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Utilizing Multiple Methods to Remediate Groundwater in Heterogeneous Soils: Three Florida Case Studies

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ABSTRACT: Heterogeneous soils add complexity to remediating petroleum constituents in groundwater. When sand is interbedded with clay, the clay can serve as a reservoir for contaminants, causing concentrations to rebound once active remediation is discontinued. A sequence of multiple remediation methods may be required to complete the reduction of dissolved contaminants to below regulatory levels. Three sites from Central Florida were selected for this case study. All three sites are underlain by layers of sand and clay, and have been impacted by dissolved benzene, toluene, ethylbenzene, and xylenes (BTEX) constituents in groundwater. Treatment at each of these sites began with a mechanical remedial method, which initially reduced concentrations but experienced rebound once treatment was discontinued. The mechanical methods were followed by in situ chemical oxidation (ISCO) and bioremediation. In all three cases injected fluids liberated adsorbed contaminants from the clay layers, allowing treatment chemicals and/or microbes greater contact with the target parameters. However, there appears to be a correlation between the thickness of clay within the impacted zone and the difficulty in obtaining final closure. A sequence of remedial methods appears to be required to reduce BTEX concentrations to below action levels.

INTRODUCTION

Mechanical remedial methods such as air sparge/soil vapor extraction (AS/SVE) are often successful in removing dissolved BTEX constituents from groundwater in relatively homogeneous sandy soils. However, an increase in the heterogeneity of the soils, especially a larger quantity of clay, reduces the long-term effectiveness of mechanical remediation. Numerous remediation methods are available which claim to be effective for remediating hydrocarbons in heterogeneous soils, but there are few unbiased published case studies. The purpose of this paper is to determine if any of the tested methods is effective alone at remediating groundwater in interbedded sand and clay.

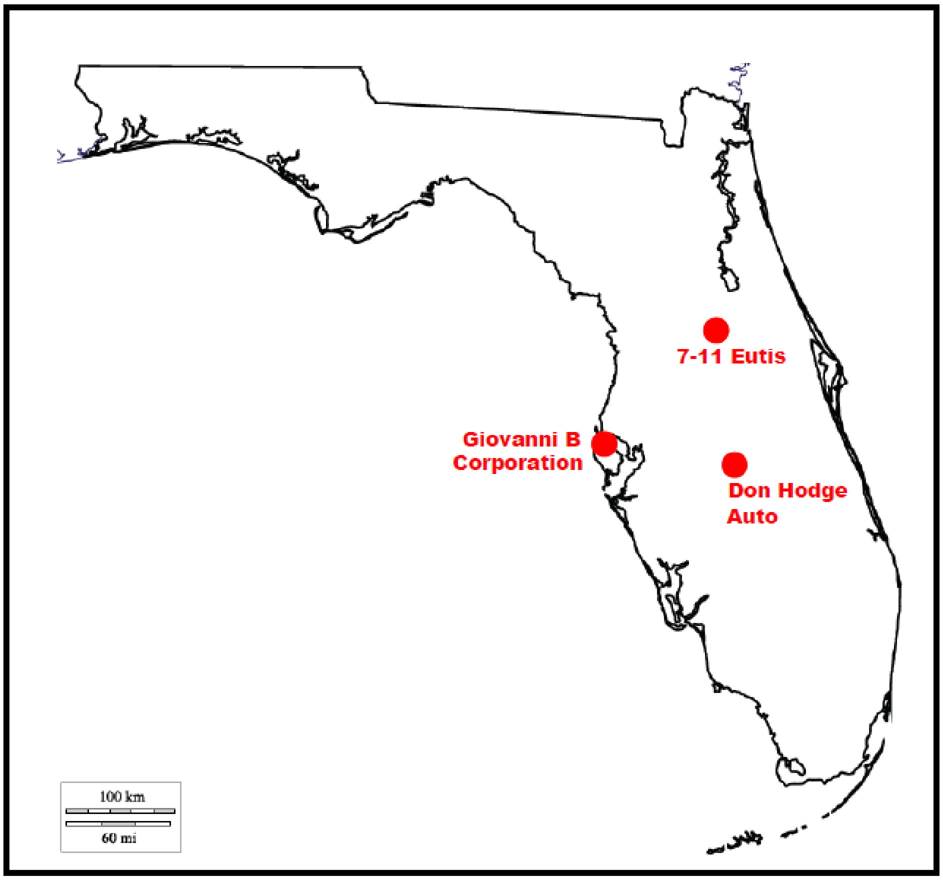

FIGURE 1. Site location map.

Three sites in central Florida were selected as case studies to determine the effectiveness of a variety of methods in sequence for remediating BTEX in heterogeneous soils. These sites are 7-11 Food Store #27972 in Eustis (7-11 Eustis), Giovanni B Corporation in Clearwater, and Don Hodge Auto Service in Frostproof (see Figure 1). Treatment at each of these sites began with a mechanical remedial method such as AS/SVE or pump and treat, which failed to permanently reduce concentrations to below regulatory standards. The mechanical methods were followed by ISCO and bioremediation by injection in sequence or combination. Information on all three of these sites is available in the public record.

METHODS

Site Lithology. Lithologic columns from the three selected sites are illustrated on Figure 2. In all three cases, the upper portion of the impacted aquifer is composed of sand or silty sand. The upper sand unit is underlain by clay at depths ranging from 6 feet [1.8 meters (m)] below land surface (bls) to 25 feet (7.6 m) bls, and the thickness of the clay within the impacted aquifer ranges from 5 feet (1.5 m) to 15 feet (4.6 m). The lithology of 7-11 Eustis consists of 4 feet (1.2 m) of sand underlain by 11 feet (3.4 m) of silty sand and 15 feet (4.6 m) of clay with another sand layer below that. Giovanni B Corporation's lithology consists of 25 feet (7.6 m) of sand underlain by a minimum of 5 feet (1.5 m) of clay. The lithology of Don Hodge Auto is 6 feet (1.8 m) of sand with 8 feet (2.4 m) of clay followed by a minimum of 26 feet (7.9 m) of sand.

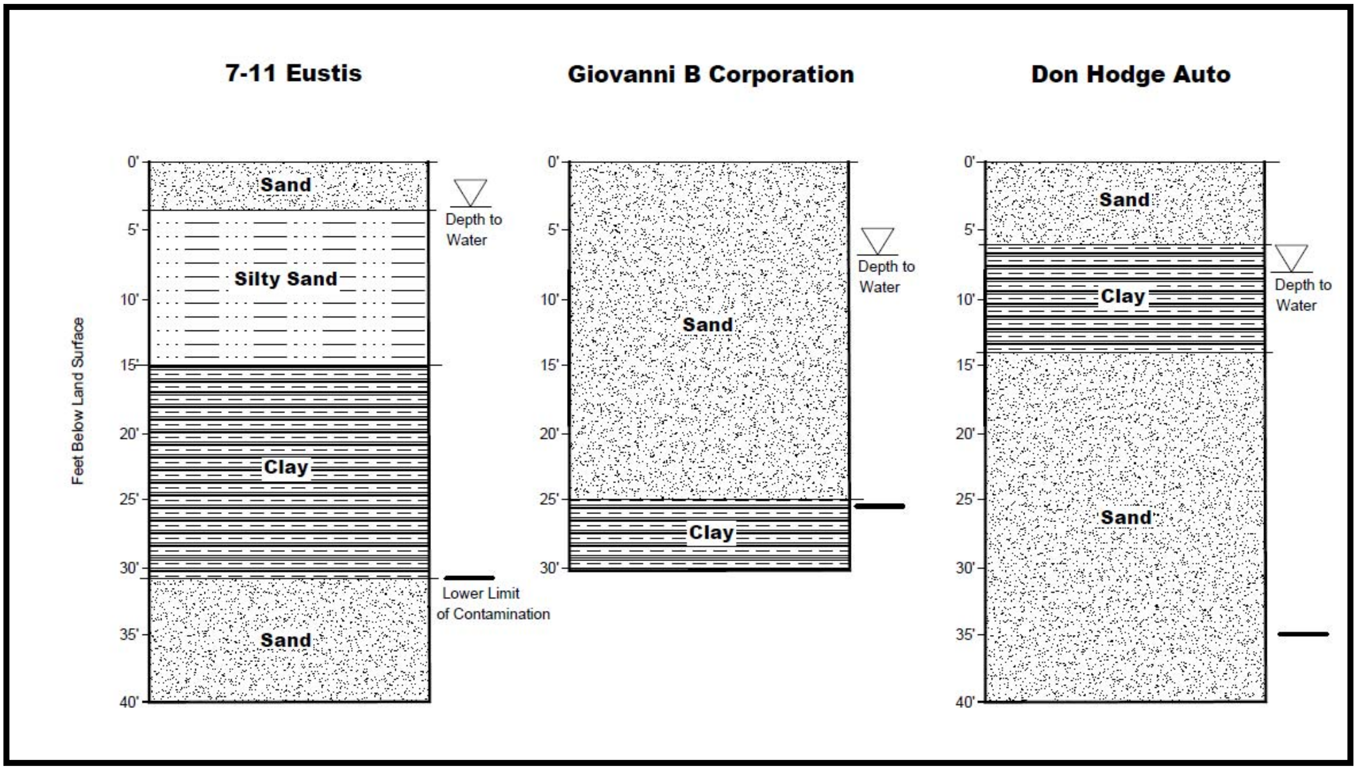

FIGURE 2. Lithologic columns of all three sites.

7-11 Eustis. BTEX contamination in the groundwater at this former service station site was originally detected in 1986. The benzene plume delineated by 1990 covered approximately 11,400 square feet (ft^2) [1,059 square meters (m^2)] with a maximum concentration of 2,100 micrograms per liter (ug/L) benzene and a maximum depth of 30 feet (9.2 m) bls. The average depth to water is 3 feet (0.9 m) bls and 15 feet (4.6 m) of the impacted zone consists of clay.

Remediation of 7-11 Eustis began in April 1993 with the operation of a pump and treat system. This system operated until March 1995 at which time the system permanently ceased operations. Additional attempts to remediate the benzene plume are listed in chronological order below.

1) March 1997 - injection of 100 pounds [45.4 kilograms (kg)] calcium oxyhyroxide
2) September 1998 – excavation of 400 tons (363,000 kg) of soil
3) September 2000 - nutrient-enhanced biosparge system startup
4) September 2006 - nutrient-enhanced biosparge system shut down
5) June 2007 – first of three sodium persulfate and iron injections

This site is currently undergoing natural attenuation with benzene concentrations gradually decreasing and slightly greater than Groundwater Cleanup Target Levels (GCTLs).

Giovanni B Corporation. BTEX contamination in the groundwater at this former service station site was originally detected in 1993. Two separate xylenes plumes were delineated by 1994 – one originating from the underground storage tanks (USTs) and the other from the fuel dispensers. Together they covered approximately 6,000 ft^2 (557 m^2) with a maximum concentration of 6,150 µg/L total xylenes and a maximum depth of 30 feet (9.2 m) bls. Only the lowermost portion of the impacted zone consists of clay.

Remediation of Giovanni B Corporation began in December 2005 with the operation of an AS/SVE system. This system operated until May 2007 by which time the BTEX parameters had been reduced to below GCTLs. However, by February 2009, the total xylenes concentration had increased to 903 µg/L, compared to the Florida total xylenes GCTL of 20 µg/L. Two injection events were conducted to address contaminant rebound: 1) April 2009 – injection of 100 gallons (378.5 L) of nutrients and microbes, and 2) March 2010 – injection of 50 gallons (189.3 L) of 10% hydrogen peroxide.

The final injection completed the active remediation of the site. By March 2013, all BTEX contaminants had been reduced to below their respective GCTLs. The site was officially closed in August 2013.

Don Hodge Auto Service. BTEX contamination in the groundwater at this active automobile repair facility was originally detected in 1991. The total xylenes plume was delineated by 2000 with an aerial extent of approximately 16,000 ft^2 (1,486.4 m^3) with a maximum concentration of 7,900 ug/L total xylenes and a maximum impacted depth of 35 feet (10.9 m) bls. A 9-foot (2.7 m) thick clay layer occupies the upper portion of the impacted zone.

Remediation of Don Hodge Auto Service began in June 2003 with the operation of an AS/SVE system. This system operated until June 2015 by which time the BTEX parameters had been reduced by approximately one order of magnitude but the period of AS/SVE remediation had been characterized by strong fluctuations of BTEX concentrations, indicating that AS/SVE has been only moderately effective for remediating the plume. In January 2017, approximately 2,400 gallons (9,085 L) of 5% hydrogen peroxide and 2,800 gallons (10,599 L) of microbes, nutrients, and surfactant were injected.

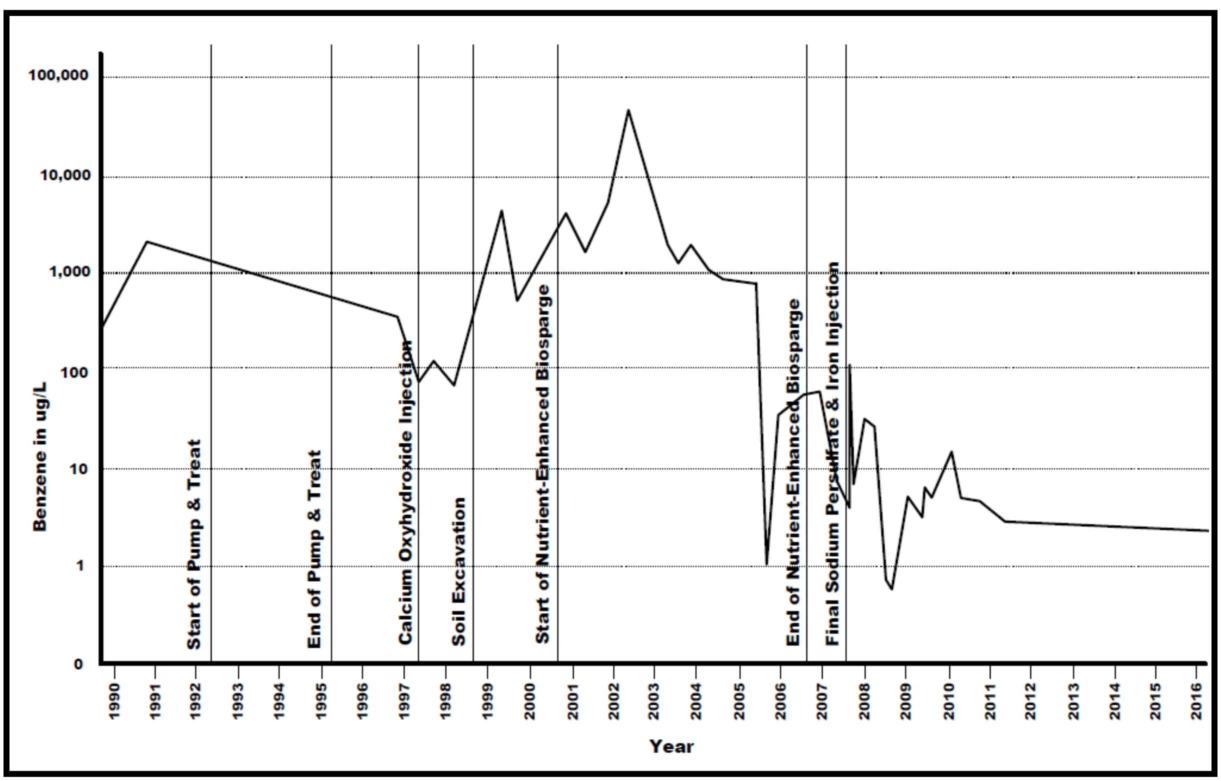

FIGURE 3. Benzene concentrations in MW-1, 7-11 Eustis.

RESULTS AND DISCUSSION

7-11 Eustis Results. Three years of pump and treat remediation at 7-11 Eustis resulted in an 84 percent reduction in benzene concentrations in the source area. Calcium oxyhydroxide injection further reduced the benzene concentration by an additional 79 %, but the September 1998 soil excavation liberated BTEX constituents that had been adsorbed in the clay layer, which increased the benzene concentration by six times. Biosparging with periodic nutrient injection further increased the benzene concentration initially, but after six years of system operation, the benzene concentration was reduced by 88 percent. The injection of sodium persulfate continued the progress of bringing benzene at 7-11 Eustis below the GCTL. A graph of benzene concentrations over time for this site is shown on Figure 3.

Giovanni B Corporation Results. A year and half of AS/SVE remediation resulted in a nearly 100 percent reduction in xylenes concentrations in the source area. However, approximately one year after the system ceased operating the concentrations began to rebound. The subsequent injection of microbes and nutrients reduced contaminant concentrations by 99.5 percent within six months but rebounded again by the end of a year. Although an injection of hydrogen peroxide reduced contaminant concentrations to below the GCTLs within three months, rebound occurred three times over the following three years before remediation was considered complete. A graph of xylenes concentrations over time for this site is shown on Figure 4.

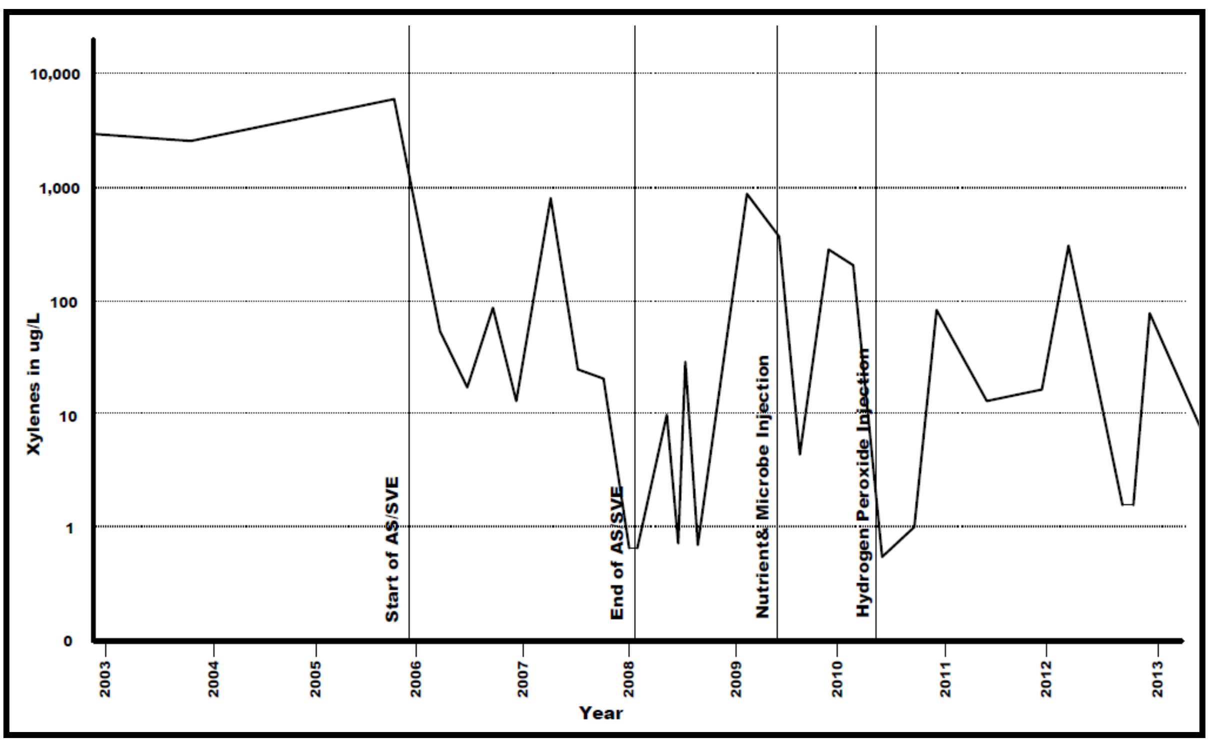

FIGURE 4. Xylenes concentrations in MW-14, Giovanni B Corporation.

Don Hodge Auto Service Results. At the Don Hodge Auto Service an AS/SVE system operated for 12 years, successfully reducing BTEX concentrations by 86 percent. Although the basic concentration trend during the 12 years of AS/SVE remediation was downward, the xylenes concentrations fluctuated wildly between sampling events. The variation in the top of the water table is approximately 5 feet (1.5 m), most of which occurs within the clay layer. It appears likely that the rising water table entering the smear zone liberated contaminants adsorbed in the clay, increasing the dissolved concentrations after each rise in the water table. The January 2017 injection event increased contaminant concentrations by introducing surfactants. Subsequent sampling events will show whether the increased availability of dissolved hydrocarbons will eventually lead to reduction in concentration without rebound. A graph of the xylenes concentrations at Don Hodge Auto Service is included as Figure 5.

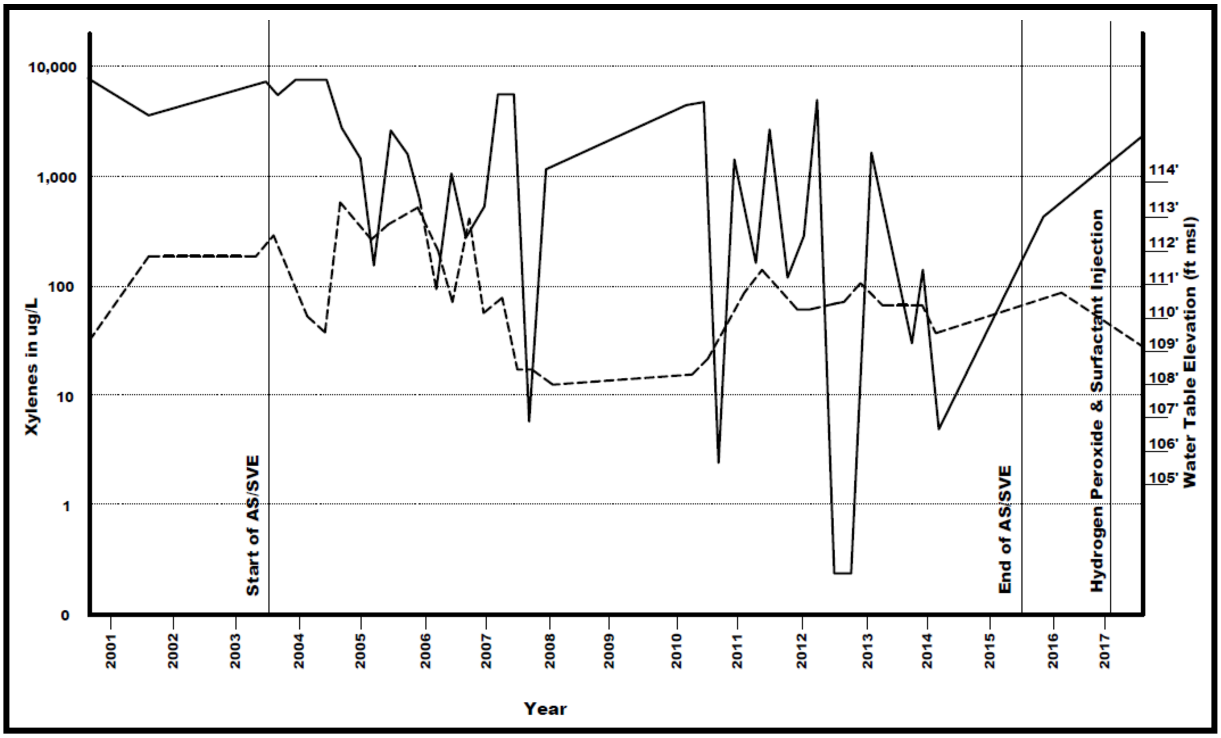

FIGURE 5. Xylenes concentrations in MW-1, Don Hodge Auto Service.

Effectiveness of Remedial Methods in Heterogeneous Soils. The following remedial methods were employed at one or more of the three sites selected for this case study: pump and treat, AS/SVE, calcium oxyhydroxide injection, nutrient-enhanced biosparge, sodium persulfate and iron injection, nutrient and microbe injection, and hydrogen peroxide injection. All of these methods were effective in reducing BTEX concentrations temporarily. However, in every case, rebound eventually occurred due to residual contaminants trapped in clay soils. It is significant that the only one of the three sites to achieve closure to date is the site with the smallest amount of clay in the impacted zone.

Based on the results of the three case studies, there is no single remedial method that is likely to permanently reduce BTEX concentrations in soils with a significant clay layer in the impacted zone. In the case of the Don Hodge Auto Service site, even 12 years of continuous AS/SVE was unable to effect reduction of contaminant concentrations without rebound. The remedial methods that appear to be most effective for sites with heterogeneous soils are the methods that include the injection of chemicals or nutrients or both in sequence. These methods liberate contaminants adsorbed in clay layers, facilitating cleanup by either chemical reactions or bioremediation. In theory the combination of surfactants, hydrogen peroxide, microbes, and nutrients employed at Don Hodge Auto Service could be the optimum method for remediating a site with a significant clay layer, but the final proof of that theory will have to wait on future sampling events.

CONCLUSIONS

The author conducted a study of three BTEX-impacted sites in central Florida with varying amounts of clay interbedded with sand to determine the optimum remedial method or combination of methods for reducing dissolved hydrocarbons in heterogeneous soils. The three sites selected – 7-11 Eustis, Giovanni B Corporation, and Don Hodge Auto Services – have been remediated by mechanical and injection methods. Of the three only Giovanni B Corporation has achieved closure to date.

All three sites were first remediated by mechanical means (pump and treat or AS/SVE) and experienced rebound of contaminant concentrations once the mechanical remedial method ceased operations. The mechanical methods were followed by a sequence of injections of nutrients and microbes, and chemicals, such as calcium oxyhydroxide, hydrogen peroxide, and sodium persulfate. In most cases these injections were followed by a reduction of contaminant levels but multiple injection events were typically required to reduce the rebound concentrations. Two injection events were sufficient to bring the Giovanni B Corporation site to closure, which had the lowest amount of clay in the impacted zone. 7-11 Eustis required two injection events combined with six years of nutrient-enhanced biosparge to bring concentrations down to natural attenuation levels. The long-term effectiveness of the injection of hydrogen peroxide, surfactants, and nutrients at Don Hodge Auto Service will be determined by future sampling events.

There appears to be correlation between the thickness of clay in the impacted zone and the difficulty of achieving permanent contaminant concentration reductions. Many methods appear to have some effect but there is no one remedial method that will complete the restoration of a site with significant clay with a single application. Multiple methods strategically employed that initially mechanically remove mass from relatively large porous media followed by flushing or destroying contaminants from the clay layers biologically or chemically appear to have the greatest impact on BTEX concentrations in heterogeneous soils.

REFERENCES

Bienkowski, L.S. 2015. *Task 2 Report, Don Hodge Auto Service*. Ellis & Associates, Inc., Jacksonville, FL.

Contaminant Cleanup Target Levels. 62-777 FAC Table 1.

Friedloi, M., and B. Wallace. 1998. *Tank Closure and Soil Assessment Report, 7-Eleven Store #27972*. RUST Environment & Infrastructure, Altamonte Springs, FL.

Green, M, and S.L. Schomer. 1992. *RAP Implementation/Start-up and As-built Report, 7-Eleven Store #27972*. SEC Donohue Environment & Infrastructure, Altamonte Springs, FL.

Hentschel, P., and T Hauschild. 2013. *Remedial Action Interim Report/No Further Action Request, Giovanni B Corporation*. Groundwater & Environmental Services, Inc., Pinellas Park, FL.

Kiely, R. 2006. *Startup Report, Giovanni B Corporation*. Envirotrac Environmental Services, Tampa, FL.

Lewis, M.K. Jr. 2010. *6th and 7th Quarter PARM Report, Giovanni B Corporation*. Envirotrac Environmental Services, Tampa, FL.

Machado, A.H. 2016. *Task 2 RA Letter Report, 7-Eleven Store #27972*. Advanced Environmental Technologies, LLC, Lakeland, FL.

Patrick, J.E., and S.L. Schomer. 1990. *Contamination Assessment Report, 7-Eleven Store #27972*. Westinghouse Environmental and Geotechnical Services, Inc., Altamonte Springs, FL.

Pondo, D. 1994. *Contamination Assessment Report, Giovanni B Corporation*. Metcalf & Eddy, Tampa, FL.

Smar, M., and S.L. Schomer. 2011. *Supplemental Site Assessment Report, 7-Eleven Store #27972*. AECOM, Orlando, FL.

Sorenson, K.C. 2001. *Level 4 Supplemental Site Assessment Report, Reliable Auto*. Advanced Environmental Services, Inc., St. Petersburg, FL.

Sorenson, K.C. 2014. *Year 11-2nd Quarter O&M Report, Don Hodge Auto Service*. Advanced Environmental Services, Inc., St. Petersburg, FL.

Sorenson, K.C., and L.L. Shaetz. 2003. *Remediation System Installation and Startup Report, Reliable Auto (aka Don Hodge Auto Service)*. Advanced Environmental Services, Inc., St. Petersburg, FL.

Sweeney, M., and S.L. Schomer. 1997. *Results of ORC Installation and Six Month Monitoring Program, 7-Eleven Store #27972*. RUST Environment & Infrastructure, Altamonte Springs, FL.