Chemosphere 162 (2016) 1–7

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Effects of bioaugmentation on sorption and desorption of benzene, 1,3,5-trimethylbenzene and naphthalene in freshly-spiked and historically-contaminated sediments

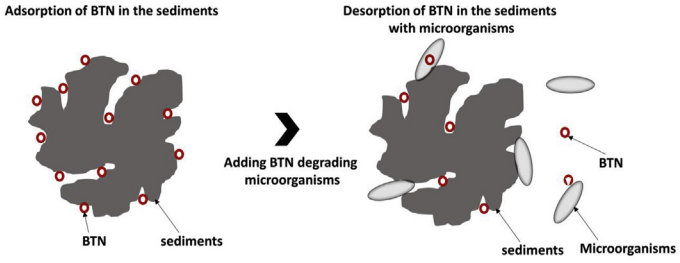
YueHua Li [a, b], Liang Chen [a, c, *], YuLong Liu [d, **], Fei Liu [e], Paul H. Fallgren [f], Song Jin [f]

[a] State Key Laboratory of Hydraulic Engineering Simulation and Safety, Tianjin University, Tianjin 300072, PR China
[b] School of Environmental Science and Engineering, Tianjin University, Tianjin 300072, PR China
[c] School of Civil Engineering, Tianjin University, Tianjin 300072, PR China
[d] Research Institute of Safety and Environment Technology, China National Petroleum Corporation, Beijing 102206, PR China
[e] Beijing Key Laboratory of Water Resources and Environmental Engineering, China University of Geosciences, Beijing 100083, PR China
[f] Advanced Environmental Technologies, LLC, Fort Collins, CO 80525, USA

HIGHLIGHTS

- Bioaugmentation influences aging of petroleum compounds in sediments.
- Microorganisms accelerate desorption of sequestrated benzene, 1,3,5-trimethylbenzene and naphthalene.
- Bioaugmentation influences desorption similarly in freshly- and historically-contaminated sediments.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 23 November 2015
Received in revised form
13 July 2016
Accepted 15 July 2016

Handling Editor: Chang-Ping Yu

Keywords:
Aged petroleum hydrocarbons
Bioaugmentation
Groundwater
Rebound
Sediments
Sorption and desorption

ABSTRACT

This work investigated the frequently observed “rebounds” of contaminants of concern in groundwater systems. Specifically, influences of bioaugmented microorganisms on the sorption and desorption of representative petroleum constituents [benzene, 1,3,5-trimethylbenzene and naphthalene (BTN)] were studied in freshly-spiked and historically–contaminated sediments. Capable microorganisms were enriched and supplemented to contaminated sediments to enhance biodegradation of petroleum hydrocarbons. In freshly-spiked sediments, when petroleum-degrading microorganisms were added, concentrations of dissolved petroleum constituents appeared to increase initially, and 12.4, 14.0 and 20.0 mg/kg of benzene, 1,3,5-trimethylbenzene and naphthalene, respectively desorbed from the sediments into the water phase. In the historically-contaminated sediments, the augmentation of petroleum-degrading microorganisms led to the desorption of 0.023–0.059, 0009–0.016, and 1.731–2.763 mg/L of previously sequestrated BTN into the water phase, and also triggered the desorption of 0.051 –0.223, –0.133–2.630, and 2.324–1.200 mg/kg of previously sequestrated BTN as the methanol extraction quantity. The mechanisms of the enhanced desorption at the presence of microbes remain to be determined; however, we presumed that microbially produced constituents such as biosurfactants and cell mass could have attributed to the partition of petroleum compounds from the sediments. Findings from this study may partially explain “rebounds” of certain petroleum constituents into the groundwater during *in situ* bioremediation practice, although such immediate rebounds sometimes are

* Corresponding author. School of Civil Engineering, Tianjin University, Tianjin 300072, PR China.
** Corresponding author.
E-mail address: liangchen@tju.edu.cn (L. Chen).

http://dx.doi.org/10.1016/j.chemosphere.2016.07.051

weak, and the desorbed constituents can be eventually biodegraded under proper biogeochemical conditions.

1. Introduction

Bioremediation is a common strategy for treating subsurface matrices impacted by organic contaminants such as petroleum compounds (Cerniglia, 1993; Alexander, 1995, 1999, 2000; Mehrdad et al., 2008; Jeon and Madsen, 2013). The success of bioremediation relies heavily on the bioavailability of the contaminants (Semple et al., 2004). For sites that are historically contaminated, the bioavailability of petroleum hydrocarbons tends to decrease with time due to "aging" (Alexander, 1995, 2000). The "aging" of contaminants may result in the sorption of contaminants into soils and the loss of bioaccessibility (Semple et al., 2004). This "aging" and the resultant sorption are also evident in the declined solvent extractability of contaminants in soils (Hatzinger and Alexander, 1995; Morrison et al., 2000; Semple et al., 2003, 2004; Smolders et al., 2009; Ma et al., 2012). Sorption and entrapment of petroleum compounds within soils are influenced by soil organic matter (SOM) content, type of soil inorganic constituents, activity of soil microorganisms, and the contaminant types and concentrations (Carroll et al., 1994; Reid et al., 2000; Sverdrup et al., 2002; Smolders et al., 2009; Shchegolikhina et al., 2012). Chen et al. (2013) reported that toluene biodegradation rates decreased due to the "aging" effect, and humic materials might have caused toluene sorption. Chung and Alexander (2002) observed that the extent of sorption of phenanthrene and atrazine was influenced by soil properties such as porosity, cation-exchange capacity, particle surface area, and clay mineralogy. Further evidence of contaminant sorption by soils was reported by Kottler and Alexander (2001), who observed 22–58% of polycyclic aromatic hydrocarbons were not solvent extractable from soil soon after spiking the compounds, and that percentage increased to 47–77% after 28 days.

Sorption of contaminants by soils can substantially affect microbial degradation activities. Meanwhile, the presence of microorganisms could also impact contaminants sorption and desorption in soils (Guthrie and Pfaender, 1998; Kastner et al., 1999). Xu and Zhang (1994) found that addition of microorganisms increased DDT desorption from soil. Nam and Alexander (2001) also reported that the percentage of a compound that could be sequestered was determined by the initial rate of its biodegradation. Much of the compounds became biologically unavailable (sequestered) when the initial rates of biodegradation were slow, and little remained if the soil had higher degradative activity. Similarly, Vessigaud et al. (2007) observed that biodegradation controlled the diffusion and sequestration of fluoranthene-degrading residues (parent compound and degradation products) by transforming it into more polar molecules that could diffuse into the intra-aggregate pore water and might become bound to the matrix or entrapped in the microporosity.

The role of contaminant-degrading microorganisms on compounds sequestration in sediments has not been well determined. During the remediation of petroleum hydrocarbons contaminated sites, the "rebounds" of target contaminants are frequently observed (Cunningham et al., 2001; Korotkevych et al., 2011; Ponsin et al., 2014). We hypothesized that some of these "rebounds" may be caused by the desorption of sequestrated petroleum hydrocarbons due to the augmentation of microbial populations and their activities; and the sites containing freshly spilled and "aged" contaminants would have different patterns of "rebounds". To test our hypothesis, representative petroleum hydrocarbons, benzene, 1,3,5-trimethylbenzene and naphthalene (BTN), were selected in this study. Pristine and contaminated sediments were used to represent sites containing freshly spilled and aged contaminants to investigate possible microbial influence on compounds sequestration and desorption during the implementation of bioaugmentation techniques.

2. Materials and methods

2.1. Groundwater and sediments

Groundwater used in the study was collected from a domestic well located on campus of China University of Geosciences in Beijing, PRC (Table 1). Groundwater was filtered through 0.2-μm membrane filters to remove microorganisms, and then purged with nitrogen (N_2) to reduce the dissolved oxygen (DO) to <1 mg/L to resume the anaerobic condition of the groundwater, unless otherwise stated. Petroleum hydrocarbon-contaminated sediments collected from an oilfield in Liaoning Province, China, were used as the microbial source in this study. Total petroleum hydrocarbon (TPH) content of the sediments was measured to be 42.1 g/kg. Pristine (uncontaminated) sediments used in this study were collected near the contaminated sediments location. All sediments were crushed and screened by 2-mm sieve, and characterized by using an X-ray diffractometer (D/MAX 2500, Akishima, Tokyo, Japan; Table 2).

2.2. Sorption, desorption and methanol extraction of BTN from uncontaminated sediments without microorganisms

To prepare BTN stock solution, groundwater (1 L) was added to a 1-L amber glass bottle sealed with PTFE-lined cap. Benzene (liquid, 100 mg), 1,3,5-trimethylbenzene (liquid, 20 mg) and naphthalene (solid, 20 mg) were added into the bottle. The bottle was shaken vigorously for 12 h at 25 ± 1 °C for complete solubilization. All chemicals used were analytical grade (Beijing Chemical Reagent Company, Beijing, China).

Seven 100-mL containers were set up with groundwater and BTN stock solution at ratios of groundwater: stock solution (volume ratios) of 90:10, 80:60, 60:40, 40:60, 20:80, 10:90, and 0:100. The containers were sampled before the sediments addition. The containers were amended with 25 g of uncontaminated sediments and incubated at 25 °C (rotating shaker set at 150 rpm) for 48 h. Then the containers were centrifuged at 2000 rpm for 15 min, and 1 mL of the supernatant water phase was collected to determine the BTN concentrations. The remaining water phase was discarded, and the weight of container with water and/or sediments was measured before/after every step during the sorption experiment to calculate the residual water phase volume in the sediments.

The containers were replenished with fresh groundwater and the above procedures were repeated to continue the desorption tests after each incubation period of 48 h. The concentration of BTN in the water phase from the sorption and desorption experiments was determined by a gas chromatograph-mass spectrometer (GC-MS) equipped with a purge and trap injector according to U.S.