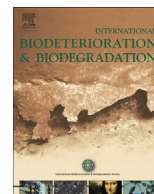




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Field scale ex-situ bioremediation of petroleum contaminated soil under cold climate conditions



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ABSTRACT

Assessment and development of strategies for ex-situ bioremediation in cold climates, such as in Canada, is of great importance for decontamination of contaminated soils. The purpose of this paper was to evaluate and compare the use of microbial consortia inoculant and mature organic compost as bioaugmentation and biostimulation strategies to enhance ex-situ bioremediation of a soil contaminated with heating oil under cold weather conditions. The soil was impacted as a result of leakage from an above-ground storage tank and had an initial TPH concentration was $940 \pm 127 \mu\text{g g}^{-1}$ (dry weight). Aerobic biopiles of 16 m^3 each were constructed and subjected at a field scale to microbial consortia inoculant and 10:1 ratio of mature organic compost. Two biopiles (S) contained only soil as control, two biopiles (S + C) contained soil and compost to assess the individual effect of compost addition, two biopiles (S + M) contained soil and microbial consortium to assess the individual effect of microbial consortium addition, and three biopiles (S + C + M) contained soil plus compost and microbial consortium to assess the combined effect of compost and microbial consortium addition. Over a 94 days period, composite soil samples for each biopile were collected and analysed for total petroleum hydrocarbons (TPHs), volatiles (F1), semi volatiles (F2) and non-volatiles (F3) fractions, microbial counting, and pH. Additionally, field measurements including soil temperature, moisture content, carbon dioxide (CO_2) and oxygen (O_2) were carried out. Although the ambient temperature varied from $-3.5 \text{ }^\circ\text{C}$ to $-24.1 \text{ }^\circ\text{C}$, the internal soil temperatures for the different experimental setups maintained above freezing conditions. Results showed that biocell inoculated with microbial consortia and amended with 10:1 soil to compost ratio under aerobic conditions performed the best, degrading 82% of total petroleum hydrocarbons (TPHs) with a first-order kinetic degradation rate of 0.016 d^{-1} , in cold weather conditions. The average removal efficiencies for TPHs after 94 days for S, S + M, S + C treatments were 48%, 55%, and 52%, respectively. Statistical analysis indicated significant difference ($p < 0.05$) within and between the final measurements for TPHs and significant difference between the treatment with combined effect (S + C + M) and the control (S) as well as individual treatments of S + C and S + M. The study concluded that the combination of microbial consortia inoculation and mature compost as a biostimulant in a 10:1 soil to compost ratio would be an appropriate bioremediation approach for this type of soil matrix, reaching values below provincial standard regulations in just 40 days after construction. This research aimed to provide valuable knowledge to practitioners about cost-effective and existing strategies for ex-situ bioremediation during cold weather conditions.

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1. Introduction

Fossil fuels represent by far the largest source of energy for human activities, accounting for 81% of all energy consumption worldwide in 2010 (IEA, 2012). During storage, transportation and combustion of fossil fuels massive amounts of contaminating

hydrocarbons are released into the surrounding environment with fuel spill being the major contamination route. In Canada, approximately 60% of contaminated sites involve petroleum hydrocarbon (PHC) contamination (Sanscartier et al., 2009b). Health risks and environmental impacts associated with the exposition of petroleum hydrocarbons have resulted in soil contamination to be considered as a major concern over the last few years. Soil contamination is not only a social and sanitary issue, but is also an economical concern, since it implies major costs related to decreasing productivity and monetary depreciation of the impacted sites for future use (Juwarkar et al., 2010). Several sites,

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among brownfields, require extensive cleanup to prevent further migration of contaminants into water, soil, air and consequent threat to human health. In Canada alone, it has been estimated that there are around 22,000 federal sites contaminated with hydrocarbons, accounting for multibillions of dollars in environmental liabilities and remediation costs (McKie, 2012).

Consequently, bioremediation of petroleum contaminated soil has become an important environmental activity, as an economical and environmentally viable solution to restore the environment to background levels in warm and temperate climates. It is also increasingly viewed as an appropriate remediation option in cold climates (Sanscartier et al., 2009b). Bioremediation exploits the ability of microorganisms to degrade or detoxify effectively and economically hydrocarbon-contaminated soils, mitigating the risk to human health and the environment (Adekunle, 2011).

Ex-situ bioremediation, as a main remediation technology to treat a wide range of hydrocarbons has been a topic of considerable research interest over the last couple of decades. In most cases the treatment of contaminated soils through ex-situ bioremediation involves two main strategies: bioaugmentation whereby petroleum degrading microorganisms are added to the soil matrix, and biostimulation, which introduces essential nutrients or biosurfactants to stimulate microbial petroleum degradation (Sayara et al., 2010). Both biostimulation and bioaugmentation can be accomplished separately or in combination, by introducing hydrocarbon-degrading bacteria and/or amending contaminated soil or sludge with a heterogeneous additional material such as compost (Namkoong et al., 2002; Yuan et al., 2009; Kriipsalu and Nammari, 2010; Adekunle, 2011). Calvo et al. (2009) reviewed the application of bioemulsifiers as a biostimulation strategy and reported that they could emulsify hydrocarbons and enhance their water solubility and increase the displacement of oily substances from soil particles. Łebkowska et al. (2011) applied multiple indigenous microorganism inoculation in soil polluted with diesel oil and aircraft fuel, obtaining 80–98% removal efficiencies of total petroleum hydrocarbons (TPHs). Lin et al. (2010) used a combination of bioaugmentation and biostimulation with the reseeded strategy for ex-situ bioremediation of petroleum contaminated soil. The diesel contamination was efficiently removed by about 70% (as total petroleum hydrocarbon) over a period of 4 weeks.

Nevertheless, successful bioremediation of petroleum hydrocarbons in soil has remained a challenge, especially under cold climate conditions. Due to the cold Ontario climate, ex-situ bioremediation techniques are limited to times of the year when temperatures are above freezing, i.e. April to November, as temperature plays an important role in controlling the nature and the extent of microbial metabolism and hydrocarbon bioavailability (Zhang et al., 2008). Over the previous years, various methods have been employed to effectively degrade petroleum contaminated soil under different approaches in cold weather conditions (Aislabie et al., 2006; Chang et al., 2011; Kauppi et al., 2011; Sanscartier et al., 2011). Chang et al. (2011) reported 55% decrease in TPHs concentrations of field aged petroleum contaminated soil by nitrogen amendment at a sub arctic site with temperatures between 4.7 and 10 °C. Mohn et al. (2001) reported on degradation of 2109 mg kg⁻¹ down to 195 mg kg⁻¹ of diesel in small-scale biopiles experiments after one year in the Canadian Arctic Tundra, using nutrients amendment, bulking agents and cultures of cold tolerant hydrocarbon degraders. Kauppi et al. (2011), successfully degraded 2700 mg kg⁻¹ of diesel-fuel contaminated clay loam soil in a boreal climate using field scale biopiles with forced aeration and adding nutrients amendment or woodchips as a bulking agent to increase porosity over a period of 11 month. Sanscartier et al. (2009b) were able to decrease 10,000 mg kg⁻¹ to approximately 300 mg kg⁻¹ of TPH concentration of diesel-contaminated soil by humidifying the

Table 1
Characteristics of the contaminated soil.

Parameter	Value	Method
<i>Physical characteristics</i>		
>19 mm (% by wt.)	8.1 ± 0.1	ASTM D2487
<19 to>4.75 mm (% by wt.)	12.7 ± 5.0	
<4.75 to>2.00 mm (% by wt.)	13.4 ± 1.7	
<2.00 to>0.425 mm (% by wt.)	49.9 ± 5.0	
<0.425 to>0.075 mm (% by wt.)	17.9 ± 2.3	
<0.075 mm (% by wt.)	3.3 ± 0.6	
<i>General inorganics</i>		
pH	7.79 ± 0.02	EPA 150.1 pH probe @ 25 °C
<i>Hydrocarbons</i>		
F1 (C6–C10) (µg g ⁻¹)	27 ± 4	CCME PHC,
F2 (C10–C16) (µg g ⁻¹)	455 ± 67	CWS Tier 1-GC-FID
F3 (C16–C34) (µg g ⁻¹)	442 ± 62	
F4 (C34–C50) (µg g ⁻¹)	ND	
TPHs (C6–C50) (µg g ⁻¹)	924 ± 127	

The values are expressed as mean ± standard deviation.

ND: non detectable.

TPHs are the sum of all the fractions.

air in an aerated/heated biopile system with average outdoor temperatures of 5.8 and 11.7 °C for 10 months. Such findings confirm the feasibility of different strategies such as biostimulation by nutrients amendment, application of bulking agents, and forced aeration, and bioaugmentation by inoculation of microorganisms to degrade hydrocarbons at significant rates under cold weather conditions (Margesin and Schinner, 2001).

Assessment of bioremediation strategies for cold climates, such as in Canada, and methods of enhancing cold weather degradation is of a great importance for an efficient and cost-effective decontamination of polluted soils. Considering above, the main objective of the present study was to assess and compare biodegradation of TPHs, as well as different fractions including readily volatile (F1:C6–C10), semi volatile (F2:C10–C16), and non-volatile (F3:C16–C34) fractions in petroleum contaminated soil using a biopile technology system under cold weather conditions. Field scale biopiles were constructed and subjected to microbial consortia inoculation and mature compost as bioaugmentation and biostimulation strategies and monitored over a period of 94 days (November 2012 to February 2013).

2. Materials and methods

2.1. Contaminated soil

Soil impacted with petroleum hydrocarbons was obtained from a contaminated site in Val-des-Bois, in the Outaouais region of Quebec, Canada, located on the eastern shores of the Du Lièvre River, north of Buckingham. Throughout the site history, heating oil (C14–C20) was stored in an above-ground storage tank (ASTs) as a fuel source. Due to leakage, the AST was removed in 1994 and subjected to environmental assessment. Current results showed a contaminated area between 11.5 and 13.5 m below the surface, covering an approximately area of 1600 m². Initial samples indicated that petroleum hydrocarbon in soil were higher than the criteria C, based on guidelines issued by Quebec provincial regulations, with an estimated concentration between 6000 and 15000 µg g⁻¹ dry weight (WESA, 2012).

The contaminated soil was excavated and transported to a treatment facility in Moose Creek, Ontario, Canada in October 2012, which had a pH of 7.79 ± 0.02 (Table 1). The impacted soil was classified as sand, according to the USDA classification system and based on grain size analysis of five composite random samples

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