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Environmental Site Assessment of Al-Daura Refinery - Evaluation of Soil Pollution with Petroleum Products

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Abstract

The environmental site assessment (ESA) of Al-Duara refinery in Baghdad, Iraq. The hydrocarbon content of the soil around this site was measured to evaluate the extent of the organic pollution depending on selected parameters of potential pollution. Soil samples were collected at various depths from almost all locations including: production units, storage tanks, the landfill lagoons at the outside boarder of the refinery and the residence location. For the evaluation of the nature of soil layers, a monitoring well was drilled in the vicinity of a wide dumping area for heavy untreatable materials. Some field tests were carried out to evaluate the hydrocarbon-in-soil (Turbidimetric); Laboratory analysis for the soil extracts and types of hydrocarbon pollutants. The soil texture was evaluated using the standard methods to assess any future remediation efforts. It appeared that storage tanks location is among the serious polluting sources via leakage or overflow of the drainage collection channels. The pollution sometimes extended deep in the soil, 80-100 cm. The maintenance and rehabilitation operations contribute much pollution by heavy sludge containing hydrocarbons, mud and inorganic matter. Most of the pollution load is directed towards the wastewater treatment plant. The dumping location and over more than 30 years resulted in deep penetration of hydrocarbons so that the location is considered the most effective source of pollution. Finally, the fall out is a major means of pollution of remote area within the site by heavy hydrocarbons.

Recommendations for remediation of the most polluted locations in the site were listed at the end of this study. However, the company is continuously doing actions to remove polluted soil and replace it with clean soil.

Keywords: Soil Pollution, Environmental Assessment, Pollution by Hydrocarbons.

التقييم البيئي لموقع مصفى الدورة – تقييم تلوث التربة بالمنتجات النفطية

الخلاصة

تم تقييم الآثار البيئية لعمليات التصفية الجارية في مصفى الدورة على الأرض. وفي البحث الحالي نقدم دراسة لتعيين المحتوى الهيدروكاربوني في التربة ضمن أرجاء المصفى كجزء من عمليات التقييم البيئي للمواقع. تم جمع عينات تربة على أعماق مختلفة ومن أغلب أماكن الوحدات الإنتاجية والخزانات ومواقع ردم المخلفات والموقع السكني المجاور. وأشتمل العمل على مراقبة نسب الهيدروكاربونات في أعماق وصلت إلى ثمانية أمتار لبيان مدى نفوذ التلوث إلى الأعماق عند أسوأ منطقة في الموقع وهي مكان ردم المخلفات الثقيلة. تم إجراء فحوصات حقلية لتحديد الهيدروكاربونات في التربة، وأجريت تحاليل مختبريه تفصيلية لتراكيز وأنواع الهيدروكاربونات في

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عينات المياه والتربة. تم دراسة تركيب بعض عينات التربة بما فيها التدرج الحجمي للدقائق في مختبرات معتمدة باستخدام الطرق القياسية لأهميتها في أعمال المعالجة اللاحقة.

دلت القياسات الحقلية والتحاليل المختبرية على انتشار المواد الهيدروكاربونية في العينات. تنتشر الهيدروكاربونات حول الخزانات الإستراتيجية جراء السكب وطفح القنوات المعدة لاستلام مخلفات تفريغ الخزانات من المياه وتمكنت من النفوذ إلى عمق 80 – 100 سم. أما عينات التربة المأخوذة على أعماق وصلت إلى 8 م في منطقة الحفر النفطية فقد تبين نفوذ الهيدروكاربونات إلى أعماق 4 م وتتدرج من نحو 2% عند التربة بعمق 50 سم وحتى عشرات الأجزاء من المليون عند عمق 3-4 م. ومع ذلك تبين نظافة المياه الجوفية المأخوذة من بئر المراقبة من الزيوت.

Introduction

Petroleum refining is among the most important industries utilizing the crude oils and contributing very much to the production of large number of essential materials in the modern life. Petroleum refining is one of the largest industries in Iraq and have great impact on other activities of the country in industry, health, heating and power generation etc. Refining uses so many technologies to extract the light fractions and reforming of some cuts into more valuable products. Catalytic cracking and vacuum distillation are involved to reduce the required operating temperature of the petroleum processing. One of the major costs of the refining industry is the emission of various pollutants to the environment, represented by air water and soil. However, potential environmental hazards associated with refineries have caused increased concern for communities in close proximity to them.

Petroleum refineries separate crude oil into a wide array of petroleum products through a series of physical and chemical separation techniques. These techniques include fractionation, cracking, hydrotreating, combination / blending processes, and manufacturing and transport. The refining industry supplies several widely used everyday products including petroleum gas, kerosene, diesel fuel, motor oil, asphalt, and waxes [1].

The past production practices may have led to spills on the refinery property that now need to be cleaned up. Bacteria that may use the petroleum products as food are often effective at cleaning up petroleum spills and leaks. Many residuals are produced during the refining processes, and some of them are recycled through other stages in the process. Other residuals are collected and disposed of in landfills, or they may be recovered by other facilities. Soil contamination including some hazardous wastes, spent catalysts or coke dust, tank bottoms, and sludges from the treatment processes can occur from leaks as well as accidents or spills on or off site during the transport process [1].

Vasariviceus et al [2] studied the soil pollution by hydrocarbons from military operations and presented a model for the prediction of the path and tendency of oil products infiltration into the soil over a period of 50 years. Palmroth et al. [3] presented detailed phytoremediation investigation based on the assessment of soil contamination by hydrocarbons and other organic and inorganic pollutants. Wcislo [4] reviewed the polyaromatic hydrocarbon (PAH) contamination levels in environmental media in Poland, including surface soil. This research is important in order to better define exposure estimates in the general population and to examine the relationship between levels of PAHs in the environment and the subsequent development of health effects.

During the refining processes various waste materials are produced. Most of these wastes are sent for the treatment within the refinery. However, the heavy liquid wastes that are difficult to be treated by the water treatment system are sent into a dumping location located outside the south-east fence of the refinery. Most frequently, the furfural content exerts a large harmful effect on the living matter in the wastewater biological treatment basins and such waste fluids are transported to the dumping location. The accumulated heavy liquids are left for natural drying especially during the severe hot weather in summer time of Iraq. The drying continues until the soil cracks. The soil and the hydrocarbon residues are burned to minimize the hydrocarbon content of the location. New soil is then brought from other places and laid on the dried petroleum residues and mixed with the aid of shovel to furnish a new dumping location. To speed the degradation, the dried lagoon contents are burned to get rid of the petroleum residues [5].

The present work is a field and laboratory investigation for the evaluation of soil pollution with petroleum hydrocarbons inside and outside the Al-Daura refinery site.

Materials and Methods:

1-Sampling Plan

In the beginning the team wanted to take samples from any abnormal or a potential pollution source. This included some temporary cases where the staff of the refinery insisted that they are not a predominant situation. During the progress of the work, it appeared that such temporary cases were solved and wastes or spills were removed or treated. Thus, the analyses of the samples did not cover such remediated situations.

Fig. 1 shows a schematic map of the site showing the sampling locations. The most acceptable practices [6] are applied during the sampling process to ensure that the sample is representative and undergo the least contamination in an area of soil. Surface and deep soil samples were collected from specific locations of potential pollution. Soil samples were taken not directly from the location covered with hydrocarbons but from the nearby (~ 100-150 cm) aside from it and at certain depth to search for potentials of pollution. The sampling covered the whole refinery area with focus on storage tanks surroundings, flare surrounding, maintenance operations and heavy residues dumping location outside the refinery.

2- Soil Layers Sampling:

Another set of samples were taken during the drilling operation of the monitoring well at depth ranged between 0.5 to 8.0 m. A sampling interval of 50 cm was chosen and a total of 15 samples were taken during the drilling.

3- Total Petroleum Hydrocarbon Analysis:

The total petroleum hydrocarbon content in soil samples was measured by the PetroFLAG system. The PetroFLAG™ System was supplied from Dexsil® Corporation, USA. The method is recommended by ASTM standard screening procedure [7]. This method is based on the extraction of hydrocarbons from soil with a suitable solvent (mostly methanol), filtration under pressure and mixing of the filtrate with an aqueous solution of special detergent to aid the emulsion formation between water and the hydrocarbon. The emulsion is then measured by turbidimetric technique. The system includes a balance, extraction tubes, cartridge filtration, a stopwatch and vials containing special developer reaction bottle and the measuring-reading equipment.

As in any refinery, the most expected pollutants for soil and water are the crude oil, the intermediate fractions and final hydrocarbon products. The most applicable technique for hydrocarbon analysis is the Gas chromatography, GC, employing capillary column separation and detection with flame ionization (FID). The Gas Chromatographic measurements of the soil extracts were carried out on a computer controlled Agilant system equipped with a hydrogen generator and Auto sample Injector. The separation was carried out on a capillary column 100 m x 0.25 mm. The operating conditions were as follows: Injector Temperature, 350^{o}C; column, temperature programming 40 -300 ^{o}C at a rate of 10 ^{o}C/min. The carrier gas was nitrogen at a flow rate recommended by the manufacturer. The detector was FID working a temperature of 350 ^{o}C using hydrogen flow rate of 30 ml/min and air ar a flow rate 300 ml/min [5].

The most relevant sample preparation procedures were applied [8]. The soil samples were extracted using the soxhlet extraction process ASTM. A weighed soil sample (20-25 g) was thoroughly mixed with an equal amount of anhydrous sodium sulphate to aid dehydration and then placed in the extraction cellulosic thimble. The thimble was inserted within the soxhlet apparatus and hexane or dichloro-methane was employed for the extraction. Experiments indicated that the extraction requires 16 -20 hr. In the next day the soxhlet system is dismantled and the flask containing the hydrocarbon extract was fitted in the RV-05 IKA Rotary Evaporation System to aid the concentration and solvent recovery. The evaporation continues until the volume of the solution is reduced to 4-7 ml volume. The concentrate was kept in a glass vial until the time of GC analysis.

4- Soil Texture determination:

Soil samples are first ground in porcelain mortar and sieved to pass 1-mm sieve. A portion of the sieved soil, 50 g was equilibrated with about 200 ml of water for 15 min and 100 ml of 5% calgon (sodium hexametapho-sphate) solution was added. The mixture was then transferred into

1000-ml graduated cylinder and water was added to the mark. The cylinder was covered tightly with Parafilm and mixed thoroughly by inversion upside down for 10 times keeping the hand on the parafilm.

The Bouyoucos hydrometer and the thermometer were immersed in the water soil suspension at the first 40 s for specific gravity and temperature measurements. The temperature measurement must accompany the specific gravity measurements to correct the hydro-meter reading (for every degree above 20° C, an increment of 0.2 is added to the initial hydrometer reading).

Another temperature and specific gravity reading were taken after two hrs time. To determine the percent of soil separates, use the formulas below [9]:

% sand=100-[first hydrometer reading (corrected for temp.)/sample wt. (50 g)] x 100

% clay= [second hydrometer reading (corrected for temp.)/sample wt. (50 g)] x 100

% silt = 100 - (% sand + % clay)

Results and Discussion:

1- Hydrocarbon Contents of Well-Soil Samples:

The soil sampling assured the recovery of minimally disturbed samples. Beyond a depth of 6 m, the soil was fine-grained and appeared as thick slightly greenish mud. The total hydrocarbon content of the soil samples at the various depths of the monitoring well, as determined by the extraction turbidimetric method, are shown in Fig. 2. It is clear that the depth at which maximum hydrocarbon content (≥ 2%) was that of 0.5 m. This refers to the continuous mixing of the surface soil with soil from other places so that the surface is always diluted compared with the 0.5 m deep soil. The high hydrocarbon contents of soil often occur after long time exposure and maintenance operation at the specific location [3].

Due to the operations carried out at the surface soil within the area of the dumping lagoons, the hydrocarbon contamination was concentrated in the first 150 cm. This was best reflected by the oil content of water samples taken from this well. This will be the subject of a detailed investigation on the water quality in the site.

Hydrocarbons are associated with sorbed organic matter in the soil and hence the strength of the hydrocarbon sorption will vary according to the nature of the hydrocarbon and the organic matter content of the soil [10]. Fig. 3 illustrates the manner in which the hydrocarbons will be sorbed in a soil. The amount of pores in the soil matrix is dependent the soil classification. Clays generally have high percentages of small pores, whereas sand has a low percentage [11].

The hydrocarbon rich soil samples of the drilled well (depth 50 -100 cm) were extracted with hexane and after concentration by vacuum evaporation the extract gave the chromatogram shown in Fig. 4.

Excluding the signal of the solvent, hexane, the profile of the detected hydrocarbons greatly resemble the gas oil chromatogram shown in Fig. 5, being measured under the same operating conditions and to that of a standard gasoil cited in the literature [12].

The GC signals of rest of the soil sample extracts did not show distinct peaks for hydrocarbons even at the ranges of carbon number of C25 and C36. Again the extraction of a considerable amount of soil and evaporation of the solvent under vacuum to dryness resulted in a resinous material. The occurrence of resinous materials may be produced by the long term effect of hydrocarbons being catalyzed by the soil minerals [13]. They could not be analyzed with regular GC separation. The resins are evaluated by fractionation with liquid chromatographic Techniques [14].

Similar profile could be recorded for all the extracts from the dumping lagoons samples. The continuous work on mixing the soil at this location with clean soils from other places and turning them upside down aid the prevention of the evaporation of these middle distillates from soil.

2- Total Petroleum Hydrocarbon in Soil Determination

Some of the collected samples were not analyzed because the locations were remediated by the company and the residues were removed from the area immediately after the sampling. The total hydrocarbon contents are shown in table (1). The samples representing the soil within the dumping lagoons, confirmed those related to the soil layers of the monitoring well digging. However some variation from those exhibited in Fig. 2 above, because the surface soil within the lagoons area is always subject to mixing with clean soil and burning and exposed to the summer sun of Iraq for several seasons. Fig. 6 shows the chromatogram of the hydrocarbons extracted from the surface soil of the dumping lagoon location. It is clear that the heat of sun evaporated the light and semivolatile hydrocarbons and only resinous material is left there.

At a depth of 2 m, the soil sample taken within the new project area exhibited almost no hydrocarbon content. This is related to the difficult penetration of hydrocarbons down to such depths [11]. Some samples were characterized by an average hydrocarbon content of about 1000 ppm (0.1%). The sample taken at the flare location indicated relatively high hydrocarbon content even at a high dilution.

It is worthwhile that the pollution did not reach the ground water. This was proved by analyzing water samples from this well daily over three weeks.

At a depth of 25 cm within the flare area, the hydrocarbon content was less than 0.1%. For the samples that represent the area around the fire accident happened to the light naphtha tank on December, 2007, with its surface area covered by fire extinguishing residues, the hydrocarbon content were relatively lower than those for other tanks. Meanwhile the land was covered by mixtures of chemicals, soot, foam and some hydrocarbons.

The area between the ware houses and storage tanks exhibited moderate hydrocarbon content in the soil (0.07%). The samples taken at an average distance of 2-3 m from the area covered by a spill from storage tanks and at certain depths, yet indicated moderate to high hydrocarbon contents (0.08 – 1.48%). The soil at the pipeline between new project and main street (Taken at the storage tanks side) exhibited relatively high hydrocarbon content (< 1.5%) indicating the large impact of the storage tanks spills on the pollution of soil. Sample 29 of the area around the TEL tank, exhibited the lowest hydrocarbon content of the soil. This is related to the ignorance of the area for several years period and the role of the metal plate roof of the unit in preventing or making the deposition of hydrocarbons the least in the refinery locations.

However, the model proposed by Vasarevecius et al [3] could not be applied for the Daura location because of the extreme difference in climatic conditions with that dominant in Lithuania. In some cases a small distance between samples gave high differences in the hydrocarbon contents. The choice of these samples was according to the use of the location. However, the sample at the surface of the location exhibited the highest values (~ 1.3%).

The extraction of the soil with methanol and the drying of the solvent at ambient temperature resulted in heavy and highly viscous residue which can be sorted as resinous material. However, the long exposure time of the soil to the heat radiation from the flare and the action of atmospheric oxygen and soil mineral components can account for a catalytic conversion of middle distillates into resinous material [15-17].

The soil contamination with hydrocarbons might have adverse effects on cultivation. Recent studies on the effects of hydrocarbon contamination on the plant ecosystem concluded that more research is required in this area [18].

3- Texture Analysis:

The soil texture data is often correlated with the contaminants concentration of soil [19]. The soil classification can be evaluated once the percentages of sand and silt is determined using the texture Triangle analysis chart [20]. The soil type is an important factor in determining the extent of hydrocarbons as well as their evaporation [11]. The soil samples exhibited different soil textures and the results are listed in Table 2. For remediation purposes the texture of soil mixtures has to be determined to aid the process of leaching and washing.

Conclusions:

1. Storage tanks location is among the serious polluting sources via leakage or overflow of the drainage collection channels. The pollution sometimes extended deep in the soil, 80-100 cm.
2. Maintenance and rehabilitation operations contribute much to pollution by heavy sludge

containing hydrocarbons, mud and inorganic matter. Most of the pollution load is directed towards the wastewater treatment plant.

3. The dumping location and over more than 30 years resulted in deep penetration of hydro-carbons so that the location is considered the most effective source of pollution.

4. The detailed chemical analysis proved that the soil is contaminated with relatively heavy hydrocarbons of resinous nature, except for the dumping location in which middle distillate hydrocarbons could be detected.

Recommendations:

The following steps are recommended here for the remediation based on the findings above.

1. Stopping the deterioration of the land by repairing the leaks and stop any leakages from pipes, tanks and reactors. However, the main efforts should be directed towards the light oil production units, because the lube oil plants are only partially in operation.
2. Removal of free phases, apparent hydrocarbon spilled on the ground at the loading stations, desalting and distillation units.
3. Overall checking and repair of the manholes and piping networks.
4. Virtually and apparently contaminated soil must be excavated down to a depth of few centimeters, and collected from various locations in a central place to be washed out.

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Table (1) Hydrocarbon Contents of Soil Samples.

No.	GPS	Depth, m	HC Content, ppm
1	N 33 16 28” E44 26 30”	60 cm	19800
2	N33 16 28" E44 26 30.”	120 cm	4580
3	N 33 16 27.5” E 44 26 31”	100 cm	5680
4	N 33 16 28” E44 26 30”	surface	14780
5	N:33◦16´.45”, E44◦25´37”	2m	not detected
6	N:33◦16´.50”, E:44◦25´.51”	Surface	1173
7	N:33◦16´.53”, E:44◦25´.43”	surface	1080
8	N:33◦16´.53”, E:44◦25´.43”	30 cm	1150
9	N:33◦16´.56”., E:44◦25´.53”	Surface	Very high
10	N:33◦16´.55”, E:44◦25´.53”	25 cm	4883
11	N 33 16 43” E 44 25 50”	surface	740
12	N 33 16 43” E 44 25 50”	Surface	2047
13	N 33 16 43” E 44 25 50”	100 cm	877
14	N 33 16 37” E 44 26 05”	Surface	893
15	N 33 16 45” E 44 25 919	70 cm	13140
16	N 33 16 46” E 44 26 00”	70 cm	987
17	N 33 16 47” E 44 25 41”	50 cm	2277
18	N:33◦16´.45”, E 044◦25´39”	Under pipes	15213
19	N:33◦16´.48”, E 044◦25´.47”	25 cm	1288
20	N 33 16 31”; E 44 26 16”	Surface	4120
21	N 33 16 42” E 44 25 41”	60 cm	1076
22	N 33 16 42” E 44 25 42”	Surface	1397
23	N 33 16 29” E 44 25 901	Surface	51
24	N:33◦16´.48”, E 44◦25´.48”	surface	14080
25	N:33◦16´.50”, E 44◦25´.51”	25 cm	500
26	N:33◦16´.48”, E 44◦25´.48”	30 cm	1423
27	N:33◦16´.48”, E 44◦25´.51”	Surface	1930
28	N:33◦16´.48”, E 44◦25´.51”	30 cm	408
29	N:33◦16´.50”, E 44◦25´.49”	Surface	1070
30	N:33◦16´.50”, E 44◦25´.49”	30 cm	533
31	N:33◦16´.55”, E 44◦25´.42”	Surface	320
32	N:33◦16´.56”, E 44◦25´.716	30 cm	34
33	N 33 16 42” E 44 25 41”	surface	795
34	N 33 16 14” E 44 25 49”	Surface	2850
35	N 33 16 48.4” E 44 26 46”	Surface	765
36	N 33 16 16” E 44 26 55”	surface	675

Table (2) Soil Texture Results of Soil of Al-Daura Site

Sample Description*	Sand %	Silt %	Clay%	Soil Type
Surface soil between API tanks and CT	46	26	28	Sandy clay Loam
Soil at 25 cm depth between API and CT	65.4	18	16.6	Sandy Loam
Surface soil near Engeco unit and CT	49.6	22.8	27.6	Sandy clay Loam
Soil at 30 cm depth near Engeco unit and CT	45.2	27.2	27.6	Loam
Land between CT and rectangular basins	54.8	19.6	25.6	Sandy clay Loam
under the pipeline adjacent to warehouse	68.4	10.0	21.6	Sandy Loam
2 m depth at the new prcject area	33.4	31.8	34.8	Clay Loam
3m North from Tank 2100 crude oil	49	26	24	Sandy clay Loam
Surface soil Near discarded TEL tank	50	26	24	Sandy clay Loam
Subsurface RO pumps chamber, Near outer fence by the river	43	23	34	Sandy clay Loam
Between the slop tanks and CT	49	23	28	Sandy clay Loam
Surface soil from the dumping lagoon	31	32	37	Clay Loam
area between tanks and warehouse	38.4	46	15.6	Loam
Surface soil near Tank 2114	26.4	54	19.6	Silt Loam

*CT = Cooling Tower, RO = Reverse Osmosis.

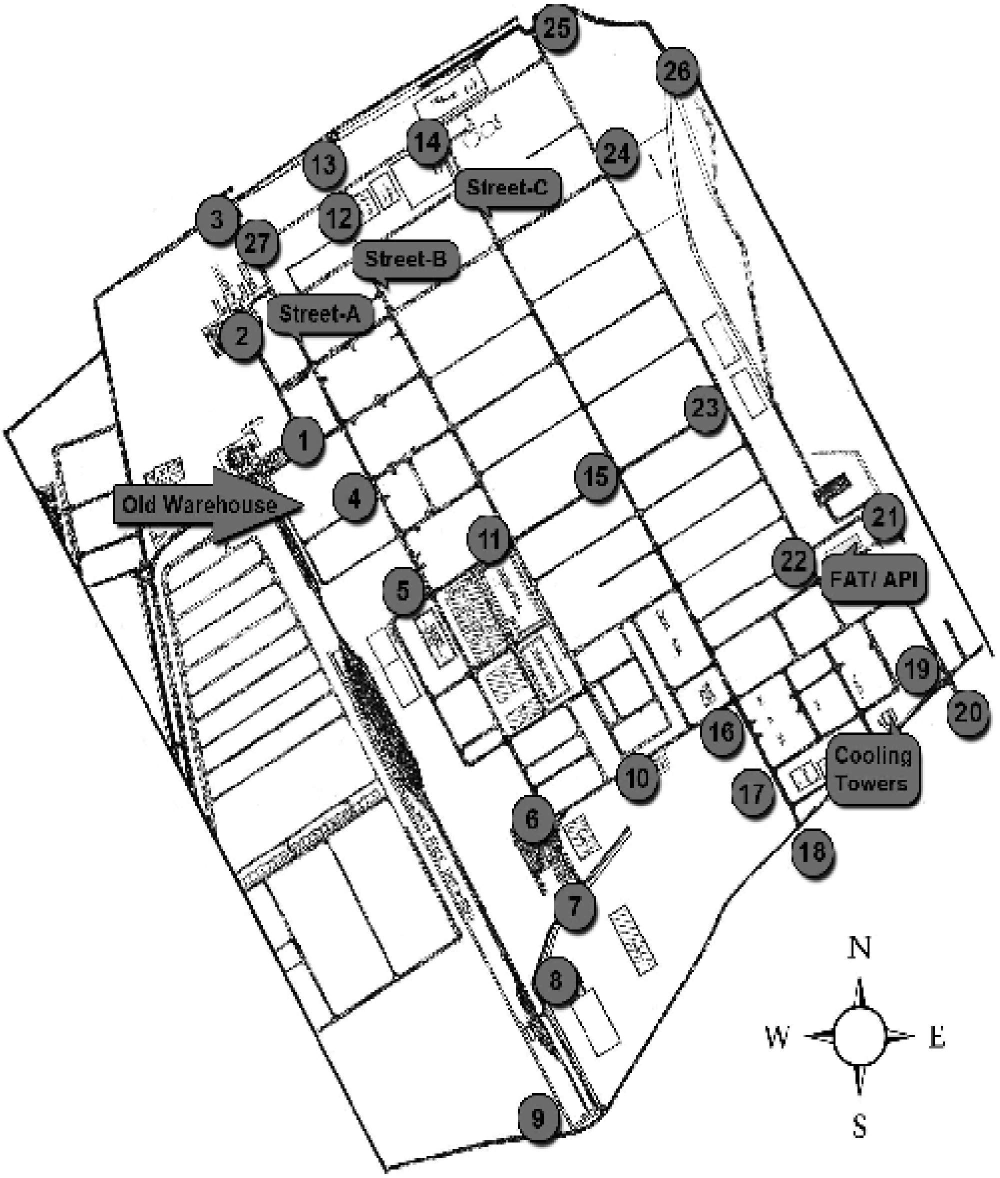

Figure (1) Schematic Map of the Site Showing the Sampling Locations

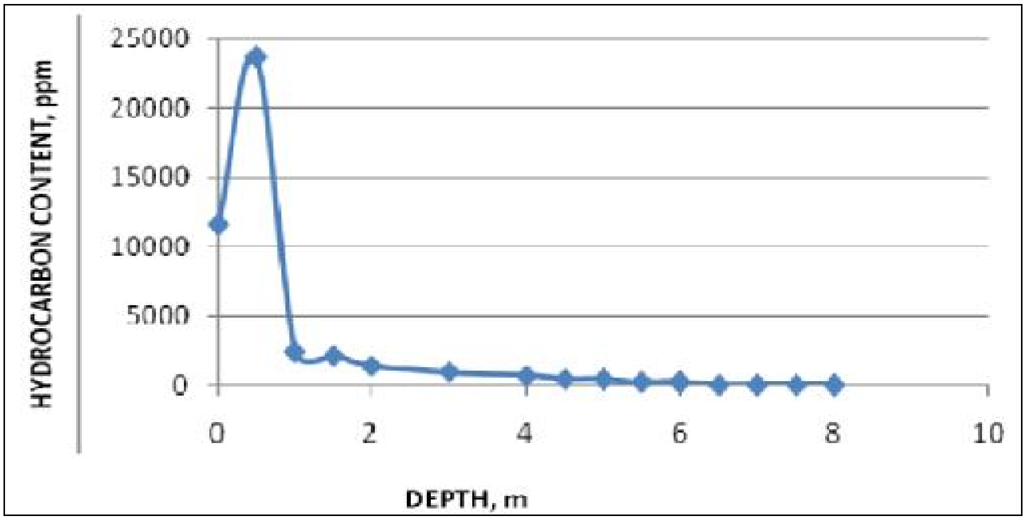

Figure (2) Distribution of Hydrocarbons with the Depth of Soil

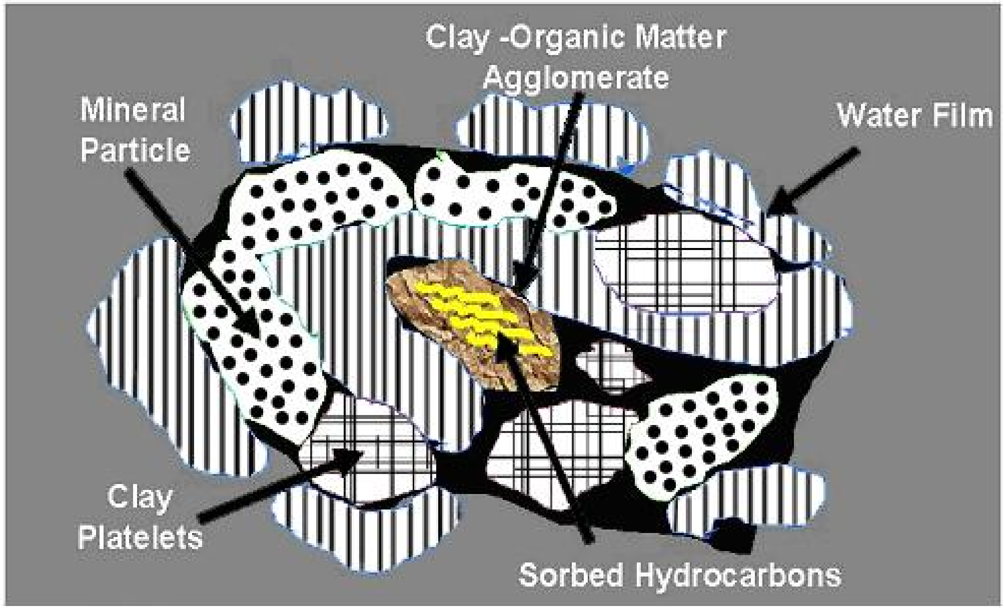

Figure (3) Hydrocarbon Interfractions with Soil [11].

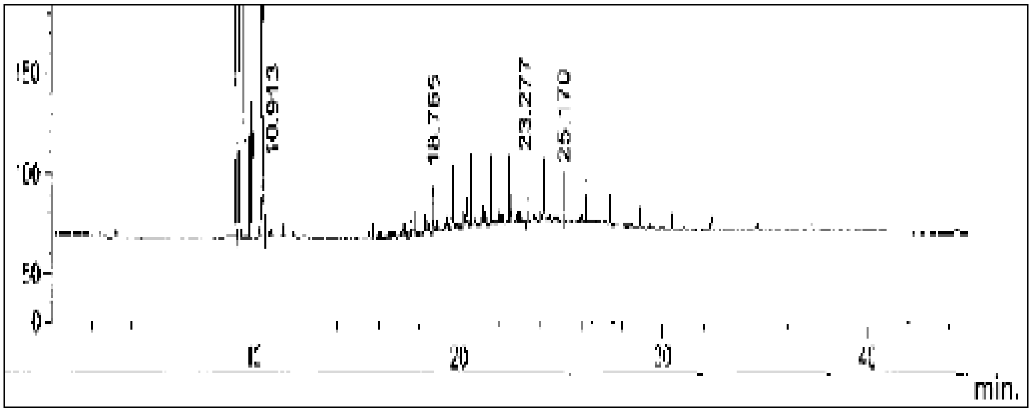

Figure (4) Gas Chromatogram of Hexane Extract of Soil at 100 cm Depth in Dumping Lagoons

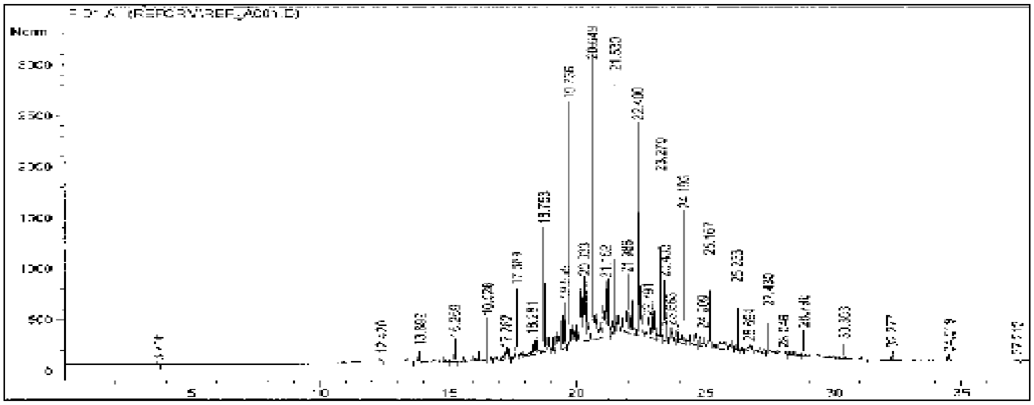

Figure (5) Gas Chromatogram of Dehydrated Gasoil

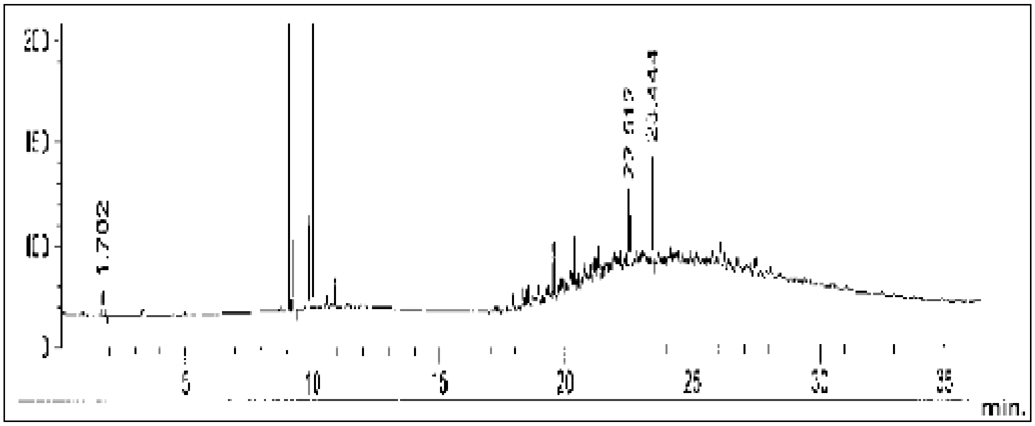

Figure (6) Gas Chromatogram of Hexane Extract of Surface in Dumping Lagoons