



Research article

Application of an in-situ soil sampler for assessing subsurface biogeochemical dynamics in a diesel-contaminated coastal site during soil flushing operations

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ABSTRACT

Subsurface biogeochemistry and contaminant dynamics during the remediation of diesel-contamination by in-situ soil flushing were investigated at a site located in a coastal region. An in-situ sampler containing diesel-contaminated soils separated into two size fractions (<0.063- and <2-mm) was utilized in two monitoring wells: DH1 (located close to the injection and extraction wells for in-situ soil flushing) and DH2 (located beyond sheet piles placed to block the transport of leaked diesel). Total petroleum hydrocarbon (TPH) concentrations and biogeochemical properties were monitored both in soil and groundwater for six months. A shift occurred in the groundwater type from Ca-HCO₃ to Na-Cl due to seawater intrusion during intense pumping, while the concentrations of Ni, Cu, Co, V, Cr, and Se increased substantially following surfactant (TWEEN 80) injection. The in-situ sampler with fine particles was more sensitive to variations in conditions during the remedial soil flushing process. In both wells, soil TPH concentrations in the <0.063-mm fraction were much higher than those in the <2-mm fraction. Increases in soil TPH in DH1 were consistent with the expected outcomes following well pumping and surfactant injection used to enhance TPH extraction. However, the number of diesel-degrading microorganisms decreased after surfactant injection. 16S-rRNA gene-based analysis also showed that the community composition and diversity depended on both particle size and diesel contamination. The multidisciplinary approach to the contaminated site assessments showed that soil flushing with surfactant enhanced diesel biodegradation, but negatively impacted in-situ diesel biodegradation as well as groundwater quality. The results also suggest that the in-situ sampler can be an effective monitoring tool for subsurface biogeochemistry as well as contaminant dynamics.

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1. Introduction

Global increases in the use of liquid fuel products has led to increasing contamination of soils and groundwater by leaking underground storage tanks and pipelines. Of particular concern are total petroleum hydrocarbons (TPH), which are composed of various compounds including alkanes, aromatic hydrocarbons,

methyl *t*-butyl ether (MTBE), ethanol, butanol, and inorganic compounds (e.g., sulfur or nitrogen). All of these compounds are harmful or even toxic to microbes, plants, and animals, and can be a source of long-term soil and groundwater contamination (Hentati et al., 2013; Plaza et al., 2005). Thus, the remediation of TPH-contaminated subsurface environments is an area of active research. The regulatory guidelines governing TPH in soil and the remedial goal for TPH in groundwater near industrial and commercial sites in South Korea are 2000 mg kg⁻¹ and 1.5 mg L⁻¹, respectively (Ministry of Environment, 2015).

Among the many strategies and techniques used in the remediation of TPH-contaminated soils and groundwater, in-situ soil

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flushing (washing) is considered to be an effective technology as it is comparatively simple and can lead to relatively fast treatment (Rao et al., 1997). Typical applications of in-situ soil flushing involve the placement of sheet piles and trenches to block further transport of leaked fuels, followed by the leaching of fuel contaminants by injecting water into the contaminated zone combined with down field groundwater extraction. Extraction efficiency can be enhanced by the addition of surfactants or alcohols such as ethanol (Long et al., 2013; Mulligan et al., 2001; Rao et al., 1997). The extracted groundwater containing the leached fuel contaminants can be further treated by ex-situ physical and chemical separation or other treatments.

During in-situ soil flushing, groundwater quality can be easily monitored by analyzing the extracted groundwater. However, it is much more challenging to monitor in-situ soil decontamination due to the difficulty in obtaining representative soil samples from subsurface environments. Soil samples can be collected using a soil core drilling machine; however, this approach is expensive and does not provide truly representative samples, as soil cores cannot be collected repeatedly from precisely the same location. In-situ soil samplers provide an alternative approach for monitoring biogeochemical dynamics in subsurface environments during in-situ contaminant remediation; these samplers are easily deployable in monitoring wells and can be tailored for the investigation of specific in-situ processes. For example, in-situ sediment samplers have been shown to provide important information on the differences between the microbial communities attached to sediment and those suspended in groundwater (Alfreider et al., 1997; Flynn et al., 2008).

Organic contaminants such as petroleum hydrocarbons can strongly bind to the soil matrix, especially fine particles. Thus, the distribution of soil particle sizes in subsurface environments can strongly affect the transport of TPH. For example, the efficiency of oil removal by soil flushing is much greater in sand than in clay loam (Abdel-Moghny et al., 2012). The performance of in-situ soil flushing is limited by the presence of low-permeability zones, and not very effective in low permeability soils with high clay content (US EPA, 2006). In addition, during the transport of non-aqueous phase liquids (NAPLs) such as petroleum-based fuels through subsurface soil and sediment, high levels of hydrocarbons can be retained in soil macro- and micro-pores in the form of residual saturation, which can act as a long-term source of groundwater contamination (Feng and Aldrich, 2000; Kwon et al., 2016a). Although much is known about the effects of soil flushing processes on TPH release from contaminated soil and sediment in general, the effects of many fundamental biogeochemical processes relevant to changes in subsurface environments have yet to be elucidated, especially in the context of groundwater quality and microbial community dynamics. Moreover, few studies have conducted intensive coincident monitoring of geochemical and microbial community dynamics.

In this study, we investigated the geochemical and microbial dynamics during the remediation of subsurface diesel contamination using an in-situ soil sampler. The specific goals of this study are: (1) understand how soil flushing changes subsurface biogeochemical processes; and (2) determine if an in-situ sampler can effectively monitor and reflect subsurface biogeochemical processes.

2. Material and methods

2.1. In-situ soil flushing at a diesel-contaminated coastal site

The study area is located in the eastern part of South Korea in a coastal region (Fig. 1) that has been contaminated over a 10-year

period by a leaking diesel fuel tank. The total amount of leaked diesel is estimated to be about 40,000,000 kg and the contaminated area is approximately 31,000 m². Local offices and storage buildings occupied over 42% of the contaminated area. The subsurface material at this site is primarily composed of sand (>95%), with minor amounts of silt and clay (<1%).

To remediate the contaminated sediments in this area, a regional engineering consortium applied an in-situ soil-washing technique. In early 2014, the consortium constructed a total of 162 injection and extraction wells for full-scale remedial action. Well pumping was initiated on July 6, 2014 (day 20) and continued to December 19, 2014 (day 170). The contaminated groundwater was collected from the surrounding extraction wells at a rate of 1000 ton d⁻¹ and separated into free phase NAPL and groundwater. After treatment for the soluble organic contaminants, the treated water was diluted by fresh water and re-injected into the injection wells. On August 25, 2014 (day 60), approximately one volume of surfactant (i.e., 0.1–0.2% TWEEN 80) and two volumes of groundwater were mixed and then injected, and the injection continued for 2.5 months. The total injected volume of groundwater and surfactant in this period was 32,307 m³ and 14,156 m³, respectively.

To test the feasibility of subsurface biogeochemical monitoring, we selected two monitoring wells, identified as DH1 and DH2. Wells DH1 and DH2 were approximately 10 m apart. DH1 was close to the injection and extraction wells for in-situ soil flushing, and thus was expected to be directly influenced by the transport of leaked diesel; DH2 was located beyond the sheet piles, which blocked the transport of leaked diesel (Fig. 1).

2.2. Preparation of in-situ soil samplers containing diesel-contaminated sediments

To collect representative subsurface sediment samples during in-situ flushing, we used an in-situ sampler containing diesel-contaminated sediments. In addition, to examine how sediment particle size affects the distribution of TPH and the dynamics of soil microbial communities, we prepared two in-situ soil samplers containing sediments separated into two size fractions [500 g each of <0.063-mm fine fraction (= 100% silt and clay) and <2-mm bulk fraction (= 99.4% sand and 0.6% silt and clay)] (Fig. 1). The diesel-contaminated subsurface sediments were obtained from the core material (approximately 40 kg) retrieved at the depths of 3–5 m in the vicinity of the underground diesel fuel tank (of monitoring wells DH1 and DH2) on May 28, 2014. The sediment was stored at 4 °C until further treatment. The size distribution of the sediments was analyzed according to ASTM Standard D402. Size classification was achieved by sieving into <2-mm (consisting of the coarse, medium, fine, and very fine sand, silt, and clay fractions) and <0.063-mm (silt plus clay fractions) sizes. The contaminated sediments were placed in a fine stainless wire cloth with 0.02-mm openings (635 mesh T316 Stainless, TWP INC, Berkeley, USA) and submerged in the wells at a depth of 4–5 m below the surface (mbsf) on June 12, 2014. Our aim was to design an in-situ sampler that allowed dissolved ions, TPHs, and bacteria to pass through, but retained fine particles (e.g., <0.063 mm). The sediment containers of the in-situ samplers with two size fractions were placed in the well at 30 cm apart in depth (Fig. 1).

2.3. Sampling sediments and groundwater

The in-situ samplers were removed from wells DH1 and DH2 at day 0 (<1 h), 11, 26, 53, 61, 76, 92, 99, 123, 136, 150, and 171 between June and December 2014, and sediment samples (ca. 10 g) were collected from the middle of the sampler using a sterilized spatula; and placed in sterile centrifuge tubes for analysis of sediment pH,

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