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Comparison of landfarming amendments to improve bioremediation of petroleum hydrocarbons in Niger Delta soils

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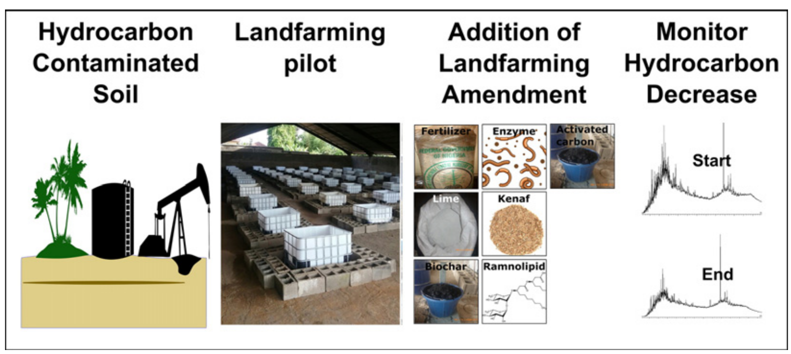
HIGHLIGHTS

- Seven amendments to improve landfarming of hydrocarbons in soils were tested.
- Most were no better than NPK nutrients except biochar combined with rhamnolipid.
- Attenuation rate and hydrocarbon carbon number inversely related.
- Heavier hydrocarbons contributed more to removal due to higher initial concentration.
- Aromatics attenuated faster than aliphatics for all hydrocarbon fractions.

GRAPHICAL ABSTRACT



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ABSTRACT

Large scale landfarming experiments, using an extensive range of treatments, were conducted in the Niger-Delta, Nigeria to study the degradation of oil in contaminated soils. In this work the effect of nutrient addition, biosurfactant, *Eisenia fetida* (earthworm) enzyme extract, bulking and sorption agents and soil neutralization were tested. It was found that these treatments were successful in removing up to 53% of the total petroleum hydrocarbon in the soil within 16 weeks. A comparison between treatments demonstrated that most were no more effective than agricultural fertilizer addition alone. One strategy that did show better performance was a combination of nutrients, biochar and biosurfactant, which was found to remove 23% more Total Petroleum Hydrocarbons (TPH) than fertilizer alone. However, when performance normalized costs were considered, this treatment became less attractive as a remedial option. Based on this same analysis it was concluded that fertilizer only was the most cost effective treatment. As a consequence, it is recommended that fertilizer is used to enhance the landfarming of hydrocarbon contaminated soils in the Niger Delta.

The attenuation rates of both bulk TPH and Total Petroleum Hydrocarbon Criteria Working Group (TPHCWG) fractions are also provided. These values represent one of the first large scale and scientifically tested datasets for treatment of contaminated soil in the Niger Delta region. An inverse correlation between attenuation rates

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and hydrocarbon molecular weight was observed with heavy fractions showing much slower degradation rates than lighter fractions. Despite this difference, the bioremediation process resulted in significant removal of all TPH compounds independent of carbon number.

1. Introduction

The Niger Delta (Nigeria) is a region with a substantial history of oil and gas extraction going back to the 1950s. Oil spills have occurred in the region as a result of sabotage crude oil theft, operational spills, and refining under very primitive conditions (Lindén and Pålsson, 2013; Moffat and Linden, 1995; UNEP, 2011). In 2012, the International Union for Conservation of Nature Niger Delta Panel (IUCN-NDP) reviewed the biodiversity recovery of selected impacted areas within the Niger Delta and provided recommendations on remediation and rehabilitation processes for oil impacted sites in the Niger Delta (IUCN-NDP, 2013). One of the main features of the work was focused on reviewing and recommending techniques to improve the effectiveness of landfarming as a method to enhance the degradation of oil in contaminated soil (IUCN-NDP, 2013).

Landfarming is a well-practiced technique used in the Delta, and elsewhere, to bioremediate crude oil contamination in soils to reduce the oil concentration and the associated risks to human health and the environment (Maila and Cloete, 2004). It reduces the concentration of petroleum hydrocarbon constituents principally by bacterially-mediated biodegradation, although volatilization, abiotic processes and fungal-mediated processes may also play a part (Maila and Cloete, 2004). It typically involves the spreading of excavated contaminated soils in a thin layer on the ground surface of a treatment site and stimulating aerobic microbial activity within the soils to accelerate naturally occurring biodegradation processes (Brown et al., 2017; Khan et al., 2004; Vidali, 2001). The removal of petroleum hydrocarbons using landfarming has been applied at large scale with good rates of success (Brown et al., 2017; Heitzer and Sayler, 1993). In addition, field experiments after real oil spills have also shown that the degradation rates can be enhanced with similar methods as those presented in this study (Zabbey et al., 2017; Atlas and Bragg, 2013). The technology is attractive for use in the Niger Delta because it is simple to implement, requires little in terms of infrastructure or equipment, is effective in reducing hydrocarbon concentrations in the environmental conditions founds in the Delta (a warm, wet tropical forest), has less detrimental impacts to surrounding communities than certain more aggressive remedial options, and provides employment opportunities for local communities. As landfarming is most effective when environmental conditions permit microbial growth and activity, its application often involves the enhancement of certain environmental parameters including moisture content, pH and availability of oxygen and nutrients (Brown et al., 2017; Liu et al., 2017).

Weathering of petroleum hydrocarbon contamination in soils is a process characterized by the attenuation of lighter, more volatile and water soluble hydrocarbons and a concomitant increase in the proportion of heavier and more structurally complex components in the residual oil. A number of studies have shown weathered hydrocarbons to be more difficult to biodegrade (Atlas and Bragg, 2013; Björklöf et al., 2008). This is caused by a change in the physical and chemical characteristics of the oil and an increased tendency to bind to soils (Björklöf et al., 2008; Brassington et al., 2010; Huesemann et al., 2004; Liu et al., 2012; Semple et al., 2003). As a result, weathered hydrocarbons are thought to show reduced bioavailability for biodegradation processes. Most remediation of onshore oil spills in the Niger Delta is performed on soils that have undergone some degree of weathering. Therefore, understanding and optimizing landfarming conditions to improve the removal of weathered hydrocarbons is an important step to improving remediation strategies of contaminated soils. The aim of this study was to assess various strategies used to improve the effectiveness of landfarming on oil contaminated soils.

Data is presented from a large scale mesocosm trial performed in Nigeria using an extensive range of treatment variations. To our knowledge, this is the first time a large number of landfarming enhancement technologies have been compared together in a scientifically robust manner to identify the best remedial options available to enhance removal of hydrocarbons from soils using landfarming.

2. Experimental methods

2.1. Test soils and crude oils

The effectiveness of various landfarming treatments were tested on clean, sieved (5 mm mesh) Niger Delta agricultural top soil, sourced from Port Harcourt, combined with fresh Bonny Light Crude oil. The soil used in this work originated from the "low land forest" eco-zone in the Niger Delta. The lowland rain forests areas in the Niger Delta are the most habituated of the ecozones. They are typically cleared of native woodland vegetation and most of the former lowland rainforest is now derived savannah. The land does not flood during the wet season, apart from the areas adjacent to surface watercourses or within surface water flood plains. Soils are typically red clayey silt or silty clay.

2.2. Methods for hydrocarbon analyses in soils

Solvent extractable material (SEM) was measured gravimetrically following Soxhlet extraction with dichloromethane according to Standard Method 5520 E (Clesceri et al., 1998). Total petroleum hydrocarbon (TPH), C_8 to C_{40}, was determined by Gas Chromatography (GC) after extraction with acetone and hexane (1:1) according to a USEPA 8015B method (USEPA, 1996a). TPHCWG of hydrocarbon fractions was measured using USEPA 8015B after fractionation into aliphatic and aromatic fractions using a Rapid Trace Solid Phase Extraction Column (Biotage, Uppsala, Sweden). Hydrocarbons measured by the TPHCWG method were grouped into 14 fractions based on equivalent carbon (EC) numbers (TPHCWG, 1997). For determination of Gasoline Range Organics (GRO) in the carbon chain range of C_5–C_{10} by was performed by headspace GC-FID using the extraction method as described in US EPA method 5021 (USEPA, 2014). Volatile hydrocarbons were quantified using headspace GC–MS according to USEPA methods 8260 (USEPA, 1996b). Semi-volatile hydrocarbons were determined by GC–MS according to USEPA 8270 (USEPA, 1998) after dichloromethane extraction. All work described above was performed by Jones Environmental Laboratories, Flintshire, UK.

2.3. Methods for non-hydrocarbon analysis in soils

pH was determined using Metrohm automated probe analyser after extraction of soil using one part solid to 2.5 parts deionised water. Soil metals were determined by Inductively Coupled Plasma - Optical Emission Spectrometry (ICP-OES) using US EPA Method 200.7 (Martin et al., 1994) following acid digestion of dried and ground solid samples using Aqua Regia refluxed at 112.5 °C. Organic carbon was quantified by combustion in an Eltra Total Organic Carbon furnace/analyser following method USEPA 415.1 (USEPA, 1974). Total nitrogen was determined using the Kjeldahl method (Bradstreet, 1954). Anion and cation concentrations in soil pore-water were determined by a Thermo Aquakem Photometric Automatic Analyser. The experimental procedures