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Hydrogeophysical imaging of deposit heterogeneity and groundwater chemistry changes during DNAPL source zone bioremediation

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ABSTRACT

Robust characterization and monitoring of dense nonaqueous phase liquid (DNAPL) source zones is essential for designing effective remediation strategies, and for assessing the efficacy of treatment. In this study high-resolution cross-hole electrical resistivity tomography (ERT) was evaluated as a means of monitoring a field-scale in-situ bioremediation experiment, in which emulsified vegetable oil (EVO) electron donor was injected into a trichloroethene source zone. Baseline ERT scans delineated the geometry of the interface between the contaminated alluvial aquifer and the underlying mudstone bedrock, and also the extent of drilling-induced physical heterogeneity. Time-lapse ERT images revealed major preferential flow pathways in the source and plume zones, which were corroborated by multiple lines of evidence, including geochemical monitoring and hydraulic testing using high density multilevel sampler arrays within the geophysical imaging planes. These pathways were shown to control the spatial distribution of the injected EVO, and a bicarbonate buffer introduced into the cell for pH control. Resistivity signatures were observed within the preferential flow pathways that were consistent with elevated chloride levels, providing tentative evidence from ERT of the biodegradation of chlorinated solvents.

1. Introduction

Chlorinated dense nonaqueous phase liquids (DNAPLs), such as trichloroethene (TCE), are amongst the most problematic manmade sources of groundwater contamination (Pankow and Cherry, 1996). They have been widely used by a range of industries over many decades, and through spills, leaks, uncontrolled releases and disposal now contaminate the subsurface in many industrialized areas (Pankow et al., 1996; Tait et al., 2004; Rivett and Clark, 2007). Due to their density DNAPLs can migrate through the water table and spread under the influence of gravity until residual levels are attained and capillary trapping prevents further movement, or an impermeable layer is reached, at which point lateral spread or pooling can occur (Schwille, 1988; Parker et al., 2003). Many chlorinated solvents are characterized by low solubilities and a resistance to biodegradation and natural attenuation (Pankow et al., 1996), and can therefore remain in the ground for many decades. Their solubility, although low, is sufficient to exceed regulatory limits (Ajo-Franklin et al., 2006), and so remediation of contaminated sites is therefore generally required. Where DNAPL is present below the water table or at significant depths in-situ remediation strategies, including biostimulation, are often the only viable option (Kueper et al., 2003; ITRC, 2007). Biostimulation often involves the introduction of an electron donor, such as acetate or emulsified vegetable oil (EVO), to stimulate the activity of microbes involved in reductive dechlorination, and to generate the low redox potentials

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required for dehalorespiration. The ultimate goal of bioremediation is the complete transformation of chlorinated solvents to a nontoxic daughter product such as ethene.

Robust characterization and monitoring of DNAPL source zones is essential for remedial design, optimization and performance assessment (Kavanaugh et al., 2003; Brusseau et al., 2007). Where significant subsurface heterogeneity exists, conventional intrusive investigations and groundwater sampling can be insufficient, as the information they provide is restricted to vertical profiles at discrete locations, with no information between sample points. Therefore significant uncertainty can remain, in relation to the lithological variability, and in the distribution of DNAPL, electron donor or other amendment fluids. In order to mitigate this problem complementary geophysical ground investigation methods are now emerging (US EPA, 2004), as they have the advantage of producing spatial or volumetric information on subsurface variability, and can be sensitive to changes caused by the injection of amendment fluids (Lane et al., 2004; Hubbard et al., 2008; Williams et al., 2009). Examples of field scale geophysical monitoring of DNAPL bioremediation experiments are, however, rare; previous studies are described by Daily and Ramirez (1995), who used cross-hole electrical resistivity tomography (ERT), with a spatial resolution of a few m^2, to monitor methane electron donor injection at a TCE contaminated site, and Lane et al. (2006) who applied cross-hole radar methods for monitoring spatial and temporal distribution of EVO at a TCE and dichloroethene (DCE) contaminated site.

In this study cross-hole ERT was used as a means of imaging the subsurface during a pilot-scale experiment to monitor the bioremediation of a TCE source zone. The geophysical study formed a component of a wider experiment, which was designed to test the hypothesis that enhanced anaerobic bioremediation by reductive dechlorination can result in the effective treatment of chlorinated solvent DNAPL source areas (Zeeb et al., 2008). The specific objectives of the geophysical imaging described in this paper were to assess the efficacy of cross-hole ERT as a means of characterizing geological and hydrogeological heterogeneity, and monitoring changes in groundwater chemistry associated with the injection of EVO electron donor and bicarbonate buffer used for pH control, and chloride released through the biodegradation of chlorinated solvents. Here a novel experimental design was employed, involving ERT arrays and multilevel groundwater sampling (MLS) arrays installed in closely spaced boreholes, which formed monitoring transects in both the source and plume zones. These arrays were designed to complement the detailed geochemical point sampling by providing geophysical imaging at a resolution (i.e. dm^2) approaching that of the geological heterogeneity indicated during drilling.

2. Electrical resistivity tomography (ERT)

ERT is a geophysical imaging technique that is used to generate 2D and 3D models, or images, of the resistivity distribution in the subsurface. Data collection and processing methodologies are widely described in the literature (e.g. Slater et al., 2002; Bentley and Gharibi, 2004; Cassiani et al., 2006), and so only a brief description is provided here. ERT surveys involve making a large number of four-point direct current (DC) electrical measurements (consisting of pairs of current and potential electrodes) using computer controlled automated measurement systems and multi-electrode arrays. These data are inverted to produce images of the subsurface; this is typically achieved by using regularized nonlinear least-squares algorithms (e.g. Loke and Barker, 1996) in which the forward problem is solved using either finite element or finite difference methods. ERT electrodes can be deployed either as surface or borehole arrays, or as a combination of the two. Cross-hole imaging was selected for this study to ensure that ERT image resolution was maintained with depth. The superior depth resolution that can be achieved using cross-hole relative to surface imaging is particularly important when characterizing and monitoring complex ground conditions and processes, where information is required at the scale of the heterogeneities. Cross-hole imaging, unlike surface array imaging, can potentially resolve layers that are in the order of tens of centimetres thick at depths of more than 10 m (e.g. Kemna et al., 2004).

2.1. Application to hydrogeophysical investigations

The use of ERT as a ground imaging technique is based on the petrophysical relationships linking resistivity, hydrogeological and geological parameters (e.g. Revil et al., 1998; Lesmes and Friedman, 2005). The degree of fracturing, porosity, tortuosity, mineralogy, saturation, temperature and groundwater resistivity all affect the resistivity of subsurface materials, thereby providing the basis for using ERT for geological and hydrogeological investigations. The use of ERT for characterizing subsurface geology is well documented, with many examples of investigating unconsolidated saturated sediments (e.g. Froese et al., 2005; Kilner et al., 2005), such as those found at the research site detailed in this study. Generally, the major lithological effect on resistivity in these types of sediments is the proportion and type of clay minerals (Shevnin et al., 2007), with increasing clay content causing a decrease in resistivity. The close link between resistivity and many important hydrogeological parameters and properties has led to the increased use of ERT for hydrogeophysical investigations, where it has been used to study groundwater quality (Ogilvy et al., 2009), moisture content (Zhou et al., 2001) and in-situ remediation (Daily and Ramirez, 1995). Of particular significance for hydrogeophysical investigations are the Archie equations (Archie, 1942) that link resistivity with pore fluid conductivity, saturation and porosity. When used in time-lapse mode ERT can provide spatial or volumetric information on changes in the subsurface, which, assuming a fixed geology, are usually related to changes in saturation (both water and non-wetting phase NAPLs), temperature, and the composition of the pore fluid. In some cases, quantitative estimates can be made of seepage velocities (Sandberg et al., 2002; Wilkinson et al., 2009), spatial moments (Binley et al., 2002; Singha and Gorelick, 2005; Looms et al., 2008), hydraulic conductivity (Binley et al., 2002), and tracer mass and concentration (Singha and Gorelick, 2006; Deiana et al., 2007; Oldenborger et al., 2007; Deiana et al., 2008).

2.2. DNAPL contamination and bioremediation

Examples of the use of ERT to detect and monitor DNAPL in the subsurface are relatively sparse and can be divided into