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The impact of lead co-contamination on ecotoxicity and the bacterial community during the bioremediation of total petroleum hydrocarbon-contaminated soils ☆

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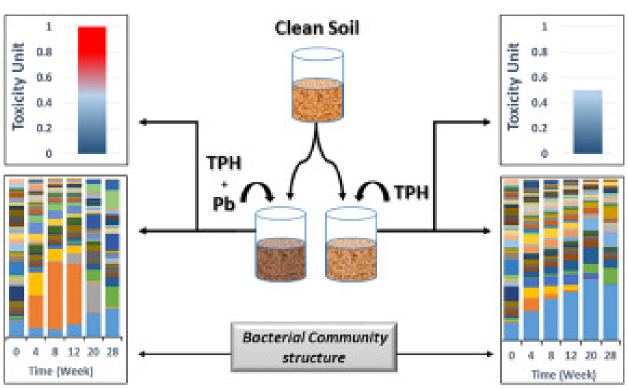
Highlights

- Biostimulation is effective in remediating co-contaminated soils.
- Co-contamination has a deleterious impact on the efficacy of bioremediation.
- The presence of lead (Pb) increases the associated ecotoxicity.
- Bacterial diversity is influenced by the presence of pollutants and nutrients.

Abstract

The continued increase in the global demand for oil, which reached 4,488 Mtoe in 2018, leads to large quantities of petroleum products entering the environment posing serious risks to natural ecosystems if left untreated. In this study, we evaluated the impact of co-contamination with lead on the efficacy of two bioremediation processes, natural attenuation and biostimulation of Total Petroleum Hydrocarbons (TPH) as well as the associated toxicity and the changes in the microbial community in contaminated soils. The biostimulated treatment resulted in 96% and 84% reduction in TPH concentration in a single and a co-contamination scenario, respectively, over 28 weeks of a mesocosm study. This reduction was significantly more in comparison to natural attenuation in a single and a co-contamination scenario, which was 56% and 59% respectively. In contrast, a significantly greater reduction in the associated toxicity of in soils undergoing natural attenuation was evident compared with soils undergoing biostimulation despite the lower TPH degradation when bioassays were applied. The earthworm toxicity test showed a decrease of 72% in the naturally attenuated toxicity versus only 62% in the biostimulated treatment of a single contamination scenario. In a co-contamination scenario, toxicity decreased only 30% and 8% after natural attenuation and biostimulation treatments, respectively. 16s rDNA sequence analysis was used to assess the impact of both the co-contamination and the bioremediation treatment. NGS data revealed major bacterial domination by Nocardioides spp., which reached 40% in week 20 of the natural attenuation treatment. In the biostimulated soil samples, more than 50% of the bacterial community was dominated by Alcanivorax spp. in week 12. The presence of Pb in the natural attenuation treatment resulted in an increased abundance of a few Pb-resistant genera such as *Sphingopyxis* spp. and *Thermomonas* spp in addition to *Nocardioides* spp. In contrast, Pb co-contamination completely shifted the bacterial pattern in the stimulated treatment with Pseudomonas spp. comprising approximately 45% of the bacterial profile in week 12. This study confirms the effectiveness of biostimulation over natural attenuation in remediating TPH and TPH-Pb contaminated soils. In addition, the presence of co-contaminants (e.g. Pb) results in serious impacts on the efficacy of bioremediation of TPH in contaminated soils, which must be considered prior to designing any bioremediation strategy.

Graphical abstract



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Introduction

The demand for petroleum products continues to increase; in 2018, world oil demand increased by 1.5 million barrels a day, 1.6% higher than the average in the last decade (IEA, 2018). Inevitably, during the exploration, recovery, storage and transport of such large quantities of petroleum products, vast amounts of petrogenic hydrocarbons enter the environment causing serious land contamination (Varjani, 2017).

Total Petroleum Hydrocarbons (TPHs), the main component of crude oil comprise a broad family of short (C_8–C_{16}) and long-chain (C_{17}–C_{40}) aliphatic hydrocarbons and a minor group of aromatic compounds (1–5 rings), which largely comprise carbon and hydrogen (Abbasian et al., 2015). TPHs are classified as priority contaminants due to their direct and indirect effects on the ecosystem if left untreated. When TPH seeps into the soil, low molecular weight volatile compounds evaporate, while other compounds attach to particles in the soil where they may remain for years or enter the groundwater, causing deleterious effects on the environment and human health. The high toxicity of TPH has a direct effect on the soil biota as well as severe disorders of the human immune system, central nervous systems, kidneys, liver and spleen (ATSDR, 2011).

Various chemical and physical techniques can be used to treat TPH contaminated soil, such as soil washing, soil vapour extraction, incineration and solidification (Jasmine and Mukherji, 2019; Chen et al., 2019). These techniques, are however relatively expensive due to operational costs and damage the natural properties of the soil (Xu and Lu, 2010). In contrast, bioremediation, which recruits indigenous biological agents to breakdown the contaminants, represents a simple, environmentally safe and cost-effective technique of contaminated soil remediation (Ron and Rosenberg, 2014). Indeed, the first response to any soil contamination takes place naturally through the action of the indigenous microflora of the soil. This process, natural attenuation occurs when biodegradation of the contaminant occurs without any enhancement or human interference (Yu et al., 2005). The biodegradation rate can be accelerated by biostimulation where nutrients such as carbon, phosphorus, nitrogen and oxygen are added to the contaminated soil in order to stimulate the microflora and thus accelerate the biodegradation process (Andreolli et al., 2015). Biostimulation has been successfully applied to many contamination cases resulting in a significant decrease in TPH concentration. For example, higher degradation rates (78–90%) of TPH have been reported using biostimulation in comparison with natural attenuation (61–77%) (Khudur et al., 2015; Qin et al., 2013).

However, one major challenge impacting the efficiency of the bioremediation of TPH contaminated soil is co-contamination with heavy metals (Thavamani et al., 2012; Olaniran et al., 2013a; Liu et al., 2017; Khudur et al., 2018b). Lead (Pb) is likely to be present alongside TPH as a co-contaminant in many aged oil spills, as Pb was widely used as a fuel additive (Khudur et al., 2018a). A recent study confirmed that Pb concentrations (50–1750 mg kg^{-1}) were detected in 58 surface soil samples collected from the Melbourne metropolitan area, Australia (Laidlaw et al., 2018). According to Australian guidelines established by the National Environment Protection Council, the Health

Investigation Levels (HILs) of soil Pb is 300mg kg^{-1} for residential areas with gardens or accessible soil and 1500mg kg^{-1} for industrial or commercial areas (NEPC, 2011).

Lead is highly toxic to soil biota since it has a higher affinity for oxygen and thiol groups, enabling it to displace essential metals from their binding site (Bruins et al., 2000; Nouha et al., 2016). We recently reported an elevation in ecotoxicity in aged TPH-heavy metals co-contaminated soils (Khudur et al., 2018a). The presence of Pb in TPH-contaminated soil affects the structure of the microbial community which underpins any biodegradation process. Although numerous recent studies have addressed the biodegradation of TPH in co-contaminated environments, very little is known about the changes in dynamics of natural soil microbial communities' during the bioremediation of co-contamination scenarios (Varjani, 2017; Liu et al., 2017). Understanding changes in the microbial communities' structure following exposure to contaminants represents a crucial step in designing an effective bioremediation strategy (Chen et al., 2015; Klimek et al., 2016).

The aim of this study was to evaluate the efficacy of bioremediating TPH in TPH-Pb co-contaminated soils and to assess the subsequent impact of the remediation process on soil ecotoxicity. In addition, 16S amplicon sequencing was employed to assess the changes in the microbial community during the remediation process. In our previous studies, we evaluated the efficacy of natural attenuation and biostimulation in remediation TPH-contaminated soil by creating an optimal C:N:P molar ratio in the biostimulated soils (Khudur et al., 2015). We have also reported that TPH and heavy metal co-contamination significantly elevated the remaining ecotoxicity of the weathered, naturally attenuated soils (Khudur et al., 2018a). Here, we are reporting, for the first time, the efficacy of natural attenuation and biostimulation in remediating TPH-Pb co-contaminated soils, using RemActiv a commercially available biostimulator. In addition, to the best of the authors' knowledge, the impact of Pb co-contamination together with biostimulation on the associated ecotoxicity and the soil bacterial community during the bioremediation process has not been previously reported. Thus, here we aim to achieve a solid basis upon which to design appropriate bioremediation strategies for co-contaminated soils.

Section snippets

Experiment design

Clean pastureland soil from Victoria, Australia, was collected using a sterilized shovel and placed in clean plastic buckets and then transported to RMIT University, Bundoora campus (Schinner et al., 2012). The collected soil was stored overnight at ambient temperature and then sieved using a 6 mm sieve. The soil was divided into six sub-samples (12 kg each) to set up the experimental treatments. The treatments included (i) Natural attenuation (NA) and (ii) Biostimulation (BS) of TPH only …

Assessing TPH concentration

The efficacy of bioremediation in the reduction of TPH in the TPH contaminated and TPH-Pb co-contaminated soils is shown in Fig. 1A. A significant reduction in TPH concentration was observed in both bioremediation strategies implemented, natural attenuation and biostimulation, for single and co-contamination scenarios.

The biostimulation strategy resulted in significantly higher efficiency in terms of bioremediation of TPH contaminated soil in comparison with natural attenuation. In a single …

Conclusion

This mesocosm study concluded that the presence of Pb as a co-contaminant has negatively impacted the efficacy of TPH bioremediation, especially after biostimulation, in co-contaminated soils. The biostimulation treatment showed 84% reduction in TPH concentration representing a 24% greater reduction than natural attenuation in co-contaminated soils. However, this represents 11% less reduction in comparison to TPH- only contaminated soils. Also, in co-contaminated soils, the biostimulation …

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: …

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