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Temporal evolution of the geoelectrical response on a hydrocarbon contaminated site

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ABSTRACT

A geoelectrical campaign was initiated in 2009 to delineate a massive hydrocarbon spill, which occurred because of a petroleum pipe breakdown in 2009. These measurements have been compared with both field resistivity measurements made in 2009 and with laboratory measurements. From a physicochemical point of view, a hydrocarbon contamination has to be understood as a spatially and temporally varying object, responsible for a change in geoelectrical response. To evaluate the signal measured on site, geoelectrical laboratory measurements were performed on the petroleum oil extracted from the site during two stages of degradation. On the laboratory scale, the non-degraded oil shows an increase in resistivity, normalized chargeability and quadrature conductivity with oil content, whereas the degraded oil indicates a slight decrease in resistivity, but no modification of the phase-lag and chargeability parameters. In the field, resistivity measurements performed in 2009, just after the pipe breakdown, show weak changes in resistivity measured over the contaminated area. However, between 2009 and 2012, biodegradation of the oil has led to a clear decrease in the resistivity within the impacted zone. No variations in normalized chargeability or quadrature conductivity were measured in 2012 between the contaminated and the non-contaminated areas, despite the presence of biofilms. In the field, the studied hydrocarbon contamination under degradation appears not to modify the capacitive part of conduction, but rather it concerns the ohmic part of conduction. The field and laboratory measurements led us to the conclusion that to choose the most discriminatory and efficient geophysical parameters, it is necessary to have a priori information about the oil (i.e. non-degraded or partially degraded). In the present case study, there is no need to acquire chargeability and phase-lag parameters to locate the contamination in the field, as they do not undergo any change. On the other hand, the resistivity parameter is a good indicator of the presence of partially degraded oil. We also note that normalized chargeability values have been very useful in characterizing the geometry of the clayey-silt roof.

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1. Introduction

To study the features of a contaminated site, direct measurement techniques, such as groundwater and soil analysis, are usually employed, because both soil and water samples can provide a good inventory of the contamination. Nevertheless, drilling and piezometric wells may be misplaced and may not intercept the contamination. To be as complete and reliable as possible, the number of monitoring wells has to be multiplied, thereby increasing the cost of the study. In conjunction with these techniques, geophysics can offer continuous images of subsurface contamination. For this purpose, geophysics has to consider the geology, the contaminant type, and the limitations of the site (e.g., accessibility, electromagnetic noise level, presence of buried objects).

Among all geophysical methods, geoelectrical ones are used and, more specifically, resistivity and Induced Polarization (IP) techniques

in both spectral and temporal domains. At low frequencies (<100 Hz), IP response is linked to the electrical double layer, as described precisely, for example, by Leroy et al. (2008) or Revil and Florsch (2010). It has been proven that IP is sensitive to textural parameters (Leroy et al., 2008), to structural parameters such as grain size and pore throat that can lead to permeability of the media (Revil and Florsch; Weller et al., 2010; Zisser et al., 2010), or to the interaction between the matrix and fluid (Börner et al., 1993; Schmutz et al., 2010).

The presence of organic contaminants in the medium affects both the ohmic and the capacitive conductions in the IP response, because when contaminants enter the rock matrix, surface reactions occur between the contaminants and the soil grains. Such surface reactions include sorption reactions, in which contamination adheres to the mineral surface (King and Olhoeft, 1989) or interacts with the mineral surface (Vaudelet et al., 2011a). These phenomena are affected greatly by the saturation conditions (Breede et al., 2012). The measured IP response depends on the distribution, concentration, and types of contaminants present in the subsurface (Kemna et al., 2012).

Many organic pollution geophysical studies indicate that hydrocarbon oils are electrically resistant and tend to increase the resistivity (Benson et al., 1997; Sauck, 2000). Martinho et al. (2006) have also noticed an increase in the chargeability with the presence of organic compounds, such as toluene and benzene. Several Spectral Induced Polarization (SIP) studies have shown an increase in both resistivity and phase with the presence of hydrocarbon oil (Börner et al., 1993; Schmutz et al., 2010; Vanhala et al., 1992). King and Olhoeft (1989) linked the phase increase to the polymerization of toluene on montmorillonite. Aromatic hydrocarbons, being hydrophobic, are strongly adsorbed on the solid phase, especially on organic matter.

In contrast, other SIP studies noted a decrease of both resistivity and phase with the increase in hydrocarbon content (Revil et al., 2011; Vanhala, 1997; Weller and Börner, 1996). Revil et al. (2011) assumed that the presence of polar molecules can also decrease global resistivity. The most common polar components are asphaltenes, which decrease the resistivity of the medium (Chow et al., 2004). Some organic molecules are involved in the Stern layer and will be adsorbed at the mineral surface, which decreases polarizability (Schwartz and Furman, 2012).

Revil et al. (2011) have linked these trends with the wettability of the oil. Fresh oil is mostly non-wetting (the oil is not in contact with the mineral surface) towards the grain, which increases the polarizability, whereas some oils containing more polar components become wetting (the oil may be in contact with the mineral surface) towards the grain, which decreases polarizability.

Thus, hydrocarbon contaminations have to be understood as spatially and temporally varying objects from the physicochemical point of view (Sauck, 2000). At the laboratory scale, Ustra et al. (2012) noted that, with time, the influence of clay content becomes dominant towards the toluene content. Measurements on columns were undertaken by Abdel Aal et al. (2004) investigating the evolution of the IP response of diesel-amended sediments. The evolution of the biotic and abiotic columns was monitored over a period of 36 weeks. While the biotic column showed a temporal increase of real and imaginary conductivity, the abiotic column showed no evolution of IP response. Diesel biodegradation modified both the electrolytic and interfacial conductivities via the release of byproducts and through the alteration of the mineral surface and bacterial development (Abdel Aal et al., 2010). Abdel Aal et al. (2006) performed measurements on soil cores and noted that the smear zone affected by bacterial biodegradation showed an increase in the imaginary part of the conductivity, but only weak changes on the real part of the conductivity. The presence of bacterial biofilms, acting as colloidal particles (Poortinga et al., 2002), increases the response associated with the polarization of the electrical double layer. In the field, Atekwana and Atekwana (2010) noticed that the smear zone and the capillary fringe, which are the zones most affected by the free-phase hydrocarbon degradation, show higher bulk electrical conductivities. The byproducts of degradation may cause an increase in fluid mineralization, via the increase in the amount of total dissolved solids, and decreased resistivity (Atekwana et al., 2004).

This study shows (i) the IP response at the laboratory scale for fresh oil and for oil under (bio)-degradation, both taken from the hydrocarbon contaminated site, (ii) the temporal evolution of the Direct Current (DC) response on the contaminated site between 2009 and 2012 and (iii) the IP response on the contaminated site in 2012. The response to the fresh and partially degraded oil at the laboratory scale constitutes an analog to the temporal evolution of this oil in the field. For help with their interpretation, field measurements made in 2012 are compared both with field measurements made in 2009 and with the laboratory analog.

2. Materials and methods

2.1. Methods

The DC resistivity is measured thanks to Ohm's law, where the resistivity is equal to the ratio between the measured potential and the injected intensity *modulo* a geometric factor. This resistivity is responsive to both electrolytic and surface contributions. This method does not differentiate between the contribution of the electrolytic conductivity and the contribution of the surface conductivity. However, the differentiation can be performed using IP methods.

At low frequencies, the IP effect consists of the capacitive behavior of the media. It is measured as a residual voltage at the termination of an applied current (time domain) or as a phase shift between the injection and tension sine (frequency domain).

In the time domain, the total chargeability m (in ms) expresses the magnitude of the IP effect,

$$m = \frac{1}{V_0} \int_{t_0}^{t_f} V_t(t) \cdot dt \qquad (1)$$

where V_0 is the measured voltage (at the time of the current cut-off $t < t_0$) during application of the current and V_t is the residual voltage measured at time t, between t_0 and t_f after termination of an applied current.

The chargeability parameter depends upon both surface polarization mechanisms and global conduction mechanisms. The normalized chargeability, defined by the chargeability divided by the resistivity, characterizes the magnitude of the polarization only (Slater and Lesmes, 2002). This parameter is directly proportional to the quadrature conductivity of SIP.

In the frequency domain, the IP response may be presented in terms of magnitude and phase or in terms of real and imaginary components of the impedance. We choose to compute complex resistivity ρ^* (in $\Omega \cdot m$):

$$\rho* = |\rho| e^{-i\theta} = |\rho| (\cos\theta + i\sin\theta) \qquad (2)$$

with $|\rho|$ the resistivity magnitude (in $\Omega \cdot m$) and θ the phase (in mrad).

SIP measurements can also be expressed in terms of complex conductivity σ^* (in $S \cdot m^{-1}$):

$$\sigma* = \frac{1}{\rho*} = |\sigma| e^{i\theta} = \sigma' + i\sigma'' \qquad (3)$$

with σ' the in-phase conductivity and σ'' the quadrature conductivity:

$$\sigma' = |\sigma| \cdot \cos\theta \qquad (4)$$

and

$$\sigma'' = |\sigma| \cdot \sin\theta. \qquad (5)$$

The in-phase conductivity is linked to the ohmic and dielectric losses and is sensitive to changes in fluid chemistry. The quadrature conductivity is linked to the capacitive conduction, which allows the material polarization to be described. At low frequencies (<100 Hz), the quadrature conductivity is sensitive to physicochemical properties at the fluid–grain interface (Kemna et al., 2000; Slater and Lesmes, 2002).

The global methodology adopted consists of geophysical and geochemical experimental laboratory measurements associated with field measurements. Geoelectrical laboratory experiments were performed using the oil extracted from the contaminated site. The measurements were undertaken to evaluate the response that may be measured in the field and to ensure the feasibility of the field campaign. Two kinds of oil were analyzed: (i) the free phase, which seems not to be degraded and named Oil F; and (ii) the top of an oil recovery tank, where active biodegradation occurs (called Oil B). The field campaign consisted of complex resistivity, seismic, ground penetrating radar, and low-frequency electromagnetic measurements. Seismic, ground penetrating radar and low-frequency electromagnetic measurements were not adapted to the problems on this site. Taking into account the environmental constraints, only (complex) resistivity permitted the delineation