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المعالجة البيولوجية للنفايات الصناعية السائلة الخطرة في حفرة التبخر، في الوفرة جنوب دولة الكويت

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الخلاصـة

أجريت المعالجة البيولوجية لتلوث الترسبات الزيتية في موقع طريق الوفرة / ميناء عبدالله كيلو ١٤ في جنوب دولة الكويت. وأظهرت محتوى النفايات مستويات عالية من المعادن والمواد الكيميائية الأخرى. المستويات عالية من المواد العضوية تم تحديدها بواسطة تحليل مطلب الأكسجين الكيميائي ومطلب الأكسجين البيولوجي، وكذلك مجموعة الكربون العضوية، وكذلك إجمالي المواد الهيدروكربونية البترولية. إن الأساليب التي استخدمت في هذه الدراسة كانت عبارة عن حرث وتقليب التربة الملوثة بالإضافة إلى التنفيس البيولوجي، وزيادة المادة الحيوية (biosparging)، وتسميد التراكم الحيوي. وبعد سنة واحدة من الحرث والتقليب تمت زراعة الموقع حيث تبين بالمعالجة بأن كمية ثمانية معادن ثقيلة أنخفضت بنسبة 81%. وكما أظهرت مراقبة المركبات العضوية، ومطلب الاكسجين الكيميائي، ومطلب الأكسجين البيولوجي، وإجمالي المواد الهيدروكربونية البترولية، والمركبات العضوية أن هناك انخفاضاً إجمالياً بعد انقضاء سنة واحدة بنسبة 71.1%، 89.47%، 94.17%، 95.36% على التوالي. خلاصة البحث تبين بأن المعالجة البيولوجية كانت فعالة جدا، ومن الممكن معالجة النفايات السائلة الخطرة بهذه الطريقة الآمنة نسبياً.

Bioremediation for hazardous liquid industrial waste at evaporation pit, Wafra south of Kuwait.

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ABSTRACT

Bioremediation of oily sludge contamination was conducted at the Wafra/Mina Abdullah site (Kilo 14) in southern Kuwait. The waste contents originally showed high levels of trace metals and other toxic chemicals. High levels of organic parameters were indicated by analyses of chemical oxygen demand, biological oxygen demand, total organic carbon, and total petroleum hydrocarbons. Methods used in bioremediation included *in-situ* and *ex-situ* approaches such as: landfarming, bioventing, bioaugmentation, biosparging, and composting (biopiling). After one year of landfarming treatment, eight heavy metals showed overall reduction of 81%. Of the organic compounds monitored, chemical oxygen demand, biological oxygen demand, total petroleum hydrocarbons, and total organic carbon showed one-year reductions of 71.1%, 89.47%, 94.17%, and 95.36%, respectively. In summary, bioremediation was shown to be very cost effective method in this research, it is highly recommended to be implemented in heavy oily sludge area.

Keywords: Bioremediation; chemicals; heavy metals; landfarming; oily sludge.

INTRODUCTION

Kuwait is located in the Middle East bordered by Iraq from north and west; Saudi Arabia towards the south and bounded by Arabian Gulf towards the east. The geographical coordinates of Kuwait are 29’ 30” N latitude and 45’ 45” E longitude. Total area of Kuwait is 17, 8182km^2. The shore line is about 500 km including all the nine islands (Fig. 1). Kuwait is a desert country which is falling in the arid environment meteorological system, accordingly the rainfall is scarce and does not reach 400 mm in a year. Summer temperature ranges from 35 °c to 52 °c. Winter temperature ranges from 27 °c to -2 °c (for extreme cases). The State of Kuwait is a very flat country and shows very simple geological structures, since it is located between the Arabian Peninsula in the west and south-west and Zagross orogeny to the north and north-east. Wadi Al-Batin which is a former fluvial channel bounds Kuwait from the west.

Jal Az-Zor escarpment is a noticeable geomorphic terrain towards the north west of Kuwait Bay. Many geologists have written about the surface and sub-surface geology of Kuwait such as (Cox, 1932; Mitchel, 1957; Milton, 1967 and Al-Sarawi, 1995). The study area is located along the Alwafra/ Mina Abdulla road at Kilometer 14 south of Kuwait. The area is about 1 km² and four meters deep and it once received about 5000 m³ daily for five years of industrial hazardous liquid waste and untreated sewage in one of the largest unlined evaporation pits in the State of Kuwait. The main objectives of this study are to evaluate the level of contamination in the study area, bioremediate the soil contaminated with toxic materials and hydrocarbons, landfarming the contaminated site, produce compost from the contaminated soil, and create a national park for the public by planting trees.

The scope of work included developing, enhancing and augmenting bioremediation program work plans for contaminated soil by hazardous industrial oil and untreated sewage. The collected samples were geochemically analyzed to identify trace metals, total petroleum hydrocarbons, and total percentage of organic content before and after treatments. The aim of this study was to rehabilitate the soil contamination that has taken place in the past 5 years through the bioremediation and landfarming process. The study will concentrate on the removal of contaminants, reducing their toxicity and using the end product for composting.

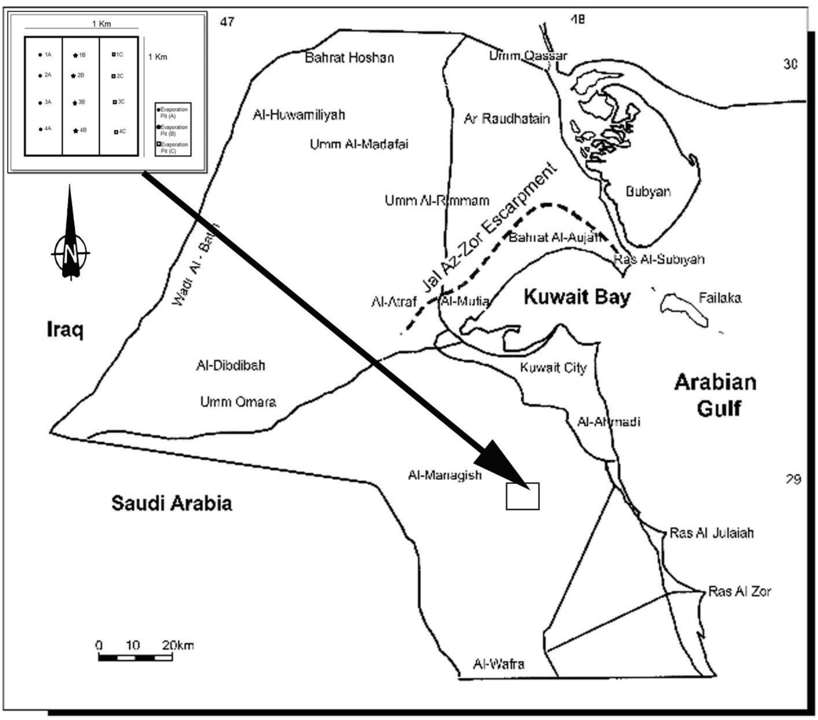

Fig. 1. Location map of Kuwait showing the study area

MATERIALS AND METHODS

The study area located in AlWafra, 60 Km south of Kuwait City (Fig. 1). It was established in 2009 to collect most of the untreated sewage and used oil. Three unlined evaporation pits about $1Km^2$ were constructed for collection of approximately $4500m^3$ daily mixture of untreated industrial and domestic wastes (Fig. 2). The site contains 20 billion gallons of industrial waste and untreated sewage and is about 4 meters deep.

Fig. 2. Accumulation of used oil and untreated sewage in the evaporation pits.

Sample site

A total of twelve (12) contaminated soil sludge samples were collected from the three pits in the study area to determine the physical-chemical parameters such as (temperature, pH, COD, BOD, salinity, dissolved oxygen, moisture content, and nutrient concentration). Moreover the determination of trace metals (TM), total petroleum hydrocarbons (TPH), and total organic carbons (TOC) was carried out.

Sample collection

A. *Sludge samples (before treatment)*

A total of twelve (12) sludge samples were collected from 3 evaporation pits (A, B, and C) (Fig. 4) in the field by aluminum boxes. For sludge samples, trace metals, TPH, COD, BOD, pH, Temperature, and TOC were determined. All collected samples were stored in aluminum boxes and were delivered the same day to the analytical laboratory for analysis.

B. *Soil samples (after treatment)*

A total of twelve (12) soil samples were collected from the treatment cell, four (4) samples each. They were analyzed after 120 days, 240 days, and one year. The treatment cell is a mixture of sludge contaminated soil, clean soil, shredded plants and bacteria (indigenous microbes). The samples were stored in plastic bags and transported to the laboratory for chemical analysis.

Analytical methods

A. *Trace metals (TM) analysis*

For the determination of metals, samples were subjected to microwave digestion using conc. nitric acid, filtered and the filtrate was used for metal analysis using ICP-OES (Varian Vista – MPX) and / or ICP-MS, (project No. GS 01/05). Inductively Coupled Plasma analyzer (ICP) uses emission spectroscopy to qualify elements in a sample. Atomic emission spectroscopy is a technique for detecting and measuring chemical elements in samples. The technique measures the intensity of light emitted by atoms of the elements of interest at a specific wavelength.

Approximately 0.3 g of sample was taken in a clean and dry container (microwave digestion Teflon tube). Concentrated nitric acid (approximately 10 ml) was added to the tube and heated in a microwave oven. The solution was then filtered in another container through 0.45 micron Whatman filter paper number 42 and the insoluble residues on the filter paper were rinsed with 10% nitric acid. The residue was then discarded. The filter paper blank and nitric acid blank were also prepared similarly. The filtrate was then quantitatively transferred into 50 ml volumetric flask and volume was made up to 50 ml by using 1% nitric acid. The method used is based on the U.S. EPA Method 200.7 (USEPA 2001).

B. *Total petroleum hydrocarbons (TPH) analysis*

EPA Method 418.1 (USEPA 1978), infrared spectrophotometry has been one of the most widely used methods for the determination of TPH in soils, recommended by EPA 1978. This method is applicable to the quantitative determination of TPH's in the soil by extraction and subsequently FT-IR measurement of the extract. TPH was measured in soil by placing the 5 g subsample into a glass flask and adding 0.5 g anhydrous sodium sulphate, 0.5 g silica gel and 25 ml dichloromethane. The flasks were shaken for 30 min at 100 rpm (Schawab *et al.*, 1999). After shaking, the soil–solution extract was filtered through Whatman filter paper, dichloromethane was evaporated and the filtrate was dissolved in carbon tetrachloride (CClч) and analyzed for TPH in infrared light (USEPA method 418.1, 1978) with a Perkin Elmer 16F PC FT-IR (USA).

The calibration plot is used to calculate the concentration of hydrocarbons in soil by using the following equation:

Mg/kg = I.V / a.w

Where:

I = integral value from IR measurement

V = volume of the extract (100ml)

a = slope of the calibration curve

w = weight of the soil sample (about 5g to 10g)

C. *Total organic carbon (TOC) analysis*

The rapid dichromate oxidation of organic carbon was determined according to the methods described by Schumacher (2002). The best known of the rapid dichromate oxidation methods is the Walkley-Black procedure which has been the “reference” method for comparison to other methods in numerous studies. In this procedure, potassium dichromate ($K_2Cr_2O_7$) and concentrated H_2SO_4 are added to between 0.5 g and 1.0 g (although the range may be up to 10 g depending on organic carbon content) of soil or sediment. The solution is swirled and allowed to cool (note: the sample must be cooled as a result of the exothermic reaction, when the potassium dichromate and sulfuric acids are mixed) prior to adding water to halt the reaction. The addition of H_3PO_4 to the digestive mix after the sample has cooled has been used to help eliminate interferences from the ferric (Fe 3+) iron that may be present in the sample although in most cases, this step is not necessary (Tiessen & Moir, 1993).

Experimental design

The evaporation pits were dried by pumping the liquid while the sludge was transported to the bioremediation site by backhaul Truck (Fig. 3). The sludge was mixed with clean soil, shredded plants, organic sludge from the sewage treatment plant, nutrient and brine (Fig. 4). The compost mixture collected into piles and aerated with blowers and vacuum pumps. Air was supplied to the mixture of the biopile system through 2 inches plastic pipes and pumps to increase the oxygen. The biopiles system remains for one month. Then the compost was transferred into an open site and left for four months in piles up to 3m height, 2.5m width and 300m length, with regular excavation and landfarming using mixing equipment and Cutter-Bucket combos. The caliche layer was prepared for the landfarming treatment cell to prevent the leakage of the contamination to the subsurface. More than 300 tons of compost was prepared for use in landfarming and landscape planting of the site for environmental enhancement.

Field monitoring and treatment

The landfarming was monitored daily to control the temperature and moisture content (35-85%), and the concentration of oxygen, carbon dioxide, and volatile organic compounds. The landfarming treatment cell was shoveled with addition of brine and moisture. Tilling of the treatment cell was done at a regular interval of once per week to maintain aeration for the microbial community at the bioremediation sites. This was done with the help of a tractor attached with cultivator. Regular watering of the treatment cell was done each day with a water tanker to keep the soil moisture and to not become dry as a result of the landfarming process (tilling, evaporation, and plowing). The treatment cell was covered with plastic sheet for air emission control. The bioremediation process was monitored by analyzing the sludge and soil samples from time to time. The parameters analyzed were TPH and its various fractions, moisture content, microbial count, pH and selected heavy metals. Moreover, salinity, PH, DO, and nutrient concentration were measured.

Following is a listing with photographs of biological treatments used in this research. Methods used closely follow those given in the literature review.

- Bioremediation consisted of using microbes and nutrients (fertilizers), and controlling moisture and temperature (Fig. 5).
- Bioventing added air (oxygen) and nutrients to enhance the growth of indigenous bacteria for bioremediation (Fig. 6).
- Bioaugmentation used sources of seed organisms to enhance bioremediation.
- Landfarming used excavation, spreading, and frequent tilling of soil to enhance bioremediation.
- Composting also used above bioremediation methods but including the addition of organic components such as wood chips and sewage sludge.

Modeling and statistical application

"STATISTICA V.6™" software was used to analyze data obtained from sample analysis Uni-variant analysis: Descriptive statistic (Maximum, Minimum, and Average) and uni-variant plots (Bar graphs).

Fig. 3. Extraction of sludge by backhaul Truck.

Fig. 4. Bioremediation treatment cell.

Fig. 5. Enhancement of anaerobic bacteria growth by adding nutrient (plants).

Fig. 6. Bioventing in-situ bioremediation technique.

RESULTS AND DISCUSSION

Trace Metals (TM) in Sludge.

The study was implemented on (12) samples (table 1) of sludge (before treatment) which were collected from (3) evaporation pits (A, B, and C). The chemical analysis reveals that vanadium ranging from 5.755 µg/g to 24.897 µg/g, the highest was found in pit 1C. While the permissible limit for (V) in soil is 130 µg/g (Canadian Council of

Ministers of the Environment, 1999). The V concentration was below the permissible limit. Chromium ranging from 0.581 µg/g to 12.303 µg/g, the highest was found in pit 1C. While the permissible limit for (Cr) in soil is 100 µg/g (ECC, 1986). The Cr concentration was below the permissible limit. Iron ranging from 61.544 µg/g to 1070.471 µg/g, the highest was found in pit 1C. While the permissible limit for (Fe) in soil is 200 µg/g (FAO, 1996). The Fe concentration was above the permissible limit. Nickel ranging from 0.261 µg/g to 4.559 µg/g, the highest was found in pit 1C. While the permissible limit for (Ni) in soil is 30-75 µg/g (ECC, 1986). The Ni concentration was below the permissible limit. Copper ranging from 0.161 µg/g to 21.149 µg/g, the highest was found in pit 1C. While the permissible limit for (Cu) in soil is 50-140 µg/g (ECC, 1986). The Cu concentration was below the permissible limit. Zinc ranging from 2.696 µg/g to 106.044 µg/g, the highest was found in pit 1C. While the permissible limit for (Zn) in soil is 150-300 µg/g (ECC, 1986). The Zn concentration was below the permissible limit. Cadmium ranging from 0.068 µg/g to 0.181 µg/g, the highest was found in pit 3B. While the permissible limit for (Cd) in soil is 1-3 µg/g (ECC, 1986). The Cd concentration was below the permissible limit. Lead ranging from 0.255 µg/g to 11.717 µg/g, the highest was found in pit 1C. While the permissible limit for (Pb) in soil is 50-300 µg/g (ECC, 1986). The Pb concentration was below the permissible limit. Figures (7) show the average of metals concentration before and after treatment. Al-enezi et al. (2004) studied the heavy metals content of municipal wastewater and sludges in Kuwait. They found that Cr, Hg, Pb, Ni, Cd, Zn and Cu are the common metals found in Kuwait's raw wastewater and sludge.

Total petroleum hydrocarbon (TPH)

The chemical analysis reveals that total petroleum hydrocarbon (before treatment) ranging from 54.100µg/g to 2100 µg/g, and the highest was found in pit 1C (Table 2). After treatment the TPH was reduced to an average of 44.738 µg/g. The analysis of the sludge contaminated soil (pH 6.0-7.5, moisture content 6.370% and organic carbon content of 0.883%) showed an initial average TPH concentration of 766.840 µg/g at the start of the experiment. There was a significant reduction in TPH concentrations about 46.8% and 81.7% after 120 days and 240 days, respectively. After one year with bioremediation (landfarming) treatment the TPH was reduced to about 94.17% to an average of 44.738µg/g (Table 2). Figure (8) show the average TPH concentration before and after treatment. Atlas (1981) studied microbial degradation of petroleum hydrocarbons and reported that a very diverse group and microbes have the ability to degrade petroleum hydrocarbons. Al-awadhi *et al*. (1996) studied landfarming of oil-lakes- contaminated soil as a result of the Gulf war in Kuwait. They reported that within 15 months of landfarming treatment the oil contamination was reduced by more than 80%.

Table 1. Chemical analysis of trace metals before and after landfarming treatment.

ID Unit	V (µg/g)		Cr (µg/g)		Fe (µg/g)		Ni (µg/g)		Cu (µg/g)		Zn (µg/g)		Cd (µg/g)		Pb (µg/g)	
	Before	After	Before	After	Before	After	Before	After	Before	After	Before	After	Before	After	Before	After
1A	5.755	4.751	0.581	0.252	110.169	53.361	0.261	0.193	0.161	0.075	3.191	1.851	0.089	0.016	0.591	0.178
1B	7.900	5.158	4.455	2.124	728.316	451.192	2.532	2.025	0.605	0.237	3.814	2.156	0.071	0.014	0.831	0.353
1C	24.897	19.316	12.303	9.438	1070.471	624.131	4.559	3.324	21.149	17.162	106.044	78.721	0.177	0.165	11.717	9.081
2A	7.038	4.823	2.223	1.258	454.954	312.045	1.829	0.930	0.419	0.159	3.183	1.820	0.087	0.021	0.733	0.194
2B	6.727	3.422	0.850	0.232	248.310	112.068	0.350	0.216	0.229	0.110	3.012	1.652	0.068	0.011	1.013	0.750
2C	7.319	4.917	1.400	0.960	373.288	152.364	0.754	0.538	0.414	0.155	3.472	2.100	0.076	0.015	0.255	0.104
3A	7.101	4.592	0.974	0.428	141.667	55.230	1.073	0.847	1.003	0.746	8.753	6.318	0.121	0.055	0.661	0.211
3B	8.229	6.543	1.226	0.751	162.066	56.817	1.699	1.163	1.474	1.028	12.570	8.261	0.181	0.147	0.534	0.163
3C	6.817	4.287	0.533	0.216	61.544	26.128	0.873	0.461	0.599	0.216	5.797	3.689	0.074	0.014	0.413	0.119
4A	7.064	4.748	1.995	0.843	374.939	209.344	1.460	0.822	0.499	0.140	3.604	2.142	0.083	0.032	0.754	0.199
4B	6.669	4.193	1.752	0.754	317.515	134.261	0.859	0.364	0.297	0.112	2.696	1.561	0.075	0.014	1.047	0.682
4C	6.062	4.095	0.723	0.436	175.283	55.746	0.712	0.320	0.353	0.271	2.760	1.585	0.084	0.020	0.768	0.290
Mean	8.465	5.903	2.418	1.474	351.543	186.890	1.414	0.933	2.267	1.700	13.241	9.321	0.099	0.081	1.610	1.027
PEL	130*		100**		200***		30-75**		50-140**		150-300**		1-3**		50-300**	

A, B, and C are evaporation pits.
(*) limits described by Canadian Council of Ministers of the Environment (1999).
(**) limits described by European community commission (ECC) (1986).
(***) limits described by Food and Agriculture Organization (United Nations) (FAO) (1996)

Table 2. Chemical analysis for sludge evaporation pits after treatment.

ID Unit	TPH (µg/g)		TOC (%)	
	Before	After	Before	After
1A	294	45.652	0.020	0.022
1B	229	44.812	0.230	0.031
1C	2100	44.628	0.218	0.027
2A	491	45.925	0.817	0.064
2B	178	43.324	0.273	0.046
2C	698	46.167	0.261	0.035
3A	1640	43.598	0.915	0.071
3B	910	44.068	0.231	0.038
3C	749	45.511	0.253	0.047
4A	1660	43.473	7.260	0.042
4B	199	43.661	0.005	0.059
4C	54.100	46.043	0.110	0.016
Mean	766.840	44.738	0.883	0.041

A, B, and C are evaporation pits.

Table 3. Physical and biological parameters before and after landfarming treatment.

ID Unit	PH		DO (ppm)		COD (ppm)		BOD (ppm)	
	Before	After	Before	After	Before	After	Before	After
1A	6.4	8.1	9.2	6.1	733	220	244	22
1B	6.8	7.9	9.8	6.0	652	187	189	25
1C	7.5	8.5	11.2	6.2	684	196	235	23
2A	6.0	8.2	10.3	5.8	751	214	196	20
2B	6.9	7.6	9.9	6.3	623	190	204	24
2C	6.5	8.1	10.8	5.9	693	205	178	25
3A	7.1	7.7	11.7	6.1	712	231	215	19
3B	6.6	8.3	10.1	5.7	684	182	270	21
3C	6.5	8.4	9.5	6.4	778	211	186	24
4A	6.9	7.9	9.3	6.2	675	226	239	18
4B	7.3	8.2	10.6	5.9	734	179	254	27
4C	6.7	8.1	11.1	6.1	656	186	193	26
Mean	6.766	8.083	10.291	6.058	697.917	202.250	216.917	22.833

A, B, and C are evaporation pits.

Total organic carbon (TOC)

The chemical analysis reveals that total organic carbon (before treatment) ranged from 0.005% to 7.260%, and the highest was found in pit 4A (Table 2). After one year of Landfarming treatment the TOC was reduced to about 95.36% to an average of 0.041%. Figure (8) shows TOC concentration before and after treatment. Figure (8) shows the average TPH concentration before and after treatment. Also Balba et al. (1998) studied bioremediation oil contaminated soil in Kuwait (2160 m^3). They found that TPH levels were 39400 mg/kg and reduce to 81.7% after one year.

COD and BOD

The chemical and biological oxygen demand analysis showed that the chemical oxygen demand before treatment ranged from 623 ppm to 778 ppm and the average was 697.917 ppm (Table 3). After one year of treatment the COD was reduced by about 71.1%. The biological oxygen demand before treatment ranged from 178 ppm to 270 ppm and the average was 216.917 ppm. After one year of treatment about 89.47% reduction was found in BOD (Table 3).

PH

During the experimental period, the initial pH of the soil was neutral to slightly alkaline in the range of 6.0-7.5. During the landfarming treatment in the first month, a slight increase to moderately alkaline was observed, raising the pH to 7.9 to 8.2, possibly due to the addition of the nutrients (Table 3). The nutrients applied in this study used a Carbon : Nitrogen : Phosphorous (C:N:P) ratio of 100 : 9.5 : 2.5.

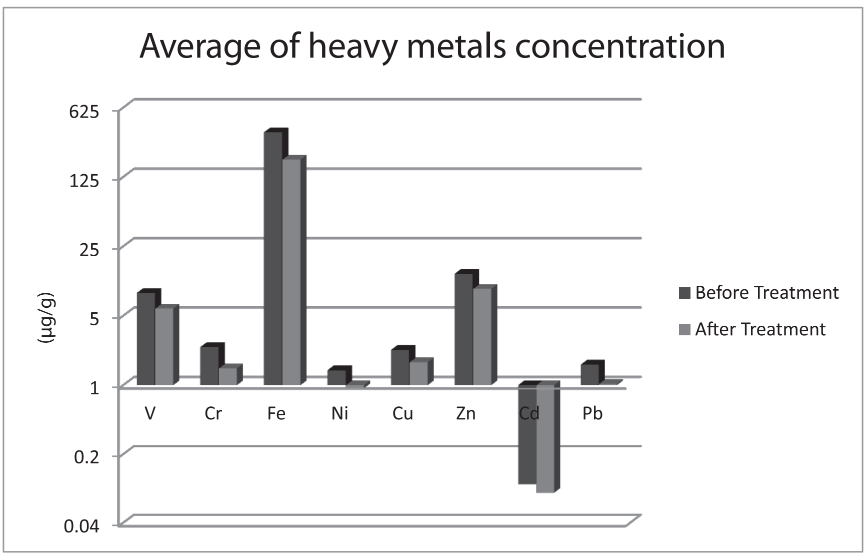

Fig. 7. Average of heavy metals concentration before and after treatment.

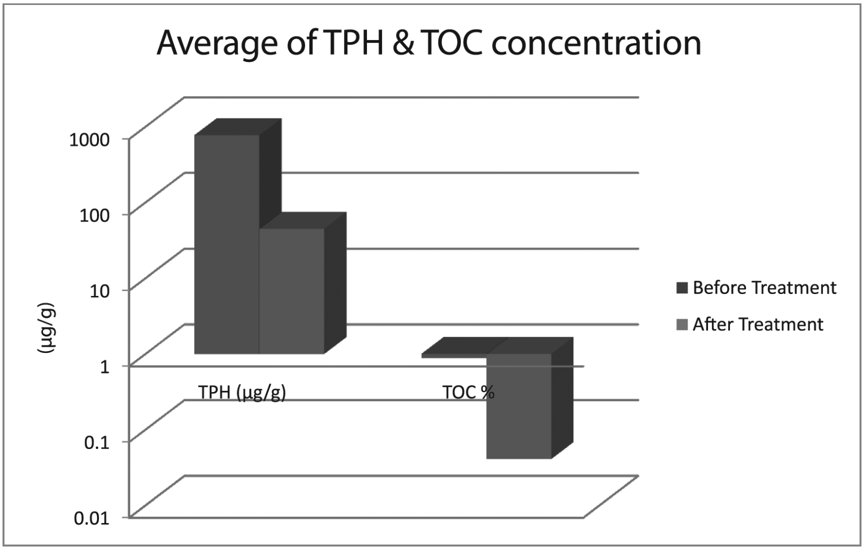

Fig. 8. Average of TPH & TOC concentration before and after treatment.

SUMMARY AND CONCLUSION

A variety of remediation methods was tested and proved successfully in this research. *In situ* techniques worked very well by adding nutrients, oxygen, and microbes. Landfarming used a number of natural processes to provide degradation of any contaminants present in the soil. This proved to be a simple, cost effective technique of remediating contaminated soil on the site. Addition of oxygen and nutrients worked well through bioventing, biodegradation, bioaugmentation, and biosparging. *Ex situ* or off site treatments using the above methods also worked well, especially for landfarming and biopiling (composting), and through the use of sewage sludge. The adequate PH ranges between 6.5 – 7.5 and temperature between 30 – 45 °C to enhance the microbial growth. Nakasaki et al. (1993) studied the effects of pH control on composting. They reported that the range of pH values suitable for bacterial development is 6.0–7.5, while fungi prefer an environment in the range of pH 5.5–8.0. While Nester et al. (2001) found that most bacteria which degrade petroleum hydrocarbons are found in soil, it has an optimal range of temperature from 25 °C to 45°C.

The sample analysis for both soil and sludge revealed that the contamination levels were very high at some sites, especially evaporation pit C. The chemical analysis indicates that these levels were much reduced by treatment and over time. Eight metals were monitored and results after one year indicated an 81% overall reduction. The data also indicate that treated trace metals were much lower than ECC and Canadian Council permissible limit. Iron (Fe) was above the FAO permissible limit, which indicated the source of this relatively non-toxic component was sewage rather than industrial. Of the organic compounds monitored, COD, BOD, TPH, and TOC showed one-year reductions of 71.1%, 89.47%, 94.16%, and 95.36%, respectively. In summary, bioremediation was shown to be very effective in this research, providing hope for remediation of these common waste pits found throughout the Arabian Gulf oil production sites.

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