Application of Electromagnetic Exploration Techniques to Oil Contaminated Soil

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ABSTRACT

The effectiveness of various electromagnetic geophysical exploration techniques for detecting near-surface soil contaminants is described in this paper. Field tests were carried out at an industrial site containing light nonaqueous phase liquid (LNAPL) in soil. Experiment results show that the combination of ground penetrating radar (GPR), electrical resistivity and electromagnetic (EM) exploration provide a means of mapping of subsurface contamination. The region where radar signal amplitude is decreased and high resistivity appears is illustrated over areas with LNAPL contamination under site specific circumstances. Electrical resistivity and EM exploration gave the similar resistivity distribution. A relationship between contaminant saturation and dielectric constant is established based on chemical analysis and modeling of mixed material. It is necessary to understand the geophysical properties of hydrocarbon contaminants for detecting them by geophysical techniques successfully.

Key words: ground penetrating radar (GPR), electrical resistivity, soil contamination

INTRODUCTION

Nonaqueous phase liquids (NAPLs) are hydrocarbons that exist as a separate, immiscible phase when in contact with water and/or air. NAPLs are typically classified as either light nonaqueous phase liquids (LNAPLs) which have densities less than that of water, or dense nonaqueous phase liquids (DNAPLs) which have densities greater than that of water. The most common LNAPL related soil and groundwater contamination problems result from the release of petroleum products (Newell, et al., 1995).

When an LNAPL is spilled at or near the ground surface, it generally infiltrates vertically downward through the vadose, or unsaturated zone as discrete accumulations of the contaminants until it reaches the top of the capillary fringe. These accumulations do not necessarily stay in one place, but they can migrate laterally as a continuous, free-phase layer along the upper boundary of water-saturated zone due to gravity and capillary forces. In general, lateral migration may be expected to be greatest in the direction of groundwater flow (i.e., maximum decrease in water table elevation). These LNAPLs become a continuous source of groundwater contamination. Although LNAPLs have low solu-

bilities, large volumes of groundwater are contaminated by free-phase product slowly dissolving into the surrounding groundwater.

Unambiguous detection of LNAPLs in the subsurface using noninvasive geophysical methods has been growing interest in recent years. The electrical parameters of permittivity and conductivity of LNAPLs may be detectable from the surface for geophysical methods. Air and LNAPLs both have low conductivities and the relative permittivities of 1 and 2-3, respectively, compared to high conductivity and a relative permittivity for water of 81 (Daniels, 1996). Lucius et al. (1992) summarized the electrical properties of toxic substances.

Ground penetrating radar (GPR), electrical resistivity and electromagnetic (EM) exploration are geophysical techniques that are commonly used to investigate the subsurface for engineering and environmental purposes. All of techniques have been widely applied over the last few decades to address the growing need for a non-invasive and cost-effective way to assist in the characterization of contaminated sites. Olhoeft (1986) gave an overview of electrical methods for detecting hydrocarbons. Low frequency techniques particularly are applied to locating high-conductivity fluids, while high-frequency techniques such as GPR are suitable for detecting con-

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trasts in permittivity. Benson and Mustoe (1998) used multiple geophysical methods including electrical resistivity, GPR and very low-Frequency electromagnetic induction (VLF) to help map groundwater contamination produced by hydrocarbons leaking from underground storage tanks. Guy et al. (2000) demonstrated EM data showed low conductivity anomalies correlated with areas of contamination but there were no discernable correlations between GPR data and the extent of contamination at a creosote site. GPR response appears to be discrepant and site dependent in controlled LNAPL spill experiments. An increase and a decrease in GPR amplitude was observed by Campbell et al. (1996) and Redman et al. (1994), respectively. A number of contaminated site studies have identified anomalous GPR responses related to areas of known contamination (Daniels et al., 1995; Grumman et al., 1995; Sauck et al., 1998). Electrical resistivity and EM exploration have been successful in some cases for delineation and monitoring areas that have anomalous subsurface conductivity associated with contamination (Buselli et al., 1990; Monier-Williams, 1990).

In this paper, GPR, electrical resistivity and EM surveys were conducted at a former industrial site due to petroleum contamination within near-surface soil produced by filling oil contaminated soil. The objectives of these geophysical studies were (1) to map the extent of hydrocarbon contamination within near-surface materials, and (2) to estimate the relationship between dielectric constant and oil saturation.

SITE DESCRIPTION

The site of these geophysical investigations is a former industrial site where oil contaminated soil was partly filled. The region filled with oil contaminated soil is indicated by gray color in Figure 1(a). There are three boreholes drilled in this site, and their locations are shown in Figure 1. Two boreholes Q-11 and J-22 are located in the filled area. Based on the core samples at the site, shallow subsurface stratigraphy consists of several layers and the depth of each layer is position dependent, as shown in Figure 1(b). The information revealed by borehole D-11 shows silty deposition down to 5.3 m except a thin sand layer of 60 cm lying at 2 m. But there is a thicker sand layer with the thickness of 2 or 3 m shown by boreholes Q-11 and J-22 due to filled structure.

An investigation combining GPR, electrical resistivity and EM exploration was undertaken at the study site. Two kinds of survey lines were designed at the site (Figure 1). We first conducted a preliminary survey along line F-1. The length of line F-1 is 200 m. Keeping a spacing of 20 cm off line F-1, site F-2 is designed as a cell, 20×30 m as shown in Figure 1. There are 21 lines from line A to line U, running in north-south direction. There are 31 lines, line 0 to line 30, in the west-east direction. The line spacing is 1 m. The established mesh intersects contaminated and uncontaminated areas.

SOIL SAMPLE AND ANALYTICAL RESULTS

Soil samples were obtained from drilling. These three boreholes for the core sampling, D-11, J-22 and Q-11 are shown in Figure 1. The bored core samples in the acrylic tubes were cut to make 161 sample units. Each sample unit was cylinder-shaped, with a diameter of 50 mm and a length of 10 cm. The water content, oil content and resistivity were measured using these soil sample units. A simplified explanation of several soil samples for each sample location shown in Figure 1 is provided in Table 1.

The relative permittivity of soil samples was measured by Time Domain Reflectometry (TDR: TRIEM-ES, P2D) and then the volumetric water content was calculated by Topp's Equation (Topp et al., 1980). Resistivity of soil samples was measured by a bipolar electrode arrangement. The solvent extraction/infrared analysis method was used to measure the concentration of oil content of each soil sample.

The porosity of a rock or soil is simply the fraction of the material volume, i.e. pore space. In quantitative terms the porosity is

ϕ defined as (Fitts, 2002)

$$\phi = \frac{V_v}{V_t} \quad (1)$$

where V_v is the volume of voids in a total volume of material V_t . The volumetric water content is the fraction of space occupied by water in a given volume of material V_t :

$$\theta = \frac{V_w}{V_t} \quad (2)$$

where V_w is volume of water. If a void space of the soil is occupied by air, water, and contaminants, the void volume V_v is equal to

$$V_v = V_a + V_w + V_c \quad (3)$$

where V_a and V_c are the volume of air and contaminants. By dividing the total volume of material, eq. (3) can be altered to

$$\phi = \phi_a + \phi_w + \phi_c \quad (4)$$

where ϕ_s (silt/sand), ϕ_a (air), ϕ_w (water), and ϕ_c (contaminant) are constituent proportions of soil void space. The respective saturations of air (s_a), water (s_w), and contaminant (s_c) are given by

$$s_a = \frac{\phi_a}{\phi}, \quad s_w = \frac{\phi_w}{\phi}, \quad s_c = \frac{\phi_c}{\phi} \quad (s_a + s_w + s_c = 1) \quad (5)$$

We started the estimation of porosity from calculating the masses and volumes of the four phases (air, water, oil and solids). Then we proceeded to determine the unknown quantities as saturation of air (s_a), water (s_w), and oil (s_c). Some parameters, like densities of

(a)

(b)

Fig. 1 (a) Plan view of the experiment site. The preliminary survey line F-1 (heavy line) is 20 cm off from site F-2. Oil contaminated soil was filled in the region indicated by gray color. (b) Borehole stratigraphy. Locations of samples in Table 1 are marked on the borehole sections using S1, S2 and so on.

Table 1 Results of soil sample analysis. Samples correspond to the locations indicated on Figure 1(b).

Samples No.	Lithology	Porosity ϕ	Resistivity (Ωm)	Relative permittivity ϵ_r	Volumetric water content θ	Mass fraction of oil (ppm)
S1	sand	35.9% *	5.0	12.3	23.5%	2900
S2	silt	40.8% **	4.3	22.4	36.65%	2800
S3	silt	41.3% **	5.3	17.7	31%	38000
S4	silt	35% *	5.7	7.7	15.45%	34000
S5	oily silt	45.4% **	71.6	16.1	29%	79000
S6	oily silt	39% *	14.7	14.4	26.6%	29000
S7	sand	36.4% *	35.6	13.3	25%	1900
S8	sand	37.7% *	16.8	15.2	27.6%	6000

* Type I; ** Type II

Fig. 2 The relationship between relative dielectric constant and oil saturation by CRIM model. Dashed line indicates soil Type I ($\phi=0.368$, $s_a=0.27$). Solid line denotes soil Type II ($\phi=0.427$, $s_a=0.02$). Circle and diamond symbols correspond to soil samples of Type I and Type II respectively which are provided in Table 1.

water ($\rho_w=1 \text{ g/cm}^3$), mineral solids ($\rho_s=2.65 \text{ g/cm}^3$), and oil ($\rho_c=0.85 \text{ g/cm}^3$), should be known beforehand.

The porosity ϕ of the silt/sand deposition is given by eq. (4) which ranges from 35% to 45%. In order to simulate the electromagnetic properties of the soil made of silt and sand, and partially saturated with air (gas), water and contaminant, we separate the soil samples (Table 1) into two types according to the porosity. Type I gives an average porosity of 0.368, and Type II of soil samples has a higher average porosity equal to 0.425. The average of air saturation (s_a) in soil Type I is 0.27 and that is 0.02 in soil Type II.

Complex refractive index model (CRIM) (Wharton et al., 1980) is commonly used for analyzing petrophysical parameters which for a three-component system of soil particles, air, and water can lead to the relative dielectric constant of the mixed material as

$$\sqrt{\epsilon_r} = (1-\phi)\sqrt{\epsilon_s} + \phi s_w \sqrt{\epsilon_w} + \phi(1-s_w)\sqrt{\epsilon_a} \quad (6)$$

where ϵ_s , ϵ_w , and ϵ_a denote the relative dielectric constant of soil particles, water and air. The three-component CRIM model has been used in several studies (Alharthi and Lange, 1987; Roth et al., 1990), and other researchers have used a four-component version to model sand-clay mixtures (Wharton et al., 1980).

It is important to establish a relationship between relative dielectric constant and oil saturation (s_c) under typical field conditions. CRIM model shows a relationship between water saturation and dielectric constant. The analytical results of soil samples gave porosity and air saturation. The oil saturation is equal to subtraction of air and water saturation from 1 ($s_c=1-s_a-s_w$). Then a relationship between oil saturation and dielectric

constant can be obtained by CRIM equation. For $\epsilon_w=81$, $\epsilon_a=1$, and $\epsilon_s=5$ (Daniels, 1996) the relationship between oil saturation and dielectric constant is established in Figure 2. The dashed line corresponds to soil Type I ($\phi=0.368$, $s_a=0.27$) and the solid line denotes soil Type II ($\phi=0.427$, $s_a=0.02$). For instance, if $s_c=0.2$, $s_w=1-s_a-s_c$, would be equal to 0.53 in Type I and 0.78 in Type II, giving relative dielectric constants of 11.1 and 18.8, respectively. The dielectric constants of soil samples measured by TDR are shown by circles (Type I) and diamonds (Type II). They show the correlation with CRIM model. Soil fully saturated with water ($s_w=1$) has a permittivity of 25.9 (Type II, $\phi=0.427$). Carcione and Seriani (2000) simulated the electromagnetic properties of a soil model and tested with the results of the Borden experiment (Greenhouse et al., 1993). They showed the permittivity of 25 of soil with a porosity of 0.4 fully saturated by water. The relationship of dielectric constant and water saturation, air saturation, and contaminant saturation will help understanding the electromagnetic properties of soil under site specific circumstances. Also, it gives the theoretical limitation of detection by geophysical exploration.

GPR SURVEY

GPR data were acquired using a GPR system (RAMAC, MALÅ Geoscience, Sweden) with 100 MHz antennas. This system employs physically separable transmitter and receiver antennas, which were set at a fixed separation of 1 m. Four GPR experiments were conducted at the site in the different time. Radar data were collected along line F-1, and on 21 running north-south lines (line A-U) in October 2003 and 27 running

west-east lines (line 2–28) in January 2004 at site F-2, respectively. The standing surface water present at the site due to the heavy rain previous night had an effect on the GPR response in the first three experiments. The quality and repeatability of GPR data depends on the short-term rainfall history prior to data collection (Daniels et al., 1995). Only GPR data acquired from the last experiment, in May 2005 survey are discussed and presented in this paper. Profile data were collected in a common offset configuration. Survey lines (west-east running) are 20 m long with measurement increment every 0.1 m.

Figure 3 shows GPR profiles acquired along line 11 and line 22. Three borehole locations were marked in this figure. The GPR reflection from the interface of the upper silt and the upper sand layer is continuous and horizontal along line 22, which is marked by dashed line in Figure 3(b), but it is discontinuous and ambiguous along line 11 (Figure 3(a)). Information from borehole J-22 show that the thickness of the upper silt layer is 1.8 m. The average EM wave velocity in the upper silt and

sand layer could be estimated as 0.13 m/ns and correspond to ϵ_r of 5.3. This velocity was used to modify two way travel time to depth for GPR data.

Borehole information show us that lower silt and the lower sand layer is contaminated by oil. It corresponds to the depth ranging from 2.5 m to 5.5 m. The water table depth is denoted at 4 m in the boreholes. Note that the water table does not yield a radar reflection in GPR profiles. Annan and others (1991) reported that mapping and detecting the water table with GPR is effective when the ratio of the transition zone thickness to the radar wavelength is in the range of 0 to 0.3. In this case the radar wavelength of the 100 MHz center frequency antenna was approximately 1.3 m in the subsurface soil. The thickness of transition zone should be more than 0.4 m in consideration of the soil texture (pore size distribution, sorting of grain size) at this site. It may be reasonable explanation for the absence of radar reflection from the water table.

Some studies have reported the attenuation of GPR signal amplitude in the vicinity of gasoline concentration

Fig. 3 GPR profiles along line 11 (a) and line 22 (b). Borehole locations are marked and the dashed line in (b) denotes the interface between the layers of upper silt and upper sand.

Fig. 4 Instantaneous amplitude displayed by horizontal slices of GPR data at depths of 2.5 m, 3 m, 3.5 m, and 4 m. Borehole locations are marked. The areas marked by dashed circles with decreased signal amplitude correspond to the high resistivity region marked by dashed circles in Fig. 6.

in the capillary fringe above the water table and describe a 'shadow', or 'halo', or 'muted' appearance coincident with areas of known contamination (e.g., Daniels et al., 1992; Grumman and Daniels, 1995; Maxwell and Schmok, 1995; Sauck, et al., 1998). It is ambiguous of GPR response to the contaminants from the vertical profiles due to radar signal attenuation and the absence of radar reflection from the water table at this site. This means it could be difficult to distinguish whether the 'shadow', or 'halo', or 'muted' appearances exist in the vertical profiles as described by above literatures.

GPR data acquired along parallel lines permit to establish three-dimensional image. Horizontal slice may be a good way to display the amplitude increasing or decreasing clearly with depth and, further, to show context clue to the distribution of contamination. The amplitude shown in horizontal slices in Figure 4 was instantaneous amplitude (Taner et al., 1979) obtained from complex trace calculated using the Hilbert transform.

ELECTRICAL RESISTIVITY AND EM SURVEY

Electrical resistivity and EM techniques both measure electrical properties of the earth, and hence both are used for many of the same applications. Haryu et al. (2004) has reported electrical resistivity and EM survey along line F-1 and in site F-2. We summarize the results as described by Haryu et al. (2004).

A west-east resistivity profile was acquired along line F-1 using an electrode spacing of 2 m. The shallow subsurface extending to depth 5–7 m shows the resistivity from 5 to 20 Ωm , and 2–5 Ωm below. Relatively high resistivity layer indicates sandy soil, and low resistivities correspond to the clayey soil and mudstone below. There are four boreholes along line F-1. Compared to the borehole information, the high resistivity distribution ($>10 \Omega\text{m}$) above the water table corresponds to an oil plume.

Electrical resistivity and EM data were acquired in north-south direction (line A-U) along 21 lines with the length of 30 m. The data show similar results from both

Fig. 5 Vertical slices of resistivity distribution by electrical resistivity survey along line D (a), line J (b), and line Q (c) in site F-2. Borehole locations are marked in figures.

methods but EM data gave smaller resistivity values than electrical resistivity data. Figure 5 shows vertical resistivity distribution along lines D, J, and Q at site F-2 by electrical resistivity. The vertical profiles indicate the resistivity change from low to high along the direction from west to east. It resulted in GPR data acquired in the direction with resistivity increasing, that is in west-east direction, perpendicular to the direction of electrical resistivity and EM data acquisition.

The horizontal slices of resistivity distribution observed by electrical resistivity survey are shown in Figure 6. High resistivity ($>10 \Omega\text{m}$) areas distributed similarly in the slices of different depths. It located from 5 m to 30 m in the north-south direction and from 5 m to

20 m in the west-east direction.

DATA INTERPRETATION AND DISCUSSION

Organic contaminants like crude oil generally have a high electrical resistivity and low dielectric constant. Theoretical studies and studies on controlled and laboratory experiments report high resistivities and enhanced GPR reflection from the water table in areas having hydrocarbon contamination (Campbell, et al., 1996; Daniels et al., 1992; Redman et al., 1994). However, studies at real sites often reach quite different results (Daniels et al., 1992; Grumman and Daniels, 1995; Maxwell and Schmok, 1995; Sauck et al., 1998). They

Fig. 6 Horizontal slices of resistivity distribution by electrical resistivity survey at depths of 2.5 m, 3 m, 3.5 m and 4 m. The location of borehole D-11 is marked. High resistivity ($>10 \Omega\text{m}$) zone corresponds to the filled region, the gray zone in Fig. 1. The areas marked by dashed circles coincide with the areas of decreased GPR signal amplitude marked in Fig. 4.

reported a distinctive, 'muted' or 'fuzzy' GPR response over an LNAPL contaminant zone. They suggested the attenuation of EM wave be caused from contaminant vapor effects (Grumman and Daniels, 1995) and from the effective roughness of mixture with water and gasoline (Maxwell and Schmok, 1995). Further, it has been reported that areas of hydrocarbon contamination have high resistivity (Benson and Mustoe, 1998) and low conductivity (Guy et al., 2000). However, different results obtained by Monier-Williams (1995) and Gajdos and Kral (1995) show high apparent conductivities and low apparent resistivities. Sauck (1998) explained the changes from electrically resistive to conductive behavior with time caused by hydrocarbon spills due to biodegradation.

In this study, we interpret results by integration of different geophysical data including GPR response, resistivity and borehole information. The decreased GPR signal areas in Figure 4 have quite similar locations when compared with the high resistivity regions in Figure 6. For example, the low amplitude areas marked

by dashed circles in Figure 4 correspond to the same locations with high resistivity ($>10 \Omega\text{m}$) in Figure 6. The borehole information revealed that high level oil contaminated depth begins from about 2.5 m (see Table 1). The areas of low amplitude and high resistivity marked by dashed circles are verified as the contaminated areas known from borehole information. Further, the high resistivity ($>10 \Omega\text{m}$) zone, from 5 m to 30 m in the north-south direction and from 5 m to 20 m in the west-east direction, in Figure 6 corresponds to the filled region, and larger area may imply expansion of contamination. In addition, we expect the zone around borehole D-11 ranging from a depth of 3 m to 3.5 m in Figure 4 as a contaminated area, because of the decreased GPR signal and known borehole information, whereas low resistivity is shown at this area in Figure 6. The stratigraphy in the area around borehole D-11 is silty, but the region of high resistivity shows sandy structure as described in the previous section. The silty layer has a lower resistivity than the sandy layer as shown in Table 1. This explains why the area around borehole D-11 is oil

contaminated but has low resistivities.

According to site specific circumstances, we suggest the contaminated zone be mapped by decreased radar signal amplitude and high resistivity. The reason for the decrease in the signal amplitude is explained by the roughness of mixture with water and LNAPL. Results of soil sample analysis show that the water and LNAPL do not distribute throughout the vadose zone in a uniform, homogeneous manner in this case. The reflected signal becomes more dispersed due to the roughness of the mixture with water and LNAPL.

CONCLUSIONS

The exploration and determination of the hydrocarbon contamination (in this case LNAPL) is one of difficult issues in environmental studies. Some field investigation report results that contradict the controlled experiment, but in practice some results should be considered as different aspects of the same phenomenon. Geophysical techniques combining GPR, electrical resistivity and EM exploration provided an effective noninvasive method for locating the LNAPL contamination in the subsurface by paying close attention to site specific circumstances.

A relationship between the contaminant saturation and the dielectric constant is established from chemical analysis of soil samples and theoretical modeling of a mixed material. It is necessary to understand the geophysical properties of hydrocarbon contaminants for detecting them by geophysical techniques successfully. The correlation between the decreased radar signal amplitude, high resistivity distribution and LNAPL contamination is illustrated by GPR and electrical resistivity data. The resistivity results are quite similar from electrical resistivity and EM survey. The EM method should be suitable for preliminary survey because of portability, survey speed, and high data density.

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石油系土壌汚染調査における電磁探査技術の適用性

鹿 琪・佐藤 源之

要 旨

地下浅層に油系土壌汚染が存在する工場跡地において、物理探査技術の有効性を検証するために実験を行った。実験結果により、地中レーダ (GPR) および電磁気探査の組み合わせは汚染問題の検証について有効な手段である事を示した。

電気探査及び電磁探査による比抵抗構造は大局的に一致している。比抵抗が高くレーダ信号が弱い領域は LNAPL による汚染区域と考えられる。また、土壌サンプルにより、CRIM モデルを用いて誘電率、土壌油分含有量との関係性を調べた。このように、汚染物質の物理性質を把握することにより、物理探査技術を用いて土壌汚染構造の検証が可能であると考えられる。