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Investigation of chlorinated solvent pollution with resistivity and induced polarization

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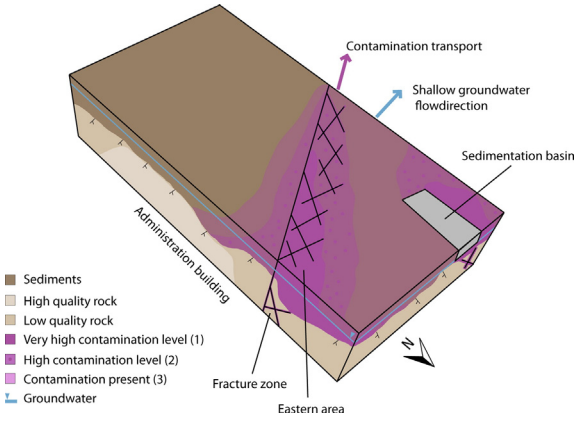
HIGHLIGHTS

- Chlorinated solvent pollution has been mapped with geoelectrical (DCIP) methods.
- Geoelectrical (DCIP) measurements are used to map hydrogeological settings.
- Time domain induced polarization show potential for *in situ* remediation monitoring.

GRAPHICAL ABSTRACT



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ABSTRACT

Globally, an enormous number of polluted areas are in need of remediation to prevent adverse effects on health and environment. *In situ* remediation and especially the monitoring thereof needs further development to avoid costly and hazardous shipments associated with excavation. The monitoring of *in situ* remediation actions needs easier and cheaper nondestructive methods for evaluation and verification of remediation degree and degradation status of the contaminants. We investigate the Direct Current resistivity and time-domain Induced Polarization tomography (DCIP) method and its use within the context of a DNAPL (Dense Non-Aqueous Phase Liquids) contaminated site in Varberg, Sweden, where an *in situ* remediation pilot test has been performed by stimulated reductive dechlorination by push injection. Our results show that the DCIP technique is an emerging and promising technique for mapping of underground structures and possibly biogeochemical spatial and temporal changes. The methodology could in combination with drilling, sampling and other complementary methods give an almost continuous image of the underground structures and delineation of the pollutant situation. It can be expected to have a future in monitoring approaches measuring time lapse induced polarization (IP), if more research is performed on the parameters and processes affecting the IP-signals verifying the interpretations. The IP technique can possibly be used for verification of the effectiveness of *in situ* remediation actions, as the current sampling methodology is inadequate.

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1. Introduction

In Sweden, >80,000 sites are assumed as or have been identified as contaminated in the national ongoing environmental risk assessment (SEPA, 2014). Presently in Sweden, large numbers of polluted areas are remediated by excavation. The contaminated masses are deposited at landfills, or in best case off-site treatment efforts are applied. *In situ* remediation and especially the monitoring thereof needs further development to avoid the costly and somewhat hazardous shipments associated with excavation. Therefore the recommendation issued by SEPA (2014) is to increase the use of alternative methods *versus* the common "dig and treat" approach. The monitoring of *in situ* remediation actions needs easier and cheaper nondestructive methods for evaluation of remediation degree and degradation status of the contaminants.

Within the TRansparent Underground Structures research project (TRUST), we investigate the Direct Current resistivity and time-domain Induced Polarization (DCIP) tomography method and its use within the context of DNAPL contaminated sites (see Johansson et al., 2015). DCIP is a non-invasive and non-destructive geoelectric measurement method that among other things has high potential for providing indirect evidence of contaminant degradation status. It is an emerging and promising technique for 2D, 3D and 4D mapping of underground hydrogeochemical structures. Examples of application areas are landfill characterization (*e.g.* Gazoty et al., 2012a, 2012b; Leroux et al., 2007; Cardarelli and Bernabini, 1997), spatial and temporal distribution of contaminants such as ions pollutants leaching from landfill sites (*e.g.* Dahlin et al., 2010; Acworth and Jorstad, 2006; Chambers et al., 2006; Dahlin et al., 2002) and characterization of DNAPL polluted sites (*e.g.* Cardarelli and Di Filippo, 2009; Deceuster and Kaufmann, 2012; Orozco et al., 2012; Power et al., 2014). Furthermore, gas migration within landfills (e.g Rosqvist et al., 2011), and underground CO_2 migration (Auken et al., 2014) including chemical changes resulting therefrom (Doetsch et al., 2015) have been monitored with DCIP.

In this study we have investigated a highly polluted old industrial (textile and mechanical industry) area, contaminated with trichloroethene (TCE) and 1,1,1-trichloroethane (TCA), both DNAPLs, as well as its degradation products and cyanide, chromium, zinc and cadmium (Tornberg et al., 2008). At the investigated site, a small pilot test has been carried out for remediation of the DNAPLs by stimulated reductive dechlorination. DNAPLs are specifically problematic pollutants to delineate with conventional methods due to their dense properties and ability to move independent of the groundwater flow direction, their harmfulness and, risks for unforeseen spreading when drilling and pumping. The fact that chlorinated solvents have been used in almost every little village's dry cleaning facilities means that the pollutant situation is far from unique, and it is highly important to develop better investigation tools, remediation research and cheap non-invasive monitoring techniques.

The aim of this study was to investigate possible uses, benefits and limitations of the DCIP technique divided into the following research questions;

1. Understanding of the underground system;
   Can we improve the understanding of the hydrogeological system (water conditions, pollutant situation, soil conditions, bedrock quality and tectonic structures) by DCIP measurement studies?
2. Understanding of the pollutant situation;
   Can we delineate chlorinated solvents source areas, *i.e.* does DCIP signals correlate to free phase pollutant concentrations measured in groundwater samples?
   Can we delineate pollution plumes associated with TCE pollutants, *i.e.* does DCIP signals correlate to concentrations of TCE and its metabolites in groundwater samples?
   Can we identify an area where *in-situ* remediation test is carried out by carbon source injection, *i.e.* does DCIP signals correlate to the area?
3. Time lapse measurements; Is it possible to monitor changes within a stimulated *in situ* remediation test area, in time steps, and thereby visualize degradation and bioactivity through time, *i.e.* can changes in the area be seen in repeated DCIP measurements and verified by enhanced biological activity?

2. Background and study area

The study area located in Varberg, southwest Sweden, has a history of polluting activities starting in the late 19th century with textile manufacturing using Cr, Zn and Cd for >60 years. The business then turned into precision mechanics with surface finishing of metals and use of zinc, chromium, cyanide and chlorinated solvents (TCE and TCA). The chlorinated solvents are the most problematic contaminants at the site with levels in groundwater of >20 mg/L of TCE and/or its metabolites. Except for handling of chemicals within the manufacturing processes, the pollutants have been spread *via* a leaky wastewater drain pipe, into a concrete sedimentation basin used for treatment of wastewater, and is now refilled with rubble, concrete and soil (Tornberg et al., 2008; Larsson and Hübinette, 2003).

A new railroad tunnel beneath the city is planned in the groundwater flow direction from the site, and the pollution problem has become critical to deal with, as lowering of the groundwater table during the construction will increase the groundwater flow gradient and faster transport of pollutants will occur.

The site outline is presented within Fig. 1 with a sketch of the general groundwater flow towards the sea in W and SW, extent of known bedrock fratures, the contaminant plume delineated from traditional investigations such as drillings, probe soundings and groundwater wells sampled, all performed before our measurements started in 2014 and forming the background baseline for our measurements. Geological profiles are shown in Fig. 2.

Varberg is located below highest shoreline with dominating sediments of postglacial sands and wave-washed gravel (Påsse, 1990). At the site the sandy sediments are overlain by fill, consisting of soil, brick and concrete pieces. The thickness of sediments at the site varies between 2.7 and 5.3 m (Magnusson and Samuelsson, 2004, see Fig. 2). The sediment thickness increases westwards from the site towards the harbour with up to *c.* 18 m of quaternary sediments, and fine grained sediments such as clays interfinger the sandy sediments and divides the gravelly aquifer in two parts (Hübinette and Bank, 2011). Underneath the sediments, dominating bedrock is granodiorite, patches of gneissic banded granite and charnockite. Exposed bedrock north of the site shows five dominating orientations of fractures and lineaments 1) NW, 2) NNW, 3) N, 4) NE and 5) E (SGU, 2006). The fracture system creates a zigzag transport pattern within the bedrock and allows for downwards movement of the DNAPL pollutants. Both ground and bedrock surfaces are dipping westwards (Fig. 2b) with a low point in the bedrock in the central part of the site (Fig. 2a) (Tornberg et al., 2008).

The fill, sandy sediments and fractured bedrock together form a heterogeneous unconfined aquifer. These highly permeable sediments and the bedrock fracture system provide fast transport pathways. Estimates of hydraulic conductivity by test pumping and slug tests show values in the order of 10^{-5}–10^{-8} m/s (Florén, 2015; Davidsson, 2013) Precipitation in Varberg is about 870 mm/year (SMHI, 2013) and evapotranspiration is estimated to >550 mm (Karlqvist et al., 1985). Runoff is calculated to be 300 mm (Karlqvist et al., 1985), which estimates the net infiltration to the aquifer to <20 mm per year.

In January 2013 a pilot study for *in situ* remediation by injection of electron donor and carbon source, HRC Primer® (glycerol tripolylactate, glycerine and lactic acid) and 3DMe® (water, neutralized fatty acids, glycerol tripolylactate and Hydrogen Release Compound Partitioning Electron Donor (Regenesis, US Patent 7667062 B2)), was performed to stimulate already existing bacteria in the subsurface to perform reductive dechlorination by anaerobic degradation. The injected stimulus is described as a long-term controlled release of lactic, organic and fatty

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