The conception of an experimental model for ex situ bioremediation of soils contaminated with petroleum hydrocarbons
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Abstract. This paper presents how to design an experimental model that can be used in the research and optimization of the "biopile" method for ex situ bioremediation of soils contaminated with petroleum hydrocarbons. Soils are collected from the site of a former depot of petroleum products. The experimental model proposed by the authors of this paper includes several "biopile" cells that are reduced in size. Laboratory experiments will be made using the proposed model at constant parameters (pH, temperature, nutrients ratio) and variable parameters (duration and intensity of soil aeration, humidity, the amount of microorganisms). Based on the results obtained from the experiments and their interpretation, optimal parameters of ex situ bioremediation technology will be established.

Key Words: soil pollution, biopile, bio-decontamination, mineral oil, pollutant reduction.

Introduction. Remediation using the Biopile method (Figure 1) is a technology that derives from both land farming and the composting method. After excavation, polluted soil is stored in piles on an impermeable surface, with a small slope and then covered with a membrane to retain gases inside the pile (Vidali 2001).

The installation is provided with perforated pipes, placed at the base of the pile; it can operate for introducing fresh air from outside into the pile to perform its aeration or to extract the air loaded with volatile substances from the pile (Pasca & Pasca 2002).

Figure 1. Schema biopile (Meuser 2013).
It is a short term technology applicable for weeks to a few months. The technology is suitable for destruction of mono-halogenated VOC and of hydrocarbons from fuels. It can be applied also for treatment of certain halogenated VOC, SVOC, pesticides, with variable efficiency. The method is applied for soils with high contamination of volatile substances (Doboș 2011).

Ex situ biopiles were used successfully for bioremediation of soils polluted in combination with the bio-stimulation method (heating, nutritive substances and aeration) and the bio-augmentation method (Aislabie et al 2006).

Decontamination of soils polluted with polycyclic aromatic hydrocarbons (PAH) using the biopile method presents high yields of extraction of 80-90%, in a relatively short time of treatment (9-12 months) (Micle & Neag 2009) and soils polluted with mineral oils were treated in a proportion of 70% in 7 months since the start of the experiment (Lecomte 1995).

Reduction of hydrocarbons in the soil was conducted on biopile piles. The biopile piles of 0.6 m³ were bio-stimulated with nutritive substances and aerated (66-75%) (Seklemova et al 2001) and the pile of 30 m³ which was not aerated obtained lower yields of 50% (Chaineau et al 2003).

Iturbe et al (2004), built a biopile system of 100 m³ to study the biodegradation ability of PAH, using indigenous microflora. After 2 months of treatment they obtained a yield of 85% (Iturbe et al 2004). A remediation efficiency of 80% was obtained also on a 27 m³ biopile, thus proving the capacity of the biopile method in bioremediation of soils polluted with TPH (Iturbe et al 2007).

Gogoi et al (2003) worked with soil polluted with TPH in cells of 500 kg with various treatments. Soil was amended with nutrients and inoculated with a microbial consortium isolated from soils contaminated with hydrocarbons. The system was aerated one hour a day at a rate of 100 m³/h. At the end of the 365 days of operation, remediation in the cells was 75% with a degradation rate of 90 mg/kg/day (Gogoi et al 2003).

Pollutant reduction (TPH) was extremely efficient, registering a reduction of pollutant from 2,870 mg kg⁻¹ down to 616 mg kg⁻¹ (40 days), 457 mg kg⁻¹ (125 days) and 357 mg kg⁻¹ (221 days) using biopile (Heely et al 1994). Case studies in the case of an intensively aerated biopile of 3.20 m, led to the decrease of TPH of 8200 mg kg⁻¹ to 4200 mg kg⁻¹ (200 days) and down to 3,900 mg kg⁻¹ (300 days), at a depth of 1-2 m and from 6,600 mg kg⁻¹ to 2,300 mg kg⁻¹ (200 days) and down to 1,600 mg kg⁻¹ (300 days) at a depth of 0-1 m (values estimated approximately) (Koning et al 2001).

After the critical analysis of the biopile method of depollution of soil polluted with hydrocarbons, it was concluded that the biopile method is the most adequate because yields of over 95% can be obtained.

The concept of the experimental model derives from a few imposed requirements: the realization is made by a simple small sized construction; to allow, in laboratory conditions, biological treatment of soils polluted with petroleum hydrocarbons, to allow the variation of physic-chemical parameters of soil (duration and intensity of soil aeration, humidity, the amount of microorganisms).

**Description of the experimental model.** Based on the study on the critical analysis of the treatment method of soils polluted with hydrocarbons and of the innovative solution, the experimental model was designed through which the treatment of soil is conducted in laboratory conditions. An experimental model was made which is based on the biopile treatment method, it is the most effective in terms of the degree of depollution (over 95%), for a large range of contaminants and it can be designed to be effective for any association of the site and petroleum products.

The conceptual diagram of the proposed experimental model (Figure 2) seeks to achieve two main objectives:
- treatment of soils polluted with petroleum hydrocarbons;
- variation of the main physic-chemical parameters of the soil (duration and intensity of soil aeration, humidity, the amount of microorganisms).
Experiments on pollutant extraction (TPH) from polluted soil are conducted in 8 cells biopile. Each cell has different parameters (Table 1) which are studied to emphasize the optimum conditions to obtain a depollution with a yield as high as possible.

Table 1

<table>
<thead>
<tr>
<th>Biopile cells</th>
<th>pH</th>
<th>TPH (mg/kg)</th>
<th>The amount of nutrients C/N/P</th>
<th>Temperature (°C)</th>
<th>The amount of microorganisms (mL/100 kg)</th>
<th>Humidity (%)</th>
<th>Soil aeration (h/day)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell 1</td>
<td></td>
<td>100–500</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Cell 2</td>
<td></td>
<td>100–500</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Cell 3</td>
<td></td>
<td>100–500</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Cell 4</td>
<td>7</td>
<td>7,000</td>
<td>100:10:1</td>
<td>20–25</td>
<td></td>
<td></td>
<td>4</td>
</tr>
<tr>
<td>Cell 5</td>
<td></td>
<td>100–500</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Cell 6</td>
<td></td>
<td>500–1,000</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Cell 7</td>
<td></td>
<td>500–1,000</td>
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<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Cell 8</td>
<td></td>
<td>500–1,000</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1</td>
</tr>
</tbody>
</table>

Soil samples are brought into the laboratory and prepared to carry out the experiments. They are mixed and sieved through a sieve with the hole diameter of 2 mm. Soil samples thus prepared are placed in a plastic tank with a lid (L x l x h = 50 x 35 x 25 mm), placed on a slightly sloping surface. At the bottom of the biopile cells it is put a layer of permeable gravel with a thickness of 5 cm. In this layer of gravel are introduced two perforated PVC pipes, with a diameter of 2 cm, which carry out soil aeration.
The soil is improved by administering necessary nutrients for the proper development of microorganisms. The amount of nutrients is dosed adequately at the beginning of the experiment, with the optimum report C/N/P 100:10:1.

Hydrocarbon extraction from the polluted soil is achieved with microorganisms selected from the soil under study (indigenous microflora). After the soil was improved with microorganisms and nutrients, it is mixed to achieve a homogenization of the polluted soil which is subjected to treatment/decontamination.

Aeration is achieved with a Fini AMICO compressor. Pipes are arranged horizontally in the biopile pile, at a distance of 17 cm from one another and at 9 cm from the edges of the tank. The debit of air introduced is 10 m$^3$/min. The quantity of air introduced varies depending on the cell in which it is applied (1 h/day, 4 h/day). This air flow will be divided in 10 minutes long aeration phases. The oxygen content in the soil will be measured and monitored with the WTW Multiline ids – 3430 multiparameter.

Humidity is monitored throughout the experiment using the humidity sensor, achieving a humidity of 40%, respectively 60%, depending on the cell where the experiment is carried out.

During the entire period of the experiment, the laboratory temperature will remain constant (20–25°C) and the pH of the soil will remain constant (7).

Weekly, of the 8 biopile cells are taken soil samples (10 g) and leachate (10 mL) which is analyzed to observe the variation in the quantity of hydrocarbons during the experiment (4 weeks). At the end of the testing period, graphs will be drawn to emphasize the optimum parameters of the biopile treatment method to obtain yields as high as possible.

In designing the experimental model for laboratory conditions, a simple construction was chosen, reduced in size and with low cost of implementation.

**Conclusions.** Biological soil treatment is a method of interest being less expensive than other methods; plus, it protects the organic matter in the soil. The achievement of the proposed experimental model will allow biologic treatment of soils polluted with petroleum hydrocarbons, in laboratory conditions. Also, the model will allow the variation of physic-chemical parameters of the treated soil. Based on the results of the experiments and their interpretation, will be established the optimum parameters of the ex-site bioremediation technology, that can be applied when using the biopile bioremediation method at a pilot scale.

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