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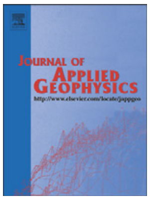

Temporal GPR imaging of an ethanol release within a laboratory-scaled sand tank

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ABSTRACT

Within the last decade efforts in geophysical detection and monitoring of fossil fuel releases into the subsurface have shown increasing success, including the ability to geophysically detect and delineate enhanced and natural biodegradation and remediation activities. The substitution of biofuels, such as ethanol, for fossil fuels is becoming persistent in the national and international marketplaces making it subject to the same types of accidental releases and exposure scenarios currently associated with the transport and storage of fossil fuels. Thus, there is interest from both academics and regulators to investigate the feasibility of applying geophysical methodologies to biofuel releases. In this study, we performed experimental and numerical investigations on the feasibility of using ground penetrating radar (GPR) to monitor the migration of an ethanol release. A tank scale model of a closed hydrologic system was prepared with Ottawa sand and instrumented with an automated gantry measurement apparatus for time-lapse measurement of zero offset and coincident GPR reflections on multiple horizontal planes. Measurements were acquired in the unsaturated and saturated zones throughout the injection and transport of the ethanol release. The results of the monitoring suggest a measureable contrast within both time and frequency domains of the GPR data coincident with the ethanol release and subsequent migration. We conclude that the monitoring of ethanol in a sand matrix at various levels of saturation is possible with GPR.

1. Introduction

Ethanol use has grown in recent years due in large part to its inclusion in fuels as an emission reducing fuel oxygenate. The additional draw to ethanol has been to replace methyl tertiary-butyl ether (MTBE) as a fuel oxygenate in reformulated gasoline (up to 10% by volume in gasoline) due to the solubility of MTBE in groundwater and its carcinogenic effects (Wheals et al., 1999). “Flex Fuel” vehicles utilizing E85 (85% ethanol, 15% gasoline) have also contributed to the use and distribution of ethanol. While ethanol is not considered to be directly harmful to human health, its use has indirect consequences through secondary environmental effects.

The potential for ethanol to also impact groundwater has been the focus of many studies in recent years. Researchers have found that the addition of ethanol to the subsurface in the presence of benzene, toluene, ethylbenzene, and xylenes (BTEX) reduces natural attenuation of the harmful BTEX compounds because ethanol is preferentially biodegraded (Corseuil et al., 1998; MacKay et al., 2006; Powers et al., 2001; Ruiz-Aguilar et al., 2003). Additionally, ethanol has cosolvency effects on existing non-aqueous phase liquids (NAPL) allowing transport and partitioning of harmful and otherwise immobile chemicals in the subsurface (Da Silva and Alvarez, 2002; Frietas, 2009; Gomez and Alvarez, 2009; McDowell et al., 2003). Gasoline mixed with ethanol has also been shown to penetrate clay layers that would otherwise be impenetrable to gasoline alone (Stallard et al., 1997). Multiple studies have shown the degradation of ethanol to result in methane production at potentially hazardous levels (Frietas et al., 2010a,2010b; MacKay et al., 2006).

Physical property differences between ethanol and water suggest that geophysical imaging methods such as resistivity, induced polarization, and high frequency electromagnetic methods can provide a means of differentiating between water-saturated pore spaces and ethanol-saturated pore spaces in the subsurface (Glaser et al., 2010, 2011; Henderson et al., 2010; McNaughton et al., 2009; Personna et al., 2011a,2011b). Lucius et al. (1992) showed ethanol to be miscible in water, with a density of 0.79, a relative dielectric permittivity (dielectric) of 25 (at 20 °C), and a frequency dependent response. When compared with the dielectric of water (80), there should be sufficient contrast to detect the presence of ethanol (Glaser et al., 2010, 2011; Henderson et al., 2010). Additionally, even though ethanol is known to be miscible in water, laboratory experiments have shown that ethanol can be retained within the capillary fringe rather than it being infiltrated into the saturated zone (Frietas and Barker,

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2009; Glaser et al., 2010, 2011; Henderson et al., 2010). A reduced dielectric permittivity should be observed in areas where ethanol and water readily mix, while areas previously occupied by air (dielectric of 1) in the pore space should also demonstrate an increase in the dielectric due to the wetting of pore space by the ethanol (Farmani et al., 2008; Hagrey and Muller, 2000).

In this work we investigated an ethanol release with ground penetrating radar (GPR), with the main objective to demonstrate that the method is capable of discerning the movement of ethanol within a sandy host environment. GPR is a high resolution electromagnetic geophysical method capable of discerning contrasts in bulk dielectric based on varying volumetric mixtures of soil, air, water, and ethanol. The release was conducted in a highly controlled setting, i.e., a sand tank with known geometrical, hydrological, and electrical parameters (Bano, 2006; Benedetto, 2010; Birken and Versteeg, 2000; Loeffler and Bano, 2004; Mazella and Majer, 2006; Versteeg, 2004; Versteeg and Birken, 2001). Mixing models and FDTD modeling were also completed in an effort to understand the individual contributions to the resulting GPR profiles from ethanol mixing and tank geometry (Bano et al., 2009). The GPR models were run with different dielectric values to span the range of expected laboratory conditions. For the laboratory experiment, we used variations in reflected electromagnetic wave amplitude, two-way travel time of the wave, and power spectra to make assessments of ethanol in the saturated zone, unsaturated zone, and within the capillary fringe. With this laboratory study, the GPR method will be shown to have sufficient sensitivity to warrant additional investigations at the field scale.

2. Ground penetrating radar wave propagation and analysis

GPR utilizes electromagnetic radio waves in a frequency range of 50 Mhz–2 Ghz, which are emitted by a transmitting antenna. The resulting reflected and refracted waves are recorded by a receiving antenna that measures the voltage amplitude over time, called a trace. GPR is used extensively to investigate both near surface soils as well as a range of engineered structures such as roads and bridges. Propagation and reflection of the radio wave through any medium depend on the dielectric and other electromagnetic properties of the medium.

GPR measurements can be made in a transmission configuration or a reflection configuration. The transmission configuration generally measures the volume of the earth (or sample) located between the two antennas, usually borehole to borehole. The reflection configuration relies on reflections resulting from contrasts in the dielectric properties of the soil for antennas on the same soil plane, for example on the ground surface or within a single borehole.

2.1. Wave propagation theory

The propagation of GPR energy into the subsurface can be described using Maxwell's equations (Carcione, 1996; and Greaves et al., 1996). The factors which control the arrival time and shape of the waveform are the velocity and attenuation of the wave in the medium (Davis and Annan, 1989). The velocity (V_m) of radio waves is dependent upon the relative dielectric permittivity (ε_r) and the relative magnetic permeability (μ_r) in proportion to the speed of light in free space ($c = 299.8$ mm/ns):

$$V_m = \frac{c}{\sqrt{\left[\left(\frac{\varepsilon_r \mu_r}{2}\right) \times \left[\left(1+P^2\right)+1\right]\right]}} \tag{1}$$

where the loss factor is represented by P, generally expressed as:

$$P = \frac{\sigma}{(2\pi f) \times (\varepsilon_r \varepsilon_0)}. \tag{2}$$

Frequency is denoted as f, σ is the electrical conductivity, and ε_0 is the dielectric permittivity of free space (8.854×10^{-12} F/m). In non-magnetic materials, μ_r is approximately 1 (Reynolds, 1997). Eqs. (1) and (2) are valid only for real values of permittivity and conductivity. Furthermore, Davis and Annan (1989) show that in low-loss geologic materials, P is approximately zero, reducing Eq. (1) to:

$$V_m = \frac{c}{\sqrt{\varepsilon_r}}. \tag{3}$$

For heterogeneous materials of contrasting dielectric values, a reflection coefficient (R) at a given interface with no signal loss, and for the case of normally incident signal only, is quantified by Conyers and Goodman (1997):

$$R = \frac{\sqrt{\varepsilon_a} - \sqrt{\varepsilon_b}}{\sqrt{\varepsilon_a} + \sqrt{\varepsilon_b}} \tag{4}$$

where the relative dielectric constants of the layer on either side of the interface are represented by ε_a and ε_b, respectively. The transmission coefficient, T, is simply:

$$T = 1 - R. \tag{5}$$

It is important to note that the transmission coefficient calculation as presented is also only valid for vertically incident waves. GPR signals are attenuated through multiple mechanisms. Every reflection at a dielectric interface results in some loss of energy available for the deeper reflections, thus attenuating the signal (Reynolds, 1997). Signal loss due to refraction along a high velocity interface, such as that of air and the ground surface, can also occur (Rucker and Ferré, 2003). Additional modes of signal loss include: signal scattering due to inhomogeneities within the medium (Benedetto, 2010; Doolittle and Collins, 1995); scattering from thin layers or point sources like cobbles (Davis and Annan, 1989); objects that have dimensions comparable to the signal wavelength, i.e., Mie scattering (Bano, 2006; Reynolds, 1997), or the many facets of pore specific conditions, including but not limited to, pore structure, pore shape, pore fluid distribution, and pore fluid chemistry (Bano, 2004; Chen and Or, 2006; Cosenza et al., 2003; Endres and Bertrand, 2006; Friedman, 1998; Jones and Friedman, 2000; Kenyon, 1984; Sen et al., 1981, 1984; Shen et al., 1985; and Tyc et al., 1988).

The frequency-dependent attenuation factor (α) that provides the attenuation for a specific frequency for a given medium with known conductivity, magnetic permeability, and dielectric properties is represented by:

$$\alpha = 2\pi f \left\{ \left(\frac{\varepsilon_r \mu_r}{2}\right) \left[\left(1 + \frac{\sigma^2}{(2\pi f)^2 \times \varepsilon_r^2}\right)^{1/2} - 1 \right] \right\}^{1/2}. \tag{6}$$

Again, assuming a low loss medium, Davis and Annan (1989) show that Eq. (6) can be simplified as:

$$\alpha = \frac{\left(1.69 \times 10^3\right) \cdot \sigma}{\sqrt{\varepsilon_r}}. \tag{7}$$

The attenuation factor is given in dB/m, and gives the rate at which electromagnetic energy is dissipated into the ground. Additionally, the wave amplitude will decrease inversely with distance from the source due to geometric spreading (Annan, 2001). Since the presence of pore water will affect both the dielectric and the conductivity of the medium, the attenuation factor of a soil has been used to estimate soil water content (Olver and Cuthbert, 1988). Within the context of this study, the relative attenuation factor differences and reflection amplitude values are likely to be the most descriptive variables for detecting the presence (or absence) of ethanol throughout the soil column.