



Bioremediation of creosote contaminated soil in both laboratory and field scale: Investigating the ability of methyl- β -cyclodextrin to enhance biostimulation



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ABSTRACT

We investigated the bioremediation of 16 polycyclic aromatic hydrocarbons (PAH) in historically creosote contaminated soil using both laboratory and field scale experiments. We found that nutrient amendments and circulation of methyl- β -cyclodextrin (CD) solution enhanced soil microbial degradation capacity. In the laboratory experiment, the degradation of lower molecular weight, 2–3 ringed PAHs was achieved already by circulating nutrient solution and the use of CD mainly increased the desorption and removal of larger, 4–5 aromatic ringed PAH compounds. The 1% CD concentration was most feasible for bioremediation as most of the extracted PAH compounds were degraded. In the 5% CD treatment, the PAH desorption from soil was too fast compared to the degradation capacity and 25% of the total PAH amount remained in the circulated solution. Similar lab-scale results have been generated earlier, but very little has been done in full field scale, and none in freezing conditions. Although freezing stopped circulation and degradation completely during the winter, PAH degradation returned during the warm period in the field test. Circulation effectiveness was lower than in the laboratory but the improved nutrient and moisture content seemed to be the main reason for decreasing soil PAH concentrations. It also appeared that PAH extraction yield in chemical analysis was increased by the CD treatment in field conditions and the results of CD-treated and non-treated soil may therefore not be directly comparable. Therefore, a positive effect of CD on PAH degradation velocity could not be statistically confirmed in the field test. Based on our results, we recommend initiating the bioremediation of PAH contaminated soil by enhancing the microbial degradation with nutrient amendments. The CD seems to be useful only at the later stage when it increases the solubilisation of strongly absorbed contaminants to some extent. More investigation is also needed of the CD effect on the PAH yield in the chemical analysis.

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1. Introduction

Creosote has been used for decades in wood impregnation processes to preserve and waterproof wooden structures like railway sleepers, telephone poles and bridge and pier deckings. The annual creosote production has been up to 100,000 tons in both Europe and in the USA, the main use being in the wood preservation industry (Melber et al., 2004), where the spills in wood

preserving plants or releases from treated wood products have contaminated the soil. The European Soil Data Centre estimated that wood and paper industry contributes to nearly 4% of soil contamination in Europe and that polycyclic aromatic hydrocarbons (PAHs) have a role in nearly 11% of all soil contamination (Panagos et al., 2013). In Finland, approximately 6% of all contaminated land sites are contaminated by creosote and most of these are in the premises of old sawmills (Sunni et al., 2007).

Coal tar creosote is a dark, oily liquid formed by fractional distillation of crude coal tars in 200–400 °C. It consists of a complex mixture of several hundred chemicals, of which only 20% are present in amounts greater than 1%. The composition of creosote varies

Abbreviations: PAH, polycyclic aromatic hydrocarbon; CD, cyclodextrin.

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depending on the origin of the coal and the distillation process, but six major compound classes can be sorted out: i.e. aromatic hydrocarbons, tar acids/phenolics, nitrogen-, sulphur- or oxygen containing heterocycles and aromatic amines (Melber et al., 2004). The main compound class of creosote is the PAHs, which can make up approximately 85% of creosote composition (Mueller et al., 1989).

PAHs are hydrophobic organic compounds with a low aqueous solubility, high melting and boiling points and low vapour pressure (Zhang et al., 2006). They are composed of fused, aromatic rings, whose biochemical persistence arises from the dense clouds of π -electrons on both sides of the ring structures, making PAHs resistant to nucleophilic attack (Zhang et al., 2006). These properties promote PAH accumulation in the solid phases of the terrestrial environment and make them highly persistent in the soil (Zhang et al., 2006). PAH compounds are also resistant to the utilization by bacteria, which can degrade chemicals only when these are dissolved in water (Harms and Bosma, 1997; Johnsen et al., 2005). Despite this and although PAHs may also undergo adsorption, volatilization, photolysis and chemical degradation, microbial degradation is the major elimination process of PAHs in the soil (Koivula et al., 2004; Haritash and Kaushik, 2009). Many bacterial (*Pseudomonas aeruginosa*, *P. fluorescens*, *Mycobacterium* spp., *Haemophilus* spp.) and fungal (*Pleurotus ostreatus*) species can degrade PAHs in contaminated soils or sediments (Haritash and Kaushik, 2009), but the degradation rate in natural environmental conditions is extremely slow, if not completely lacking, due to the high recalcitrance of PAHs (Breedveld and Sparrevik, 2000). This is often due to the slow rate of contaminant desorption from soil particles, i.e. poor bioavailability, rather than the inability of degradation by microorganisms (Breedveld and Karlsen, 2000).

In many European countries, specific guideline values have been set for the most relevant soil and groundwater contaminants. The values are based on the ecological or health risk, but these values vary in different countries (Carlon, 2007). According to the decree on the Assessment of Soil Contamination and Remediation Needs, published by the Finnish Government in 2007 (214/2007) the threshold value (i.e. assessment is needed when the value is exceeded) for the sum concentration of 16 PAHs in soil is 15 mg kg^{-1} d.w., the lower guideline value (residential use of soil) 30 mg kg^{-1} d.w. and the higher guideline value (industrial use of soil) 100 mg kg^{-1} d.w. Guideline values have also been given for seven individual PAHs (anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(k)fluoranthene, phenanthrene, fluoranthene and naphthalene): for these, the threshold, lower, and higher guideline values are 1, 5 and 15 mg kg^{-1} d.w., respectively, except for the high cancer risk benzo(a)pyrene, for which the threshold value is 0.2 and the lower guideline value 2 mg kg^{-1} d.w.

As the water solubility of contaminants is the rate-limiting factor for microbial degradation, additives have been used to enhance contaminant availability. Surfactants are surface tension reducing compounds, which increase the solubility and bioavailability of hydrophobic organic compounds by accumulating at the interface of two phases that have distinct polarity (Singh et al., 2007). In the bioremediation of contaminated soils, both synthetic and biosurfactants have been used. One group of the biosurfactants are cyclodextrins (CDs), which are oligosaccharides formed in the enzymatic degradation of starch by bacteria (Wang and Brusseau, 1995; Semple et al., 2007). The three most common cyclodextrins are α -, β -, and γ -CD, which contain 6, 7, and 8 monomeric glucopyranose units, respectively (Ravelet et al., 2002). These compounds are torus-shaped molecules with a hydrophilic exterior, which provides them with high aqueous solubility and a hydrophobic cavity, into which organic contaminants can be encapsulated (Ravelet et al., 2002; Del Valle, 2004; Papadopoulos

et al., 2007). Other advantages of using CDs in bioremediation are their biodegradability, non-toxicity, and relatively stable physicochemical properties in a range of solution chemistry conditions (McCray and Brusseau, 1998; Ko et al., 1999). Of the three CD homologues, β -CDs or their derivatives are the most studied in soil remediation because they are the cheapest option (Wang and Brusseau, 1993; Badr et al., 2004; Viglianti et al., 2006), they are suitable in size for the encapsulation of aromatic hydrocarbons, and their water solubility can be chemically enhanced (Petitgirard et al., 2009). However, the evidence of the remediation effect of β -CDs in PAH contaminated soils is mainly based on laboratory studies (Viglianti et al., 2006; Petitgirard et al., 2009; Zhang et al., 2012; Sun et al., 2013) and the implementation of the procedures in pilot and full scale experiments is lacking although these are essential to understand how remediation treatments can be up-scaled and how they function in realistic field conditions.

Here we present results from a study, where we investigated whether biodegradation of PAHs in contaminated soil can be enhanced using methyl- β -cyclodextrin. Based on the literature we hypothesized that CD would have a positive effect on PAH removal in lab conditions, but the aim was to test whether this effect could be replicated in field scale in cold conditions. We first tested the cleaning efficiency of varying CD-concentrations (0%, 1% and 5%) in creosote contaminated soil in the laboratory, and then based on the results of the laboratory test, we chose the most effective CD concentration (1%) for a field-scale bioremediation test of the same soil. In both tests, soil remediation was based on the degradation capacity of the indigenous microbial community, stimulated using nutrient amendments.

2. Materials and methods

2.1. Experimental soil

The soil used in this study was collected from a former impregnation plant area in Mikkeli, Eastern Finland, where railway sleepers had been treated with creosote oil from the beginning of the 20th century until 1982 (Sunni et al., 2007). The restoration was started in 2002 using mainly soil excavation and off-site disposal. The excavated soil was stored at the waste disposal site of Metsäsairila Ltd in Mikkeli, where the field experiment of this study was implemented. The texture of the soil was sand with a total C content of 7.6 g kg^{-1} soil (d.w.) and total N content less than 1 g kg^{-1} soil (d.w.) (analysed in Ramboll Analytics, Lahti). The total PAH concentration was around 1000 mg kg^{-1} soil (d.w.) with highest concentrations composed of 3-ringed fluoranthene and 4-ringed pyrene (Table 1).

2.2. Chemicals used in the remediation treatments

In the laboratory and field experiment, a 50% (w/v) aqueous stock solution of methyl- β -cyclodextrin (Cawasol W7 M TL, Wacker Chemie AG, Germany) was used as a surfactant. In the field test, 75% calcium peroxide (CaO_2) (IXPER[®]75C, Solvay S.A, Belgium), consisting of 17% (by weight) molecular O_2 , was used as an oxygen source. CaO_2 is a powdery chemical, which decomposes and releases oxygen in the presence of water ($2\text{CaO}_2 + 2\text{H}_2\text{O} \geq 2\text{Ca(OH)}_2 + \text{O}_2$) (Nykänen et al., 2012). The N sources used in the field experiment were methylene urea (Yara Suomi Oy, Finland) and saltpetre (YaraBela Suomensalpietari Se+, Yara Suomi Oy, Finland) which contains 14.5% $\text{NH}_4\text{-N}$ and 12.5% $\text{NO}_3\text{-N}$. Using methylene urea in soil bioremediation is recommendable because it releases N slowly and the overdose of ammonia and excessively high pH values can be avoided (Peltoala et al., 2006). Ammonium nitrate (NH_4NO_3) solution was used as a

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