



## Petroleum sludge bioremediation and its toxicity removal by landfill in gunder semi-arid conditions

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### ABSTRACT

In this investigation, petroleum sludge landfilling was carried out in order to assess the biodegradation degree and the final product quality. The microbial analysis showed a good microorganism proliferation which reinforces the biodegradation process. The total mesophilic and thermophilic microflora evaluated symmetrically as they increased at the intermediate stage and decreased at the final. The C/N and  $\text{NH}_4^+/\text{NO}_3^-$  ratios decreased while the polymerization degree increased at the end of the landfilling process. The total polyphenols and total petroleum C<sub>6</sub> to C<sub>22</sub> hydrocarbons were removed by 71.6% and 73% respectively, and that affected the reduction of the phytotoxicity in a positive way. All these changes are in agreement with the efficiency of the bio-transformation process and showed that petroleum sludge and filling reduced the toxic organic compounds and led to a stable final product.

### 1. Introduction

Petroleum refineries fractionate the crude oil in general by thermal cracking, to obtain liquefied petroleum gas, kerosene, diesel oil, naphtha, and other residual fuel oil (World Bank Group, 1998). This kind of industry is not environment friendly, it is responsible for the emission of toxic compounds, such as benzene, toluene, and xylene in the magnitude of 2.5 g released per ton of crude petroleum processed, and other volatile organic compounds ranging from 0.5 to 6 kg/t of crude oil (World Bank Group, 1998).

This industry sector uses a large quantity of water during the refinery process. After intensive usage, water becomes polluted by the contact of different petroleum products. As wastewater must be treated before its release into the surrounding area, a significant amount of sludge is generated in petroleum refineries. Sludge production in China is estimated to 3 million tons/year (Wang et al., 2012). In general, petroleum refinery can generate a huge amount of petroleum sludge estimated between 3 and 5 kg per ton of crude oil processed (Van Oudenhoven et al., 1995).

Petroleum sludge is considered as a hazardous waste, because the presence of toxic compounds, heavy metals, solids, water-in-oil emulsion, and other recalcitrant residues (Khalil et al., 2018). It is characterized by a total petroleum hydrocarbon rate in the range of 5–86.2% (Tahhan et al., 2011; Liu et al., 2012). As a general classification, Reddy et al. (2011) categorized the petroleum-derived hydrocarbons into aliphatic, aromatic, nitrogen sulfur-oxygen and asphaltenes. Petroleum sludge can be generally characterized by 40–52% alkanes, 28–31% aromatics, 8–10% asphaltenes and 7–22.4% resins (Hu et al., 2013; Lin et al., 2018).

Given the high concentration of toxic substances in petroleum sludge and referring to the literature, landfilling cannot be considered as a sustainable approach to sludge management. As a disadvantage, in situ landfilling is a temporary solution (contaminants remain in the environment) and permanent monitoring is necessary. Also, ecological agriculture strongly prohibits the use of untreated sludge on lands, unless heavy metals and organic compounds are within tolerated limits. In other words, technical approaches must be employed to reduce the sludge volume, or recovering the fuel from the sludge and disposing the

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unrecoverable residues from petroleum refinery (Pinheiro and Holanda, 2009; da Silva et al., 2012).

Our study focused on disposal approach, especially bioremediation by landfilling. Petroleum sludge landfilling is not recognized as a clean method for treatment nor valorization. The purpose of this study is primarily the evaluation of quality and maturity of the final product. This will provide information on the organic matter biotransformation and its stability. The objective is also the biodegradation assessment of some pollutants such as hydrocarbons and polyphenols. The content of these compounds at the end of the process is necessary to determine the landfilling efficiency on toxicity reduction.

## 2. Material and methods

### 2.1. Sludge landfilling and sampling

A typical wastewater treatment plant in petroleum refinery generates two types of sludge. The first one is produced from the flocculation-flotation process and the second from the biological treatment. Our study is concerned by the biological type which is stored in an internal landfill. The petroleum refinery is located in Mohammedia (Morocco) which is characterized by an annual temperature average of 18 °C, 400 mm of rainfall and 80% of humidity.

Three stages were monitored:  $T_0$  as the raw matter and sludge stored for  $T_i$  (4 months) and  $T_f$  (14 months). The sampling was carried out by taking a dozen of samples at several points of each windrow (width, length, surface and depth), these samples are carefully mixed to make a homogeneous sample. Then samples of 1 kg were obtained for each stage ( $T_0$ ,  $T_i$  and  $T_f$ ) by quartering method. The three samples of 1 kg each were kept at  $-20$  °C until analysis. Three replicates were made for each analysis.

### 2.2. Pollutant extraction

Extraction of total polyphenols was carried out by a hydroethanolic mixture with different ethanol proportions (0%, 25% and 50%) according to Macheix et al. (1990). 1 g of fresh sample was introduced in stirring tube with 40 mL of solvent mixture and agitated for 2 h. Purification of the extracts was made after filtration by ethyl acetate.

For the extraction of total petroleum hydrocarbons (TPH), 4 g of fresh samples were mixed with 4 g of  $MgSO_4$  until the obtention of a dry matter. The mixture is then putted in a cellulose cartridge for extraction by Soxhlet apparatus for 12 h with 120 mL of dichloromethane and diethyl ether (1:2 v/v). By the end of 12 h, all the TPH was extracted as the solvent was clearly colorless in the Soxhlet. After the extraction, the solvent was evaporated using a rotary evaporator. Purification of the extracts was performed by adding a silica gel and  $Na_2SO_4$ . The extract is weighed to determine the amount of TPH.

### 2.3. Analytical methods

#### 2.3.1. Physico-chemical analyses

The pH was measured on an aqueous extract of each sample at room temperature (1 g/10 mL of distilled water) as stipulated in the French standard procedure AFNOR NF T90-008. Organic matter (OM) was calculated after calcination in a muffle furnace at 600 °C for 6 h. Total organic carbon (TOC) was determined by titration after a reaction in a potassium dichromate solution (CEAEQ, 2012). Total Kjeldahl nitrogen (TKN) was assayed in 0.5 g samples by using the classical Kjeldahl procedure, by steam distillation according to standard AFNOR (1975). Ammonium ion content was assayed by alkaline distillation and nitrates after reduction by Dewarda alloying. All analyses were carried out in triplicates.

#### 2.3.2. Determination of total polyphenols

Analysis of total polyphenols concentration was carried out by the

Folin-Ciocalteu (FC) reagent, using the method of Singleton (1999). 1 mL of the extract and 9 mL of distilled water is added to 0.5 mL of the FC. After 5 min, 0.5 mL of sodium carbonate (20%) is added and the solution is placed in darkness for 2 h (Bonnaillie et al., 2012). The absorbance was measured in quartz bowl (volume 4 mL, optical distance 1 cm) by a spectrophotometer at 765 nm, and total polyphenols content was calculated by the regression equation established by the caffeic acid.

#### 2.3.3. Total petroleum hydrocarbon analysis

**2.3.3.1. Gas chromatography analysis (GC/FID).** The gas chromatography analysis was performed with a Hewlett-Packard gas chromatograph, equipped with a capillary Rtx-5 column (30 m × 0.32 mm ID × 0.25 μm). The carrier gas was the helium used at 2 mL/min flow rate. The oven temperature program was: 40 °C held 3 min, then increased to 300 °C at 15 °C/min. The TPH extracts were injected in splitless mode and the injector was set to 250 °C (Cortes et al., 2012). The GC/FID is performed as qualitative analysis, as performed by Rojas-Avelizapa et al. (2006) and Suganthi et al. (2018). The identification of the TPH was done by the RESTEK chromatogram library. The removal rate was determined using the formula:

$$\text{Removal rate(\%)} = 100 \times (A_i - A_f) / A_i \quad (1)$$

$A_i$  is the pic area of each hydrocarbon at the initial stage and  $A_f$  is the pic area of each hydrocarbon at the final stage.

**2.3.3.2. Infrared spectroscopy analysis.** The total petroleum hydrocarbon extracts were analyzed in the wavenumber range between 3000 and 2900  $cm^{-1}$  (Schwab et al., 1999; Brassington et al., 2010), with an infrared spectrophotometer VERTEX 70. The removal rate was determined using the formula:

$$\text{Removal rate(\%)} = 100 \times (Abs_i - Abs_f) / Abs_i \quad (2)$$

$Abs_i$  is the absorbance in the initial stage and  $Abs_f$  is the absorbance in the final stage.

#### 2.3.4. Microflora analysis

The microflora analysis was carried out by using sludge extract agar prepared according to El Fels et al. (2014a). After filtration, 15 g of agar was added and the filtrate was sterilized at 120 °C for 15 min. Samples from different sludge stages were mixed, suspended in physiological water (10 g in 100 mL) and homogenized by vortexing. A serial dilution up to  $10^{-9}$  was made. Cultivable microflora was enumerated by plating and spreading 0.1 mL of aqueous samples from  $10^{-4}$  to  $10^{-9}$  on the sludge extract agar, and the petri dishes were incubated at 28 °C and 45 °C. Identification of bacteria, actinobacteria and fungi was performed by comparison of morphological aspect of each specie and confirmation by a binocular microscope.

### 2.4. Phytotoxicity test

20 seeds of cress and lettuce were placed on a filter paper in petri dishes, then watered with 5 mL of water-soluble extracts of the different landfilling stages and the petri dishes were placed in darkness at 25 °C. The water-soluble fraction was made from 10 g of fresh sample in 100 mL of distilled water. Three replicates were made. The germination index (GI) was calculated using the following equation (Zucconi et al., 1981):

$$GI(\%) = (NSe \times LRe) / (NSw \times LRw) \times 100 \quad (3)$$

$NSe$ ,  $NSw$  are number of seeds germinated in water-soluble extracts and distilled water, respectively (after 24 h); and  $LRe$ ,  $LRw$  are the length of roots in soluble extracts and distilled water, respectively (after 72 h).

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