Measuring the dielectric properties of soil–organic mixtures using coaxial impedance dielectric reflectometry

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ABSTRACT

Contamination of soils with non-aqueous phase liquids (NAPLs) is frequently produced by accidental spills and storage tanks or pipes leakage. The main goals dealing with soil and groundwater contamination include determining the extension of the affected zone, monitoring the contaminant plume and quantifying the pollution degree. The objective of this work is to evaluate the potential of dielectric permittivity measurements to detect the presence of NAPLs in sands. Tested samples were fine, medium, coarse and silty sand with different volumetric contents of water and paraffin oil. The dielectric permittivity was measured by means of a Coaxial Impedance Dielectric Reflectometry method in specimens with either known fluid content or at different stages during immiscible displacement tests. A simplified method was developed to quantify the amount of oil from dielectric permittivity measurements and effective mixture media models. Obtained results showed that groundwater contamination with NAPL and the monitoring of immiscible fluid displacement in saturated porous media can be clearly identified from dielectric measurements. Finally, very accurate results can be obtained when computing the contamination degree with the proposed method in comparison with the real volumetric content of NAPL ($r^2 > 90\%$).

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1. Introduction

Soils are porous media composed by multiple phases including mineral particles, water and air. Organic contaminants in contact with soils divide between the water, air and mineral phases and sometimes form a separate phase which is recognized as non-aqueous phase liquid (NAPL). Detection of NAPL and the monitoring of contaminated plumes are of fundamental importance in geoenvironmental studies.

The effort to detect organic contaminants in soils is increasing since the last ten years. The monitoring and mapping of contaminated soil frequently involve geophysical prospecting methods as complement of conventional geological, chemical and hydrological studies (Garcia–Gonzalez et al., 2008; Knight, 2001). Non-destructive geophysical techniques help to rapidly detect contaminants in soils and to monitor the extension of polluted areas (Carcione et al., 2003). Electric methods based on the propagation/reflection of electromagnetic waves have been successfully used in the past to detect organic contaminants in soils (Ajo-Franklin et al., 2006; Shinn et al., 1998). However, detection is restricted to sites with spatial and temporal variations of the complex dielectric permittivity of soils and high contamination levels (Brewster et al., 1995; Hwang et al., 2008; Kaya and Fang, 1997; Wilson et al., 2009).

Dielectric measurements are useful to estimate water and oil content within porous media (Liu and Mitchell, 2009; Persson and Haridi, 2003; Topp et al., 1980). The presence of liquid organic contaminants in soils can be identified due to their different dielectric, physical and chemical properties with respect to pore water (Ajo-Franklin et al., 2006; Moroizumi and Sasaki, 2008; Son et al., 2009). Although previous studies showed that NAPL affects the complex permittivity of soils, little concern has been given to the effect of medium heterogeneities and the spatial variability of NAPL saturation on computed concentrations of contaminants.

The purpose of this study is to investigate the correlation between the volumetric content of NAPL and the dielectric permittivity of sandy soils. The objectives include the analysis of the dielectric behavior of saturated sands contaminated with NAPL, the monitoring and detection of anomalies and trapped oil by measuring the dielectric permittivity with a Coaxial Impedance Dielectric Reflectometry sensor, and the development of a simplified procedure to determine the volumetric content of NAPL by means of dielectric permittivity measurements. These will provide new results useful to interpret geophysical field data and to quantify the presence of organic contaminants by means of non-destructive dielectric measurements.

2. Complex permittivity of soils

The relative permittivity (κ^*) of heterogeneous porous media is a complex parameter defined as:

$$\kappa^* = \frac{\varepsilon^*}{\varepsilon_0} = \kappa' - j \; \kappa'' \tag{1}$$

where the real and imaginary components are:

$$\kappa' = \frac{\varepsilon'}{\varepsilon_0} \tag{2}$$

$$\kappa'' = \frac{\varepsilon''}{\varepsilon_0} + \frac{\sigma_0}{\omega}\frac{1}{\varepsilon_0} \tag{3}$$

and $j = \sqrt{-1}$, ε' and ε'' are real and imaginary permittivity, respectively, ε_0 is the free space permittivity (8.85×10^{-12} F/m), ω is the angular frequency and σ_o is the conductivity at zero frequency (DC).

The permittivity of soils depends on the volumetric content and permittivity of each phase, particle shape and orientation, phase distribution, interaction between phases and the frequency of measurements (Endres and Knight, 1991; Robinson and Friedman, 2002; Wobschall, 1977). In the megahertz frequency range a relaxation of soil permittivity arises when electrical charges accumulate at interfaces and borders (Arulanandan and Smith, 1973). This mechanism can be very important in fine soils (silts and clays) but has irrelevant significance in water-wet sands (Rinaldi and Francisca, 1999; Saarenketo, 1998), dry soils (Ulaby et al., 1990), and soils saturated with NAPL (Francisca and Rinaldi, 2003).

Effective medium models consider that the effective complex permittivity (κ^*_{eff}) of phase mixtures depends on the permittivity and volumetric content of each of phase (Kärkkäinen et al., 2000; Van Beek, 1967). Different mixing models predict different effective permittivity for the same soil. The simplest mixture formulae consider that:

$$\kappa^{*c}_{eff} = \sum_i \frac{V_i}{V} \kappa^{*c}_i \tag{4}$$

where κ^*_i and V_i are the permittivity and volume of the 'i' phase respectively, V is the total volume, and c is a constant that varies from -1 to 1 depending on the geometrical arrangement of the components (Lichtenecker and Rother, 1931). The fitting parameter c usually ranges from 0.4 to 0.8 for soil mixtures (Jacobsen and Schjønning, 1995), when $c = 0.5$ the model is known as Complex Refractive Index Mixture (CRIM) and applies for isotropic mediums, and if $c = 1/3$ the Eq. (4) becomes equal to the Looyenga's model (Van Beek, 1967). Francisca and Rinaldi (2003) compiled effective medium models frequently used for analyzing the dielectric permittivity of soils. These models can be used to predict the dielectric properties of contaminated soils by extending them to four-phase mixtures (particles, water, contaminant and air). Recently, Seyfried et al. (2005) presented a simplified equation to determine the water volumetric content of soils from the measured dielectric permittivity as follows:

$$\theta_w = A\sqrt{\kappa^*_{eff}} + B \tag{5}$$

where A and B are fitting parameters. These authors successfully fitted the model to 19 different soil samples capturing the influence of saturation and soil texture on dielectric permittivity.

The displacement of immiscible liquids inside the pores modifies the complex permittivity of soils. The dielectric properties of NAPL are characterized by $\kappa' \approx 2$ and $\kappa'' \approx 0$ due to the low polarizability and conductivity of these liquids, which results in a low capacity to dissolve in water and associate with other elements. The NAPL displaces either air ($\kappa' = 1$; $\kappa'' = 0$) or water ($\kappa' \approx 78.5$ and $\kappa'' > 0$) when penetrating in the soil pores and consequently modifies κ^*_{eff} (Darayan et al., 1998; Kaya and Fang, 1997; Son et al., 2009). When NAPL ($\kappa' \approx 2$) displaces water ($\kappa' \approx 78.5$) in saturated soils, the change in dielectric permittivity is easily detected (Rinaldi and Francisca, 2006). However, detection of immiscible displacement becomes difficult in the case of unsaturated soil when a liquid with $\kappa' \approx 2$ replaces the air ($\kappa' = 1$) in the soil pores. In this case, the initial dielectric properties of the soil and a complimentary measurement of

contamination are required. The phase distribution and wetting conditions can also affect the permittivity of soil–water–organic mixtures (Santamarina and Fam, 1997). This phenomenon is significant when testing clay–water–oil mixtures given that the presence of oil and the mixing order can affect the development of the diffuse double layer around clay particles.

Commonly employed techniques to measure soil's dielectric permittivity in the megahertz frequency range include time domain reflectometry (TDR), ground penetrating radar (GPR), resonant frequency modulation devices, coaxial transmission lines, and impedance-network analyzers. The three first techniques can be applied both, at field or laboratory scale, while the last ones provide very accurate laboratory data (Benson and Bosscher, 1999; Cassidy, 2007; Francisca and Rinaldi, 2003; Knight, 2001). There were many efforts to correlate and upscale dielectric permittivity measurements from the field to the laboratory with limited success (Brewster et al., 1995; Carcione et al., 2003). The main identified problems are related to the spatial variability of soil properties and biodegradation effects (Johnston et al., 2007; Kao and Prosser, 2001,).

Geoenvironmental applications of geophysical techniques based on electromagnetic wave propagation frequently include detection of contaminant in soils and monitoring of contaminant plumes. The spatial variability of soil dielectric impedance, which is mainly controlled by the spatial variability of soil type, porosity and moisture content, is the main limiting factor for the identification of dielectric features that could be associated with contaminants in soil pores. However, if the dielectric permittivity of an aquifer at a given place is known before the infiltration of organic contaminants, any change in dielectric properties can be associated with the contaminant migration.

3. Laboratory methods

3.1. Dielectric permittivity measurements

The complex dielectric permittivity was measured with the Hydra Probe Coaxial Impedance Dielectric Reflectometry probe manufactured by Stevens Water Monitoring Systems Inc. This sensor has four metal rods or tines; three of them define a cylindrical measurement volume of 2.5 cm in diameter and 6 cm in length, and the fourth rod is in the center of this sensing volume [Fig. 1(a)]. The tines are used to measure the complex dielectric permittivity and a temperature sensor is placed in the probe head. The probe can be placed under the water table and in contact with contaminant substances for very long times without suffering damage.

The equipment generates a 50 MHz electromagnetic wave that propagates as a planar waveguide to the tines and reflects from the sensing volume creating a standing wave as in a coaxial transmission line. The reflected signal depends on the probe impedance which in turn depends on the dielectric properties of the material in the sensing volume (Campbell, 1990).

The sensor accuracy to measure the real and imaginary component of the dielectric permittivity of the material in the measurement region is $\pm 1\%$ (or ± 0.5). The dielectric permittivity response of the sensor is linear with the contact length of the rods with the tested material (Seyfried et al., 2005), which allows identifying anomalies when the measurement are conducted by gradually inserting the probe in the soil.

The dielectric permittivity is determined from three measured voltages, but unfortunately, the equation that relates the voltages to the dielectric permittivity is not provided by the manufacturer (Blonquist et al., 2005). The volumetric water content, soil salinity, soil conductivity and pore water conductivity are computed from the real and imaginary dielectric permittivities (Stevens Vitel, 1994). The soil type under test should be selected given that the equipment has three specific calibration curves, for sand silt and clay, to compute moisture content.