Evaluation of geophysical methods for the detection of subsurface Tetrachloroethylene in controlled spill experiments
Aldo Mazzella*, United States Environmental Protection Agency and Ernest Majer, Lawrence Berkeley National Laboratory.

Summary
A controlled Tetrachloroethylene (PCE) spill experiment was conducted in a multi-layer formation consisting of sand and clayey-sand layers. The purpose of the work was to determine the detection limits and capability of various geophysical methods. Measurements were made with ten different geophysical techniques before, during, and after the PCE injection. This experiment provided a clear identification of any geophysical anomalies associated with the presence of the PCE. During the injection period all the techniques indicated anomalies associated with the PCE. In order to quantify the results and provide an indication of the PCE detection limits of the various geophysical methods, the tank was subsequently excavated and samples of the various layers were analyzed for residual PCE concentration with gas chromatography (GC). This paper presents some of the results of five of the techniques: cross borehole complex resistivity (CR) also referred to as spectral induced polarization (SIP), cross borehole high resolution seismic (HRS), borehole self potential (SP), surface ground penetration radar (GPR), and borehole video (BV).

Introduction
Tetrachloroethylene (PCE), typically used as a dry cleaning solvent, is a predominant contaminant in the subsurface at Superfund Sites. PCE is a Dense Non-Aqueous Phase Liquid (DNAPL) that migrates downward into the earth, leaving behind areas of residual saturation and free product pools on areas of low permeability. While the maximum dissolved phase concentration of PCE is fairly low, on the order of 200 parts per million (ppm), this is above the maximum contaminant level (MCL) for drinking water of 0.005 ppm. Hence, the non-aqueous phase product in the subsurface acts as a continuous source of ground water contamination. Typical ground water remediation techniques, such as the pump and treat method of the drinking water, have not proven very effective for aquifer cleanup. For example, it is estimated that it would take more than 50 years of the pump and treat method to clean up some contaminated sites.

Effective remediation requires the location of the non-aqueous phase PCE product in the subsurface. The purpose of the current research is to evaluate the use of geophysical methods to detect and delineate the subsurface non-aqueous phase PCE. Since the geophysical methods have a wide range of response to natural formation variations, it is difficult to clearly identify the response due only to the presence of PCE at existing hazardous waste sites. In order to identify any geophysical anomaly associated with the PCE, a series of controlled spill experiments have been conducted. In these experiments, measurements with a number of geophysical methods were made before, during and after the injection of a DNAPL, such as PCE, into the subsurface.

One of the first such experiments was conducted at the Canadian Forces Base at Borden. The activities were conducted with the University of Waterloo and the US Geological Survey (USGS). Additional experiments were also conducted at the Oregon Graduate Institute with the USGS. A number of different geophysical methods were tested in each of these experiments with good success (Greenhouse et. al., 1993). While geophysical anomalies were observed, none of the methods by itself provided a unique signature for the presence of PCE. The existence of vertical steel walls and tanks to contain the migration of the PCE, prevented the evaluation of a number of other geophysical methods. The purpose of the current research is to evaluate these other geophysical methods for detecting the presence of PCE.

Method
In order to evaluate the other geophysical methods of interest, the current experiment was conducted in a non-metallic, fiberglass tank that was housed in a special building at the University of California Richmond field station that had been designed to minimize electromagnetic coupling and electrical noise interference for geophysical research. The tank is about 2.4 meters in diameter and 1.8 meters in depth and was constructed with PCE resistant resin (Figure 1). Two other barriers were in place outside the tank to prevent any PCE leakage to the outside environment. A multi-layer formation consisting of sand, and sandy-clay layers was engineered and constructed in the tank. It was necessary to consider various factors in the construction of the formation. These included a balance between the presence of clay needed for the induced polarization response for the CR method and high formation resistivity needed for the surface GPR method. In addition, the layers had to have good hydraulic conductivity to allow the PCE to penetrate into the formation within a reasonable time period of a few days and the ability to effectively allow the removal of air in the
Evaluation of geophysical methods for the detection of subsurface Tetrachloroethylene in controlled spill experiments

formation pore space for good seismic signals. The final configuration was developed after a number of experimental laboratory studies were conducted.

The layers in the tank were constructed with a washed, well sorted medium grain sand, Unimin 20/30. This is composed of over 99.8% SiO2. A Calcium Montmorilllite clay, Az-1, obtained from the National Clay Repository at Purdue University was used for the development of the clay/sand layers. The central portion of the tank, with a diameter of about 1 meter, was the primary area for the experiment. The formation in this area consisted of three layers, a 53 cm thick upper sand layer, a 20 cm thick 3% clay/sand layer overlying a 30 cm thick 6% clay/sand layer (dry weights). Beneath this, a 3 cm thick solid clay layer was placed to prevent the PCE from migrating directly to the bottom of the tank. The formations were fully saturated during construction with 0.001 molar calcium chloride distilled water. The conductivity of the water was about 230 microseimens/cm. A number of wells and probes were placed in the formation during construction to accommodate the different geophysical methods (Figure 2).

Spill Experiment

After construction the formation was monitored and allowed to stabilize for about two months. About 10 days before the injection experiment began, daily monitoring established a background baseline and noise level for the different geophysical methods. In order to track the migration of the PCE with downhole video and subsequent excavation, a dye, Oil Red O, was added to the PCE at a concentration of 3 gms/liter.

In May 2004 the injection experiment began. Eighty-five (85) liters of PCE were injected over a period of 26 hours into the subsurface at a fairly constant rate of about 3.7 liters/hour. The PCE was injected through a tube at a depth of about 6 cm below the surface. The injection tube extended 24 cm above the surface. This allowed a sufficient head to develop to overcome surface tension effects and the PCE flowed into formation. Ten different geophysical methods were utilized to monitor the subsurface properties before, during and after the injection. These methods were cross borehole high resolution seismic, cross borehole complex resistivity, borehole self potential, surface ground penetrating radar, borehole video, prototype very early time transient electromagnetic, prototype high frequency electromagnetic sounder, prototype directional borehole radar, cross borehole radar tomography, and a prototype borehole dielectric tool. Only some results from the first five of these methods are discussed in this paper. These research activities involved six (6) scientists from the USGS, three (3) scientists from LBNL, and two (2) EPA scientists.

Initial results showed significant changes in the responses of most of the geophysical methods during the PCE injection period. Partial data from the surface 900 MHz GPR, seismic (40 Khz) and CR methods are shown in Figures 3, 4, and 5. These are data just before, during and just after the PCE injection. The two-way travel times of the GPR data in Figure 3 have been converted to depth using a relative dielectric constant of 24 to 27. These values were previously obtained from time domain reflectometry measurements and from a common mid-point GPR survey and are in agreement with the borehole dielectric tool responses. The GPR data in Figure 3b, 9 hours after the spill started, show a significant anomaly at about the 50 cm depth. This is at the top of the 3% clay
Evaluation of geophysical methods for the detection of subsurface Tetrachloroethylene in controlled spill experiments

About 33 liters of PCE had been injected at this time. The borehole video indicated that 4 hours after the spill started, a 4 cm thick layer of PCE had accumulated and spread across the 3% clay/sand layer at the 50 cm depth.

Data from the cross borehole seismic (40kHz dominant frequency) and CR methods at depths around 50 cm (top of 3% clay where it seemed that the PCE pooled before breaking through) are shown in Figures 4 and 5. These are data taken also just before, during and after the PCE injection. The cross borehole seismic travel times are plotted in Figure 4. A peak increase of about 5% in travel time is observed during the injection. Note that there was a slight residual in the travel times after the PCE flowed through this level in that the travel times did not return all the way to the original values. The seismic amplitude also became attenuated during this time.

The CR data in Figure 5 represents the cross borehole dipole – dipole response at 10 Hz. Changes in both the amplitude and phase are observed during the injection. About a 700% increase in the resistivity amplitude and a 40% decrease in the phase are observed at the peak of the injection period.

At about 9 hours into the spill, the borehole video indicated that the PCE had found a seam through the block clay near the CR receiver and most of the PCE migrated to the bottom of the tank shortly after the injection stopped. Through subsequent analysis it was estimated that about 67 of the 85 liters of PCE injected ended up at the bottom of the tank. Most of the post spill responses are therefore measuring residual PCE.

**Conclusion**

Anomalies were observed with multiple geophysical methods indicating changes occurred in different physical properties of the formations with the presence of PCE. A monitoring approach utilizing multiple different geophysical methods could provide unique detection and identification of subsurface PCE.

**References**


**Notice**

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy. Mention of trade names or commercial products does not constitute endorsement or recommendation by EPA for use.
Evaluation of geophysical methods for the detection of subsurface Tetrachloroethylene in controlled spill experiments

Figure 3a,b,c  Surface 900 MHz GPR data before, during and after the PCE injection. Note the anomaly at the 50 cm depth in 3b, at the top of the 3% clay layer. The post injection data in 3c shows a residual anomaly.

Figure 4. Cross borehole seismic travel time (Y axis in microseconds) at different depths from 46 to 54 cm at different times before, during, and after the PCE injection.

Figure 5. Cross borehole complex resistivity amplitude and phase at 50 cm depth, 15 cm dipole length, 10 Hz frequency. Data was taken just before, during and after the PCE injection.