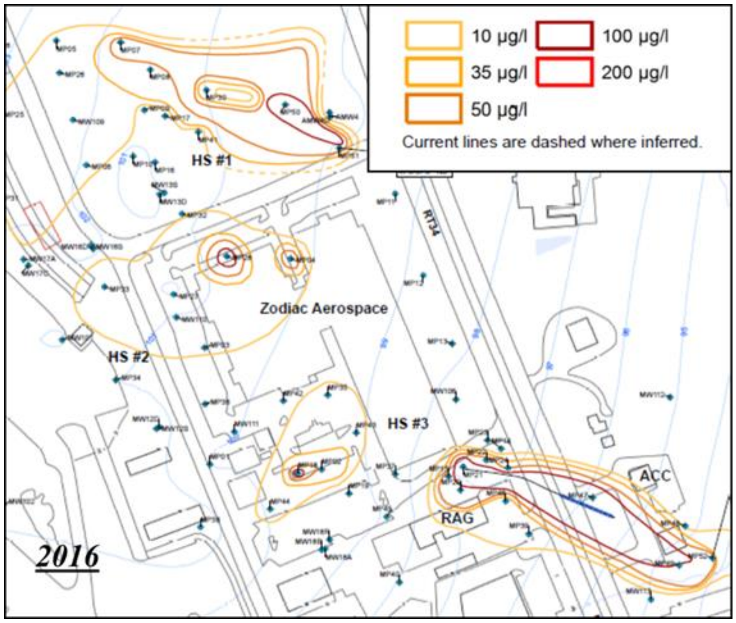

FIGURE 4. TCE plume extent at after six years of the in situ treatment process

Amendment Injections. The field parameter data collected after each amendment injection campaign served as the primary source of data for assessing effectiveness of amendment distribution and amendment persistence. Effective distribution of the electron donor amendment (EVO) resulted in DO decreases to below 1 mg/L and ORP decreases to negative values in the range -50 mV to -200 mV. Distribution of the sodium bicarbonate resulted in increases in aquifer groundwater pH, typically from initial pH values of 4 to 5 SU up to 6 to 7 SU. Additionally, effective sodium bicarbonate distribution was also indicated by specific conductivity increases, since the bicarbonate is ionic in nature. Assessment of this field parameter data indicated several interesting results. First, vertical variation in the amendment indicators was observed at different vertical positions in the same MP, even though the same amendment volume was injected at roughly the same time into each of the three stacked screens. This result suggests that amendment likely flowed vertically, as well as horizontally, at the time of injection. Second, the field parameter data indicated a range of times for appearance of amendment effects at downgradient monitoring points ranging from quick (several days) to upwards of six months after the time of injection (see Exhibit 13). Both of the above observations are indicators of heterogeneity in the lithology. Finally, the field parameter data was used over longer periods of time to understand the persistence of both the bicarbonate and the electron donor components of the amendment in the aquifer. Persistence varied with the level of effectiveness of amendment distribution at the respective monitoring point. Generally, for areas where good distribution was achieved, the electron donor was found to persist for 18 months to three years. The spatial variation in amendment injection flow rates achieved per standard 10-foot screen length for the constructed injection wells are shown in the site map of Figure 5. The data on this map indicate that amendment injection flow rates of 0.5 to 2.0 gallons per minute (gpm) per 10-foot screen were achieved over a vast majority of the site. The data also indicate some areas where lower flow rates (less than 0.5 gpm per 10-foot screen) occurred with greater frequency, thus indicating generally less permeable lithology, at least in the aquifer vertical zones targeted for treatment. The notable areas of lower injection flow rates include the southeastern portion of HS #3 and limited areas of HS #1 and HS #2. Some of these less permeable areas were subsequently selected for repeat amendment injection using direct inject with pneumatic fracturing, to improve distribution.

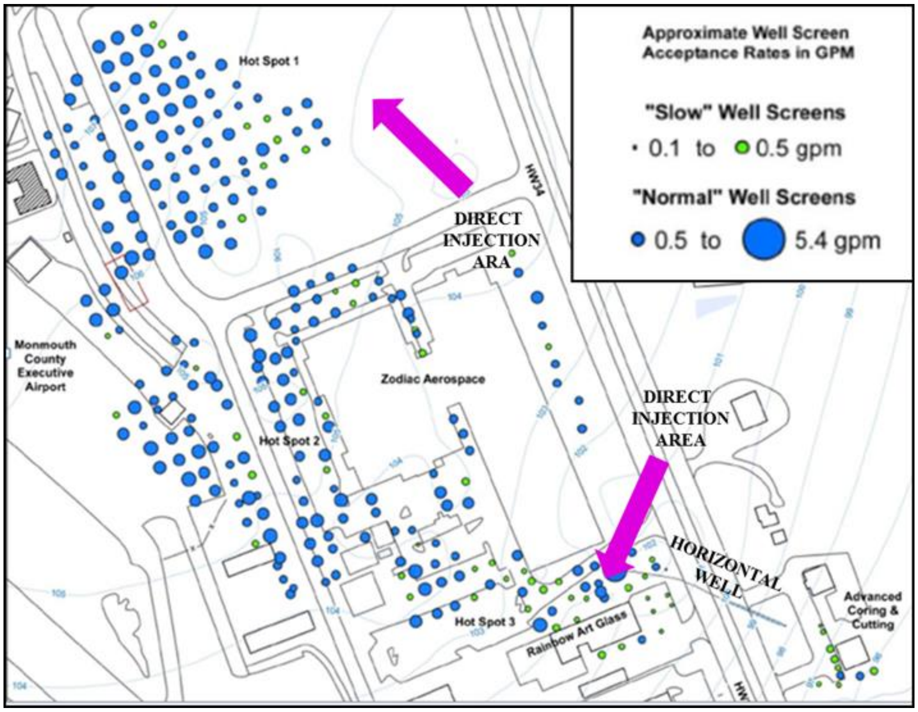

FIGURE 5. Injection well screen amendment acceptance rates across the site

CONCLUSIONS

The large-scale TCE and 1,1-DCE plume at the MDI Superfund site represented a significant challenge for full plume in situ treatment, due to size, vertically and laterally varying lithology across the site, dilute contaminants, and the native aquifer conditions of pH and DO. A cost-effective approach for in situ treatment was implemented over the last six years at the site and has resulted in reduction of the plume area by roughly 70 percent.

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