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Remediation trials for hydrocarbon-contaminated sludge from a soil washing process: Evaluation of bioremediation technologies

F.J. García Frutos[a,*], R. Pérez[a], O. Escolano[a], A. Rubio[b], A. Gimeno[b], M.D. Fernandez[c], G. Carbonell[c], C. Perucha[d], J. Laguna[d]

[a] CIEMAT, Avda. Complutense 40, 28040 Madrid, Spain
[b] IGME, La Calera 1, 28760 Tres Cantos, Madrid, Spain
[c] Laboratory for Ecotoxicology, INIA, Crta. de la Coruña Km. 7.5, 28040 Madrid, Spain
[d] HERA AG AMBIENTAL, Isla de Hierro 7, 28703 S. Sebastián de los Reyes, Madrid, Spain

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ABSTRACT

The usual fate of highly contaminated fine products (silt-clay fractions) from soil washing plants is disposal in a dump or thermal destruction (organic contaminants), with consequent environmental impacts. Alternative treatments for these fractions with the aim of on-site reuse are needed. Therefore, the feasibility of two technologies, slurry bioremediation and landfarming, has been studied for the treatment of sludge samples with a total petroleum hydrocarbon (TPH) content of 2243 mg/kg collected from a soil washing plant. The treatability studies were performed at the laboratory and pilot-real scales. The bioslurry assays yielded a TPH reduction efficiency of 57% and 65% in 28 days at the laboratory and pilot scale, respectively. In the landfarming assays, a TPH reduction of 85% in six months was obtained at laboratory scale and 42% in three months for the bioremediation performed in the full-scale. The efficiency of these processes was evaluated by ecotoxicity assessments. The toxic effects in the initial sludge sample were very low for most measured parameters. After the remediation treatments, a decrease in toxic effects was observed in earthworm survival and in carbon mineralisation. The results showed the applicability of two well known bioremediation technologies on these residues, this being a novelty.

1. Introduction

A particular contaminated site may require a combination of procedures to achieve optimum remediation. Biological, physical and chemical technologies may be used in conjunction to reduce the contamination to a safe and acceptable level. The selection of appropriate technologies depends on the nature of the contaminant(s) and site characteristics, regulatory requirements, costs, and time constraints. The successful treatment of a contaminated site depends on the proper selection, design and adjustment of remediation technology operations based on the properties of the contaminants and soils and on the performance of the system [1].

Soil washing uses liquids (usually water, occasionally combined with solvents) and physical processes to scrub soils. This process separates fine soil (clay and silt) from coarse soil (sand and gravel). Because hydrocarbon contaminants tend to bind and sorb to smaller particles, separating the smaller soil particles from the larger ones reduces the volume of contaminated soil. Soil washing is cost effective because it reduces the quantity of material that would require further treatment by another technology. This smaller volume of soil, which contains the majority of clay and silt particles, can be further treated by other methods. The usual destination of these fractions is disposal in a dump or thermal destruction (in cases of organic contaminants), with consequential environmental impacts. The development of alternative, environmentally friendly treatments for these fractions allowing for their on-site reuse is therefore needed.

Bioremediation is an attractive approach for cleaning up petroleum hydrocarbons because it is simple to maintain, applicable over large areas, cost effective and leads to the complete destruction of the contaminants. These treatments have emerged as a "green" alternative for treating these environmental contaminants [1]. Conventionally, on-site technologies such as landfarming, composting and soil piles have been employed; the most advanced ex situ methods such the use of bioreactors provide better control to enhance the hydrocarbon degradation process.

One of the difficulties of developing bioremediation strategies lies in achieving results in the field that are as good as those in the laboratory. To date, most of the reported experiments on soil bioremediation have been performed in the laboratory (in

* Corresponding author at: Energy Department (CIEMAT) Building 20, E20.P1.12 Avda. Complutense 40, 28040 Madrid, Spain. Tel.: +34 91 346 6529; fax: +34 91 346 6269.
E-mail address: garcia.frutos@ciemat.es (F.J.G. Frutos).

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well-controlled conditions), whereas field-scale experiments have remained scarce [2]. In this context, treatability or feasibility studies are used to determine whether remediation will be effective in a given situation. The extent of the study varies depending on the nature of the contaminants and the characteristics of the site. For sites contaminated with common petroleum hydrocarbons (e.g., gasoline and/or other readily degradable compounds), it is usually sufficient to examine representative samples for the presence and level of an indigenous population of microbes, nutrient levels, the presence of microbial toxicants, and sample characteristics such as pH, porosity, and moisture. Hydrocarbon bioremediation can be promoted by stimulation of the indigenous microorganisms by introducing nutrients and oxygen (biostimulation) [3].

The objectives of remediation processes are usually based on threshold levels of soil contaminants. Bioremediation has proven to be successful in numerous applications for petroleum-contaminated soils. However, during bioremediation processes, changes in bioavailability and metabolite yield can occur. Therefore, questions remain as to the efficiency of bioremediation in lowering soil toxicity. Consequently, it is necessary to incorporate ecotoxicity assessments to evaluate the treatment efficiency.

Due to the scarcity of studies on biological treatment of these classes of residues [4,5], the possibilities for the treatment of sludge (fine fraction) from a soil washing plant at a site contaminated with hydrocarbons using bioremediation technologies were studied in this paper. Chemical and biological assays were combined to evaluate the efficiency of the sludge remediation technologies.

2. Materials and methods

2.1. Sample characterisation

The studied sample was the sludge fraction from a soil washing plant at a site contaminated by hydrocarbons. The site was previously contaminated due to hydrocarbons storage tanks, mainly diesel hydrocarbons. The physical properties of the sample were determined according to the standard procedures [6]. The grain size characterisation was performed by elutriation using Cyclosizer equipment.

The hydrocarbon content was determined by gas chromatography–flame ionisation detection (GC-FID), measuring aliphatic and aromatic hydrocarbon fractions in the range of C_{10}–C_{40} according to the ISO 16703:2004 norm [7]. The total petroleum hydrocarbon (TPH) content was also determined with an infrared (IR) method using the portable InfraCal model HATR-T2 analyser in accordance with EPA method 1664 [8].

In selected samples, gas chromatography coupled to mass spectrometry, GC-MS, was performed to determine the major fractions affected by biodegradation. Samples of 1 g were extracted with hexane. Analyses by GC/MS were performed on the hexane extracts, conducted in scan mode with two columns, CP-Sil 8CB and HP-5MS, to evaluate organic compounds between n-C_6 and n-C_{35}. An Agilent 5890/5972 A GC/MS chromatograph was used for this process.

2.2. Soil sample

Control soil for ecotoxicity testing was collected from the surface layer of a field located near Madrid (Spain). Soil was air-dried and sieved (2 mm mesh). The main physicochemical characteristics of this soil were as follows: clay, 7.8%; silt, 18.8%; sand, 73.4%; pH, 7.3 and organic C, 1.09%. This soil fulfilled the conditions outlined by the OECD [9] for use as a control soil in microbial assays (sand >70%, pH 5.5–7.5 and organic carbon content ranging from 0.5 to 1.5%).

Table 1
Test program of treatability study.

Treatability test	Level of treatment	
	Laboratory	Pilot/Demonstration
Bioslurry	• Microcosms (Respirometry) • Stirred reactors	Air-lift pilot plant
Landfarming	• Microcosms (Respirometry) • Mesocosms (Columns + Trays)	Field (landfarming cell)

2.3. Treatability study

Two different treatment scales were studied: a laboratory scale and a pilot or demonstration scale (Table 1).

2.3.1. Bioslurry

For the bioslurry treatability tests, the optimal conditions were established at the laboratory scale by respirometry, first in microcosms and then in reactors. The experimental design covered different pulp densities and C:N:P ratios, i.e., natural (control), 100:10:1 and 100:10:0.5, in accord with previous work reported in the literature [10,11], and different treatment times.

Bioreactor simulation tests were performed by respirometry at different conditions in the microcosms, following the oxygen consumption and carbon dioxide production with a Micro-Oxymax respirometer (Columbus Instruments) equipped with an IR sensor for CO_2 and a paramagnetic sensor for O_2.

The respirometer houses 20 independent chambers that allow for the simultaneous measurement of oxygen consumption and carbon dioxide production in each chamber. The chambers are 250 mL ISO flasks with magnetic stirring and contain 20 g of sample for each study condition.

The different treatment conditions studied were mainly pulp density (from 1% to 20% w/v) and nutrient addition compared with the control assay (without nutrients). All tests were performed in triplicate. Tests lasted 31 days, and samples were harvested periodically for the analysis of hydrocarbon content to evaluate contaminant biodegradability and biodegradation rate.

Biotreatment simulated with bioreactors was conducted at the laboratory scale using 2 L mechanically stirred reactors at 20% (w/v) pulp density. The addition of nutrients and treatment time were tested. Based on the results obtained at the laboratory scale, the optimal conditions were applied at the pilot scale in a BiOEIMCO air-lift plant equipped with three 60 L reactors.

2.3.2. Landfarming

The optimal landfarming conditions were established by treatability tests in microcosms monitored by respirometry and were applied to mesocosms in packed columns and trays. The experimental design covered different C:N:P ratios and humidities with respect to the water holding capacity (WHC). The microcosm assays were performed as described above with the following differences. The humidity of the samples was adjusted to 60%, 70% or 80% of WHC, different C:N:P ratios were tested by nutrient addition, and the duration of the tests was 61 days.

Due to the fine texture of the initial sample, the material was amended with barley straw to facilitate its aeration and handling. Amended assays (with nutrients) were compared with the control assay (without nutrients). All samples were turned over weekly to facilitate aeration. Samples were harvested periodically, and the hydrocarbon content was analysed.

Landfarming mesocosms assays were performed at the optimal conditions obtained at the microcosm scale, using a two-step treatment simulation at field temperature. The first step was performed in packed columns with 4 kg of sludge, monitored daily and