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Experimental study of electrical complex resistivity in a 2D multiphase porous medium under non-isothermal conditions: application to soil

remediation monitoring

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Running title: Induced polarization under non-isothermal conditions

Highlights

- Proper application of spectral induced polarization (SIP) in samples polluted by dense non-aqueous phase liquids (DNAPL)
- Temperature and saturation separately affect electrical complex resistivity
- Temperature has a considerable effect on Cole-Cole parameters in a 2D tank
- Saturation changes have a remarkable impact on phase shift
- 2D tank results on complex resistivity confirm 1D cell findings

Abstract

In this decade, electro-geophysical methods are widely used in different environmental subjects. Study on soil remediation polluted by DNAPLs has become a certain need for all countries. Geoelectrical methods show their potential to facilitate evaluating decontamination processes. Our challenge in this study was to understand how coupled temperature and saturation changes affect electro-geophysical parameters in a contaminated 2D sample. The primary objective was to evaluate the efficiency and potential of spectral induced polarization (SIP) for monitoring the recovery of dense non-aqueous phase liquids in contaminated porous media. A set of 2D tank experiments investigated the impacts of temperature and saturation changes on the electrical complex resistivity of a saturated porous medium under nonisothermal conditions. The measurements were made with a coal-tar and water fluid pair in a porous medium that has been simulated by 1 mm glass beads. We circulated hot water around the tank and used an immersion heater to heat the porous medium in the tank at different stages. The SIP technique (also called complex resistivity) was used to measure the complex electrical resistivity of a medium in the frequency domain. The experimental results for a simple drainage case were validated using numerical modeling. The complex electrical resistivity was used to obtain the saturation field before and after imbibition. For this purpose, the generalized Archie's law obtained for the same fluid pair with 1D cells (with a vertical flow) was used. Our results from electrical resistivity measurements for saturation fields are in accordance with 2D tank images and can illustrate the saturation change with pointwise resistivity measurements. The results show that saturation change has the main role in electrical resistivity variation compared to temperature (5 to 7%). We also studied the effects of temperature change on the Cole-Cole parameters and the results confirm our previous findings with the same variation trend in these parameters. The results from varying electrical complex resistivity from the 2D tank (with vertical and horizontal flow) in the laboratory conditions will help us to understand the coupled temperature and saturation effects on complex resistivity in a real polluted site case.

*Keywords

Induced polarization, modeling, pollution, resistivity, environmental

1 Introduction

2 Pollution of soil due to dense non-aqueous phase liquid (DNAPL) spilling has become a 3 crucial issue of concern during the last decade, causing numerous geo-environmental (e.g., 4 mitigation of DNAPLs on aquifers) problems (Habiyakare et al., 2022). In polluted sites, 5 DNAPLs sink below the water table and cause dissolved plumes in the subsurface (Pankow 6 and Cherry, 1996; Power et al., 2014). After many years of research, soil contamination due to 7 DNAPLs is still one of the main challenges for the environmental and geophysical 8 communities (Kavanaugh, Suresh and Rao, 2003). DNAPLs, including coal tar (CT), 9 contaminate soils and water widely when released in the subsurface, mainly from industrial 10 parks. DNAPLs are part of a group of immiscible fluids that pass through the saturated and 11 unsaturated zones due to gravity and capillary forces, via migration passages made of pores, 12 until stopping at an impermeable stratum (Illangasekare et al., 1995; Power et al., 2014). 13 Various remediation techniques can be used in these sites to depollute them. These include 14 pump and treat, thermal, bioremediation, and chemical methods (Beyke and Fleming, 2005; 15 Hadley and Newell, 2012). Pump-and-treat is the common and first remediation technique to 16 remove the free product. After pumping, the residual saturation can be removed using 17 biochemical remediation techniques.

18 Geophysics has shown the value of the long-term monitoring of clean-up processes (e.g., 19 Brewster et al., 1995; Wilson et al., 2009). Of all the geophysical methods, electromagnetic 20 methods and especially electrical methods are useful for studying the processes involved 21 during clean-up and degradation of pollutants (Chambers et al., 2004; Mao et al., 2016; Deng 22 et al., 2020), 3D imaging of a jet fuel spill (Orozco et al., 2021; Helene, Moreira and Bovi, 23 2020). The electro-geophysical methods, especially direct current (DC) resistivity, induced 24 polarization (IP) techniques, and spectral induced polarization (SIP) have proved their 25 potential in geological conditions, in saturated conditions (e.g., Weller, Nordsiek and

Debschütz, 2010; Slater et al., 2014; Revil et al., 2015; Kremer et al., 2016b; Sparrenbom et 26 al., 2017), unsaturated conditions (e.g., Maineult, Jougnot and Revil, 2018), oil, gas, and 27 28 mining industries (e.g., Friedrichs et al., 1999), soil pollution and remediation monitoring 29 (e.g., Pearce and Zuluaga, 2004; Ling and Zhang, 2017; Liao et al., 2018) and civil 30 engineering (e.g., Susanto et al., 2013). In a multiphase porous medium, various combinations 31 of different parameters such as temperature, saturation, level of compaction, and pore fluid 32 properties can have a similar effect on the measurements (e.g., Sen and Goode, 1992; Denicol 33 and Jing, 1998). Therefore, studying the effect of each parameter to determine their role and ideally, their related simultaneous effects on geophysical parameters is necessary. 34

35 Sadowski (1988) performed the first laboratory study on different clay samples with 36 different pollutants and different concentrations. He found that the SIP method could be used 37 to identify organic contaminants in clayey soils for different concentrations. Börner, Gruhne 38 and Schön (1993) contributed to laboratory research on the effects of organic liquid 39 contaminations on different rock types and sandstones. They demonstrated that both the real 40 and imaginary parts of complex resistivity are affected by the properties of the contaminant 41 filling the pores. They noted that in some cases the various contaminants could be 42 differentiated using the imaginary part and emphasized the effect of interactions between pore 43 fluid characteristics and pores' internal surface structure.

The existence of organic contamination can affect capacitive conduction in the IP response due to the surface reaction between contamination and soil grains (Vaudelet *et al.*, 2011). IP responses in these contaminated soils are related to the saturation (Breede *et al.*, 2012), distribution, concentration, and types of contaminants present in soil (Kemna *et al.*, 2012). Previous studies demonstrate that the presence of contaminants (Gasoline and leachate results of acidification by organic and carbonic acids) in the soil increases resistivity (Benson, Payne and Stubben, 1997; Sauck, 2000). Martinho, Almeida and Matias (2006) reported an 51 increase in chargeability due to the presence of pure organic contaminations (gasoline, 52 isopentane, toluene, and benzene) in clayey soil. Many studies based on the SIP method have 53 shown an increase in amplitude and phase shift due to the existence of hydrocarbons (Vanhala, 54 Soininen and Kukkonen, 1992; Börner *et al.*, 1993; Schmutz *et al.*, 2010). However other SIP 55 studies reported a decrease in amplitude and phase shift in different media consisting of 56 different pure and dissolved hydrocarbons (Weller and Börner, 1996; Vanhala, 1997; Revil, 57 Schmutz and Batzle, 2011).

Recently, Iravani et al. (2020a, 2020b) evaluated how temperature and saturation 58 59 changes in two DNAPLs (coal tar and chlorinated solvent) affect complex electrical resistivity 60 and relative permittivity. They used an oven to study temperature change and a 1D drainageimbibition system to study how saturation changed these two parameters. They carried out 61 experiments in cylindrical cells filled with glass beads (to simulate soil/porous media) and 62 63 saturated with water and CT. The relationship between electrical resistivity and relative 64 permittivity with saturation was investigated for two different coupled immiscible fluids, 65 water/coal-tar and canola oil/salty ethanol. They found that temperature changes these two geophysical parameters linearly for the coal tar (CT) and chlorinated solvent coupled with 66 water. The effect of saturation change was not linear, and they did state that the generalized 67 68 Archie's law could fit their experimental data well. Colombano et al. (2020) investigated the 69 relationship between resistivity and saturation for chlorinated solvent. They used a drainage-70 imbibition protocol to monitor electrical resistivity and permittivity. Their objective was to 71 estimate residual saturation indirectly. They used the complex refractive index model (CRIM) 72 and generalized Archie's law to model their permittivity and resistivity data, respectively. They reported that estimated electrical resistivity presented less correlation with the 73 74 experimental data compared to permittivity. They concluded that temperature and saturation 75 affect the electrical resistivity and relative permittivity of the medium. Colombano et al.

(2021) investigated the imbibition of a mixture of chlorinated solvents by thermal and chemical enhancements (Colombano *et al.*, 2021). They found that the temperature has no effect on DNAPL recovery that could be linked to the actual properties of the oils. However, coal tar is more viscous than most chlorinated solvents. Moreover, the dynamic viscosity of coal tar decreases significantly with temperature (Philippe *et al.*, 2020). To the best of our knowledge, no other work is reported on electrical resistivity measurements for coal tar.

Blondel *et al.* (2014) compared lab and field results in clayey and clayey-silt soils. They quantified a resistivity increase, normalized chargeability, and decreased resistivity in measurements with oil not degraded. When it is partially degraded they saw no changes in the phase-lag and chargeability parameters. Gourry, Jeannot and Proust (2001) also showed that under certain conditions, oil pollution like this can increase chargeability because of high resistivity and it is rather related to wettability. This statement was not confirmed by Blondel *et al.* (2014).

89 Orlando and Palladini (2019) carried out a series of laboratory tests to study how 90 resistivity and permittivity vary in a 2D cell in a multiphase (two-phase) system. The 91 contamination they used was HFE-7100 (hydrofluoroether) in saturated (with pure, dissolved, 92 and gaseous phases) grained glass bead (GB) porous media. They used geophysical 93 measurements and physico-chemical constraints to monitor these parameters over a long 94 period. Different geophysical techniques were used to propose empirical models to monitor 95 water saturation in a multiphase porous media contaminated by DNAPL. Photographs were used to confirm the geophysical data. Low frequency IP can detect the pure HFE-7100 phase. 96 97 In another study, Orlando and Renzi (2015) studied how electrical resistivity varies in ground 98 due to the existence of DNAPL in a laboratory study. Like Orlando and Palladini (2019), they 99 used a GB-packed medium contaminated with HFE-7100 in a saturated condition. They found 100 that different DNAPL phases have different impacts on the physico-chemical characteristics of 101 the contaminated medium. In their study, free and dissolved DNAPL caused a change in 102 electrical resistivity. They concluded that electrical resistivity coupled with the GPR technique 103 can be used for characterizing the contaminated sites because neither technique could be 104 sensitive enough for a long time.

105 It is known that the presence of organic contaminants modifies the impedance of the fluidgrain interface through clay polymerization (Olhoeft, 1985; Vanhala, 1997). The presence of 106 107 organic pollutants as free phase changes the distribution of the conductive aqueous phase in 108 the pore space (like air can do in unsaturated media). As SIP response changes with varying water/air saturation (e.g., Ulrich and Slater, 2004; Titov et al., 2004; Binley et al., 2005, 109 110 Maineult et al. 2018), free-phase hydrocarbons should also produce a measurable SIP effect. A 111 few studies have been conducted on complex resistivity laboratory measurements either on 112 artificially contaminated soil or rock samples or on field samples from contaminated sites. 113 Even though the results differ in the observed effects, they proved that the SIP response of 114 soils and rocks is measurably influenced by hydrocarbon contamination. However, a detailed 115 understanding of how contamination affects SIP response is still lacking. Many studies did not 116 pay sufficient attention to controlling the nature or distribution of contaminants in the porous 117 media, and on assessing independently the effects of dissolved, adsorbed, and free-phase, 118 leading sometimes to what appear to be contradictory results. Here, we assumed that the SIP 119 response of the contaminant obeys a law similar to what Vaudelet et al. (2011) observed above 120 a plume of chlorinated solvents, i.e., a Cole-Cole like behavior.

121 The study of geophysical parameters during remediation by the thermal enhanced 122 method is influenced by two environmental parameters: temperature due to soil heating 123 (Iravani *et al.*, 2020a, Philippe *et al.*, 2020) and saturation changes (Iravani *et al.*, 2020b; 124 Colombano *et al.*, 2020) due to contamination pumping. In previous studies, we separately

investigated the impact of temperature and saturation on electrical resistivity in contaminated
saturated porous media with CT (Iravani *et al.*, 2020a, 2020b).

127 In this study, we investigated the coupling of transient heat transfer in multiphase flow 128 with saturation variation in a 2D tank. No work to date has studied transient temperature 129 transfer and saturation changes at the same time in 2D due to its complexity. Heating in the 2D 130 tank mimics the soil heating in the field by increasing the temperature of pollutants to decrease 131 their viscosity. These phenomena increase pollutant mobility and so increase the pumping rate 132 of contamination in saturated soil during depollution. Heating the samples also changes the 133 geo-electrical parameters. The goal of the current study is to determine the saturation field in 134 the 2D tank from resistivity measurements. All these experimental studies have been done to 135 implement the SIP for monitoring the real remediation process of DNAPLs in the field.

136 It is worth noting that one of the main challenges in the monitoring of the subsurface with 137 geophysical methods is that there are few methods (e.g., boreholes) for verifying the recorded 138 geophysical response of the medium to validate the remediation process (Spies, 1996; Keys, 139 2017).

140 Theoretical background

141 Spectral-induced polarization mechanisms

142 SIP is an extension of the IP method with alternating current (AC) in the frequency 143 domain. SIP is used to study the ratio and phase shift between voltage and current of the 144 subsurface or a sample. Generally, electrical polarization in the range of studied frequencies 145 (i.e., from 0.01 to 10,000 Hz) has four main mechanisms including electrode polarization, 146 membrane polarization, Maxwell-Wagner polarization, the polarization of the diffuse layer 147 (electrical double layer, EDL) including the Stern layer polarization (e.g., Kemna *et al.*, 2012). 148 This study does not separately investigate the different mechanisms involved to characterize149 the effect of CT saturation and temperature changes.

150 Electrical complex resistivity

151 The electrical resistivity (ρ) in a homogeneous sample can be obtained by multiplying 152 resistance (R) and a geometric factor (K) related to the position of the electrodes (for current 153 injection and voltage measurement). The complex electrical resistivity reads (e.g., Guéguen 154 and Palciauskas, 1994; Ghorbani, 2007; Glover, 2015):

$$\rho_{(f)} = \rho'_{(f)} - i\rho''_{(f)} = |\rho_{(f)}| exp(i\varphi)$$
(1)

where ρ' and ρ'' are, respectively, the real and the imaginary parts of complex resistivity and $|\rho_{(f)}|$ and φ are the amplitude and the phase shift of the resistivity response (for low phase, below a few hundred of mrads, can be simplified as $\varphi \approx \rho_{(f)}'/\rho_{(f)}'$. These equations can be written in terms of the electrical conductivity, $\sigma_{(f)}$ (S.m⁻¹) which is the inverse of the electrical resistivity.

160 A very first relationship to study the effect of temperature on electrical resistivity was
161 presented by Dakhnov (1962) as:

$$\rho_w = \frac{\rho_{w_0}}{1 + \alpha (T - T_0)} \tag{2}$$

162 where ρ_w is the resistivity of water at temperature *T* and ρ_{w0} is the resistivity of water at the 163 initial temperature T_0 , and α is the temperature coefficient of resistivity (see Iravani et al, 164 2020a, for instance). We used equation (2) to obtain α value for our experiments using 165 thermocouples, with a reference temperature of T₀=20°C, where T is the temperature measured 166 by thermocouples, and ρ_{w0} is the averaged measured resistivity at 20°C. This linear 167 relationship was defined for water but can be used for DNAPL mixtures (Iravani *et al.*, 2020a).

168 In the literature, no universal model has been presented with the consideration of the 169 physico-chemical properties of each phase in a multiphase system in regard to the frequency 170 changes. The lack of a developed comprehensive model on this subject steers researchers to fit 171 the experimental data of complex resistivity with the phenomenological models. One of the 172 most used phenomenological models is presented by Cole and Cole (1941) and was derived 173 from the Debye (1929) model, which was firstly developed for relative permittivity (Pelton *et* 174 *al.* 1978).

Pelton *et al.* (1978) proposed a model for SIP spectra that exchanged relative
permittivity with electrical complex resistivity for practical applications that describes

$$\rho(\omega) = \rho_0 \left[1 - m(1 - \frac{1}{1 + (i\omega\tau)^c}) \right]$$
(3)

where ρ_0 is the low-frequency resistivity, c is the so-called Cole-Cole exponent, *m* is the chargeability, and τ is the mean relaxation time (*m* and c can be in the range of 0 and 1). If c equals 1 this equation will be similar to the Debye model. For this study, the Cole-Cole model was chosen because it has been demonstrated to be a good model to represent empirically the polarization of single grains or pores (e.g., Leroy *et al.* 2008; Niu & Revil 2016).

In this study for saturation prediction, the generalized Archie's law, which is developed
by Glover (2010) based on the classical Archie's law (Archie, 1942), was used. It is defined as
below

Archie's law
$$\sigma_{mixed} = \sigma_{(f)} \Phi^m$$
 (4)

185 and

Generalized Archie's law
$$\sigma_{mixed} = \sum_{i=1}^{n} \sigma_i \Phi_i^{m_i}$$
 (5)

186 where
$$\sigma_{(f)}$$
 (S/m) is the conductivity of the liquid inside the pores, Φ is the porosity, *i* is the
187 indication of each phase and m is the cementation exponent.

In this study, we validate mixing models and calibration curves obtained from the previous study in cells and columns (Iravani *et al.*, 2020a, 2020b) using a 2D two-phase flow 190 and comparing the results with imaging for validation. In a previous study, Iravani et al. 191 (2020a) showed both resistivity and phase are related to the temperature variation in 1D 192 columns. They found a linear relationship between temperature and resistivity. By using the 193 Cole-Cole model they also found that temperature affects the frequency domain. Irayani et al. 194 (2020b) studied the saturation impact on the magnitude and phase of electrical complex 195 resistivity. They reported the generalized Archie's law was applicable to study the multiphase 196 medium contaminated by CT. They found that saturation change has a direct impact on 197 electrical resistivity.

198 Material and methods

A laboratory-scale experiment was carried out to study temperature and saturation effects on complex electrical resistivity during the drainage and imbibition of DNAPLs in a 2D tank. The experiment was done in a saturated porous medium with an immiscible liquid pair (CT and water). In the following, we describe the essential points, experimental setup, and procedures used for this experiment.

204 2D tank setup

The 2D tank has dimensions of $30 \text{ cm} \times 50 \text{ cm} \times 7 \text{ cm}$ (height, width, depth). A schematic form of the front, back, and top of the tank is shown in Figure 1. The body of the 2D tank is made of a solvent resistant material called polyvinylidene fluoride (PVDF). The front wall of the tank with a thickness of 0.8 cm is made of a transparent glass rectangle through which pictures are taken at each stage.

A stainless-steel heating element was inserted inside the left-side of the tank to produce the non-isothermal condition. Fourteen type K thermocouples (green circles) record temperatures in the tank. The thermocouples were connected to a Campbell CR-1000 acquisition system. The temperatures were measured with sampling intervals of 30 s. For the 214 SIP measurements (Figure 1c), black and blue circles represent 30 current and 30 potential electrodes, respectively, 5 cm apart from each other. The electrical AC is injected between two 215 216 stainless steel current electrodes (diameter=65 mm) made of nickel-cobalt alloy (MP35N) and 217 the response voltage is measured between two handmade non-polarizable potential electrodes 218 (Cu/CuSO₄) with a diameter of 1 cm. Non-polarizable electrodes are used in SIP studies to 219 measure the voltage response of an injected current in a medium (e.g., Kremer et al. 2016a; 220 Abdulsamad et al., 2016). In non-polarizable electrodes, current can freely pass without 221 polarization. Among common electrode solutions, Cu/CuSO₄ was chosen based on the method 222 proposed by Maineult, Bernabé and Ackerer (2004). The solution was made using a mixture of 72.75 g of milli-Q water (ultrapure water), 26 g of CuSO₄, and 1.25 g of gelatin in 100 g of 223 224 solution. A heating shaker was used to mix the solution for one hour at 80°C.

The non-polarizable electrodes were placed outside the samples to avoid inflow interference that in turn can interfere with electrical current signals (Maineult *et al.*, 2004). A porous ceramic (Ameteksi scientific instruments) was used in the interface of non-polarizable electrodes and the saturated porous media. Each non-polarizable electrode was connected to the SIP-lab IV (manufactured by Radic Research) with a shielded cable to avoid contamination of the data due to electromagnetic coupling (Maineult *et al.*, 2004; Bagotsky, 2005).

231 Glass beads (GB) made of silica with a diameter of 1 mm, forming a pack with a 232 porosity of $40\% \pm 2\%$, were used as a porous medium. Different GB sizes were tested and this 233 size was chosen to simulate a porous medium in the lab with close physical properties to field 234 soil (e.g., the grains size and the porosity of the medium). The tap water was degassed using an 235 ultrasonic tank (VWR Ultrasonic Cleaner - USC500D). For obtaining temperature variation 236 effects in a non-isothermal condition, an immersion heater heats from the left side of the tank, 237 and heat transfers to the right side. In non-isothermal conditions, there is a possibility of 238 studying the temperature and saturation changes at the same time based on how temperature

varies across the different halves of the tank and how saturation varies during imbibition fromthe bottom middle of the tank.

241 Water is injected using five pipes located at the bottom of the tank with the possibility of 242 cutting fluid currents by using clips and clamps. The pipes are numbered and connected to a 243 peristaltic pump (Watson Marlow 530U). The tank is filled step by step by first filling 5 cm 244 bottom layer of well spread GB and then water was injected until the porous medium was fully 245 saturated. This procedure should be carried out with great patience to avoid any air bubbles 246 being captured inside the porous network. These bubbles, especially when they are connected 247 to electrodes or thermocouples, can cause significant measurement errors. This operation is 248 repeated step by step until reaching the top of the 2D tank and when all electrodes were buried 249 by GB and water (almost 27.5 cm). A camera is placed on a fixed tripod in front of the 2D 250 tank. Photos were taken during increasing CT levels and imbibition at regular time intervals of 251 three and one minutes, respectively.

Previous experiments using imaging, time-domain reflectometry (TDR), and mass balance analysis showed a 2D flow behavior (Colombano *et al.*, 2021). However, the character of the electric field can be in 3D, and therefore, our numerical model uses a 3D geometry (with a small thickness) to simulate electrical fields.

256 Experimental and measurement procedure

Twenty-seven SIP measurements of an equatorial dipole-dipole (EDD) array were performed to resolve the variations of the electrical resistivity within the 2D tank. The conventional location of measurement for an EDD array is in the middle of the rectangles in Figure 1c. The exact place of measurements with the EDD array is illustrated in Figure 2 with blue circles and the number of measurements. Before and after each experiment, measurements were made with tap water to examine the performance and accuracy of electrodes and calculate the geometric factors (K). The geometric factor for water is easy to

264 obtain because the resistivity of water is homogeneous and can be measured by a portable 265 conductivity meter and the resistance by SIP lab. Then we calculated the K factor from 266 resistivity over resistance. To validate the work, the measured geometric factors should be 267 close to each other. According to our experience, the non-similarity of the geometrical factors 268 was mainly related to non-polarizable electrodes or their surroundings. A blank test (with 269 water only) was done for at least 24 hours to have enough data to increase measurement 270 accuracy. The measurement readings may have serious errors, because of air trapping inside 271 the polarizable electrodes. With a simple test with only water, these deficit electrodes can be 272 replaced. In the next step, the experimental geometric factors (K) must be compared with ones 273 that will be estimated by a numerical model for all 27 SIP measurements.

274 After creating a stable porous medium saturated with water, CT was injected from the 275 bottom of the 2D tank (using the same points of entry where water was injected). For each step 276 of increasing DNAPL content, measurements were made until the DNAPL has been reached to 277 the middle of the 2D tank at 15 cm. This procedure was repeated three times to obtain a 278 horizontal interface between CT and water in the middle of the tank. In the experimental 279 process, obtaining a perfect flat boundary with CT was not easy due to fingering and dissimilar 280 distribution in the different parts of the 2D tank. After stabilizing the experimental setup with 281 the portion of half GB+water and the other half GB+CT, SIP measurements were made before 282 heating, in the frequency range of 0.183 Hz to 20 kHz. Resistivity and temperature were 283 measured at all levels in the 2D tank.

After heating the tank from the left side and diffusing the heat to the right side of the tank, our experimental protocol was done for drainage and imbibition of water where SIP measurements were conducted at each step. The heating time takes almost 12 hours to have a stable temperature in each thermocouple (measurement zone) of the tank. When the recorded temperature was stable, the pumping of CT from the bottom middle of the tank was started.

Due to the rapid imbibition, it was not possible to measure more than two frequencies during this dynamic stage of the experiments. Therefore, during the imbibition period, the SIP spectra were recorded in two frequencies (1.46 Hz and 2.92 Hz), close to the ones used commonly in the field scale. The resistivity calculations were, therefore, based on a single-frequency approach.

294 Iravani et al. (2020a) reported the α values (equation (2)) for the same liquids (water 295 and CT) with and without GB in 1D cells and columns. From their comparison between the α 296 value of water with other studies, normally the α value should be almost the same. We 297 calculated this value again using our 2D tank experimental data to be sure it was the same in 298 this new 2D configuration. In three levels of measurements, the top level is filled with 299 GB+water and the bottom level with GB+CT. Figure 3 is the linear regressions of the 300 resistivity data with the temperature where the y-axis is the ratio of the resistivity at reference temperature ($T_0=20^{\circ}$ C) and resistivity at T (ρ_0/ρ) minus 1, and the x-axis is the temperature 301 difference $(T - T_0)$. The slopes of the linear regression of the 9 measurements at the top with 302 303 GB+water and the bottom GB+CT are presented in Figure 3a and Figure 3b. The obtained 304 regression is in accordance with the previous finding by Iravani et al. 2020a,b. Hayashi (2004) 305 studied the linear relationship between electrical conductivity and temperature that was usually 306 used in previous studies on water with different salinity and chemical substances. Regardless 307 of the compositional and structural differences, the relationship between these two parameters 308 of all different water samples was linear. He suggested being more cautious if temperature 309 values are dissimilar to the reference temperature of 25 °C. The α values for water-saturated 310 GB (0.052) and CT saturated GB (0.075) are the same as what we found in columns (Iravani et 311 al., 2020a) but the coefficients of determination are less (GB+water 13.09% and GB+CT 312 3.28%) than columns for both.

According to Iravani et al. (2020b), the final equations for correcting resistivity for bothmultiphase systems are:

$$\rho_{(GB+water)} = \rho_{0(GB+water)} / (1 + 0.052(T - 20))$$
(6)

$$\rho_{(GB+CT)} = \rho_{0(GB+CT)} / (1 + 0.075(T - 20))$$
⁽⁷⁾

315 After validating α values in two different geometries and despite the complexity of CT 316 components, these two equations can be used in our 2D tank for temperature correction of 317 electrical resistivity in the multiphase system of GB+CT and GB+water. The porous medium 318 in the tank was subjected to heating with an immersion heater. The temperature fields (back 319 view) in the tank before and after imbibition are shown in Figure 4. We used Surfer software 320 to interpolate the data. Surfer interpolated the temperature and the corrected resistivity in the 321 range of 110 to 100,000 Ω .m with the Kriging method. In this study, for all data fitted with 322 Surfer, the same griding methods as Kriging were used with the experimental variograms 323 fitted with linear component type and no trend removal. The temperature diffuses faster in the 324 water at the top due to its higher thermal conductivity. The temperature close to the heater is almost 50°C and due to heat loss, it decreases across the tank to room temperature (20°C). 325

326 Experimental electrical resistivity value was measured and corrected with temperature 327 for three testing EDD array levels. In the scatter plots of Figure 4, each point corresponds to a 328 mean value of a square using an EDD array. Before imbibition, the first level of EDD at the bottom of the tank had almost the same resistivity. The decreasing resistivity in the middle part 329 330 of the tank after imbibition suits the CT saturation decreasing and replacing DNAPL with 331 water in this part. The points in the boundary of the tank were considered as same as their 332 neighbor points for interpolation. Our study of temperature and saturation effects has been 333 done separately for the condition that both factors want to be considered. After correcting the 334 data for the temperature effect, the impact of saturation changes could be determined. The next 335 step was pumping CT out from the middle bottom of the tank. Performing the imbibition step 336 was to simulate the thermally enhanced pump and treat technology for soil remediation. The 337 objective was to monitor how water and CT saturation vary inside the 2D tank using the 338 electrical resistivity measurements.

339 Validation of the electrical resistivity measurements

340 Before starting the main drainage-imbibition experiment, to validate the performance of 341 the electrodes, the first experiment was carried out by increasing the CT level in the tank at the 342 normal room temperature (20°C). In the following, this experiment's data will be compared to 343 the results obtained from a numerical model. After filling the half bottom of the tank with 344 GB+CT, the resistivity was measured for 24 hours, then the temperature of the 2D tank was 345 increased to 30 and 40°C by circulating hot water at a given temperature around the 2D tank to 346 keep the temperature constant in all 2D tank parts. Compared to heating elements, heating with 347 this method can result in temperature homogeneity throughout the whole tank.

348 For the main experiment, a laboratory protocol of a multiphase system was defined in the 2D tank that will be briefly described in the following. The 2D tank was filled with GB and 349 350 two different liquids, water as a conductive fluid and CT with low conductivity. At the starting 351 point, the tank was filled with GB and water layer by layer (Figure 5a). Figure 5b is the tank 352 filled with GB and water, ready for injecting CT from the 5 points at the bottom of the tank 353 (Figure 5c). It is necessary to slowly inject CT to have the relatively horizontal CT levels 354 (Figure 5d). Despite all these considerations, in one case (Figure 5e) when a bump shape CT 355 surface appears, slight injecting from the sides of the tank can balance the surface of CT in the 356 tank.

We used an open-source modeling tool BERT (Boundless Electrical Resistivity Tomography) developed by Günther and Rücker (2015) to model this simple drainage experiment. The possibility of modeling closed geometries as laboratory cells and tanks encouraged us to use this modeling code. The theory of developing this code based on the finite element method has been expanded from the previous study done by Rücker, Günther
and Spitzer (2006). In the other studies, code was developed for arbitrary electrode shapes
(Rücker and Günther, 2011) and long electrodes (Ronczka, Rücker and Günther, 2015).

The 2D tank, by using three series of EDD arrays in different levels, was modeled by BERT. The fundamental physical theory for electrical resistivity used in BERT software is based on Ohm's law (Günther et al., 2006). Assuming that the electric fields are conservative and the magnetic effects are negligible, the mathematical equations can be written as follow

$$\mathbf{J} = \sigma \mathbf{E}; \ \mathbf{E} = -\boldsymbol{\nabla} V, \tag{8}$$

where **J** (A.m⁻²) is the current density, **E** (V.m⁻¹) is the electric field intensity, σ (S.m⁻¹) is the electrical conductivity (the reciprocal of resistivity ρ), and *V* (V) is the electric potential (primary variable). The initial condition was ρ (t=0)=30 Ohm.m that corresponds to the electrical resistivity of glass beads saturated by water. Neumann boundary condition (nocurrent flow boundary condition) was imposed on all 2D tank borders. The number of irregularly structured meshes was 1036942 elements.

In the other experiment in the 2D tank, increasing CT level is also simulated centimeter by centimeter from the bottom of the 2D tank up to 18.0 cm from the 2D tank bottom. Before these steps, we simulate a homogeneous tank filled with only GB and water.

After injecting CT, the resistivity values were recorded for these different levels, until reaching the top of the middle EDD level (17.5 cm). This data allows us to study how resistivity varies in the experiment and the model according to an increase in CT level.

380 Results and discussions

381 Correction of data with temperature

In remediating polluted sites with the thermal enhanced method, the effect of temperature on the electrical resistivity should be corrected before studying the effect of saturation change on geophysical parameters. At the beginning of the experiment, the mean temperature in the tank was 20°C. The temperature varied between 19.1 and 22.8°C. The resistivity was corrected with temperature by using the relation proposed by Dakhnov (1962) and Keller and Frischknecht (1966) to have a geophysical parameter that is only controlled by saturation.

389 Electrical resistivity fields

390 The electrical resistivity variation was used to predict the water/CT saturation change. 391 The evolution of logarithmic apparent and inverted electrical resistivity (to facilitate the 392 comparison before and after temperature correction) during the imbibition test is shown in 393 Figure 6. The objective is to show how temperature changes can impact the calculation of 394 resistivity. Iravani et al. (2020a) presented how the temperature affects the resistivity value in 395 a multiphase sample. To study the effect of saturation change due to pumping, it is needed to 396 deduct the effect of the heating. Figure 6a,b show the log of resistivity before temperature 397 correction, and Figure 6c,d show the same results with temperature correction. This is the 398 starting point of our study on the saturation effect on resistivity values in the 2D tank. Figure 6 399 is cut 10 cm from the left side of the tank to eliminate the influence of the metallic heater on 400 the resistivity of the medium.

401 Validation of the setup configuration and electrical resistivity measurements using

402 BERT modeling

403 In modeling a physical phenomenon, the main constraint for a modeler is dealing with 404 boundary conditions (Stroud, 2006). The first boundary problems are the Neumann boundary 405 conditions (Broggini et al., 2017). We need a dipolar current injection to avoid overflow from 406 the boundaries of the 2D tank (Günther and Rücker, 2013). Using a reference electrode inside 407 the tank could solve this problem. This reference electrode should be placed slightly away 408 from the current and potential electrodes inside the tank. The second problem that arises for 409 Neumann domains is the non-uniqueness of the solution of the partial differential equation due 410 to a lack of calibration (Günther and Rücker, 2013). We used a calibration electrode to 411 overcome this problem, which must be placed at the boundaries of the tank. The potential at 412 this point (electrode) must be zero. The method used in the modeling was moving both 413 electrodes (reference and calibration electrodes) in the tank and checking if the results 414 obtained were consistent or not (Günther and Rücker, 2013).

To validate our experimental measurements with numerical modeling, we completed two steps: In the first step, the ratio of geometrical factors K obtained from the experiment with water on the 2D tank and model were compared in Figure 7. In both the experiment and model, the K varies around 0.45 to 0.5 for the EDD arrays with $\pm 20\%$ error. We noticed that in points close to the metal grid on both sides of the tank (e.g., points 23, 24, and 25) the difference in K factors obtained from the model and experiment are more significant because, in the model, the effects of metal grids have not been considered.

In Figure 8, after increasing the level from 2 cm (entering the influence zone of EDD measurements) resistivity was increased both in the experiment and model. Since the influence zone of all measurements begins at 2.5 cm. In this figure, the model simulation results properly coincide with the experimental data (Pearson's correlation coefficient, R=0.981). This 426 validates the setup configuration and shows that the electrodes measure the resistivity with 427 good precision. In the transaction part at a height of 3 and 4 cm, an overestimation was seen 428 compared to the experimental resistivity data. This overestimation could be due to the effect of 429 increasing CT and its direct effect on the resistivity value predicted by the model.

430 Phase variation during imbibition

During the imbibition period, both amplitude and phase shift of complex resistivity measured in the middle of the tank were affected by temperature and saturation changes. Figure 9 shows how phase shift varied with time in the pumping influence zone in the middle of the tank for two different frequencies (f=1.46 and 2.92 Hz). Nine measuring zones (we call them points in the following) in the same vertical positions were chosen from three levels of the EDD array in the middle of the tank.

437 Points 12, 15, and 18 are at the top, consisting of the water-saturated zone (GB+water). 438 The results showed that the phase shift is close to zero in these three points. These results 439 confirm previous findings on 1D cells and columns for the water-saturated cases (Iravani et 440 al., 2020b). Points 11, 14, 17, and 10, 13, 16 are at the middle and bottom of the tank with an 441 EDD array, respectively. During the imbibition (pumping process), CT was pumping from the 442 middle bottom of the tank, and therefore, water replaced CT forming the cone of depression. 443 As shown in Figure 9, imbibition increased and then decreased phase shift at points 11, 14, and 444 17 (at the middle of the tank), but it was constant at points 10, 13, and 16 (at the bottom of the 445 tank). This variation occurred sooner at point 14 than at point 11, which corresponds with our 446 expectation because imbibition firstly affected the middle points, then the side points because 447 of the cone of depression. Besides, the phase variations at points 14 and 17 happened at the 448 same time. Drops in phase shift at the end of imbibition happened later for points 11 and 17 449 compared to 14 due to the delay of the imbibition effect from the middle of the tank reaching 450 the sides. After imbibition (dominating water saturation), the phase shift dropped to a value 451 close to zero. An increase in phase was observed in all 3 points in the middle of the tank, 452 which can show the complex behavior of the spectra and phase due to the instantaneous 453 change of water/CT saturation. However, we observed a weak response to imbibition tests for 454 points 10, 13, and 16 situated at the bottom of the tank because they were not influenced by 455 imbibition.

After dominating water in each block of the measurement, the recorded values became stable and close to zero. Comparing results of the phase change between Figure 9a and Figure 9b shows that small changes in frequency have a large effect on phase shift in layers containing DNAPL. The higher frequency, the higher the phase shift.

460 Predicting saturation in 2D tank

In a previous study, Iravani *et al.* (2020b) found a relation between saturation state and the amplitude and phase shift of the SIP spectra for CT+water, to have a calibration relationship for further uses in the 2D tank (current study) and even thereafter in field studies as below (Iravani *et al.*, 2020b):

$$\sigma_{mixed} = 0.0345\theta_w^{1.8} + 10^{-3}(0.4 - \theta_w)^{5.2} + 10^{-14}(0.6)^{2.34}$$
(9)

465 σ_{mixed} was obtained from the inverse relationship with the measured resistivity of the 466 multiphase system in each zone. The only variable in equation (9) is water content (θ_w), 467 which can be present as water saturation after using this equation ($S_w = \theta_w / \Phi$).

Figure 10a,b, and Fig 10c,d show the saturation status and front image of the multiphase system before and after imbibition in the 2D tank, respectively. In Figure 10c,d, the strong reduction of DNAPL saturation in the middle of the 2D tank is due to imbibition. The complex nature of CT that sticks into the 2D tank glass wall surface did not allow image analysis to obtain the saturation. Therefore, we could not make a quantitative comparison of the saturation using these images. Figure 10 shows resistivity measurements in different influence zones with interpolation that can give us the saturation field. In Figure 10c, we expected more saturation changes before and after the pumping at the left side of the tank compared to its right side. The heating from the left side has a direct effect on the viscosity of CT and consequently, the pumping rate, but the DNAPL/water interface is not horizontal and we could not quantify and make this assumption. As observed in Figure 6d and Figure 10c, the effect of imbibition seems to be well observed particularly in both sides compared to the middle.

The CT saturation was not exactly 100% even before starting imbibition because some percentage of water was captured in the pore spaces before injecting CT. It seems the Archie model does not have enough precision (the effect of surface conductivity should not be neglected) in very high resistivity (high CT saturation) and that monitoring in a small 2D tank does not allow us to capture the cone of depression with high precision for imbibition.

486 Effect of temperature and saturation on Cole-Cole parameters

487 In the non-isothermal condition before starting imbibition when the bottom half of the 488 tank was filled with GB+CT, the complex resistivity was measured in the frequency domain 489 (e.g., under electrical impedance conditions) before and after heating. The tank was heated 490 from the left side by a heater and heat was dispersed from the left side of the tank to the right. 491 The most influenced part is close to the heater. There the temperature changed from 20 to 492 50°C, and that change affects two EDD measurements at the far-left side of the tank (No. 1 493 and 4). Figure 11 shows how the four Pelton parameters vary at 20 and 50°C. The 494 experimental data were fitted to the Pelton model by using a Matlab code by Maineult et al. 495 (2017). The fitting curves of the experimental data and model are presented in Figure 12.

496 We have already studied how these parameters vary with temperature in cells and 497 columns (Iravani *et al.*, 2020a). Studying these parameters in the 2D tank showed that ρ_0 and

498 log (τ) were decreasing with temperature increases. Raising the temperature from 20 to 50°C 499 slightly increased c but both m and c were so close to 1 that it can be considered as a constant 500 value. The fact that c is close to 1 means that the SIP response obeys a Debye model, and the 501 fact that m is close to 1 means that the medium is highly chargeable because of poor dispersion 502 of the spectra. These variations in results were the same for both measurements (1 and 4). Our 503 findings on how the four parameters changed in the 2D tank confirm the previous findings on 504 cells and columns with this difference: m was 0.89 and in the 2D tank for both measurements 505 was almost 1.

506 Conclusions

507 The main goal of this study was to move one step closer to execute the usage of electro-508 geophysical methods to monitor the saturation changes in soils contaminated by a DNAPL 509 (like CT) under non-isothermal conditions. In particular, applications of the SIP method in 510 contaminated porous media were examined experimentally in the laboratory by using a 2D 511 sample tank. Glass beads of 1 mm diameter were chosen because of similar physical properties 512 to field soil. This allows to model as much as possible actual field conditions in the laboratory.

513 After preparing the non-isothermal condition by heating from the left side of the tank and 514 correcting the temperature effect, we studied how the electrical complex resistivity varies 515 during imbibition. Changes in the electrical complex resistivity magnitude were measured and 516 the saturation field was obtained from the resistivity data. From the electrical resistivity 517 measurements, we predicted the changes of the saturation field before and after imbibition 518 accordingly. However, the proposed electrical resistivity monitoring did not produce the cone 519 of depression during imbibition with high precision. This may be related to CT's very high 520 resistivity and the small laboratory setup. The phase changes in 9 points in the middle of the 521 tank in all three levels showed that the phase significantly decreases after increasing water 522 saturation (imbibition period).

It is concluded that the temperature (5 to 7%) and saturation change due to replacing CT with water affects measured electrical complex resistivity. The effect of saturation change is much more considerable compared to the temperature. In the coupling of temperature and saturation variation after correcting resistivity with temperature (with a linear relation), the saturation effect will be the only variable in the real case with remediation surveys under the thermal-enhanced technique.

529 The effect of saturation change by injecting CT from the bottom of the tank was 530 evaluated by BERT as a numerical modeling tool. The results show that the measurements 531 with EDD arrays have been properly fitted by the BERT model. This validates our setup 532 configuration and confirms that our electrodes functioned properly. The goal of our study was 533 not to present comprehensive mechanistic modeling of the results but to present the experiment and the results that show the effect of heating and pumping on the SIP response, 534 535 and its interest for field measurements. Developing a mechanistic model is a step beyond, the 536 subject of future research.

537 The reported results showed that the proposed generalized Archie's law from the 538 previous study could predict the saturation. However, during pumping at the bottom of the 539 tank, the proposed coefficient did not perfectly reflect the saturation change. Finding the 540 proper coefficients of generalized Archie's law can lead to precise saturation estimation in 541 both high and low resistivity. These results at the bottom level during imbibition could be due 542 to placing the current electrodes in the part that is saturated with CT. Our previous finding in 543 1D cells supports also the idea that in a case where the current electrodes are located in a 544 saturated DNAPL part, SIP reports very high resistivity although the potential electrodes are 545 still placed in the water-saturated part.

546 How Cole-Cole parameters change before and after heating were evaluated for 20 and 547 50°C before and after heating. The results of this study show that Cole-Cole parameter 548 variations are in accordance with our previous findings.

We believe that the most outstanding part of our findings is the development of knowledge on how to monitor imbibition processes where both temperature and saturation change together in a 2D system. Validating our results in electro-geophysical field studies would advance their reliability in various environmental conditions. Our findings might be used for soils with similar physical properties.

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561 References

- Abdulsamad, F., Florsch, N., Schmutz, M. and Camerlynck, C. (2016) Assessing the high
 frequency behavior of non-polarizable electrodes for spectral induced polarization
 measurements. *Journal of Applied Geophysics*, 135, 449-455.
- Archie G.E., 1942, The electrical resistivity log as an aid in determining some reservoir
 characteristics: *Transactions of the American Institute of Mechanical Engineers*, 146,
 54–67.
- 568 Bagotsky, V.S. ed., 2005. Fundamentals of electrochemistry (Vol. 44). John Wiley & Sons.

- 569 Beck, A.E. (1981) Physical principles of exploration methods. *Macmillan International*570 *Higher Education*.
- Benson, A.K., Payne, K.L. and Stubben, M.A. (1997) Mapping groundwater contamination
 using dc resistivity and VLF geophysical methods–A case study. *Geophysics*, 62(1),
 80-86.
- Beyke, G. and Fleming, D. (2005) In situ thermal remediation of DNAPL and LNAPL using
 electrical resistance heating. Remediation Journal: *The Journal of Environmental Cleanup Costs, Technologies & Techniques*, 15(3), 5-22.
- 577 Binley, A., Slater, L.D., Fukes, M. and Cassiani, G. (2005) Relationship between spectral
 578 induced polarization and hydraulic properties of saturated and unsaturated sandstone.
 579 Water resources research, 41(12), W12417.
- Blondel, A., Schmutz, M., Franceschi, M., Tichané, F. and Carles, M. (2014) Temporal
 evolution of the geoelectrical response on a hydrocarbon contaminated site. *Journal of Applied Geophysics*, 103, 161-171.
- Börner, F., Gruhne, M. and Schön, J. (1993) Contamination indications derived from electrical
 properties in the low frequency range 1. *Geophysical Prospecting*, 41(1), 83-98.
- 585 Breede, K. and Kemna, A. (2012) Spectral induced polarization measurements on variably 586 saturated sand-clay mixtures. *Near Surface Geophysics*, 10(6), 479-489.
- 587 Brewster, M.L., Annan, A.P., Greenhouse, J.P., Kueper, B.H., Olhoeft, G.R., Redman, J.D.
- and Sander, K.A. (1995) Observed migration of a controlled DNAPL release by
 geophysical methods. *Ground water*, 33(6), 977-988.
- 590 Broggini, F., Vasmel, M., Robertsson, J.O. and van Manen, D.J. (2017) Immersive boundary 591 conditions: Theory, implementation, and examples. *Geophysics*, 82(3), 97-110.

- Chambers, J.E., Loke, M.H., Ogilvy, R.D. and Meldrum, P.I. (2004) Noninvasive monitoring
 of DNAPL migration through a saturated porous medium using electrical impedance
 tomography. *Journal of Contaminant Hydrology*, 68(1-2), 1-22.
- Cole, K.S., and Cole, R.H. (1941) Dispersion and absorption in dielectrics I. Alternating
 current characteristics. *Journal of Chemical Physics*, 9(4), 341-351.
- 597 Colombano, S., Davarzani, H., van Hullebusch, E.D., Huguenot, D., Guyonnet, D., Deparis, J.
 598 and Ignatiadis, I. (2020) Thermal and chemical enhanced recovery of heavy chlorinated
 599 organic compounds in saturated porous media: 1D cell drainage-imbibition
 600 experiments. *Science of The Total Environment*, 706, 135758.
- Colombano, S., Davarzani, H., van Hullebusch, E., Huguenot, D., Guyonnet, D., Deparis, J.,
 Lion, F. and Ignatiadis, I. (2021) Comparison of thermal and chemical enhanced
 recovery of dnapl in saturated porous media: 2D tank pumping experiments and twophase flow modelling, *Science of The Total Environment*, 760, 143958.
- Dakhnov, V.N. (1962) Geophysical well logging: The application of geophysical methods;
 electrical well logging. *Colorado School of Mines*.
- 607 Debye, P. (1929) Polar Molecules. *Chemical Catalog Company*, New York.
- Deng, Y., Shi, X., Zhang, Z., Sun, Y., Wu, J. and Qian, J. (2020) Application of spectral
 induced polarization for characterizing surfactant-enhanced DNAPL remediation in
 laboratory column experiments. *Journal of Contaminant Hydrology*, 230, 103603.
- Denicol, P.S. and Jing, X.D. (1998) Effects of water salinity, saturation and clay content on the
 complex resistivity of sandstone samples. *Geological Society*, London, Special
 Publications, 136(1), 147-157.
- Friedrichs, B., Matzander, U., Large, D. and Davies, M. (1999) Controlled Source ElectroMagnetic Mapping development and evaluation of its application to mineral

- exploration. Transactions of the Institution of Mining and Metallurgy. Section B. *Applied Earth Science*, 108.
- Ghorbani, A., 2007. Contribution au développement de la résistivité complexe et à ses
 applications en environnement. Mémoire de thèse de doctorat. Université Pierre-etMarie-Curie, Paris (France).
- 621 Glover, P.W. (2010) A generalized Archie's law for n phases. *Geophysics*, 75(6), E247-E265.
- Glover, P.W.J. (2015) Geophysical properties of the near surface Earth: Electrical properties.
 In Treatise on Geophysics, Schubert, G. (Editor-in-chief). Elsevier: *Oxford*, Vol. 11,
 89-137.
- Gourry, J., Jeannot, R. and Proust, E. (2001) Coupling of geophysical measurements and gas
 analyzes for the detection of pollutants organic on a former coking, *report RP-50856- FR*, BRGM.
- Guéguen, Y. and Palciauskas, V. (1994) Introduction to the Physics of Rocks. *Princeton University Press.*
- 630 Günther, T. and Rücker, C. (2013) Boundless Electrical Resistivity Tomography BERT 2–the
 631 user tutorial.
- 632 Günther, T. and Rücker, C. (2015) Boundless Electrical Resistivity Tomography BERT 2–the
 633 user tutorial.
- Günther, T., Rücker, C., Spitzer, K. (2006) Three-dimensional modelling and inversion of DC
 resistivity data incorporating topography II. Inversion. *Geophysical Journal International*, 166 (2), 506–517, https://doi.org/10.1111/j.1365-246X.2006.03011.x
- Habiyakare, T., Zhang, N., Feng, C.H., Ndayisenga, F., Kayiranga, A., Sindikubwabo, C.,
 Muhirwa, F. and Shah, S. (2022) The implementation of genetic algorithm for the
 identification of DNAPL sources. Groundwater for Sustainable Development, 16,
 100707.

- Hadley, P.W. and Newell, C.J. (2012) Groundwater remediation: The next 30 years. *Groundwater*, 50(5), 669-678.
- Hayashi, M. (2004) Temperature-electrical conductivity relation of water for environmental
 monitoring and geophysical data inversion, *Environmental Monitoring and Assessment*,
 96(1), 119–128.
- Helene, L. P. I., Moreira, C. A. and Bovi, R. C. (2020) Identification of leachate infiltration
 and its flow pathway in landfill by means of electrical resistivity tomography (ERT), *Environmental Monitoring and Assessment*, 192(4), 249.
- 649 Illangasekare, T.H., Ramsey Jr, J.L., Jensen, K.H. and Butts, M.B. (1995) Experimental study
 650 of movement and distribution of dense organic contaminants in heterogeneous aquifers.
 651 *Journal of Contaminant Hydrology*, 20(1-2), 1-25.
- Iravani, M.A., Deparis, J., Davarzani, H., Colombano, S., Guérin, R. and Maineult, A. (2020a)
 The influence of temperature on the dielectric permittivity and complex electrical
 resistivity of porous media saturated with DNAPLs: A laboratory study, *Journal of Applied Geophysics*, 172, 103921.
- Iravani, M.A., Deparis, J., Davarzani, H., Colombano, S., Guérin, R. and Maineult, A. (2020b)
 Complex electrical resistivity and dielectric permittivity responses to dense nonaqueous phase liquids' imbibition and drainage in porous media: a laboratory study, *Journal of Environmental and Engineering Geophysics*, 25(4), 557-567.
- Kavanaugh, M.C., Suresh, P. and Rao, C. (2003) The DNAPL remediation challenge: Is there
 a case for source depletion? *Environmental protection agency Washington DC*,
 EPA/600/R-03/143.
- Keller, G.V. and Frischknecht, F.C. (1966) Electrical methods in geophysical prospecting.
 Pergamon Press, Oxford.

665	Kemna, A., Binley, A., Cassiani, G., Niederleithinger, E., Revil, A., Slater, L., Williams, K.H.,
666	Orozco, A.F., Haegel, F.H., Hoerdt, A. and Kruschwitz, S. (2012) An overview of the
667	spectral induced polarization method for near-surface applications. Near Surface
668	Geophysics, 10(6), 453-468.

- 669 Kremer, T., Schmutz, M., Leroy, P., Agrinier, P. and Maineult, A. (2016a) Modelling the 670 spectral induced polarization response of water-saturated sands in the intermediate 671 frequency range (102 - 105)Hz) using mechanistic and empirical 672 approaches. Geophysical Supplements to the Monthly Notices of the Royal Astronomical Society, 207(2), 1303-1312. 673
- Kremer, T., Schmutz, M., Maineult, A. and Agrinier, P. (2016b) Laboratory monitoring of
 CO₂ injection in saturated silica and carbonate sands using spectral induced
 polarization. *Geophysical Supplements to the Monthly Notices of the Royal Astronomical Society*, 207(2), 1258-1272.
- Keys, W.S. (2017) A practical guide to borehole geophysics in environmental investigations.
 Routledge.
- Leroy, P., Revil, A., Kemna, A., Cosenza, P. & Ghorbani, A., 2008. Complex conductivity of
 water-saturated packs of glass beads, *J. Colloid Interface Sci.*, **321**(1), 103–117, doi:
- 682 10.1016/j.jcis.2007.12.031.
- Liao, Q., Deng, Y., Shi, X., Sun, Y., Duan, W. and Wu, J. (2018) Delineation of contaminant
 plume for an inorganic contaminated site using electrical resistivity tomography:
 comparison with direct-push technique, *Environmental Monitoring and Assessment*,
 190(4), 187.
- 687 Ling, C. and Zhang, Q. (2017) Evaluation of surface water and groundwater contamination in
 688 a msw landfill area using hydrochemical analysis and electrical resistivity tomography:

- a case study in sichuan province, southwest china, *Environmental Monitoring and Assessment*, 189(4), 140.
- Maineult, A., Bernabé, Y. and Ackerer, P. (2004) Electrical response of flow, diffusion, and
 advection in a laboratory sand box. *Vadose Zone Journal*, 3(4), 1180-1192.
- Maineult, A., Jougnot, D. and Revil, A. (2018) Variations of petrophysical properties and
 spectral induced polarization in response to drainage and imbibition: a study on a
 correlated random tube network. *Geophysical Journal International*, 212(2), 13981411.
- Maineult, A., Revil, A., Camerlynck, C., Florsch, N. and Titov, K. (2017) Upscaling of
 spectral induced polarization response using random tube networks. *Geophysical Journal International*, 209(2), 948-960.
- Mao, D., Lu, L., Revil, A., Zuo, Y., Hinton, J. and Ren, Z.J. (2016) Geophysical monitoring of
 hydrocarbon-contaminated soils remediated with a bioelectrochemical system.
 Environmental science & technology, 50(15), 8205-8213.
- Martinho, E., Almeida, F. and Matias, M.S. (2006) An experimental study of organic pollutant
 effects on time domain induced polarization measurements. *Journal of Applied Geophysics*, 60(1), 27-40.
- Niu, Q. & Revil, A., 2016. Connecting complex conductivity spectra to mercury porosimetry
 of sedimentary rocks, *Geophysics*, 81(1), E17–E32, doi: 10.1190/GEO2015-0072.1.
- 708 Olhoeft, G.R. (1985) Low-frequency electrical properties. *Geophysics*, 50(12), 2492-2503.
- Orlando, L. and Renzi, B. (2015) Electrical permittivity and resistivity time lapses of
 multiphase DNAPLs in a lab test. *Water Resources Research*, 51(1), 377-389.
- 711 Orozco, F.A., Ciampi, P., Katona, T., Censini, M., Papini, M. P., Deidda, G. P. and Cassiani,
- G. (2021) Delineation of hydrocarbon contaminants with multi-frequency complex
 conductivity imaging, *Science of The Total Environment*, 768, 144997.

- Pankow, J.F. and Cherry, J.A. (1996) Dense chlorinated solvents and other DNAPLs in
 groundwater: History, behavior, and remediation.
- Pearce, J.A. and Zuluaga, A. (2004) Electrical properties of soils at ism radio frequencies,
 from 1 to 40 MHz. *Journal of Microwave Power and Electromagnetic Energy*, 39(3-4),
 179-190.
- Pelton, W.H., Ward, S.H., Hallof, P.G., Sill, W.R. and Nelson, P.H. (1978) Mineral
 discrimination and removal of inductive coupling with multifrequency IP. *Geophysics*,
 43(3), 588-609.
- Philippe, N., Davarzani, H., Colombano, S., Dierick, M., Klein, P.Y. and Marcoux, M. (2020)
 Experimental study of the temperature effect on two-phase flow properties in highly
 permeable porous media: Application to the remediation of dense non-aqueous phase
 liquids (DNAPLs) in polluted soil. *Advances in Water Resources*, 146, 103783.
- Power, C., Gerhard, J.I., Karaoulis, M., Tsourlos, P. and Giannopoulos, A. (2014) Evaluating
 four-dimensional time-lapse electrical resistivity tomography for monitoring DNAPL
 source zone remediation. *Journal of Contaminant Hydrology*, 162, 27-46.
- Revil, A., Binley, A., Mejus, L. and Kessouri, P. (2015) Predicting permeability from the
 characteristic relaxation time and intrinsic formation factor of complex conductivity
 spectra. *Water Resources Research*, 51(8), 6672-6700.
- Revil, A., Schmutz, M. and Batzle, M.L. (2011) Influence of oil wettability upon spectral
 induced polarization of oil-bearing sands. *Geophysics*, 76(5), A31-A36.
- Reynolds, J.M. (2011) An introduction to applied and environmental geophysics. *John Wiley & Sons.*
- Ronczka, M., Rücker, C. and Günther, T. (2015) Numerical study of long-electrode electric
 resistivity tomography—Accuracy, sensitivity, and resolution. *Geophysics*, 80(6),
 E317-E328.

- Rücker, C. and Günther, T. (2011) The simulation of finite ERT electrodes using the complete
 electrode model. *Geophysics*, 76(4), F227-F238.
- Rücker, C., Günther, T. and Spitzer, K. (2006) Three-dimensional modelling and inversion of
 dc resistivity data incorporating topography—I. Modelling. *Geophysical Journal International*, 166(2), 495-505.
- 744 Sadowski, R.M. (1988) Clay-organic interactions. M.Sc. thesis, *Colorado School of Mines*.
- Sauck, W.A., 2000. A model for the resistivity structure of LNAPL plumes and their environs
 in sandy sediments. *Journal of Applied Geophysics*, 44(2-3), 151-165.
- Schmutz, M., Revil, A., Vaudelet, P., Batzle, M., Viñao, P.F. and Werkema, D.D. (2010)
 Influence of oil saturation upon spectral induced polarization of oil-bearing sands. *Geophysical Journal International*, 183(1), 211-224.
- Sen, P.N. and Goode, P.A. (1992) Influence of temperature on electrical conductivity on shaly
 sands. *Geophysics*, 57(1), 89–96.
- Slater, L., Barrash, W., Montrey, J. and Binley, A. (2014) Electrical- hydraulic relationships
 observed for unconsolidated sediments in the presence of a cobble framework. *Water Resources Research*, 50(7), 5721-5742.
- Slater, L.D., Choi, J. and Wu, Y. (2005) Electrical properties of iron-sand columns:
 Implications for induced polarization investigation and performance monitoring of
 iron-wall barriers. *Geophysics*, 70(4), G87-G94.
- Sparrenbom, C. J., Åkesso, S., Johansson, S., Hagerberg, D. and Dahlin, T. (2017)
 Investigation of chlorinated solvent pollution with resistivity and induced polarization, *Science of The Total Environment*, 575, 767–778.
- Spies, B.R. (1996) Electrical and electromagnetic borehole measurements: A review. Surveys
 in Geophysics, 17(4), 517-556.

- Stroud, I. (2006) Boundary representation modelling techniques, *Springer Science & Business Media*, ISBN: 978-1-84628-312-3.
- Susanto, A., Koleva, D.A., Copuroglu, O., van Beek, K. and van Breugel, K. (2013)
 Mechanical, electrical and microstructural properties of cement-based materials in
 conditions of stray current flow. *Journal of Advanced Concrete Technology*, 11(3),
 119-134.
- Titov, K., Kemna, A., Tarasov, A. and Vereecken, H. (2004) Induced polarization of
 unsaturated sands determined through time domain measurements. Vadose Zone
 Journal, 3(4), 1160-1168.
- Ulrich, C. and Slater, L. (2004) Induced polarization measurements on unsaturated,
 unconsolidated sands. Geophysics, 69(3), 762-771.
- Vanhala, H. (1997) Mapping oil-contaminated sand and till with the spectral induced
 polarization (SIP) method. *Geophysical prospecting*, 45(2), 303-326.
- Vanhala, H., Soininen, H. and Kukkonen, I. (1992) Detecting organic chemical contaminants
 by spectral-induced polarization method in glacial till environment. *Geophysics*, 57(8),
 1014-1017.
- Vaudelet, P., Schmutz, M., Pessel, M., Franceschi, M., Guerin, R., Atteia, O., Blondel, A.,
 Ngomseu, C., Galaup, S., Rejiba, F. and Bégassat, P. (2011) Mapping of contaminant
 plumes with geoelectrical methods. A case study in urban context. *Journal of Applied Geophysics*, 75(4), 738-751.
- Weller, A. and Börner, F.D. (1996) Measurements of spectral induced polarization for
 environmental purposes. *Environmental Geology*, 27(4), 329-334.
- Weller, A., Nordsiek, S. and Debschütz, W. (2010) Estimating permeability of sandstone
 samples by nuclear magnetic resonance and spectral-induced polarization. *Geophysics*,
 75(6), E215-E226.

- Wilson, V., Power, C., Giannopoulos, A., Gerhard, J. and Grant, G. (2009) DNAPL mapping
 by ground penetrating radar examined via numerical simulation. *Journal of Applied Geophysics*, 69(3-4), 140-149.
- Wong, P. and Horn, P.M. (1979) Magnetic phase-diagram of (Fe_xCo_{1-x}) Cl₂. *American Physical Society*, 24(3), 363-363.

793 **FIGURE LEGENDS**

Figure 1 Schematic layout of the a) front, b) top and, c) back of the 2D tank with the description of each element. In the front view of the tank, a double glazed transparent glass is used as a wall to better control the temperature and imaging purpose. Isothermal system includes a system of heating all around the tank using hot water. A stainless-steel heating element was inserted inside the left-side of the tank to produce the non-isothermal condition. In sub-figure c) the red square showed an EDD measurement. There are 27 EDD measurements in three different levels.

Figure 2 Equatorial dipole-dipole arrays from the front side of the tank. 10 current (metal)
electrodes and 10 potential (non-polarized) electrodes in each level can record 9 EDD
measurements. In three levels of measurements, 27 measurements can be recorded. Using
other arrays (e.g., Wenner Beta) is also possible between two levels.

Figure 3 Linear regression to find the α values for a) GB+water in the top level and b) GB+CT for the bottom level of EDD array. The obtained α values for GB+water and GB+CT are in accordance with the results of the previous study on 1D columns (Iravani et al., 2020a).

Figure 4 Back view of the temperature field in the tank under non-isothermal conditions a) before, and b) after imbibition (the plus (+) signs are the positions of the thermocouples and red dot is the pumping point at the middle bottom of the tank). A significant temperature (54 and 18 °C) difference between the right and the left side of the tank is due to using a heater at the right side of the tank (in the back view).

Figure 5 Different steps of experimenting on the 2D tank. a) filling the tank with GB and water, b) tank packed with GB and fully saturated by water, c) injecting CT from 5 points at the bottom of the tank (for the places of injecting points, please see Figure 1a), d) increasing step by step the level of the CT in the tank, e) possible bump shape of the CT surface – the flat interface is ideal, f) almost half of the tank filled with GB+CT. This stage is the starting point of the imbibition experiment.

Figure 6 Log of the resistivity field in the tank a) before imbibition, b) after imbibition without temperature correction, and c) before and d) after imbibition with temperature correction, e) inverted log resistivity before temperature correction, and f) inverted log resistivity after temperature correction (the circle (\bullet) and blue arrow signs are respectively the positions of the EDD measurements and drainage points).

Figure 7 Experimental and model ratio of K factor for 27 measurements of electrical resistivity with EDD array. Point values closer to 1 show a better model prediction of the experimental data. Points far from the walls have better results because of the metallic grid used in the side walls.

Figure 8 Variation of resistivity as a function of CT height for EDD array at the bottom level of the tank. These are apparent resistivity obtained from the average values of the 9 measurements at the bottom level of the 2D tank (measurements No: 1, 4, 7, 10, 13, 16, 19, 22, and 25; see Figure 2).

Figure 9 Variation of phase shift during imbibition period at the middle of the tank in 9 points (12, 15, and 18 at the top level, 11, 14, and 17 at the middle level, and 10, 13, and 16 at the bottom level) for the frequencies of a) 1.46 Hz and b) 2.92 Hz.

- **Figure 10** a) The measured CT saturation field before imbibition, b) front image of the 2D tank before imbibition, c) the measured CT saturation field after imbibition, and d) front image of the 2D tank after imbibition. The black lines in "a" and "c" are the counters between two areas (GB+water and GB+CT).
- Figure 11 Effect of temperature on Cole-Cole parameters (20 and 50°C) close to the heater.
 As it was shown in Figure 5, just points close to the heater have recorded the temperature around 50 °C.
- **Figure 12** Fitting curves of experimental data and model for a) point 1 at 20°C, b) point 1 at 50°C, c) point 4 at 20°C, and d) point 4 at 50°C. For more information about choosing the presented points, see Figure 11 description).

846	Data Availability Statements
847	
848	The datasets generated and analyzed during the current study are available from the
849	corresponding author on reasonable request.
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851	Declaration of interests
852	
853 854	\boxtimes The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
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856 857 858 859	□The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: