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Influence of non-aqueous phase liquid configuration on induced polarization parameters: Conceptual models applied to a time-domain field case study

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ABSTRACT

Resistivity and induced polarization (IP) measurements on soil contaminated with non-aqueous phase liquids (NAPLs) show a great variety in results in previous research. Several laboratory studies have suggested that the presence of NAPLs in soil samples generally decrease the magnitude of the IP-effect, while others have indicated the opposite. A number of conceptual models have been proposed suggesting that NAPLs can alter the pore space in different ways, e.g. by coating the grain surfaces and thus inhibiting grain polarization, or by changing the pore throat size and thus affecting the membrane polarization mechanism. The main aim of this paper is to review previously published conceptual models and to introduce some new concepts of possible residual NAPL configurations in the pore space. Time domain induced polarization measurements were performed at a NAPL contaminated field site, and the data were inverted using the Constant Phase Angle (CPA) model and the Cole–Cole model respectively. No significant phase anomalies were observed in the source area of the contamination when the CPA inverted profiles were compared with soil sampling results of free-phase contaminant concentrations. However, relatively strong phase and normalized phase anomalies appeared next to the source area, where residual free-phase presence could be expected according to the chemical data. We conclude that depending on the NAPL configuration, different spectral IP responses can be expected. In previous research, the NAPL configurations in different samples or field sites are often unknown, and this may to some extent explain why different results have been achieved by different authors. In our field case, we believe that the NAPL forms a more or less continuous phase in the pore space of the source zone leading to an absence of IP anomalies. The increase in phase and normalized phase angle observed next to the source zone is interpreted as a degradation zone. The ongoing biodegradation may have led to a fractionation of the continuous NAPL in the outer part of the original source zone, leading to residual presence of isolated NAPL droplets in the soil pores. With such NAPL configurations, an increased polarization can be expected according to the electrochemical- and membrane polarization mechanisms. More research is needed to confirm the effects of different NAPL configuration on spectral IP parameters.

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1. Introduction

A common environmental problem is the remediation of ground contaminated with non-aqueous phase liquids (NAPLs), e.g. gasoline or industrial chlorinated solvents. These chemicals are immiscible liquids that do not easily dissolve in water, and as such they often exist as an own phase in the soil (a so called free-phase product). NAPLs are divided into light non-aqueous phase liquids (LNAPLs) and dense non-aqueous phase liquids (DNAPLs), where the former have a lower density and the latter have a higher density than water. The density difference has the implication that large LNAPL spills are often found floating on the groundwater surface when released into the ground, while DNAPLs sink trough the groundwater until it reaches a less permeable layer, e.g. the bedrock. Free-phase products of both LNAPLs and DNAPLs can exist as a continuous phase in the ground or as a residual phase, where parts of the contaminant have been trapped in the soil and are not displaced due to gravitational forces (Atekwana and Atekwana, 2010; Ajo-Franklin et al., 2006).

Delineations of NAPL plumes are commonly carried out by means of borehole drilling and chemical sampling. However, there is a risk of spreading the contaminants vertically during this process and the drillings are expensive and give point information rather than a continuous picture of the contaminant plume. Geophysical methods are, in a general sense, effective tools in providing continuous information of soil properties in between individual boreholes. Resistivity and induced polarization (IP) are methods that have been considered promising in order to achieve detection of NAPLs in soils, and research

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has been carried out in both laboratory and field. The interest in the resistivity method has emerged because NAPLs are electrical insulators and can cause a rise in resistivity when their free-phase products displace water in saturated soil. This has been observed in a number of lab and field studies on different kinds of NAPLs (e.g. Chambers et al., 2004; Cassiani et al., 2009; Naudet et al., 2014; Power et al., 2015). However, at field sites, the situation is complicated by the fact that the age and degradation status of the contaminants change the groundwater chemistry. Release of ions during biodegradation, or mineral weathering and dissolution caused by organic acids released by bacteria, can cause increased groundwater conductivity (Atekwana and Slater, 2009; Atekwana and Atekwana, 2010). Thus, increased bulk resistivity due to free-phase NAPL presence could be suppressed by increased groundwater conductivity.

A possible solution to the problem of using resistivity alone for NAPL detection is to extend the measurements to include IP. IP spectra or time decays are sensitive to properties at the pore scale, such as e.g. grain sizes, grain shape, grain surface chemistry and pore throat size distribution (e.g. Slater and Lesmes, 2002a; Scott and Barker, 2003; Binley et al., 2005; Nordsiek and Weller, 2008; Titov et al., 2010). Consequently, the IP method may have a greater potential of being able to detect NAPLs in the pore space compared to resistivity alone. However, the results of previous studies in both field and laboratory scale vary and sometimes point in opposite directions. Several laboratory measurements have suggested that the presence of NAPLs in water saturated soil samples generally reduce the magnitude of the IP effect (Börner et al., 1993; Vanhala et al., 1992; Vanhala, 1997; Chambers et al., 2004; Martinho et al., 2006; Schmutz et al., 2010; Revil et al., 2011), while others have indicated the opposite (Olhoeft, 1985; Titov et al., 2004; Cassiani et al., 2009; Schmutz et al., 2010, 2012). As expected, varied results have also been achieved in field surveys of NAPL contaminated ground. For example, free-phase NAPL presence in an unconfined aquifer have resulted in decreased chargeability (Flores Orozco et al., 2012), while in another field study, high chargeability was interpreted as an indicator of LNAPLs in clayey sand (Deceuster & Kaufmann, 2012).

In contrast to several lab studies, where the relationship between NAPL concentration and IP response has been interpreted as essentially linear (e.g. Revil et al., 2011; Schmutz et al., 2010, 2012), other trends have also been observed. In some cases, a threshold value in the relationship between NAPL concentration and IP response in water saturated soil samples has been seen. Titov et al. (2004) found increasing chargeability with LNAPL content up to a saturation of 92% and decreasing chargeability with further increase of the LNAPL. A similar trend has been observed by Schmutz et al. (2012). In contrast, measurements by Martinho et al. (2006) indicated an initial decrease of chargeability values up to a LNAPL saturation of around 10% in clayey sand samples. Slightly increasing chargeability values were observed at higher contents, although all values were below the values of clean sand, in contrast to the data from Titov et al. (2004).

It is not always straightforward to compare the results from previous laboratory measurements since there can be variations in e.g. sample preparation and NAPL phases and species (Cassiani et al., 2009). Field data is often even more challenging because of different field circumstances such as geological setting and degradation status. Beyond variations in chemical, biological and geological parameters, a factor that plays a major role in understanding the effect on measured IP of NAPL in soils is the geometrical configuration of the free-phase product in the pore space. In published research, interpretation of IP data often rely on different conceptual models of NAPL configuration, which in different ways explain the observed results (e.g. Olhoeft, 1985; Titov et al., 2004; Martinho et al., 2006; Schmutz et al., 2010; Revil et al., 2011; Flores Orozco et al., 2012; Shefer et al., 2013). However, to date, there are no comparative studies discussing possible effects of different models. There is thus a need for a comprehensive discussion and comparison of various possible NAPL configurations, since this may enhance the understanding of why different results may be achieved when IP measurements are performed on NAPL contaminated soil.

The main aim of this paper is to review previously published conceptual models and to introduce some new concepts of possible residual NAPL configurations in the pore space. We will discuss how the conceptual models can be expected to influence spectral (Cole–Cole) IP parameters (Section 5). Field results from time domain IP measurements on a site contaminated with DNAPLs will be presented (Section 4) and discussed in relation to the qualitative results from the discussion of NAPL configurations. The data were inverted with both the Constant Phase Angle (CPA) model and the Cole–Cole model. The results indicate a zone of increased phase/chargeability and decreased relaxation time at a location outside the free-phase source area, while no effects can be distinguished at the locations where the highest DNAPL concentrations were observed. The increased IP effects are interpreted as possible effects of residual free-phase presence. We believe that the concentration and configuration of the DNAPL in the soil are important since this will affect the micro geometrical properties at the pore scale and the current paths through the soil.

In a wider context, we wish to raise the awareness of the importance of assumptions about the NAPL configuration in a pore space, an issue that was also stressed by Cassiani et al. (2009), and its possible impacts on spectral IP parameters. We believe that such considerations could enhance future interpretations of IP data measured on NAPL contaminated lab samples or field sites.

2. Theory

2.1. Polarization mechanisms

The mechanisms behind the spectral induced polarization response in metal-free soils are usually described in terms of electrochemical polarization, membrane polarization or Maxwell–Wagner polarization. The basis of the electrochemical polarization mechanism (also called grain polarization or Stern layer polarization) is the existence of electrical double layers (EDLs) around soil grains. The EDLs arises as a result of a negative surface charge on mineral grains surrounded by water, which leads to attachment of a fixed layer of counter-ions at the grain surface, the so called Stern layer. In addition, a second layer of counter-ions, called the diffuse layer, is formed and consists of more weakly electrostatically attracted solvent ions. When an electrical field is applied to the soil, the EDL becomes polarized (Schwarz, 1962). Modeling studies have shown that the polarization of the Stern layer is much stronger in magnitude compared to the polarization of the diffuse layer (De Lima and Sharma, 1992; Lesmes and Morgan, 2001; Leroy et al., 2008). The total polarization magnitude has been observed to be inversely proportional to the grain size of silts, sands and tills (Slater, 2002). The relaxation time increases with the characteristic grain (or pore throat) size, since this determine the possible length scale of diffusion for ions in the EDL (Slater, 2007).

The concept behind membrane polarization is that series of small ion-selective and non-selective zones exist in the pore system. The ion-selective zones can consist of negatively charged clay particles attracting a cloud of counter-ions, or narrow pores or pore-throats which to a large degree are occupied by the EDLs of the surrounding grains. When an electric field is applied to the system, ions are blocked in front of the ion-selective zones, and ions attached to mineral particles are displaced. The length ratio and relative ion transparencies between the ion-selective and non-selective zones determine the polarization magnitude, while the length of the non-selective zones controls the diffusion length scale and therefore the relaxation time (Marshall and Madden, 1959; Vinegar and Waxman, 1984; Titov et al., 2002).

The Maxwell–Wagner polarization mechanism is related to ion accumulations at borders between soil phases with different dielectrical properties, e.g. water and grains. This mechanism dominates at higher frequencies (above 1 kHz) and is also dependent on the shapes of

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