

# Elimination of Hydrocarbon Contaminants from Synthetic Waste water by Soil Filter

Houria Messrouk<sup>(a)</sup>, Mahfoud Hadj Mohammed<sup>(a)</sup>, Youcef Touil<sup>(a)</sup>, Abdelatif Amrane<sup>(b)</sup>

*(a) Laboratoire de biogéochimie des milieux désertique université Kasdi Merbah Ouargla Bp 511 30000 Ouargla Algeria*

*(b) Ecole Nationale Supérieure de chimie de Rennes, Université de Rennes 1, CNRS, UMR 6226, 11 allée de Beaulieu, CS 50837, 35708 Rennes Cedex 7, France*

**Abstract**— One of the major environmental problems today is hydrocarbon contamination resulting from the activities related to the petrochemical industry. Accidental releases of petroleum products in the environment are of particular concern. Hydrocarbon components have been known to belong to the family of carcinogens and neurotoxic organic pollutants. The present work aims to implement a system to reduce or even eliminate hydrocarbons; it consists in columns composed of a fixed bed of sand dunes of N'Goussa from the Region of Ouargla (Algerian South). Columns gave excellent results regarding the removal efficiency of hydrocarbons, since a yield of up to 99.65 % was obtained.

**Index Terms**— Hydrocarbons, synthetic waters, sand dunes, filtration.

## 1 INTRODUCTION

Petroleum-based products are the major source of energy for industry and daily life. The world production of crude oil is more than three billion tons per year. Petroleum oils can cause environmental pollution during various stages of production, transportation, refining and use. Oil pollution from industrial sources and other activities in harbour areas pose great hazard to terrestrial and marine ecosystems. Petroleum hydrocarbons pollutions, ranging from soil, ground water to marine environment, become an inevitable problem in the modern life [1, 2].

At present, a number of different technologies exist for spill removal, such as chemical precipitation, adsorption, and solvent extraction. However these methods have several disadvantages, such as incomplete oil removal, expensive equipment and monitoring system requirements, high reagent or energy requirements and generation of toxic sludge or other waste products that require disposal [3, 4].

In addition, more effective methods of treatment based on the use of sand dune, are at the origin of the biosorption technique that improves the ability to remove pollutants. The dynamic treatment comprises circulating the effluent through a granular solid on which there is a biofilm development consisting of microbial cells and exopolymers [3- 7].

The degradation of hydrocarbons by an "adsorption / biodegradation" technique which is known as "biosorption" continuous system is examined in this work. The study of the effectiveness of our system to treat wastewater containing hydrocarbons was performed on a mixture of compounds and at different concentrations. Water discharges from washing stations were simulated. This choice results from a previous work which showed high loads of organic matter mainly from some washing stations in the city of Ouargla, as shown in terms of COD, BOD<sub>5</sub>, TSS and phenol index. And hence researches are required on the least expensive ways to reduce the pollution that can cause degradation of fauna and flora [8, 9].

## 2 MATERIALS AND METHODS

### 2.1 Studied systems

#### 2.1.1 Polluted water

Mineral oils and fats derived from the distillation of petroleum are divided into two categories: aliphatic hydrocarbons and aromatic hydrocarbons. Oil is formed by hydrocarbon containing 17 to 22 carbon atoms [10]. In the present work we prepared synthetic water composed of a mixture of hydrocarbons and oils at volume percentages as summarized in Table (1),

TABLE 1

THE COMPOSITION OF THE SYNTHETIC WASTEWATER PREPARED [7].

Pollutant	Volume percentage (%)
20W50	25
15W40	25
40DIEZEL	15
5W40	10
10W40	10
90	9
GAZOIL	6

with the characteristics summarized in Table (2)

The choice of compounds and their concentrations was based on their frequency of use, and related to the concentrations which can be found in wastewater from several car washes in

the city of Ouargla (according to a survey that was we conducted). In addition, the research performed on the destruction of organic pollutants in aqueous phase concerns mainly

of Ouargla (Algeria) and are summarized in Table (3).

TABLE 2

THE CHARACTERISTICS OF SYNTHETIC WATER PREPARED

Parameter	Value
T(° C)	30
pH	6.7
CE (ms/cm)	3.8
HC (mg/l)	1000
COD (mg/l)	1100
BOD5 (mg/l)	76
TSS (mg/l)	213

aromatic compounds [11, 12].

### 2.1.2 Filtering material

This study was conducted with sand dune from the region of Ouargla (N 'Goussa). These sands have been studied previously and showed excellent efficacy in treating domestic wastewater. They have a very uniform chemical composition of high silica predominantly with a homogeneous particle size suitable for use as a filter bed [13, 14]. The physico-chemical characteristics of the sand were determined in the biogeochemistry of desert environments laboratory from the University

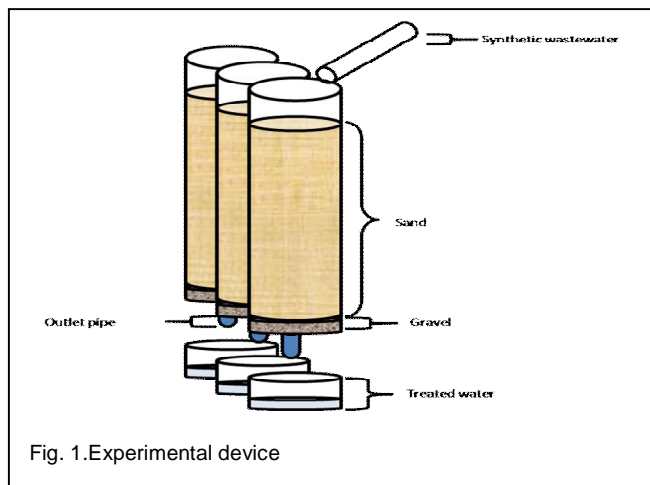
TABLE 3

THE CHARACTERISTICS OF THE SAND USED

Parameter	Value
D <sub>10</sub> (mm)	0.12
D <sub>60</sub> (mm)	0.19
D <sub>30</sub> (mm)	0.28
Coefficient of uniformity Cu	2.33
Coefficient of classification Cc	1.07
Real density (ρ true) (Kg/m <sup>3</sup> )	2960
Apparent density (ρ <sub>app</sub> ) (Kg/m <sup>3</sup> )	1785
Porosity (η) (%)	39.70
MO (%)	0.086
pH	8.46
Salt (%)	1.91
CaCo3 (%)	0.240
Conductivity CE (ms/cm)	3.3

## 2.2 Experimental device and procedure

To study the effectiveness of treatment on a fixed bed of sand dune to remove hydrocarbons from industrial wastewater, we developed the device shown in Figure (1).



We used 03 PVC columns (76 mm diameter x 700 mm height) tucked by the sand dune at a height of 600 mm. The filter material supported on a 10 mm layer of gravel was inserted to prevent the loss of particles; each filter is equipped with a drainage device, a pipe of 15 mm diameter mounted at the base of the column. These drivers must be secured in a perfect vertical position and stable to avoid any form of vibration and also to promote gravity flow. The synthetic wastewater feed having a concentration of 1g / L of oils was performed manually each hour with a flow rate of 40 ml / h for 13 hours continuously; the experiment was conducted at room temperature. Samples were collected through the outlet tube, at regular intervals of 7 days to measure the parameters which are summarized in Table (4).

## 2.3 Water analysis

To evaluate the efficacy of our columns to remove hydrocarbons from synthetic wastewater, in addition to the concentration of total hydrocarbons, the most significant parameters were analyzed, namely the chemical oxygen demand COD, the biological oxygen demand BOD<sub>5</sub> and the suspended material as indicated in Table (4)

### 3 RESULTS AND DISCUSSION

The evolution in the experimental device was followed for 10 weeks and the analysis of the treated water (output column) was started after one week of operation of the device. During this period we conducted 10 measurement\_campaigns (parameters shown in table (4)).

The removal efficiency yield (%) of filter materials in terms of oil, COD, BOD<sub>5</sub> and TSS was calculated by the following formula:

$$\text{Yield (\%)} = (X_{\text{input}} - X_{\text{output}} / X_{\text{input}}) \times 100$$

The results obtained yields, their average and standard deviations are shown in Figures (2, 3, 4, 5)

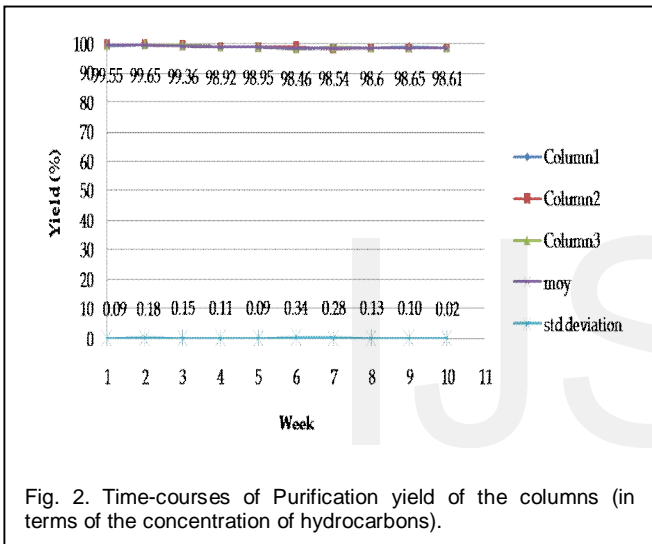
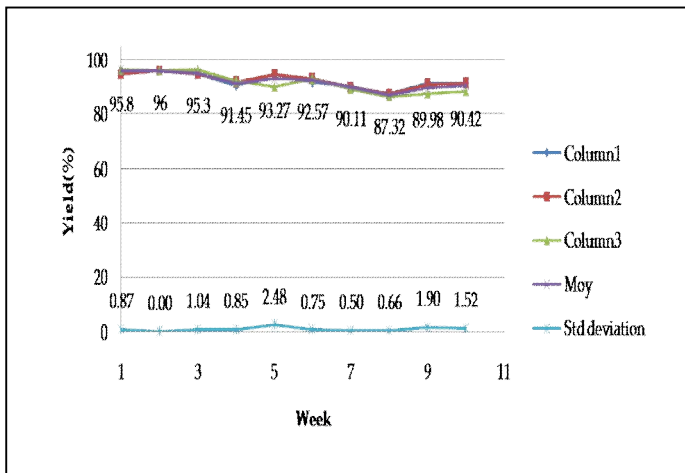


Fig. 2. Time-courses of Purification yield of the columns (in terms of the concentration of hydrocarbons).

It can be noted that the three columns of sand were efficient to remove hydrocarbons from the synthetic contaminated water, with yields between 98.5 and 99.6%; the latter which was the highest value recorded for the full period of the experiment. It can also be noted a very low decrease with time; however the yields remained very high until the tenth week (time of running).



The chemical oxygen demand corresponds to the O<sub>2</sub> content consumed by the oxidizable materials (reducing) under defined conditions. It is another way for determination of the hydrocarbons removal; however less accurate than their direct measurement. Similar observations as those drawn from the concentrations of hydrocarbons can be deduced from the COD monitoring. Indeed, the best yields were recorded during the first three weeks (96.0%) and a very low decrease was then observed, with however high removal yields until the last week of running, with 90.4% removal yield.

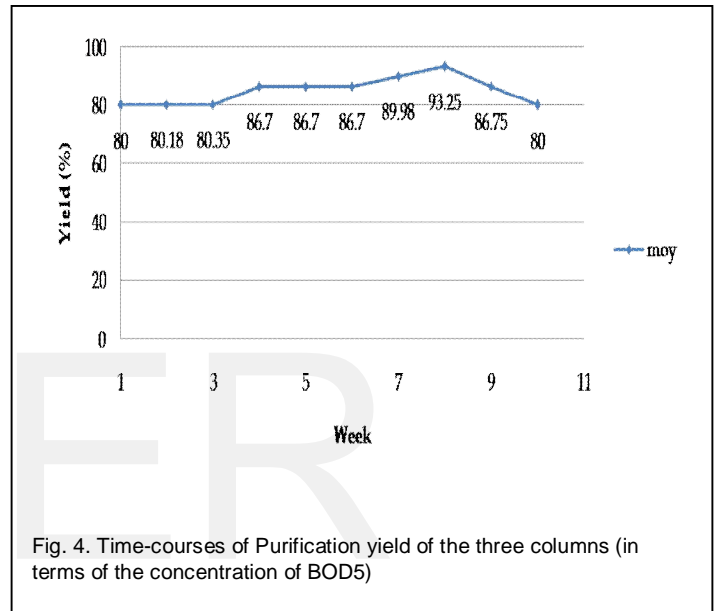


Fig. 4. Time-courses of Purification yield of the three columns (in terms of the concentration of BOD<sub>5</sub>)

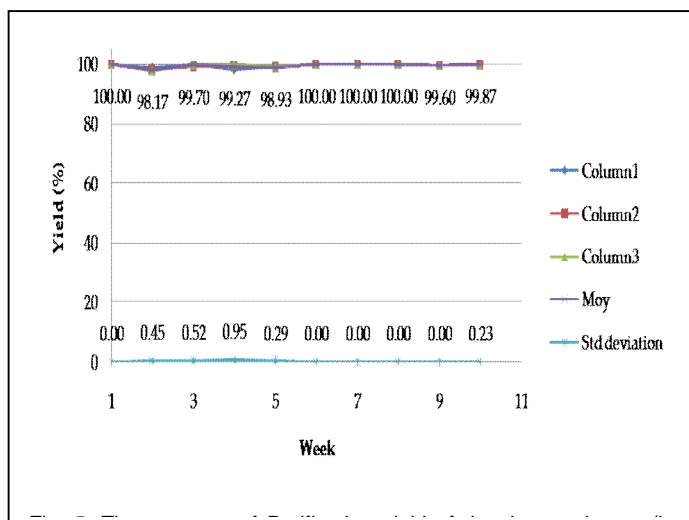
Biological oxygen demand corresponds to the amount of oxygen (mg/l) biologically consumed in five days at 20 ° C in the dark [16]. This parameter was measured for the mixture of treated water of the three columns. Registered yields were excellent and up to 93.2%. It can also be noticed a slight peak observed after eight weeks of experiment and then the BOD<sub>5</sub>

TABLE 4

THE STUDIED PARAMETERS AND THE ANALYSIS METHODS

Parameters	Methods of analysis	Units	Sources
HC	Gravimetric method	mg/L	MA.415-HGT2.0[10]
Suspended Material	Filtration on filter paper	mg/L	Norme EN872:1996[15]
COD	Method by oxidation with KMnO <sub>4</sub>	mg/L	Norme NFT90-101[11,14,16]
BOD <sub>5</sub>	Instrumental method	mg/L	Analyse de l'eau aspect réglementaires et techniques F.Rejsek, 2002[15]

values decreased to the initial removal yield (80%).



To appreciate the quality of water, it is always necessary to quantify its load of particulate matter (TSS); the removal yield of suspended solids yields (TSS) is illustrated in Figure (5), illustrating high efficiency, since the yield varied between 98.2 and 100%.

It can also be seen from the figures (2 to 5) that the results given by the three columns were almost identical, with low standard deviations showing the accuracy of the measurements.

Globally, the results obtained proved the effectiveness of sand columns to treat water contaminated by oils and hydrocarbons during a period of 10 weeks with very good performances, close to 100%. We can interpret this by two phenomena (adsorption and biodegradation), adsorption which characterizes porous materials such as sand, the oils can be adsorbed not because they are attracted to the solid surface but because the solution tends to reject it; this is what typically occurs for a hydrophobic organic compound in the presence of an adsorbent whose surface is hydrophobic [18]. By this phenomenon, oils are adsorbed on the surface of the sand, accumulate and form with time a surface layer. Besides, there is also biofilm formation on the layer, which takes about one week according to the related literature [14, 19], and which is mainly constituted of specific bacteria such as *Bacillus subtilis* and *Pseudomonas aeruginosa* [8, 20, 21].

#### 4 CONCLUSION

The amount of pollutants introduced into the environment through sanitation is undoubtedly endangering the ecological balance, if waste water from different sources is not treated prior to disposal or reuse. Among these pollutants, we directed towards hydrocarbons. Indeed, negative impacts are observed on the receiving environments such as deoxygenation of the environment due to the benefits of this type of pollutant [22]. It is from the perspective of sound management of water resources that fits our work on the study of the efficiency of the filtration, while contributing to upgrading local materials (sand dunes and others) and the preservation of the

receiving environment from the dangers of pollution cited above.

The experimental feature that we have developed is constituted by 3 columns containing a fixed bed of sand dunes (sand of N' GOUSSA, in Ouargla's region), gave satisfactory results. These have shown high efficiency of removal of hydrocarbons, since a yield up to 99.65 % was obtained. Other yields being 96 % in COD, BOD<sub>5</sub> 93 % and 100 % in TSS confirm this efficiency. Small deviations calculated between the values given by the three tested columns have confirmed these promising results.

On the other hand, the study of other models in local materials is being tested in our laboratory to refine the treatment model that could be used in the lute tale effluents, especially industrial pollution.

#### REFERENCES

- [1] O. Bordjiba, F. Bekhouche, A. Hassaine, R.Djenidi " Impact de la Pollution Par Les Hydrocarbures Sur la Qualité des Eaux Usées Dans la Région de Skikda (Nord-Est Algérie), " *European Journal of Scientific Research*, vol. 26, no. 1, pp. 87-97, 2009.
- [2] Y.Li, Y. Zhao ,S. Peng ,Q. Zhou ,L. Ma " Temporal and spatial trends of total petroleum hydrocarbons in the seawater of Bohai Bay,China from 1996 to 2005, " *Marine Pollution Bulletin*, vol. 60, pp. 238-243., 2010.
- [3] D. Nilanjana, C. Preethy "Microbial Degradation of Petroleum Hydrocarbon Contaminants, " *SAGE-Hindawi Access to Research Biotechnology Research International*, vol. 21, no. 1, pp. 1-13., 2011.
- [4] A.AbdulRahim ,M.A. Gaber " Biological Treatment of Hydrocarbon Contaminants: Petroleum hydrocarbon uptake by *Pseudomonas Alkanolytica*, " *J.KAU:Eng. Sci*, vol. 21, no. 1, pp. 39-53., 2010.
- [5] N.Janzen , S. Banzhaf , T.Scheytt , K.Bester "Vertical flow soil filter for the elimination of micro pollutants from storm and waste water " *Chemosphere*, vol. 77, no. 10, pp. 1358-1365., 2009.
- [6] Y.Touil, Y.Gherairi, M.Nefsi, M.HadjMahammed, R.Issaadi, A.Amrane "Etude comparative de l'efficacité de la filtration biologique sur un sable éolien et un sable alluvionnaire, " *Récent Progrès en Génie des procédés*, no. 101., 2011.
- [7] H. Messrouk, M. Hadj mahammed ,Y.Touil ,A.Amrane "Physico-chemical Characterization of Industrial Effluents From The Town of Ouargla (South East Algeria), " *Energy Procedia*, vol. 50, no. special, pp. 255-262., 2014.
- [8] X.Xia, H. Yu, Z.Yang, G.Huang "Biodegradation of polycyclic aromatic hydrocarbons in the natural waters of the Yellow River: Effects of high sediment content on biodegradation, " *Chemosphere*, vol. 65, no. 3, pp. 457-466., 2006.
- [9] H. Messrouk, M. Hadj mahammed, Y. Touil "Analyse quantitative des composés phénoliques dans les effluents industriels de la ville de Ouargla, " *Annales des Sciences et Technologie*, vol. 4, no. 2, pp. 95-101., 2012.
- [10] Centre d'expertise en analyse environnementale du Québec " Détermination des huiles et des graisses dans les eaux : méthode gravimétrique, MA. 415-HGT 2.0, " *Ministère du Développement durable, de l'Environnement et des Parcs du Québec*, vol.17., 2011.
- [11] C.Manole-Creanca " Procédé AD-OX d'élimination de polluants organiques non biodégradables (par adsorption puis oxydation cataly-

- tique), " PhD dissertation, L'institut National Polytechnique ., Toulouse Univ., France ., 2007.
- [12] C.Ayral " Elimination de polluants aromatiques par oxydation catalytique sur charbon actif), " PhD dissertation, L'institut National Polytechnique ., Toulouse Univ., France ., 2009.
- [13] Y.Touil, Y.Gherairi ,R. Issadi ,A. Amrane A " Biological Filtration on Sand of Dunes-Filters Fouling, " *Energy Procedia* , vol. 50, no. special, pp. 471-478., 2014.
- [14] Y.Gherairi ,A.Amrane ,Y. Touil ,M. Hadj mahammed ,F. Gherairi,L.Baemer L " A Comparative Study of the Addition Effect of Activated Carbon Obtained from Date Stones on the Biological Filtration Efficiency using Sand Dune Bed, " *Energy Procedia* , vol. 36, no. special,pp. 1175-1183., 2013.
- [15] F.Rejsek "Analyse des eaux aspects réglementaires et techniques, " Edition Centre régional de documentation pédagogique d'aquitaine ., France ., 2002.
- [16] J. Rodier, *Analyse de l'eau*. 8e édition, Dunod, Paris, pp. 59-1044, 2005.
- [17] Y.El Guamri'l, D. Belghyti'l, K. El Kharrim, S. Raweh, I. Sylla'l, et M. Benyakhef " Etude physico-chimique des eaux usées brutes de l'abattoir municipal de Kénitra (Maroc) en vu de la mise en oeuvre d'un traitement convenable, " *Sud Sciences et Technologie*, no. 16, pp. 796-5419., 2008.
- [18] C.Morlay, I. Laidin, M.Chesneau, J.P.Joly " Charbons actifs et traitement des eaux, " *L'Act. Chim* , vol. 95, no. Special, pp. 295-296., 2006.
- [19] K.Allia ,N.Taher , L.Toumi ,Z.Salem Z " Biological Treatment Of Water Contaminated By Hydrocarbons In Three-phase Gaz- Liquid-Solid Fluidized Bed, " *Global NEST Journal*, vol. 8, no. 1, pp. 9-15., 2006.
- [20] R.Latha ,R. Kalaivani " Bacterial Degradation of Crude Oil by Gravimetric Analysis, " *Pelgia Research Library USA*, vol. 3, no. 5, pp. 2789-2795., 2012
- [21] M.Peyman M.R.Serge,J. Ronald " Encapsulation of Bacteria for Biodegradation of Gasoline Hydrocarbons methods in Biotechnology:Immobilisation of Enzymes and Cells, " Second Edition ,J.M Guisan©Humana Press Inc,Totowa, NJ.
- [22] J-C.Boeglin J-C " Analyse des eaux résiduaires. Pollution industrielle de l'eau caractérisation, classification, mesure," *Techniques de l'Ingénieur, traité Génie industriel, G 1 210*, pp. 1, 1997.