Residual Hydrocarbons in a Water-Saturated Medium: A Detection Strategy Using Ground Penetrating Radar

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ABSTRACT •

- The focus of this article is to demonstrate through physical model experimentation a potential means for identifying contaminated areas where a light non-aqueous phase liquid (LNAPL) hydrocarbon has been redistributed by a rising water table in a previously hydrocarbon residual–free vadose zone using ground-penetrating radar (GPR). Analogies of the experimentation conducted in this study are situations where a rise of the water table follows leakage from a tank or pipe at depth or where an LNAPL hydrocarbon plume has migrated laterally from a surface source along the top of the saturated zone and a subsequent rise of the water table occurs. Research to date has provided insight into mechanisms that may offer the potential for LNAPL detection under certain field conditions; however, no studies have specifically focused on developing a potential detection strategy for a case in which residual hydrocarbon is present in a water-saturated medium.
- A tank model filled with gravel and sand was designed to allow GPR measurements to be made on the surface before, during, and after water and gasoline injections and fluctuations within the tank. Background GPR measurements were made initially with only water being raised and lowered in the model, and the water table was then raised and lowered beneath a volume of 219 liters of gasoline that was injected into the bottom of the tank. Measurements from the initial raising and lowering of the water with no gasoline present demonstrate the sensitivity of GPR for monitoring changes in subsurface water content and minor fluctuations of the water table. Measurements made during the raising and lowering of the water table with gasoline in the model show differences from the measurements made when only water was raised and lowered, and a comparison of the data show that reflections in GPR data can be enhanced when residual gasoline is present in a water-saturated system because there is less attenuation of the radar signal. Differences in travel times to subsurface reflections between the two stages of the experiment are also caused by the residual gasoline present in the water-saturated medium. Results of this study provide the basis for a strategy that has the potential for successful detection and delineation of LNAPL hydrocarbon-contaminated areas at field

sites where the conditions are similar to those modeled through this experimentation.

Key Words: contaminant detection, ground-penetrating radar, hydrocarbon, light non-aqueous phase liquid, vadose zone.

INTRODUCTION •

To properly remediate contaminated sites of environmental concern, it is necessary to have knowledge of the actual extent of contamination in the subsurface. A considerable amount of research in recent years has focused on the viability of using different geophysical methods to delineate and monitor areas of subsurface contamination. Groundpenetrating radar (GPR) is a technique that has been widely applied for mapping shallow stratigraphy and fractures that influence contaminant transport (Davis and Annan, 1989; Benson, 1995; Martinez et al., 1999) and for aiding in the characterization of contaminated sites by locating buried features of interest (Daniels et al., 1998; Guy et al., 1999; Radzevicius et al., 2000). Although GPR has been shown to have the potential in certain field conditions for delineating the extent of light non-aqueous phase liquid (LNAPL) hydrocarbons (Olhoeft, 1986; Daniels et al., 1995; Grumman and Daniels, 1995), further experimental research that focuses on strategies for detection in additional situations is necessary, as currently the potential usefulness of GPR prior to a survey is difficult to assess.

Analogies can be made between GPR that utilizes electromagnetic energy and the mechanical energy–based seismic reflection technique. However, with GPR, scattering occurs at impedance contrasts produced by changes in electrical properties rather than changes in elastic properties or density. The electrical properties that determine radar velocity, attenuation, and scattering from discontinuities are electrical conductivity, dielectric permittivity, and magnetic permeability. GPR works well in low loss materials (such as sand and gravel), in which displacement currents dominate and attenuation associated with conduction is low (Davis and Annan, 1989), but performs poorly in high conductivity materials (such as clays with a high cation exchange capac-

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ity). In low loss, nonmagnetic mediums, the reflection and transmission coefficients at normal incidence are governed primarily by the relative permittivity (ε_r) of the materials through which the energy is propagating. Few earth materials (unless they are ferrous) have a relative magnetic permeability that deviates significantly from unity; thus, at high frequencies and/or low conductivity, the GPR velocity can be assumed to vary inversely with the square root of the relative permittivity. Therefore, higher permittivity materials result in slower radar velocities.

The potential for GPR to detect LNAPL hydrocarbons such as gasoline can exist under certain conditions because the electrical properties of such contaminants can be quite different than water. When gasoline and air are present in the pore space of dry quartz sand, for instance ($\varepsilon_r \operatorname{air} = 1, \varepsilon_r$ gasoline = 2–3, ε_r dry quartz sand = 5), there is not a significant contrast between these fluids or materials to allow detection of the gasoline. However, in a case where gasoline exists in a system with water present (ε_r water = 81), a significant and detectable contrast can exist in areas where gasoline displaces water relative to areas where no water has been displaced. Air, gasoline, and dry quartz sand are relatively high velocity mediums for propagating radar energy when compared to water, which is a relatively slow medium. A detectable decrease in radar wave attenuation should result in areas where LNAPL hydrocarbons (electrical insulators) displace water, which is a more conductive fluid, although this can be time dependent as in some cases hydrocarbon-contaminated soils have been found to develop increased conductivity with time (Sauck, 1998).

LNAPL hydrocarbon detection strategies using GPR to date have focused on identifying anomalous responses associated with the capillary fringe and the vadose zone above the capillary fringe. Approaches have looked at changes in reflection arrival times, reflection strengths, and changes in signal character associated with areas of contamination. Reflection pull-ups have been demonstrated to be possible in areas where hydrocarbon pools develop or where gasoline displaces residual soil moisture during its descent through the vadose zone (Douglas et al., 1992; DeRyck et al., 1993). Reflection pull-downs can also result from depression of the capillary fringe in areas where contaminant pools develop due to increased travel distance; however, this effect can be counter-balanced by a relatively faster velocity of the contaminant so that no net change in travel time results (Campbell et al., 1996). Bright spots have been observed through experimental work and have been attributed to LNAPLs displacing water in the vadose zone, which can cause a higher permittivity contrast at reflecting interfaces than would exist without the contaminant presence (Redman et al., 1994; Campbell et al., 1996). Decreases in reflection amplitudes over areas of vadose zone hydrocarbon contamination have been reported by several researchers, with mechanisms being proposed that explain how an increase in bulk conductivity can result from microbial degradation of hydrocarbon plumes with time, which attenuates the radar energy–producing areas of reduced signal (Nash et al., 1997; Lucius, 2000).

Physical model and laboratory experiments have provided information relevant to GPR detection strategies for various LNAPL contaminants (Kutrubes, 1986; Lucius et al., 1990; Monier-Williams, 1995), and theoretical models involving LNAPLs have identified various circumstances under which GPR is capable of serving as a useful tool to detect and monitor these types of contaminants (Endres and Redman, 1993; Barber and Morey, 1994; Powers and Olhoeft, 1996). Previous field studies also have provided important information in regards to GPR detection strategies of LNAPL hydrocarbons that originate from spills at the surface and subsequently migrate downward through the vadose zone. However, no work to date has concentrated on GPR experimentation involving an LNAPL plume introduced at depth (with no residual hydrocarbon initially directly above the LNAPL plume) or on the GPR response over a controlled situation in which redistributed hydrocarbon exists in the pore space of a water-saturated medium. The focus of this article is to address this type of situation that often exists at contaminated field sites, through a physical model experiment, and to provide a potential detection strategy that is shown to be successful under controlled conditions.

EXPERIMENTAL METHODOLOGY •

The physical model used for experimentation consisted of a polyethylene cylindrical tank (242-cm diameter; 121-cm height) filled with pea gravel and sand. The tank model configuration is illustrated in Figure 1 as cross-sectional and plan views. A port in the center of the bottom of the tank was installed to allow for the introduction and draining of water and gasoline. Liquids were injected under a low hydraulic gradient using a standard water pump. Feeder tanks adjacent to the tank model were used for filling and also waste disposal purposes (Figure 2). A monitoring well was installed inside the tank close to the wall so that liquid levels in the tank could be monitored. Gravel (0.64- to 0.95-cm grain diameter) filled the bottom of the tank to a thickness of 16.5 cm and was overlain by 90 cm of quartz sand (0.05to 0.1-cm grain diameter). The interface between these two layers had a slight dip to allow it to be easily distinguishable from other horizontal events (such as antenna ring or bottom of tank multiples) in the data throughout the course of experimentation.

Laboratory measurements yielded average porosity values for the gravel and sand of 36 and 31% respectively, and a capillary rise of water in the sand was measured to be 11.4 cm. The gravel and sand were dry when placed in the tank, and both were assumed to be homogeneous and isotropic





layers. A GPR grid consisting of 17 parallel lines each 121 cm long and 7.62 cm apart was situated in the center of the tank over the sand medium, as shown in Figure 1B. The GPR recording system used was a GSSI SIR-10A (Geophysical Survey Systems, Inc., North Salem, NH), and the antenna was a fixed offset, bistatic antenna with a center frequency of 500 MHz (in air). The antenna was positioned at the tank surface using a track so that precise and repeatable measurements could be made along the grid lines (Figure 3). The only post-acquisition processing applied to the data were band-pass filtering to remove low frequency drift ("dewow") and a linear gain. The same processing parameters were used for all data acquired so that relative amplitude information between data acquired at different stages of the experimentation would be preserved.

Background GPR measurements were made prior to any addition of water into the tank and during an initial raising and lowering of the water table with no gasoline present. These measurements provided a baseline for comparison of measurements made during the next phase of the experiment when gasoline was introduced into the model. This initial raising and lowering of the water table also served the purpose of residually wetting the sand in the model to better simulate realistic field conditions for when the gasoline was introduced. The water table was initially raised to a maximum level of 66 cm from the bottom of the tank in three stages, and GPR measurements were made at each of these stages with water levels of 25.4, 35.6, and 66 cm (Figure 4). After measurements at the highest water level (66 cm) were made, the water was drained from the tank, leaving residual saturation of water in the sand. The water level was lowered from its maximum in stages also, and GPR measurements were made at water levels of 30.5, 13.8, and 0 cm during draining (Figure 4).

Prior to the addition of gasoline to the tank, there was \sim 7.6 cm of water at the bottom of the tank within the gravel



FIGURE 2: Experimental setup showing tank model (background left) and one of the feeder tanks (foreground right) used for filling and waste disposal purposes.



FIGURE 3: Tank model and GPR data acquisition setup. Measurements were made using an antenna positioned at the model surface guided by a track to assure accuracy along the grid lines.

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(w = geophysical water table reflection, s/g = sand/gravel interface reflection, b = bottom of tank reflection)

FIGURE 4: Two-dimensional (2D) profiles at different water levels showing effects on the GPR response of raising and lowering the model water table (no gasoline present in model).

layer. This water was the result of pore water exceeding residual saturation in the sand and gravel that migrated downward to the bottom of the tank during the 16 hr after draining the tank of water. This water remained at the time the gasoline was injected so that a spill from a tank or pipe originating at the water table and vadose zone interface or a lateral migration of contaminant along this interface would be best simulated. A volume of 219 liters of gasoline was injected into the model. The gasoline moved upward through the water and spread within the permeable gravel and initially raised the total liquid level to 22.9 cm. Water then was introduced beneath the gasoline to simulate a rise of the water table, raising the total liquid level in stages up to 73.7 cm. The water and gasoline were then drained from the tank in stages. GPR measurements were made over the grid prior to the start of the injection with 7.6 cm of water present during each stage of raising of the water table in the tank with liquid levels of 22.9, 53.3, and 73.7 cm and at each stage of drainage with liquid levels of 35.6, 7.6, and 0 cm (Figure 5).

RESULTS AND DISCUSSION •

Although 17 survey lines were recorded on the surface of the tank for each stage of raising and lowering of the water table in the experiment, only data from GPR survey line 2 are presented in this article. The results did not vary significantly from line to line as data exhibited similar trends; however, the amount of diffracted energy from tank walls



(w = geophysical water table reflection, s/g = sand/gravel interface reflection, b = bottom of tank reflection)

FIGURE 5: Two-dimensional (2D) GPR profiles at different liquid levels (water and gasoline present) showing effects of injecting gasoline into the model and then raising and lowering the water table. ns, nanoseconds.

varied somewhat and was more apparent in some of the other lines that are not presented.

GPR Response during Initial Water Table Fluctuation

GPR two-dimensional profiles at each level of the raising and lowering of the water table are presented in Figure 4. Reflections from the water saturation zone on the two-dimensional profile sections are from the top of capillary fringe above the water table. The difference in relative permittivity between the water table and capillary fringe interface was too small for GPR to detect. Therefore, the capillary fringe was imaged as the geophysical water table and not the water table itself. These background measurements made during the raising and lowering of the water table demonstrate the sensitivity of GPR for detecting small changes in the geophysical water table, although the ability of GPR to detect the water table depends on the frequency used and the material grain size that influences capillary height.

A reflection of interest on the profile sections in Figure 4 is the interface between the gravel and sand layers. When the water table was raised, this reflection became quite faint due to attenuation of the GPR signal resulting from conduction losses associated with the water. The water that was injected into the tank had a measured conductivity of 27 milli-Siemens/m. The gravel and sand reflection shows the same polarity as that of incident wave-form when the boundary between these layers was above the water table (water levels of 0 and 13.8 cm). This is due to the pore spaces being filled with air, a fast fluid, which results in the relative permittivity of the sand being greater than that of the gravel because it has a lower porosity, and therefore it is a slower medium. Conversely, the reflection from this interface shows a polarity reversal when it is submerged beneath the water table (water levels of 25.4, 35.6, 66, and 30.5 cm) because in these cases the relative permittivity of the sand is lower than that of the gravel because water is a slow fluid.

The two-way travel time of the reflection from the geophysical water table during drainage of the tank at a water level of 30.5 cm is somewhat greater than that of the geophysical water table reflection during the raising of the water table at a water level of 25.4 cm, even though the twoway travel distance to this reflector is less when the water level is at 30.5 cm. This anomalous effect is due to the residual saturation of water that remained after the peak level of water in the tank, which resulted in a slower velocity above the capillary fringe and thus a longer two-way travel time to the geophysical water table at 30.5 cm.

GPR Response during Water Table Fluctuation with Gasoline Injection

After the initial raising and lowering of the water table in the tank, 219 liters of gasoline were injected and raised the total liquid level to 22.9 cm (Figure 5). The water table was then raised in stages using a low hydraulic gradient, pushing the gasoline upwards and leaving some residual gasoline beneath the water table. It has been reported that the residual saturation of hydrocarbons in a water-saturated zone under similar circumstances is $\sim 14\%$ of the total pore volume (Wilson and Conrad, 1984). A capillary fringe (water mixed with gasoline) formed above the water table in the sand beneath the gasoline. The water saturation of this capillary fringe was <100%, and the gasoline saturation was greater than was the residual saturation of hydrocarbons beneath the water table. The reflection from the top of the capillary fringe beneath the gasoline throughout this stage of the experiment was detected by GPR as the geophysical water table in a manner similar to the initial raising and lowering of the water table with no gasoline in the tank. Also, the polarities of the reflections from the interface between the sand and gravel show the same pattern as did those during the initial (water only) stage of the experiment (Figure 4). A noticeable reflection did not occur from the gasoline layer on top of the capillary fringe because the relative permittivity difference between the gasoline and the overlying sand (with only residual water and/or gasoline) did not provide a detectable contrast.

Comparison of Data Prior to and Subsequent to Gasoline Injection

The reflections in Figure 5 from the gravel and sand boundary show stronger amplitudes when residual gasoline is present above this interface than do reflections from the same interface in Figure 4 when only water is present above this interface. Figure 6 shows side by side the two-dimensional sections at maximum water levels from Figures 4 and 5 and additionally shows representative one-dimensional traces (traces at 60 cm distance on two-dimensional sections) from these sections. The noticeable reflection differences on the two-dimensional sections and the one-dimensional traces between the measurements involving gasoline versus those involving only water were calculated using the one-dimensional traces. The reflection amplitude with gasoline present was approximately three times greater than without gasoline present, which is equivalent to an increase of 10 dB in reflection power. Although there were differences in this value depending on which trace and line number were used for calculation, measurements still exhibited similar trends, and this calculation is representative of all measurements. This difference in reflection power can be attributed to the presence of the residual gasoline (gasoline is an electrical insulator) that resulted in less attenuation of the propagated energy than when the medium was entirely saturated with water.

In addition to the enhanced reflection strength of the gravel and sand interface, the two-way travel time to this interface is decreased when gasoline is present in the pore

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FIGURE 6: Comparison of maximum water level two-dimensional (2D) sections and one-dimensional (1D) traces (traces from 60 cm distance on two-dimensional section) prior to and subsequent to gasoline injection and water table fluctuation in the model.

(s/g = sand/gravel interface reflection, b = bottom of tank reflection)

spaces of the water-saturated sand. When comparing the arrival times of the gravel and sand reflection in Figure 6 from measurements made with and without gasoline, it can be seen that the reflection with gasoline arrives ~ 3 ns faster. This observed effect makes sense, as the relative permittivity of the water-saturated zone without gasoline above the gravel and sand interface is more than that in the case where residual gasoline (assumed to be $\sim 14\%$) occupies pore spaces in the water-saturated zone. Because the sand medium has a lower relative permittivity when residual gasoline is present in the pore spaces, it has a faster velocity; therefore, a decreased two-way travel time to reflections beneath this medium resulted.

CONCLUSIONS •

Results from the physical tank model experiment show the applicability of GPR for monitoring minor fluctuations of the water table and capillary fringe. It has been demonstrated that the reflection strength of the gravel and sand interface decreased significantly as a result of increased attenuation losses when the water table was raised above this boundary in the initial stage of the experiment involving only water. However, the power of this reflection after injecting gasoline into the model and raising the water table was much greater (10 dB) as a result of decreased attenuation through the medium. Residual gasoline left behind in the pore space as a result of raising the water table also resulted in a velocity pullup (3 ns) of the gravel and sand interface reflection when compared with data acquired prior to the gasoline injection.

These experimental results have demonstrated and explained a mechanism that can potentially allow for GPR to serve as a useful tool for detecting areas of contamination in situations where zones of residual gasoline are present in a water-saturated medium. Less attenuation and faster twoway travel times to reflections beneath the saturated zone may result when compared to adjacent areas where no residual hydrocarbon contamination within the water-saturated zone exists. Accurate detection of hydrocarbon contaminants using GPR is influenced by many factors and has to date often proven to be a tough task in the field. There is much that remains to be understood regarding LNAPL hydrocarbon detection using GPR. Experimental results from this study provide the basis for a strategy that in theory is well-founded and has been demonstrated to be capable of detection under controlled conditions.

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