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Implications of soil mixing for NAPL source zone remediation: Column studies and modeling of field-scale systems

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ABSTRACT

Soil remediation is often inhibited by subsurface heterogeneity, which constrains contaminant/reagent contact. Use of soil mixing techniques for reagent delivery provides a means to overcome contaminant/reagent contact limitations. Furthermore, soil mixing reduces the permeability of treated soils, thus extending the time for reactions to proceed. This paper describes research conducted to evaluate implications of soil mixing on remediation of non-aqueous phase liquid (NAPL) source zones. The research consisted of column studies and subsequent modeling of field-scale systems. For column studies, clean influent water was flushed through columns containing homogenized soils, granular zero valent iron (ZVI), and trichloroethene (TCE) NAPL. Within the columns, NAPL depletion occurred due to dissolution, followed by either column-effluent discharge or ZVI-mediated degradation. Complete removal of TCE NAPL from the columns occurred in 6–8 pore volumes of flow. However, most of the TCE (>96%) was discharged in the column effluent; less than 4% of TCE was degraded. The low fraction of TCE degraded is attributed to the short hydraulic residence time (<4 days) in the columns. Subsequently, modeling was conducted to scale up column results. By scaling up to field-relevant system sizes (>10 m) and reducing permeability by one-or-more orders of magnitude, the residence time could be greatly extended, potentially for periods of years to decades. Model output indicates that the fraction of TCE degraded can be increased to >99.9%, given typical post-mixing soil permeability values. These results suggest that remediation performance can be greatly enhanced by combining contaminant degradation with an extended residence time.

1. Introduction

Modern contaminant hydrologists have recognized the need to address contaminants in multiple phases, in heterogeneous soil zones with highly-contrasting permeability (Brown et al., 2012; Chapman and Parker, 2005; EPA, 2003; Maji and Sudicky, 2008; Pankow and Cherry, 1996; Payne et al., 2008; Sale and Newell, 2011; Seyedabbasi et al., 2012; Suchomel et al., 2014). Current remediation technologies, many of which are based on fluid extraction and/or chemical reagent injection, may result in incomplete mass removal or limited contact between reagents and contaminants. Ultimately, such technologies have little impact on contaminants residing in low-permeability soil zones (Chapman et al., 2012; McCarty and Criddle, 2012; Neupauer et al., 2014; Payne et al., 2008; Stroo et al., 2012). An additional complication occurs at sites containing dense non-aqueous phase liquids (DNAPLs). Although the presence of DNAPLs is often inferred from groundwater concentration data, locating and remediating sparse DNAPL pools and ganglia has proven to be an enormous challenge (EPA, 2003; Pankow and Cherry, 1996; Stroo et al., 2012). In practice, currently available remediation technologies are often limited in their ability to reduce source-zone concentrations to the maximum contaminant levels (MCLs) established by the U.S. EPA (Stroo et al., 2012).

The limitations of currently available treatment technologies can be potentially overcome by using mechanical soil

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mixing techniques (Day and Ryan, 1995) to admix contaminated soils with treatment reagents including chemical oxidants (Siegrist et al., 2011) or reductants (Fjordbøge et al., 2012b; Olson et al., 2012; Wadley et al., 2005). Soil mixing has the potential to effectively treat contaminant mass in all phases, including contaminants residing in low-permeability zones. In addition, soil mixing reduces the permeability of the treated-soil zone, by blending soil strata comprising variable particle sizes and through the addition of clay as a stabilizing agent. The reduced permeability provides treatment benefits including a diminished contaminant discharge and an extended residence time (i.e., the average transit time for a particle to pass through the system). Within a system that has been modified by soil mixing, the paradigm used to describe subsurface processes is altered (as illustrated in Fig. 1). For the modified system, key attributes to be considered include (1) implications of homogenization on NAPL dissolution, (2) reactivity of granular, micro-scale (<1 mm) ZVI particles in the presence of NAPL, and (3) interaction between permeability reduction and contaminant degradation. For each of these attributes, the current state of knowledge is discussed in the following paragraphs.

The first attribute involves NAPL dissolution in porous media, a concept that has been widely studied (e.g., Brusseau et al., 2002; Farthing et al., 2012; Geller and Hunt, 1993; Imhoff et al., 1994; Johnson et al., 2003; Maji and Sudicky, 2008; Miller et al., 1990; Powers et al., 1991, 1998; Saba and Illangasekare, 2000; Sale and McWhorter, 2001). As an experimental simplification of natural systems, much of this research has involved homogeneous laboratory-scale systems. These experiments have demonstrated that water can rapidly equilibrate with NAPL in homogeneous systems (Imhoff et al., 1994; Miller et al., 1990), but that the equilibrium assumption is generally not applicable in non-uniform or field-scale systems (Brusseau et al., 2002; Gerhard et al., 2014; Maji and Sudicky, 2008; Parker and Park, 2004). The lack of NAPL–water equilibrium observed in field systems is attributed to typically-sparse distributions of NAPL and heterogeneous groundwater flow fields. However, implementation of soil mixing modifies the site conceptual model by homogenizing both soils and contaminants (Fig. 1). Although previous laboratory NAPL-dissolution studies have been conducted in homogeneous systems, implications of homogenization on NAPL dissolution have not been widely evaluated in the context of mixed-soil systems. Specific aspects unique to the mixed-soil system include field-scale homogenization and uniformly-distributed reactive media in a soil/NAPL system.

The second attribute involves the interaction between granular-scale ZVI and chlorinated solvent NAPL. ZVI-mediated degradation of chlorinated solvents has been the subject of extensive research (e.g., Arnold and Roberts, 2000; Devlin and Allin, 2005; Gillham and O'Hannesin, 1994; Grant and Kueper, 2004; Jeen et al., 2011; Klausen et al., 2003; Kouznetsova et al., 2007; Li and Benson, 2010; Liu and Lowry, 2006; Matheson and Tratnyek, 1994; Phillips et al., 2010; Reardon, 2005; Scherer et al., 1998; Taghavy et al., 2010; Zhang and Gillham, 2005). Most of the research has been conducted in the context of permeable reactive barriers, which are designed for treatment of dissolved-phase chlorinated solvents. A comparatively small body of literature addresses ZVI for treatment of NAPL-phase chlorinated compounds (Fagerlund et al., 2012; Fjordbøge et al., 2012b; Olson et al., 2012; Quinn et al., 2005; Taghavy et al., 2010; Wadley et al., 2005). Previous research into ZVI/NAPL systems has primarily been conducted in the context of injecting nm-sized ZVI particles (nZVI) into DNAPL source zones (Fagerlund et al., 2012; Quinn et al., 2005; Taghavy et al., 2010). In laboratory experiments conducted by Taghavy et al. (2010) and Fagerlund et al. (2012), effective nZVI-mediated degradation of PCE was observed, but results indicated that treatment effectiveness was contact-limited; both of these

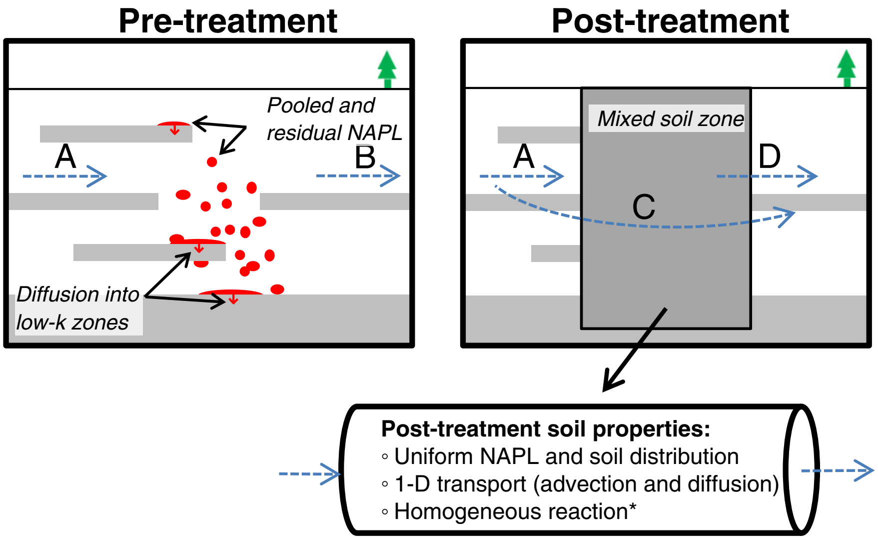

Fig. 1. Conceptual illustration of a heterogeneous NAPL source zone before and after soil-mixing remediation. Groundwater flow patterns are indicated by blue dashed arrows: prior to treatment, clean groundwater contacts the upgradient face of the source zone (A) and contaminated groundwater exits the source zone (B); after treatment, groundwater selectively bypasses the low-k treated soil zone (C) and groundwater discharge from the source zone (D) is greatly reduced. Properties of the mixed soil zone (in the case of idealized mixing) are shown. (*) The reaction is described as homogeneous, because the reactant is distributed throughout the treated soil zone, even though the reaction may be surface-mediated (as in the case of ZVI).