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# Cross-borehole geoelectrical time-lapse monitoring of in situ chemical oxidation and permeability estimation through induced polarization

Thue Bording<sup>1</sup>, Anders Kristian Kühl<sup>1</sup>\*, Gianluca Fiandaca<sup>1,2</sup>, Jørgen Fjeldsø Christensen<sup>3</sup>, Anders Vest Christiansen<sup>1</sup> and Esben Auken<sup>1</sup>

<sup>1</sup>HydroGeophysics Group, Department of Geoscience, Aarhus University, Aarhus, Denmark, <sup>2</sup>Dipartimento di Scienze della Terra 'A. Desio', Università degli Studi di Milano, Milan, Italy, and <sup>3</sup>Region of Southern Denmark

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# ABSTRACT

Worldwide, soil contamination due to industrial activities is a major issue. One method for remediation of contaminated sites is *in situ* chemical oxidation, where an oxidizing agent is injected into the contaminated soil. Normally, monitoring wells are established in the remediation area for tracking the oxidizing agent. However, wells only provide point information of the injectant spread. This issue can be addressed using cross-borehole resistivity and induced polarization tomography, by mapping the electrical properties in the entire remediation volume and by deriving, through petrophysical relations, the hydraulic properties of the medium. Here we present a proof-ofconcept study, performed over one year as part of a larger remediation project, where resistivity and time-domain induced polarization data were acquired among 10 boreholes, before and after two rounds of injection of oxidizing agents. The time-lapse resistivity models, obtained through a focusing inversion scheme that favours compact time-lapse changes, clearly show the oxidizing agent spread as highly conductive anomalies and confirmed by water conductivity measurements in boreholes. The time-lapse inversions also show spatial variability in the injectant spread, with some areas not reached. The induced polarization data quality decreased significantly just after the injection rounds, because of the decrease in resistivity and induced polarization signal level, so that induced polarization time-lapse inversions were not feasible. However, the induced polarization data were used for background characterization and to estimate permeability. In particular, there is a good match between the imaged low-permeability zones and the areas in which the injectant did not spread, identified by the time-lapse resistivity inversions. Furthermore, geological samples confirm the presence of fine-grained sediments in the estimated low-permeability zones. While time-lapse resistivity tomography may be used for documenting the injectant spread, induced polarization permeability estimates prior to injection can be used to better tailor the remediation in terms of dimension and location of injection filters.

Key words: Cross-borehole, Electrical resistivity tomography, Permeability, Timelapse monitoring.

## INTRODUCTION

Soil contamination due to past industrial activities has become a major issue in urban and natural areas worldwide.

*E-mail:	anders.	kuhl@gec	.au.dk
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Contamination can be found on the grounds of former and current dry cleaners, gas stations and chemical factories, as well as in the landfills that received waste from such sites. In Denmark alone, over 10,000 of these sites have been identified, with ten sites having been designated as 'Generational Contaminations'. These are large and complex polluted sites, which pose an active risk for either the groundwater or the environment and are expected to be problematic for generations to come if no actions are taken.

One way of addressing these contaminated sites is through in situ chemical remediation, where a remediation agent is injected into the contaminated soil, in order to break down the pollutants (Tsitonaki et al., 2010). One of the main issues with this type of remediation strategy is ensuring that the injectant spreads throughout the entire targeted volume (Pac et al., 2019). Traditionally this is verified from a number of monitoring wells with samples taken for chemical analysis in order to determine whether the injectant has reached the well or not. However, this approach does not provide any direct information on the spread of the remediation material between the monitoring wells, as it is limited by the positions of the wells and the screen sizes. As the geology of the targeted volume can often be complex, it can lead to false assumptions on the real spread of the remediation material. This can easily be the case in Danish glacial and postglacial deposits, where sand bodies can be found within clays and vice versa. The resulting large variations in permeability between the different lithologies then lead to inhomogeneous flow paths for the injectant.

Cross-borehole electrical resistivity tomography provides a possible method for mapping the spread of the injected remediation material between the observation wells when the injectant has a considerable difference in conductivity to the native pore fluid. Resistivity mapping could also be accomplished through surface measurements, but for these the vertical resolution inevitably decreases with depth. The cross-borehole approach allows for a high, constant vertical resolution throughout the volume of interest, given appropriately spaced boreholes and borehole electrodes. The usefulness of cross-borehole resistivity tomography has been demonstrated for various purposes, such as the characterization of hydrogeological properties of the vadose zone (Binley et al., 2002; Looms et al., 2008a), monitoring of unsaturated flow and transport (Looms et al., 2008b), characterization of solute transport by use of saline tracer tests (Slater et al., 2000; Perri et al., 2012), the monitoring of remediation (LaBrecque et al., 1996; Mao et al., 2015; Mao et al., 2016) and mapping of possible contaminant flow paths (Bording et al., 2019).

Increasingly, induced polarization (IP) measurements are included in resistivity measurement campaigns. The IP data can add complementary information for the characterization of the subsurface (Kemna et al., 2004) and can be measured in both the frequency domain (FD) or the time domain (TD), see Maurya, et al. (2018b) for an in-depth comparison of the strengths and limitations of both methods for field surveys. FDIP has been used in a number of environmental and remediation studies for monitoring progress and changes to the remediation effort. In many applications, it has been shown that the FDIP data can be used in the continual monitoring of activity of in situ bioremediation (e.g. Commer et al., 2011; Chen et al., 2012; Orozco et al., 2011, 2013) and monitoring chemical remediation through injection of micro to nanoscale iron (e.g. Slater and Binley, 2006a; Flores Orozco et al., 2015; Flores Orozco et al., 2019).

One of the earliest applications of IP measurements in the cross-borehole case was the FD IP study presented in Kemna, *et al.* (2004), where the target was to image hydrocarbon contamination and the lithologies present, as well as to estimate permeability at different contaminated sites. An early use of FDIP in cross-borehole was for monitoring the integrity of a permeable reactive barrier (Slater and Binley, 2003, 2006a).

The TDIP method, which allows for acquiring the IP data simultaneously with the resistivity data, has been used in cross-borehole applications since the early days (Slater and Binley, 2006b) and has recently seen increasing use. Binley *et al.* (2016) presented a TDIP study, where the goal was a hydrogeological characterization and permeability estimation of the unconsolidated sediments. Here they used the integral chargeability measurement, disregarding the spectral content present in the IP data. Similarly, Nivorlis et al. (2019) used TDIP measurements for characterization of remediation efforts, but only used integral chargeability. In Bording *et al.* (2019), the first application of full-decay spectral TDIP in cross-borehole configurations was presented for the mapping of finer sand structures in a glacial till deposit.

In surface applications, full-decay spectral TDIP has been used in a number of different studies, such as for the characterization of landfills and contaminated sites (Gazoty *et al.*, 2012a; Johansson *et al.*, 2015; Wemegah *et al.*, 2017), lithological discrimination (Gazoty *et al.*, 2012b; Rossi *et al.*, 2017), time-lapse monitoring of CO<sub>2</sub> injections (Fiandaca *et al.*, 2015; Doetsch *et al.*, 2015a) and active layer dynamics (Doetsch *et al.*, 2015b) and, more recently, for permeability estimation (Maurya *et al.*, 2018a; Fiandaca *et al.*, 2018c).

In this study, we present the application of cross-borehole resistivity and full-decay spectral TDIP tomography in a



Figure 1 (a) Location of the survey site Kaergaard Plantation in blue and Grindstedværket in red. (b) Overview of pit 3. (c) Close-up of the used electrode tubes; the lines denote the measured configurations, and the configurations presented in this paper are in yellow.

remediation project, with the aim of mapping the spread of a conductive oxidizing agent (persulfate activated by hydrogen peroxide) through time-lapse measurements, and of extracting reliable information from the IP data, both for inferring possible effects of the contamination on the IP properties and for estimating permeability. In particular, the study was performed in Kaergaard Plantation, shown in blue in Figure 1(a), one of the largest contaminated sites in Denmark, in which two rounds of injection of oxidizing agent were carried out in 2018, together with water and soil geochemical samplings and with six rounds of TDIP measurements over 10 electrode boreholes.

The oxidizing agent is often, as in this study, highly conductive, which results in a conductive anomaly with strong contrast where the agent is present. In order to avoid too smooth variations in the high-contrast time-lapse inversion models due to regularization, we used the asymmetric generalized minimum support (AGMS) norm developed by Fiandaca et al. (2015) for the time-lapse constraints. This norm favours compact time-lapse changes, and it significantly improves the size, shape and magnitude estimates of the time-lapse changes. Unfortunately, the signal level of the TDIP data decreased significantly during the injection rounds because of the high conductivity of the oxidizing agent. This caused the IP signals to become too noisy just after the injection rounds, which means that these IP data sets are not used for time-lapse inversions. However, the IP data collected before injections are used for characterizing the electrical properties of the site before remediation and for estimating permeability. The permeability estimation is carried out following the approach of Fiandaca et al. (2018c) and Maurya et al. (2018a), which applies the empirical petrophysical relations developed by Weller et al. (2015) on the inversion parameters retrieved from a full-decay spectral TDIP inversion. The choice of using the AGMS time-lapse

approach and the full-decay spectral IP inversion restricted the choice of the inversion algorithm to 2D cross-borehole inversions. These are carried out following Fiandaca *et al.* (2013) and Bording *et al.* (2019) with the AarhusInv inversion algorithm (Auken *et al.*, 2015), because these approaches are not available in 3D inversion algorithms (e.g. Commer *et al.*, 2011; Günther and Rücker, 2012; Karaoulis *et al.*, 2013; Loke *et al.*, 2014; Binley *et al.*, 2016). It is, however, not without limitations to use a 2D inversion where 3D effects are to be expected. The appropriateness and limitations of this 2D approach will be discussed in detail in the Results and Discussion sections.

## SITE DESCRIPTION

## Geological setting

The site is located in western Jutland, Denmark, in between sand dunes and 600 m from the coast. It was formed postglacially as part of a barrier coast, with a complex system of inlets, barrier-islands, lagoons and tidal inlets. This has resulted in a varied deposition of permeable sands and gravels, as well as occasional silts and clays. These coarser materials were deposited in higher energy regimes such as channels, while the finer materials were deposited when cut off from the ocean, such as by a coastal barrier. This environment means that there can be lateral and vertical variabilities in hydraulic conductivity in the area, which may influence both the distribution of present contaminants and the transport of the injected material. At a depth of 20-25 m, a clayey-silt layer underlies the site. This layer has not been penetrated by boreholes, as it is thought to be impermeable to the pollution, and it might result in contamination of deeper groundwater (Arbejdsgruppen vedrørende Kærgård Plantage, 2006). A geological description

based on core samples is available from borehole B809 in the elevation interval 2.8 m to -0.2 m, showing fine-grained sand in the top, coarsening downwards alternating between coarse sands and gravel.

## Contamination

In the period between 1956 and 1973, daily tank truck deliveries with pharmaceutical wastewater from Grindstedværket, seen in red in Figure 1(a), were deposited in Kaergaard Plantation in six separate pits. In total, an estimated 286,000 m<sup>3</sup> of wastewater was dumped at the site. The resulting groundwater contamination includes both dense and light non-aqueous phase liquids, in the form of sulphonamides, barbiturates, benzene, and chlorinated solvents and poses a health risk to humans and local wildlife. Due to the contamination leaching from the groundwater into the ocean, a bathing ban has been imposed along a 1400 m stretch of the coast (Arbejdsgruppen vedrørende Kærgård Plantage, 2005). An airborne transient electromagnetic (TEM) survey over the site in Kaergaard Plantation (Christensen and Halkjær, 2014) showed that the average resistivity in the area was around 200  $\Omega$ m, with a conductive plume found in elevations -10 m to -20 m relative to sea level, with resistivity values around 5–10  $\Omega$ m and lower, possibly due to the presence of degradation products.

## Remediation

Remediation of the site was carried out in several phases. Between 2007 and 2015, over 7500 tonnes of toxic sludge were excavated from the surfaces of pits 1 through 4. This sludge was then transported and thermally treated in Germany and the Netherlands, and thus the upper 3 m of the pits were replaced with fine sands, to prevent any immediate contact between the remaining pollution and the surface. At the same time, the Region of Southern Denmark completed a technology development project, which showed that a chemical primary treatment followed by a secondary biological treatment would be the most efficient method to clean the contamination affecting the groundwater in the area.

The chemical treatment uses persulfate activated by hydrogen peroxide, which releases free radicals that oxidize the bulk of the contamination (Tsitonaki *et al.*, 2010). Following the chemical treatment, the secondary biological treatment consists of adding Dehalococcoide bacteria, a natural soil bacterium which can break down the remaining chlorinated solvents in the soil (Region of Southern Denmark, personal communication). The second phase of the remediation began in 2017, where pit 3 was chosen as the first site for clean-up using the chosen procedure. The chemical treatments were applied by injecting the oxidizing agent in a dense hexagonal grid of injection wells covering  $\sim 1000 \text{ m}^2$  with 3 m between each injection point. In total 126 injection points were established across the pit, with each point consisting of two boreholes directly adjacent to each other with filters at two different depths. The injections were performed between elevations 3 m and 0 m in two separate filters placed at elevations 3 m to 1.5 m and 1.5 m to 0 m. Throughout the remediation process, a geochemical monitoring was performed by use of several monitoring wells established in the pit. The location of all boreholes in pit 3 and their purpose can be seen in Figure 1(b).

#### Monitoring wells for geochemistry

Thirty-three wells were established in pit 3 for monitoring geochemistry. Twenty-four of the monitoring wells have two filters at elevations 2.8 m to 1.3 m and 1.3 m to -0.2 m, while the remaining nine monitoring wells have three filters, of which only the uppermost ranging from elevation 2.8 m to -1.2 m is in our interest. Two monitoring boreholes, one of each type, are located inside the study area, i.e. B870 and B809. The geochemical data collected in the wells, together with analyses of soil samples collected at/close to B809 before/after the injection rounds, are presented in the Methodology section.

Monitoring wells are typically used to evaluate both the spread of the oxidizing agent and the degradation of contaminants, but the information that can be achieved with monitoring wells comes with two major limitations: limited lateral resolution and limited vertical resolution (Pac *et al.*, 2019). The vertical resolution of the monitoring wells is limited by the number and lengths of filters, with large filters averaging large volumes, making it impossible to tell if the oxidizing agent is present in the entire interval or just a part of it. The lateral resolution is limited by the number and spatial distribution of the monitoring wells. Looking at Figure 1(b), it is also clear that the majority of the area investigated in this study does not have a monitoring borehole in the immediate vicinity.

## METHODOLOGY

#### Geophysical survey layout, data acquisition and processing

The measurement set-up in the field consisted of ten electrode boreholes, placed within a  $55 \text{ m}^2$  area (see Fig. 1b,c). The

Activity	Date	# Quadrupoles per 3-borehole configuration
7–8 February 2018	Installation of electrodes	-
8–9 March 2018	1st measurement round	2026-2287
7-8 May 2018	2nd measurement round	1629–1708
5–6 June 2018	1st injection round	-
6–7 June 2018	3rd measurement round	1681–1860
26–27 September 2018	4th measurement round	2205-2575
29–30 October 2018	2nd injection round	-
30-31 October 2018	5th measurement round	2216-2575
14-15 March 2019	6th measurement round	2157-2575

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electrode tubes installed in the boreholes had 32 electrodes with an electrode spacing of 25 cm. The electrode tubes were made by clamping a 2 cm acid-resistant, stainless steel ring onto the 32 mm PVC tubes. Individual wires were soldered onto the metal rings, led along the tube and finally joined in a 32-pin connector.

A 12-channel ABEM Terrameter LS was used to perform the TDIP measurements. A 100% duty cycle waveform was used with 2 seconds on-time (Olsson *et al.*, 2015). Three array types were used in acquiring the measurements: (a) collinear dipole–dipole single borehole measurements, (b) parallel cross-borehole dipole–dipole arrays, (c) equatorial crossborehole dipole–dipole arrays; these are described in detail in Bording, *et al.* (2019). The measurement sequence was optimized as to maximize the time between the electrodes being used as potential after having been used as current electrodes, inorder to minimize effects of electrode polarization.

The electrodes were installed in February 2018 and data were subsequently acquired in six rounds during 2018-2019, of which there were two rounds of injections (see Table 1). During each measurement round, nine different configurations were measured, each consisting of three electrode boreholes (see the line segments in Fig. 1c). Table 1 also shows the number of quadrupoles measured for each round. The acquisition time was 12-18 hours for each round depending on the number of quadrupoles measured. The variation in the number of acquired data is mainly due to changes in contact resistance above the groundwater table, as electrodes with contact resistances above a cut-off criterion of 20 k $\Omega$  were not used. As an example of the contact resistance, Figure 2(a) shows the contact resistances measured in borehole 5 for the two injection rounds, while Figure 2(b) shows the contact resistances for non-injection rounds. The electrode resistance was high in the unsaturated zone, in the tens of  $k\Omega$ , but beneath this, contact resistances decreased, to a few  $k\Omega$ . It is also seen in Figure 2(a) that the contact resistances drop down to a few hundreds of  $\Omega$ , for a large fraction of the electrodes in rounds 3 and 5. This is immediately after the injection and is a clear indication of the injectant approaching the electrodes. The cutoff criterion of 20 k $\Omega$  for omitting electrodes in the measurements is also seen in red in Figure 2, showing that most of the deactivated electrodes are in the upper 2 m and that the number of electrodes with high contact resistance varies between the different measurement rounds.

The full waveform data were signal-processed following the methodology described by Olsson et al. (2016) with harmonic de-noising, drift correction and spike removal. Following the signal processing, the induced polarization (IP) data were imported to the Aarhus Workbench software (www.aarhusgeosoftware.dk), where a manual processing of the IP decay curves was performed, where erratic or negative decays were removed, along with gates deviating from otherwise smooth decays. The main issues were high contact resistance for some of the electrodes, capacitive coupling and poor signal-to-noise ratio in general. In general, the quality of the IP data was not high, so a large fraction of the data had to be removed. In the measurements rounds directly after injections, the highly increased conductivity further decreased the signalto-noise ratio and the quality of the IP data. Figure 3 presents the comparison on selected representative quadrupoles of the IP decays before and after the first round of injection (i.e., acquisition rounds 1 and 3), where it is clearly visible that the IP data quality decreases significantly just after the injection, due to the decrease in resistivity values, and consequently in the IP voltage level: IP data just after the injection rounds cannot be used for inversion.



Figure 2 Contact resistances measured in borehole 5. (a) Contact resistances for the injection rounds. (b) Contact resistances of the non-injection rounds. The red line shows the cut-off criteria used for no-contact electrodes.

#### Water and soil samples

In-field measurements in the monitoring wells consisted of water conductivity, pH, Redox potential and temperature, while lab analyses were carried out for a range of chemical components of which we have included perchloroethylene (PCE) and total sum of hydrocarbons. The water chemistry available within the study area is presented in Table 2. In particular, water conductivity measured in the filters and shown in Table 2 will be used for comparison to the geophysical imaging in the Results section. Unfortunately, the monitoring programme did not measure water conductivity in boreholes B809 and B870 prior to the injections, so there are no baseline values for water conductivity.

In borehole B809, soil samples were collected prior to the first injection round. After the second injection round and immediately adjacent to B809, a new set of soil samples were collected. The samples were analysed for a range of chemical compounds, of which we present PCE and total sum of hydrocarbons in Table 3. Prior to the remediation, the highest concentrations were found at elevation 2.3 m.

## Inversion

The collected data were inverted using two different approaches, all within the framework of AarhusInv (Auken et al., 2015). The first approach is an  $L_2$  time-domain IP inversion used for background and permeability estimation. The second approach is time-lapse inversions of resistivity to map conductive anomalies. The time-domain IP forward responses were calculated as described in Fiandaca et al. (2013), with a modification for buried electrodes used in Bording et al. (2019). When calculating the forward response, it is assumed that the 2D model extends indefinitely perpendicularly to the measured profile, with the electrodes modelled as points. The inversions were performed along intersecting profiles and compared to identify the effect of possible 3D structures on the models in the vicinity of the intersections. The inversions were carried out on a regular grid consisting of 81×25 cells, with cells dimensions 12.5 cm  $\times$  50 cm (thickness  $\times$  width) and standard smooth  $(L_2)$  horizontal and vertical constraints. The horizontal constraints were set to 1.075, which is the standard deviation factor of the roughness covariance matrix in the objective function; similarly, vertical constraints were set to 1.1.

The time-lapse inversions were performed following the framework of Fiandaca et al. (2015). This inversion scheme uses an iteratively reweighted least-squares (IRLS) approach, which allows for using norms in the time-lapse constraints different from the standard  $L_2$  least-squares norm. In particular, the asymmetric generalized minimum support norm (AGMS) proposed by Fiandaca et al. (2015) was used for the timelapse constraints. The AGMS norm penalizes all the time-lapse variations over the given threshold  $\sigma$  of the parameter variations (5% in the inversions presented here), regardless of the magnitude of the variation itself. In this way, big parameter variations are not over-penalized, avoiding the too smooth variations in the time-lapse inversion models that might occur when using the classic  $L_2$  time-lapse norm. The AGMS norm depends on a second setting, i.e. the expected relative area/volume of time-lapse changes,  $\alpha$ , which controls the relative weight of data and model penalizations in the objective function. The same value,  $\alpha = 0.2$ , was used when inverting all acquisition rounds against the background model, chosen



Figure 3 Selected IP data from profile E03-E05-E07. (a) Round 1, used for IP inversions. (b) Round 3, immediately after injection, kept data shown with blue lines, discarded data shown with grey lines, and negative data marked with circles. (c) Positions of electrodes, current electrodes marked with red, potential electrodes marked with blue, other electrodes in grey. Potentials were measured in pairs of electrodes with same elevation.

Table 2 Water chemistry from water samples. Injection rounds marked with grey; nm denotes values not measured

	Date	Interval masl	pН	EC	Redox	Temp.	PCE	Sum (C6H6-C35)
Filter	yy-mm-dd	(m)	-	μS/cm	mV	°C	μg/L	μg/L
B870-1	2018-05-30	1.3 - (-0.2)	nm.	nm.	nm.	nm.	1700	4800
B870-1	2018-06-06	1.3 - (-0.2)	1.7	20,100	556	12.0	nm.	nm.
B870-1	2018-09-05	1.3 - (-0.2)	4.7	402	-58	12.0	490	830
B870-2	2018-05-30	2.8-1.3	nm.	nm.	nm.	nm.	13,000	15,000
B870-2	2018-06-06	2.8-1.3	3.1	3190	540	12.5	nm.	nm.
B870-2	2018-09-05	2.8-1.3	4.6	473	-98	13.1	26,000	23,000
B809-3	2017	2.8 - (-1.2)	nm.	nm.	nm.	nm.	23,000	14,000
B809-3	2018-06-06	2.8 - (-1.2)	3.5	7100	539	10.9	nm.	nm.
B809-3	2018-09-06	2.8-(-1.2)	4.6	405	-50	11.9	nm.	nm.

as the model retrieved from the data of the first acquisition round.

In this study, the spectral IP content was parameterized using the maximum phase angle (MPA) model from Fiandaca *et al.* (2018a), which is a re-parameterization of the Cole– Cole model (Cole and Cole, 1941). The Cole–Cole model in its complex resistivity form is given as (Pelton *et al.*, 1978):

$$\rho^{*}(\omega) = \rho_{0} \left[ 1 - m_{0} \left( 1 - \frac{1}{1 + (i\omega\tau_{\rho})^{c}} \right) \right],$$
(1)

where  $\rho^*$  is the complex resistivity,  $\omega$  is the angular frequency,  $\rho_0$  is the DC resistivity,  $m_0$  is the intrinsic chargeability,  $\tau_{\rho}$  is the time constant, *C* is the frequency exponent and i is the imaginary unit. The MPA re-parameterization is then defined by the parameters:

$$\boldsymbol{m}_{\text{MPA}} = \left\{ \rho_0, \varphi_{\text{max}}, \tau_{\varphi}, \mathbf{C} \right\} , \qquad (2)$$

where  $\varphi_{\text{max}}$  is the maximum of the Cole–Cole phase spectrum, which substitutes  $m_0$ , and  $\tau_{\varphi}$  is the inverse of the

Table 3 Soil samples from B809. bd. is below detection, and nm. is not analysed

	2017	Baseline	After second injection round			
Sample depth masl (m)	PCE mg/kg	Sum (C6H6-C35) mg/kg	PCE mg/kg	Sum (C6H6-C35) mg/kg		
2.8	15	20	860	820		
2.3	16,000	7700	510	390		
1.8	320	42	190	12		
1.3	81	1.6	bd.	0.8		
0.8	220	240	bd.	0.22		
0.3	bd.	1.7	bd.	0.043		
-0.2	bd.	0.98	bd.	0.058		
-0.7	89	90	nm.	nm.		
-1.2	bd.	1.3	nm.	nm.		
-1.7	bd.	0.18	nm.	nm.		
-2.2	bd.	0.5	nm.	nm.		

frequency at which the phase peak is reached. The IP inversions were performed with the same grid and roughness constraints as the time-lapse resistivity inversions. The MPA reparameterization was used, instead of the classic Cole–Cole model, as its parameters are less correlated, resulting in better determined inversion parameters. The Cole–Cole-like spectral model was chosen because it allows an acceptable fit to the IP decays, contrary to, for example, the constant phase angle model (Van Voorhis *et al.*, 1973; Johansson *et al.*, 2015) which gives an almost doubled data fit.

#### Permeability estimation

The permeability estimation in Fiandaca *et al.* (2018c) and Maurya *et al.* (2018a) is based on the empirical petrophysical relations that link electrical properties and permeability, as derived from laboratory data by Weller *et al.* (2015) for unconsolidated samples:

$$k = \frac{1.08 \times 10^{-13}}{F^{1.12} \cdot \sigma''^{2.27}},$$
(3a)

$$k = 3.47 \times 10^{-16} \cdot \frac{\sigma_0^{1.11}}{\sigma''^{2.41}}.$$
 (3b)

Equations (3a) and (3b) were found practically equivalent by Weller *et al.* (2015) and link the permeability, k, to the formation factor, F, and the imaginary conductivity,  $\sigma''$ , (equation (3a)) or to the DC conductivity,  $\sigma_0$ , and the imaginary conductivity,  $\sigma''$ , (equation (3b)), for samples prepared with a standardized solution of NaCl with electrical conductivity equal to 100 mS/m. In Fiandaca *et al.* (2018c) and Maurya *et al.* (2018a), equation (3a) was used for permeability estimation, because the Cole–Cole model was re-parameterized with the bulk and imaginary conductivity model, which is described by the bulk conductivity,  $\sigma_{\text{bulk}} = \sigma_0 - \sigma'_{\text{surf}}(\omega = 0)$ , the maximum imaginary conductivity,  $\sigma_{\text{max}}$ , the time constant,  $\tau_{\sigma}$ , and the frequency exponent, C. The permeability in this study is computed through equation (3b), deriving the imaginary conductivity as  $\sigma'' = \varphi_{\text{max}}/\rho_0$ .

No correction for water conductivity was carried out when applying equation (3b), because the total dependence on water conductivity of equation (3b) through  $\sigma_0$  and  $\sigma''$  almost cancel out because of the exponent present in the formula (Fiandaca *et al.*, 2018b).

#### RESULTS

## Time-lapse resistivity-only inversions

The results of the time-lapse inversions of resistivity of measurement rounds 2, 3 and 4, for the lines E02\_E05\_E08 and E03\_E05\_E07 are shown in Figures 4 and 5, respectively, as resistivity sections and resistivity ratio plots. The relative positions of the two profiles are seen in Figure 1(c) where they are marked in yellow, and they overlap at the central electrode borehole E05. The dates of the measurement rounds can be seen in Table 1 and correspond to before, immediately after and several months after injections. Figures 4 and 5 also show the used electrodes in the corresponding rounds, as black dots on the electrode tube, and the position of the injection filters relative to the electrode tubes. Figure 6 shows mean interval



Figure 4 Time-lapse inversion results for configuration E02-E05-E08, rounds 2–4 against round 1. (a–c) Resistivity sections for rounds 2–4, respectively. (d–f) Ratio plots of resistivity rounds 2–4 against round 1. Water EC converted to resistivity plotted in (b and c). Ratio of water EC between rounds 3 and 4 plotted in (e).

resistivity maps and mean interval resistivity ratio plots for a selection of intervals. These maps are created in two steps: first, by computing the geometric average of the resistivity from the inversion models in the vertical direction, every 0.5 m; second, by interpolating with cubic splines the geometric averages of all the acquisition lines of Figure 1 in the *xy* plane. These mean interval resistivity maps show no significant artefacts along the 2D inversion lines, with smooth *xy* features in both the resistivity and resistivity ratio images. This partly justifies, *a posteriori*, the use of 2D inversions in this study, because no significant resistivity contrasts are present in the *xy* plane.

Both vertical profiles show, within appropriate uncertainties, the same background resistivity, as measured in rounds 1 and 2. A highly resistive layer >1000  $\Omega$ m is seen from the surface to elevation 3.5-3.6 m, which corresponds to the unsaturated zone. Between elevations 3.5 m and 2.0 m, a discontinuous area of lower resistivity ( $50-80 \ \Omega$ m) is seen. Within this area, the soils with higher concentrations of contaminants were sampled before the first injection round (Table 3). Below this elevation, the section is composed mainly of high resistivity ( $>250 \ \Omega$ m), with smaller lenses of lower resistivity ( $80-100 \ \Omega$ m). The results from the two profiles match up at their cross-over point, even though they were measured and inverted separately, as can also be seen in Figure 6, which is derived from the 2D inversion models retrieved from the ten electrode boreholes.

In Figure 4(b), measured immediately after the injection along line E02\_E05\_E08, a large conductive (<10  $\Omega$ m) area is observed in the interval from elevation 2.0 m to -1.2 m.



Figure 5 Time-lapse inversion results for configuration E03-E05-E07, rounds 2–4 against round 1. (a–c) Resistivity sections for rounds 2–4, respectively. (d–f) Ratio plots of resistivity rounds 2–4 against round 1. Water EC converted to resistivity plotted in (b) and (c). Ratio of water EC between rounds 3 and 4 plotted in (e).

This body overprints the lenses of lower resistivity seen before, but the outline of the lower resistive body between elevation 3.5 m and 2.0 m is still visible. Figure 4(c) shows the resistivity measured 2 months after injection, where the resistivity is largely in the same range as before. However, some differences are present, e.g. around elevation 3.5 m, where it is slightly more conductive. Figure 5(a-c) shows a similar story for line E03\_E05\_E07.

Figures 4(d–f) and 5(d–f) show ratio plots of time-lapse inversion results, compared with the reference model from round 1. Figure 4(d) shows that no significant changes happened between rounds 1 and 2, which was also expected. Figure 4(e) shows that the main change to occur in round 3 is the highly conductive anomaly, between elevations 2.0 m and -1.2 m with resistivities being a factor of 5–100 times lower than the background measurement. It is notable that this anomaly in both Figures 4(e) and 5(e) is most pronounced in the elevation interval between 2.0–1.5 m and 0.0 m, with the top border being relatively sharply defined, while the bottom border is less defined. This is most pronounced in profile E03\_E05\_E07 in Figure 5(e), while profile E02\_05\_08 in Figure 4(e) exhibits more lateral variation. For use in later comparisons, magenta lines have been drawn around the extent of the conductive anomaly in Figure 5(e). The lower-contrast differences between rounds 1 and 4 are clearly visible in Figures 4(f) and 5(f) and are a combined result of seasonal variations, saturation and, possibly, of the remediation process. However, these lower-contrast differences are not the focus of this study. Overall, the time-lapse inversions using the AGMS focusing scheme retrieves sharp time-lapse



Figure 6 Mean interval resistivity maps for rounds 1 and 3, along with similar mean interval resistivity ratio maps.

resistivity contrasts, without significant smearing due to the time-lapse regularization even when high contrasts are present.

Water conductivity was measured in rounds 3 and 4 (Table 2), and the corresponding bulk resistivity values were calculated assuming a formation factor of 4, a commonly use formation factor for sandy sediments. The calculated bulk resistivity values are plotted in Figures 4(b-c) and 5(b-c), agreeing fairly well with the resistivities from the inversions, considering that the actual formation factor is unknown. Since no water conductivity measurements exist from the baseline at round 1, in Figures 4(e) and 5(e) we show water conductivity ratio between rounds 3 and 4, but with colour scale flipped, to match the resistivity ratios. Also in this case there is a good agreement between water conductivity

contrasts and resistivity ratios imaged by the time-lapse inversions. This is evidence of the time-lapse inversions depicting the spread of the injectant, due to its low conductivity values.

## Induced polarization inversion and permeability estimation

The induced polarization (IP) data collected in the first measurement round before any injections were inverted using the maximum phase angle parameterization. We present the inversion of the section E03\_E05\_E07 in Figure 7. Figure 7(a– d) shows  $\rho$ ,  $\phi_{max}$ ,  $\tau_{\phi}$  and C, in that order from top to bottom, with the outline of the conductive anomaly from Figure 5(e) marked with a magenta line. The resistivity section is very similar to the resistivity-only time-lapse inversion of round 2 with the same overall structure.  $\phi_{max}$  values are relatively high



Figure 7 IP inversion results for configuration E03-E05-E07, round 1, with conductive contour overlay from Figure 4(e). (a) Resistivity section. (b)  $\phi_{max}$  section. (c)  $\tau_{\phi}$  section. (d) C section. (e) Estimated permeability section with geology from B809.

(>10 mrad) in the unsaturated zone above 3.5 m elevation. Further below, the values decrease, and are generally within the range 0.5–1.0 mrad, with zones of lower  $\phi_{max}$  present at elevations 2.0 m and 0.0 m. Contrary to what was seen in Kemna *et al.* (2004), no increased  $\phi_{max}$  values are evident at the depths where the most contaminated soils were sampled (i.e. around elevation 2.3 m; see Table 3). This finding might make it less risky to compute the permeability from the IP parameters, under the assumption that the contamination has negligible effects on IP, despite its high values.

The  $\tau_{\phi}$  section seen in Figure 7(c) shows two anomalies (>1 s) near borehole E3 at elevations 5.0 and 3.0 m. Below the groundwater table, the values are around 0.03 s to 0.30 s. The *C* section seen in Figure 7(d) shows values of 0.6 in the majority of the zone, with one zone of lower values down to 0.3, between boreholes E03 and E05 at elevation 1.0 m. The high *C* values ensure that the resolution on the time constant  $\tau_{\phi}$  is higher (Madsen *et al.*, 2017).

The permeability estimation, derived from equation (3b) through the inversion parameters  $\rho$  and  $\phi_{max}$ , is shown in Figure 7(e), along with the position of the injection filters, the outline of the conductive anomaly from round 3, and the geological description from borehole B809. In the unsaturated zone, from the surface to around elevation 3.5 m, the estimated permeability is relatively low  $(10^{-11}-10^{-10} \text{ m}^2)$ , but the

permeability estimations have limited validity in the unsaturated zone because full saturation is assumed in equation (3b). The same level of permeability is, however, seen from below the groundwater table to around elevation 2.5 m. Below this, the permeability is higher by one to two orders of magnitude, albeit with some variation. The upper boundary between high and low permeability aligns very well with a geological transition from fine to coarse-grained sand seen in borehole B809. Consequently, the spatial distribution of the oxidizing agent as determined by the conductive anomaly may be explained, with due caution because of the high concentrations of contaminants, by the permeability field derived from IP. This would be much more difficult to identify from the resistivity sections only, where the low resistivity areas depend both on the geology and on the water conductivity. In contrast, according to Fiandaca et al. (2018b) and references therein, the variability in water conductivity should have a minor effect on the IP-derived permeability estimation.

#### DISCUSSION

The main goal of this study was to assess the usefulness of cross-borehole TDIP in tracking the spread of the injected oxidizing agent in the saturated zone. The injectant used in this case had a strong conductivity contrast in respect to the present pore fluid, and its effect was clear in the initial contact resistances and the overall resistivity measurements and inversions. The time-lapse inversion results clearly showed the range and distribution of the injectant, which was visible as a clear conductive anomaly, and the extent of the oxidizing agent was backed up by water conductivity measurements in the available filters. In the resistivity sections from round 3 in Figures 4(b, e) and 5(b, e), it is seen that the upper boundary of the conductive anomaly cuts the midpoint of the upper filter in borehole B870 and at the midpoint of the upper injection filter in injection wells I102 and I108. Looking only at water conductivity and chemistry from B870 may give an indication that the injection was a success, but the resistivity indicates that the oxidizing agent barely reached the area with highest contamination at elevation 2.3 m. To get a similar level of detail would have required a greatly increased number of monitoring wells and filters. The permeability estimates from the measurements prior to the injection corroborate the story. The area with high contamination where the oxidizing agent did not go has a lower estimated permeability than the area below, making it more preferable for the oxidizing agent to flow beneath it during injection.

When comparing water conductivity measurements with the resistivity sections in Figures 4 and 5, we assume a constant, uniform formation factor, while the actual formation factors are unknown. This obviously adds uncertainty to the water conductivity estimates, but independent estimates of formation factors would again decrease this uncertainty.

The induced polarization (IP)-derived permeability estimations show a good alignment between high- and lowpermeability areas and the geological transition from fine- to coarse-grained sands, which is a possible explanation for the spatial spreading of the injectants imaged through the timelapse inversions. Bias in the permeability estimation, especially on a quantitative level, might be present because of high levels of contamination and the empirical nature of equation (3b). The resulting permeability values are thus uncertain, but the relative variations are likely reasonable. Unfortunately, we do not have any permeability estimates from other means, such as slug test, or derived from grain size distributions in the test area. Furthermore, slug tests in B870 or B809 would yield information about the average permeability in the filters, instead of direct information about the low-permeability zone.

Resistivity sections in the non-injection rounds show high values in the unsaturated zone and lower values below the groundwater table. Stripe-like structures observed in the resistivity sections were initially thought to be inversion artefacts, but their consistency and visibility in data increase our confidence in them. These structures may stem from geology, chemistry or a combination of both. The low resistivity around elevation 2.0–3.0 m is coinciding with a layer of fine-grained sands and high contamination levels. The low resistivity levels could be the effect of degradation products of PCE, trapped within the fine-grained sediments.

Prior to this study, it was not known how much the contamination would affect the resistivity in the area. Christensen and Halkjær (2014) indicate that the general resistivity of the area was unaffected above an elevation of -10 m, which is nearly 7 m below the targeted volume of this survey. This is not in contradiction to our findings, as the majority of the volume appears to have high, undisturbed resistivities, while some localized areas may have lower resistivity, possibly due to contamination.

The extent and range of the injected oxidizing agent was tracked using the resistivity data alone in time lapse, but the permeability estimation obtained before the remediation can be used to predict the injectant spread pattern and to tailor the injection accordingly. However, during the injection rounds, the IP signal was severely affected by the conductive oxidizing agent, and signal levels fell below the noise level for a majority of decays, so only IP data from the non-injectant rounds was worth considering. For applications as the one presented in this study, and similar hardware/current values, IP collection in injections rounds may be cancelled for the benefit of faster acquisition time (and less frustration in the data processing). It has the added benefit that all measurements may be closer in time and closer to the injection event.

During the measurements in May and June (rounds 2 and 3), about 30% fewer quadrupoles were measured, as the contact resistances were too high. No steps were taken, however, to resolve this issue in this study, as prior experience detailed in Bording, *et al.* (2019) showed that adding saltwater, while decreasing the contact resistance, can increase borehole effects. As the main area of interest was in the saturated zone, where electrode contact resistances were low, this reduction in data was accepted. During fall and winter, precipitation increased resulting in lower contact resistances for the remaining rounds.

In this study, we have neglected effects arising from temperature variations between measurement rounds. The limited temperature information shows little variation  $(1-2^{\circ}C;$ Table 2), so such effects are expected to be small.

In the recent past, the transition towards 3D inversions has been clear, but we have chosen to use 2D inversions in this study. This allowed us, within the same inversion code framework, to perform focused time-lapse inversions of resistivity and the spectral IP inversions, used for permeability estimation. The choice of 2D inversion comes with the problem of dealing with potential 3D effects. Some of such effects may be local conductive plumes in the *xy* plane with underestimated conductivity, and/or resistivity features outside the profile projected into the profile (Nimmer *et al.*, 2008). Borehole effects resulting in resistivity contrast directly around the boreholes are expected to be limited as electrodes were installed using direct push, with a subsequent collapse of the sandy formation around the borehole electrodes. In the unsaturated zone, this collapse is however not guaranteed. Acknowledging the limitations of 2D inversions, but also the benefits of the time-lapse focusing scheme and of the full-decay IP inversion used in this study, work on a 3D code is currently in progress within our group (Madsen *et al.*, 2019).

# CONCLUSION

Resistivity and full waveform TDIP measurements were carried out in a cross-borehole set-up at an active remediation site over the course of a year in order to test their capability in tracking the spread of the injected oxidizing agent within the target volume. The collected resistivity data were inverted with time-lapse focusing, while the induced polarization (IP) data collected prior to injection were inverted in terms of the maximum phase angle Cole-Cole re-parameterization, and subsequently used to estimate permeability through petrophysical relations derived in the laboratory for unconsolidated samples. The abovementioned set-up, processing and inversion methods allowed for a spatial coverage of the injection material not achievable by means of monitoring wells, and revealed the injected oxidizing agent did spread in the majority of the saturated zone, but that local areas were not reached by the injectant. This was unclear from the monitoring wells, but was identified easily and clearly in the resistivity sections from the time-lapse inversions, which clearly visualized where resistivity changes had occurred, and thus the extent of injected oxidizing agent. A comparison between permeability estimations from the IP inversions and the spread of the oxidizing agent furthermore showed that the oxidizing agent did not spread in the predicted zones of low permeability indicating that the first injection round may have had limited effect in terms of remediation.

As a conclusion, we find that cross-borehole resistivity and TDIP measurements are a powerful tool for mapping remediation material in the saturated zone when used in conjunction with other direct investigation methods, with significant increase in area coverage as compared with the traditional method of monitoring wells. Although permeability estimates were not used to plan the injection strategy in this study, the application of prior knowledge of the permeability field in the planning phase may lead to more efficient and successful remediations.

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# DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

# ORCID

Thue Bording D https://orcid.org/0000-0003-1932-9466 Anders Kristian Kühl D https://orcid.org/0000-0002-7512-4070 Gianluca Fiandaca D https://orcid.org/0000-0002-3395-878X Anders Vest Christiansen D https://orcid.org/0000-0001-5829-2913 Esben Auken D https://orcid.org/0000-0002-5397-4832

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