Implementing RF Heating in Fractured Bedrock to Remediate TCA DNAPL

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Abstract

The heterogeneous distribution of residual 1,1,1-trichloroethane (TCA) dense non-aqueous phase liquid (DNAPL) within discrete, poorly connected bedrock fractures renders many remedial technologies inefficient or ineffective because the DNAPL cannot be physically removed or reached to treat in situ. Some thermal technologies maintain the ability to treat a targeted volume of bedrock, thereby overcoming the physical constraints of the bedrock fracture network, but can be prohibitively expensive to implement due to the energy required to heat the rock mass. Alternatively, radio frequency (RF) heating is expected to preferentially heat groundwater and residual TCA DNAPL within the bedrock fractures, rather than the surrounding bedrock mass, making this a potentially cost-effective option for in situ thermal treatment.

An integrated RF heating and soil vapor extraction (SVE) system was designed, constructed and activated at a site impacted by TCA DNAPL in fractured crystalline bedrock. This system is believed to be the first in situ application of RF heating to treat TCA DNAPL in bedrock. RF heating is being utilized to:

- Accelerate the dissolution rate of TCA DNAPL into dissolved phase TCA.
- Accelerate the degradation of dissolved phase TCA through biotic and abiotic mechanisms (from years to days).
- Minimize hydraulic disturbances that could mobilize DNAPL or displace dissolved phase TCA.

The treatment system includes a network of nine, 100-foot deep, eight-inch diameter boreholes, an RF generator, a four-probe transmitter array, and an SVE system for control and treatment of vapors. The target groundwater treatment temperature is 50 to 60 degrees Celcius (°C). Start-up took place in December 2003 and the power output reached a maximum of 18.9 kilowatts (kW).

Monitoring data collected after the first six months of treatment indicated that elevated groundwater temperatures were measured over an area of approximately 2,000 square feet. The maximum groundwater temperature measured has been 45°C. Based on the groundwater temperature monitoring, each RF antenna has a radial heating influence of approximately 15 feet. Measurements of downhole electric field intensity show that the effective vertical radiation interval is approximately 15 feet. Therefore, the total treatment volume using four antennae is estimated at 30,000 cubic feet.

The average TCA concentration in and adjacent to the treatment area has decreased from 115,000 micrograms per liter (µg/L) to 24,000 µg/L following six months of treatment, an estimated decrease of 79%. An evaluation of the data shows both abiotic and biotic degradation of TCA to 1,1-dichloroethene (DCE), acetic acid and 1,1-dichloroethane (DCA).

Introduction

An historical release of 1,1,1-trichloroethane (TCA) occurred at the subject site. Both residual dense, non-aqueous phase liquid (DNAPL) and elevated dissolved phase concentrations have been identified in the source area. Source abatement is necessary to facilitate permanent abatement of down-gradient groundwater impacts. Source abatement depends on the ability to abate residual TCA DNAPL contained within a low yield, poorly connected, fractured crystalline bedrock. Since much of the TCA has migrated into fractured bedrock, it is not amenable to traditional or low cost remedial solutions such as air sparging and soil vapor extraction. Additionally, bedrock fractures are not highly interconnected over great distances, which makes groundwater pumping and treatment difficult to operate cost effectively. More innovative remedial technologies, such as in situ chemical oxidation (ISCO) using permanganate, are not effective at treating chlorinated ethanes, such as TCA. Site-specific, bench-scale and pilot studies and a detailed comparative analysis of remedial alternatives resulted in the selection of radio frequency (RF) heating as the recommended remedial action alternative for the site. RF heating is a technology that creates heat in the formation for the destruction and/or removal of contaminants.

Site Information

The source area is located beneath a building on the top of a hill immediately down-gradient of a groundwater flow divide. Very limited groundwater recharge occurs between the groundwater flow divide and the suspected source area. Therefore, under the building, the unconsolidated deposits are unsaturated, except for small, isolated perched water zones. Significant downward vertical hydraulic gradients have been measured in the source area.

The surficial geology at the site comprises fill over glacial till. The sandy fill has an estimated thickness of zero to 15 feet. Inspection of split-spoon soil samples from the glacial till indicated a series of high angle fractures throughout the deposit. The fractures act as planes of weakness upon which the till is easily broken apart.

Bedrock at the site is comprised of late Precambrian- to Devonian-aged rocks characterized by intensely sheared parent rocks. Remnant parent rocks include diorite, gabbro, altered basalt and schist. Northeast-trending features with secondary northwest-trending conjugate features dominate regional bedrock structure and fractures. There are no identified regional fractures transecting the site.

Results of extensive subsurface investigation and removal actions within the source area suggest that the original release migrated through the till along fracture planes to the bedrock surface, where its lateral migration appears to have been largely restricted to a bowl-shaped bedrock depression beneath the source area. Residual TCA DNAPL then migrated into bedrock fractures and appears to have been largely restricted to shallow bedrock, due to the high competency and low yield of the bedrock. The primary pathway for dissolved phase TCA migration from this shallow residual source area is generally downward via advective flow through bedrock fractures into deeper bedrock. The predominant direction of groundwater flow within the deeper fractured bedrock system is consistent with the orientation of the bedrock surface beneath the site, the orientation of local and regional bedrock structural features, and the location of a down-gradient discharge area (i.e., a sand and gravel aquifer). Ultimately, deep bedrock flow gradients become upward, eventually discharging to a sand and gravel aquifer. The primary porosity of the rock is estimated at 0.5 percent. Thus, the bedrock aquifer consists of the interconnected secondary porosity.

The historical average TCA concentration in groundwater within and around the source area was 250,000 micrograms per liter (µg/L), approximately 19 percent of TCA's aqueous solubility. The extent of residual DNAPL in bedrock fractures was delineated by coupling the results of geophysical imaging logs with data collected using immiscible-fluid absorbent liners (i.e., NAPL FLUTe liners). Residual DNAPL was noted in five of the nine boreholes where FLUTe liners were installed. TCA concentrations in groundwater from fractures containing residual DNAPL ranged from 410,000 µg/L to 640,000 µg/L (i.e., 32 percent to 49 percent of solubility). No accumulation of separate-phase product (i.e., continuous measurable layers) was detected in any of these boreholes. TCA concentrations measured in the four boreholes where DNAPL was not detected ranged from 31,000 μg/L (two percent of solubility) to 140,000 μg/L (11 percent of solubility).

Remedial Technology Selection

Extensive technology screening, laboratory treatability studies and field pilot tests were conducted to support selection of an appropriate remedial technology to abate the residual TCA DNAPL source area at the site. Results of these activities revealed three technologies that showed promise for source abatement at the site: (1) in-situ chemical oxidation (ISCO) with heat-catalyzed sodium persulfate, (2) thermal degradation at temperatures of 35 degrees Celsius (°C) or higher; and, (3) biodegradation using lactate or emulsified soybean oil plus an enrichment culture. Pilot testing of ISCO using Fenton's Reagent indicated concentration reductions followed by substantial rebound, suggesting that residual DNAPL within poorly interconnected bedrock fractures in the source area would limit the effectiveness of technologies dependant on injection of remedial additives throughout the treatment zone. Therefore, application of an in situ thermal technology was selected for implementation at the site.

Thermal treatment methods are generally best suited to source area abatement, where a high reduction in mass is desired over a relatively limited volume and time period. The primary physical and chemical properties that govern fate and transport of chlorinated solvents are temperature dependent (Table 1).

Table 1 - Thermal Effects on Chlorinated Solvent Properties

Fate and Transport Property	**Effect as Temperature Increases**
Liquid density	Decreases moderately (less than 100 percent)
Vapor pressure	Increases significantly (10 to 20 fold)
Liquid viscosity	Decreases significantly until boiling point and drops markedly upon conversion from liquid to vapor
Vapor viscosity	Increases slightly as vapor temperature increases
Diffusivity	Increases
Solubility	Increases as temperature increases
Henry's constant	Increases (more likely to volatilize in water)
Partition coefficient	Decreases (less likely to partition to organic matter in soil
Biological degradation	Increases (may decrease at higher temperatures)
Abiotic degradation	Increases

Source: USEPA, 2004

The conventional approach with thermal treatment is to use high temperatures to raise the vapor pressure of VOCs bound to soils to induce volatilization. The volatilized compounds are typically extracted via a vapor extraction system. Thermal treatment of TCA does not necessarily require as aggressive an approach as thermally induced volatilization, since TCA can be destroyed in situ through thermal decomposition.

Moderate heating of subsurface liquids can result in dramatic increases in solubility and degradation. Other properties that may enhance contaminant removal or containment, such as vapor pressure, Henry's constant and viscosity are not as critical at this site.

As shown in Table 2, there is a significant increase in abiotic degradation rates of TCA as temperature increases above 25°C. Above 55°C, the half-life of TCA is reduced to a period of days. Biological degradation is also enhanced with elevated temperatures. Biodegradation rates may increase two-fold for every 10 °C rise in temperature (USEPA, 1997). Solubility of chlorinated hydrocarbons can increase by a factor of two or more as an area is heated (USEPA, 2004).

Table 2 - TCA Abiotic Degradation Half-life as a Function of Temperature

Temperature (°C)	**TCA Half-life**	**Reference**
10	12 years	McCarty (1994)
15	4.9 years	McCarty (1994)
20	1.7 years 3.2 – 3.8 years 0.95 years	Gerkens & Franklin (1989) Klecka et al. (1990) McCarty (1994)
25	0.5 years 1 year 0.8 – 1.3 year	Dilling et al. (1975) Gerkens & Franklin (1989) Haag & Mill (1988)
40	35 days 22 – 27 days	Haag & Mill (1988) Gerkens & Franklin (1989)
55	3.6 days 4.6 days	Gerkens & Franklin (1989) Haag & Mill (1988)
60	1.2 – 3.8 days 22 days	Gerkens & Franklin (1989) Haag & Mill (1988)
80	5.5 hours 2.7 – 4.0 hours	Haag & Mill (1988) Gerkens & Franklin (1989)

A number of thermal treatment technologies were evaluated for implementation at the site: RF heating, six-phase heating, conductive heating, steam injection, and circulation of heated water. Compared to other heating methods, RF heating was attractive for use at the site because:

1. RF heating preferentially heats bipolar molecules (e.g., water), focusing the thermal energy on treating groundwater rather than heating the rock;
2. RF heating heats water within a volume of bedrock independent of the degree of fracture interconnectivity; and,
3. the efficiency of RF heating should be enhanced by site hydrologic characteristics, including the low porosity of bedrock and minimal flushing of groundwater through the source area due to minimal recharge associated with the location of the source area in the drainage basin (i.e., near a basin divide and beneath a building).

Radio Frequency Heating

RF heating technology has existed in many forms since the 1930s. The technology is commonly divided into dielectric heating and induction heating. The applications for this type of heating range from medical diathermy of muscles and joints to industrial dielectric heating techniques used to dry wood and cure adhesives. More recently, significant interest has developed in the application of this technology for environmental remediation. Thermal treatment is a relatively common and proven remedial technology when implemented in soils (Lighty, 1990). This project is thought to be the first application of RF heating in bedrock.

In-situ RF heating technology imparts heat to non-conducting materials through the application of carefully controlled RF transmissions. The technology is applied by inserting a flexible coaxial transmission line and applicator (antenna) system into one or more vertical or horizontal boreholes in the area to be treated. RF generators supply energy through coaxial lines to a multiple of electromagnetically coupled down-hole antennae, and the subsurface material between the antennae rises in temperature as it absorbs electromagnetic energy radiating from the antennae. The antennae can be placed in boreholes that are dry, wet (water) or filled with other materials depending on the application. The antennae do not require electrical contact with the formation. The dielectric heating produced by the RF antennae extends elliptically away from the antennae and into the target formation. The radial extent of the heating pattern will vary as a function of the operating frequency, the length of the RF antennae, and the electrical conductivity and dielectric constant of the target formation.

The RF borehole antenna equipment for environmental remediation are engineered to satisfy exposure guidelines established by the American National Standards Institute (ANSI) and the Occupational Health and Safety Administration (OSHA). Rules and regulations for the operation of radio frequency equipment are provided by the Federal Communications Commission (FCC).

The RF system employed at the site consists of a 27.12 megahertz (MHz) 4-channel, 19.6-kilowatt RF equipment trailer, transmission lines, four antennae, applicators, and fiber optic thermometry. The antennae are deployed in boreholes spaced approximately 15 feet apart on square. Each borehole antenna applicator is 10-feet long and 2.5 inches in diameter. Each antenna receives a maximum of 4.9 kilowatts (kW) from the RF generator module, located adjacent to the array. The RF system generates RF radiation at its ISM (Industrial, Scientific, Medicine) operating frequency of 27.12 MHz. ISM frequencies are allotted by the FCC for non-communications uses of the frequency spectrum so that this type of equipment will not interfere with communications.

A schematic of the RF heating system configuration at the site is included in Figure 1. The RF system is integrated with a soil vapor extraction (SVE) system to treat residual TCA that may be present in the vadose zone, as well as collect vapors that may be generated by the heating process. The primary components of the integrated RF/SVE remedial system are:

- RF heating system – includes four RF antennae in 100-foot deep open-bedrock boreholes configured in a square array. To treat the entire source area, the RF system was designed with four contiguous treatment cells, each consisting of a four-borehole array. The approximate distance between boreholes is 15 feet, which represents an adequate spacing between antennae to ensure that the RF fields overlap at the center of each treatment cell. The RF energy from each antenna is propagated in an elliptical shape. By applying RF energy to four wells in a square array, overlapping RF fields maximize the efficiency of groundwater heating. The antennae are 10 feet in length. Therefore, each treatment cell is heated in four vertical lifts. Monitoring wells and SVE extraction wells are used to monitor performance of the RF system.

- SVE system – consists of a vacuum blower, moisture separator, heat exchanger, and vapor phase granular activated carbon vessels. The SVE system includes a series of shallow (screened from 2 to 10 feet below grade) overburden extraction wells that serve as a sub-slab depressurization system to prevent the potential for VOC entrainment within the building. The SVE system also includes a series of deep overburden and shallow bedrock extraction wells that focus on abating residual vapor phase

VOC impacts within the vadose zone, and capturing VOCs partitioning from the bedrock aquifer as a result of heating.

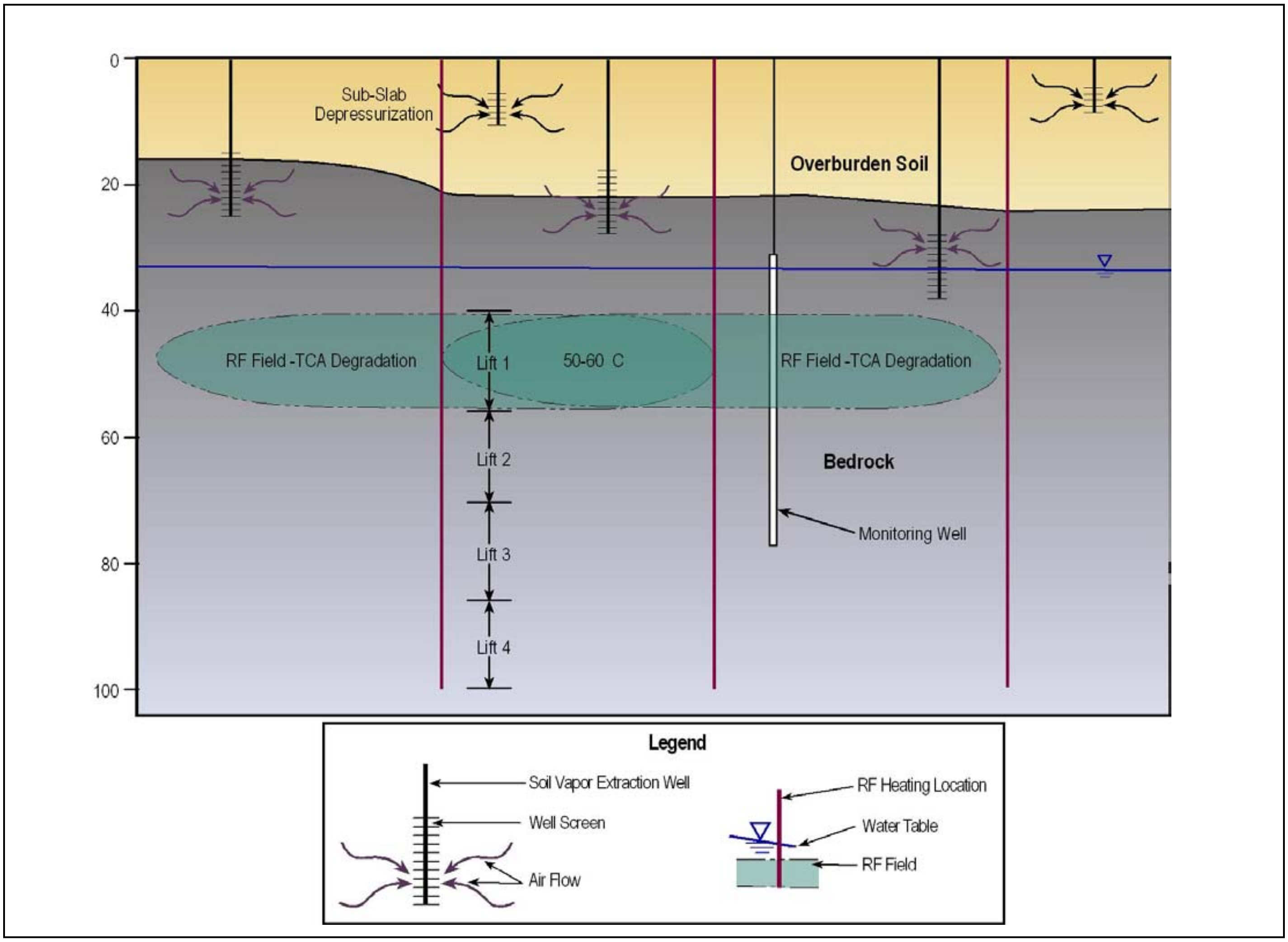

Figure 1 – Schematic of RF/SVE Remedial System Configuration

Radio Frequency Heating Implementation and Results

The SVE and RF systems were fully operational on 15 September and 9 December 2003, respectively. Both systems' operation is continuous unless system maintenance and monitoring is conducted. The following table summarizes the RF antennae configurations since system start-up.

Depth of RF Antennae (below ground surface)	Total Power	Duration
50 feet	12 kW	3 weeks
	16 kW	6 weeks
	18.9 kW (maximum power available)	3 weeks
35 feet	12 kW	12 weeks
60 feet	12 kW	2 weeks
	16 kW	5 weeks and ongoing

Down-hole electrical field measurements indicated that the RF antennae produce a vertical electrical field distribution of 15 feet and a horizontal electrical field radius of at least 15 feet. Based on the configuration of the four antennae, the RF heating system generates an overall electrical field that targets approximately 1,125 cubic yards of fractured bedrock at once.

Groundwater temperature monitoring was conducted to evaluate the rate of heating, the radius of heated groundwater and migration of heated groundwater. Temperature measurements were taken at 10-foot vertical intervals throughout the length of each deep bedrock borehole. The maximum groundwater temperature observed to date was in the monitoring well located in the center of the initial RF treatment cell; the temperature was 45.8°C and was measured at a depth of 40 feet below ground surface. Elevated groundwater temperatures have been measured over a vertical interval of 55 feet, ranging from the water table (i.e., approximately 25 feet below grade) to a depth of 80 feet below grade. Elevated groundwater temperatures have been measured over an area of approximately 2,000 square feet. The total treatment volume for using four antennae is approximately 1,100 cubic yards.

At the maximum power output (i.e., 18.9 kW), groundwater temperature increases in the heated area and near the depth of the antennae averaged 1°C per week. This rate of heating was slower than anticipated and was likely due to the low bedrock moisture content. When the antennae were raised to 35 feet below ground surface, groundwater at 60 feet cooled at an average rate of 0.5°C weekly. However, groundwater temperatures at 50 feet remained relatively constant. Although the target groundwater temperature range of 50°C to 60°C has not yet been achieved, groundwater temperatures continue to increase with continued system operation.

Groundwater temperature increases have resulted in significant reductions in VOC concentrations in source area groundwater (Figure 2). The average TCA concentration in and adjacent to the treatment area has decreased from 115,000 µg/L to 24,000 µg/L following six months of treatment, an estimated decrease of 79%.

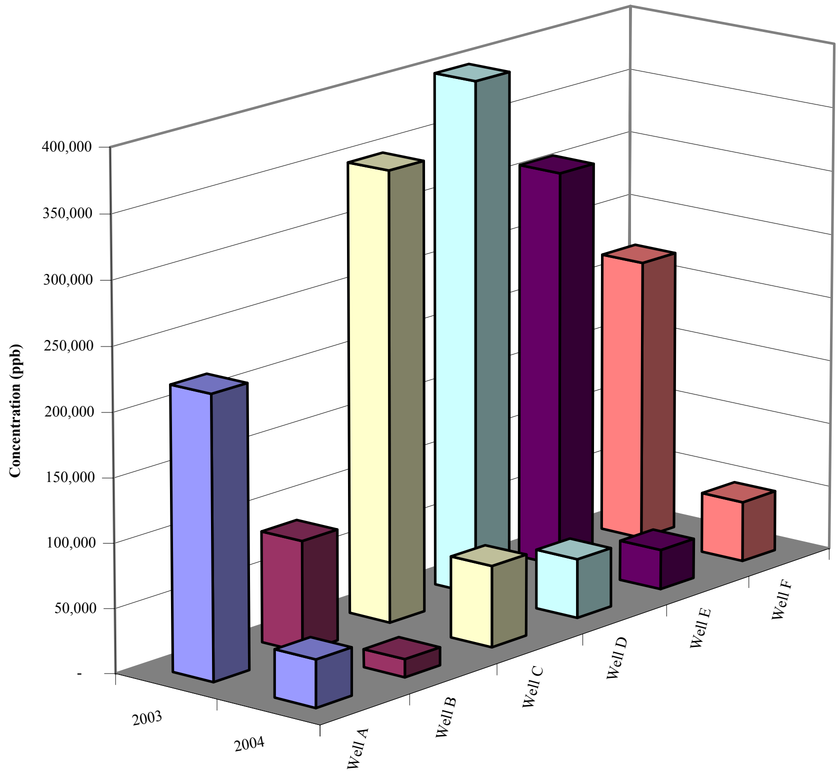

Figure 2 - TCA Concentrations in Heated Source Area Groundwater

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