



Modelling of Aqueous Petroleum Hydrocarbon in Polluted River

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ABSTRACT: This research focused on the modelling of aqueous petroleum hydrocarbon polluted river in Abonnema wharf Rivers State, Nigeria. Analytical techniques for Total Petroleum Hydrocarbon (TPH) count in water were investigated. The results obtained showed the TPH contents ranging from 9.5276 ppm to 223.8895 ppm respectively indicating the concentration of Phenolic content as most abundant with a concentration of 223.8895 ppm while C9 as the least with a concentration of 9.5276 ppm. The elevated levels of total petroleum hydrocarbons (TPHs) from Abonnema wharf have provided evidences of severe petroleum contamination as compared with the international certified relevant regulatory agencies standards. The petroleum hydrocarbon concentration contamination was modelled using MATLAB employing the Michaelis-Menten Equation. The programmed model reveals that the petroleum hydrocarbons contamination transition levels are linear and possesses positive trends for C8, C12, C16 and C25 hydrocarbons.

KEYWORDS: Petroleum Hydrocarbon, Polluted River, MATLAB Program

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I. INTRODUCTION

Oil from the petroleum industry enters the aquatic environment through several sources such as, fall outs from gas flaring, disposal of used lubrication oils, washings from oil tanks, leakages from marine vessels and out board engines, sabotage, erosion and run off from crude oil polluted lands, seepage, refinery effluents, rupture of ill maintained flow lines installations and engineering errors. Nigeria has been exploring and exploiting crude oil for decades and the consequences on the oil producing and processing areas have become highly problematic in the onshore and offshore installations. Despite improved technologies in petroleum exploration and production techniques in the last few decades, several cases of avoidable accidents have occurred resulting in hydrocarbon pollution of the environment. In most of the oil producing countries in the world, the petroleum industries have contributes greatly to aquatic environmental degradation and pollution. Even after use, the resultant gases contain un-burnt hydrocarbons discharged into air from which they can be washed by the rain and back to land and also into the sea [1]. About 9 million barrels of crude oil have been spilled in the Niger Delta since 1958, the department of petroleum resources stated that between 1989 and February, 2000 a total of 536,858.84 barrels of crude oil were spilt in 2252 incidents. Out of the number the report observes only 23,003.84 barrels were recovered, while the principal portion of 513,854.98 barrels was lost to the environment. The general feeling of the people of the area however is that oil spill incidence and quantity of oil spilt are grossly under reported particularly by official sources, this implies that the figures listed above could actually be a tip of the iceberg. A greater majority of this spilt oil gets into the streams, rivers and ocean directly or indirectly where a reasonable fraction either mixes with water or sinks into the sediment causing severe damage to benthic organisms [2].

Every human needs about 20 litres of freshwater a day for basic survival (drinking and cooking) and an additional 50 to 150 litres for basic household use [3]. However, with growing population and an overall increase in living standards, not only is the overall demand for freshwater pushing limits (one third of world now lives in areas of portable water scarcity) but increasing pollution from urban, industrial and agricultural sources is making available water resources unsuitable while increasing health risk [4]. The release of petroleum hydrocarbons and their derivatives either accidentally or deliberately to the environment is known to pose problems of increasing magnitude throughout the world [5]. Hydrocarbon contamination in the environment is a

very serious problem whether it comes from petroleum, pesticides or other toxic organic matter. The pollution caused by petroleum is of great concern because petroleum hydrocarbons are toxic to all forms of life [6]. The toxicity from hydrocarbon ingestion can affect several body organs, such as the lungs, liver and kidney. A variety of hydrocarbon pollutants in receiving water bodies are known to cause disruptions of the natural equilibrium between living species and their natural environment [7]. Hydrocarbons pose no risk on their own. However, when exposed to sunlight or nitrogen oxides, they undergo a chemical reaction. It is well known that emissions and pollution created by human beings in this industrial age are dangerous and hydrocarbons make up a large portion of these damaging compounds.

Total Petroleum Hydrocarbon (TPH) is a term used to describe a large family of several hundred chemical compounds that originally come from crude oil. Crude oil is used to make petroleum products, which can contaminate the environment [8]. There are so many different chemicals in crude oil and in other petroleum products. It is not practical to measure each one separately. It is useful to measure the total amount of TPH at a site. TPH is a mixture of chemicals, but they are all made mainly from hydrocarbons. Some of the TPH compounds can affect your central nervous system. One compound can cause headaches and dizziness at high levels in the air. Another compound can cause a nerve disorder called "peripheral neuropathy," consisting of numbness in the feet and legs, other TPH compounds can cause effects on the blood, immune system, lungs, skin, and eyes.

Scientists divide TPH into groups of petroleum hydrocarbons that act alike in soil or water. These groups are called petroleum hydrocarbon fractions. Each fraction contains many individual chemicals. Some chemicals that may be found in TPH are hexane, jet fuels, mineral oils, benzene, toluene, xylenes, naphthalene, and fluorine, as well as other petroleum products and gasoline components. However, it is likely that samples of TPH will contain only some or a mixture of these chemicals. The amount and types of compounds in a petroleum hydrocarbon released into the environment differ widely depending on the product spilled and how it weathered. This variability makes it difficult to determine the toxicity and mobility of weathered petroleum products solely from TPH results. However, an analysis of soil and groundwater can be used to approximate risk depending on the hydrocarbon range, how well the site has been characterized and the intended use of the land.

The associated impacts of oil spills in mangrove vegetation and coastal waters cannot be overemphasized in Niger-Delta [9]. Water contamination becomes a noteworthy problem because water is an essential natural resource for human life. There is various human health complications associated with petroleum hydrocarbons contamination. Some of these include carcinogenicity, genotoxicity, deoxyribonucleic acid (DNA) damage, birth defects, childhood leukaemia, infertility and miscarriages in women, sterility, skin rashes and irritation, respiratory system disorders, and cancers of different parts (organs) of the body [10]. It has been reported that workers exposed to petroleum hydrocarbons have adverse health symptoms such as headaches, eye and skin irritation, respiratory difficulties and among others [11]. The co-existence of toxic heavy metals and hydrocarbons (HCs) at many of contaminated sites poses a severe threat to the environment.

Total petroleum hydrocarbons (TPHs) are a large family of several hundred chemical compounds that originally come from crude oil. They are found in the range of C₆ through C₃₅ as mixture containing hundreds to thousands of hydrocarbons including aliphatic (straight carbon chain) and aromatic (carbon ring) compounds. Some hydrocarbon mixtures may also contain priority pollutants including volatile organic compounds (VOCs), semi-volatile compounds (SVOCs) and metals, each of which have their own specific toxicity information by Agency for Toxic substances and Disease Registry [12]. The presence of chemical contaminants in the coastal environments from many anthropogenic sources is a major threat to the marine water [13]. Large amounts of these contaminants through sewage, petroleum spills, municipal and industrial discharges, automobile wastes and vehicular emission due to incomplete combustion of fossil fuels are possibly carried by river runoffs through their estuaries into the sea [14]. Petroleum hydrocarbons are one of the major pollutants which are frequently discharged into the coastal water, though not usually regulated as hazardous wastes by Agency for Toxic substances and Disease Registry [15]. Although they are naturally present in very low concentration in the marine sediments, but larger amount comes from petrogenic and pyrogenic sources. Thus, the bottom sediment being the habitat of many aquatic organisms is recognized as reservoir of petroleum hydrocarbons in the marine environments with high risk of bioaccumulation [16]. When Total Petroleum Hydrocarbon is released into the environment through accidents, releases from industries, or as a by-product from commercial or private uses and/or into the soil through spills or leaks, it moves from the soil to the groundwater and some organisms found may break down some of the contaminants into smaller fractions while some may evaporate into the atmosphere, others will stay in the soil for a longer time and will be broken down by other organisms found in the soil causing hazardous health effect [17].

According to Environmental Protection Agency [18]. Total Petroleum Hydrocarbon releases into the environment will threaten public health and safety by contaminating drinking water, causing fire and explosion hazards, diminishing air and water quality, compromising agriculture, destroying recreational areas, destroying

habitats and food, and wasting non-renewable resources. Once the soil is polluted by petroleum hydrocarbons the recovery may take several years. Health effects from exposure to Total Petroleum Hydrocarbons depend on many factors. These include the types of chemical compounds in the Total Petroleum Hydrocarbon, how long the exposure lasts, and the amount of the chemicals contacted. Very little is known about the toxicity of many Total Petroleum Hydrocarbons compounds. Until more information is available, information about health effects of Total Petroleum Hydrocarbon must be based on specific compounds or petroleum products that have been studied.

The compounds in different Total Petroleum Hydrocarbon fractions affect the body in different ways. Some of the Total Petroleum Hydrocarbons compounds, particularly the smaller compounds such as benzene, toluene, and xylene (which are present in gasoline), can affect the human central nervous system. If exposures are high enough, death can occur. Breathing toluene at concentrations greater than 100 parts per million (100 ppm) for more than several hours can cause fatigue, headache, nausea, and drowsiness. When exposure is stopped, the symptoms will go away. However, if someone is exposed for a long time, permanent damage to the central nervous system can occur. One Total Petroleum Hydrocarbon compound (n-hexane) can affect the central nervous system in a different way, causing a nerve disorder called "peripheral neuropathy" characterized by numbness in the feet and legs and, in severe cases, paralysis. This has occurred in workers exposed to 2,500 ppm of n-hexane in the air. Swallowing some petroleum products such as gasoline and kerosene causes irritation of the throat, stomach, central nervous system, depression, difficulty in breathing and pneumonia from breathing liquid into the lungs. The compounds in some Total Petroleum Hydrocarbon fractions can also affect the blood, immune system, liver, spleen, kidneys, developing fetus, and lungs. Certain Total Petroleum Hydrocarbon compounds can be irritating to the skin and eyes. Other Total Petroleum Hydrocarbon compounds, such as some mineral oils, are not very toxic and are used in foods.

To protect the public from the harmful effects of toxic chemicals and to find ways to treat people who have been harmed, scientists use many tests.

One way to see if a chemical will hurt people is to learn how the chemical is absorbed, used, and released by the body; for some chemicals, animal testing may be necessary. Animal testing may also be used to identify health effects such as cancer or birth defects. Without laboratory animals, scientists would lose a basic method to get information needed to make wise decisions to protect public health. Scientists have the responsibility to treat research animals with care and compassion. Laws today protect the welfare of research animals, and scientists must comply with strict animal care guidelines. Animal studies have shown effects on the lungs, central nervous system, liver, kidney, developing fetus, and reproductive system from exposure to Total Petroleum Hydrocarbon compounds, generally after breathing or swallowing the compounds.

One TPH compound (benzene) has been shown to cause cancer (leukemia) in people. The International Agency for Research on Cancer (IARC) has determined that benzene is carcinogenic to humans (Group 1 classification). Some Total Petroleum Hydrocarbon compounds or petroleum products such as benzo (a) pyrene and gasoline, are considered to be probably and possibly carcinogenic to humans (IARC Groups 2A and 2B, respectively) based on cancer studies in people and animals. Most of the other Total Petroleum Hydrocarbons compounds and products are considered not classifiable (Group 3) by IARC.

II. MATERIALS AND METHODS

2.1 Study Area

Abonnema wharf is situated in Rivers State, within the southern part of Nigeria. It is an oil producing area in Rivers State which shares boundary with Degema and Rumuoji etc. Abonnema wharf is located in the Niger Delta basin between Latitude 4^o46'N and Longitude 7^o12'E with an area of 108km² [19]. The area is characterized by alternate wet and dry season, the rainy season starts in April to October with peak in September. It experiences rainfall of about 250cm which diminishes in October. The dry season lasts from November to March with the periodic dust-laden harmattan wind between December and February. The presence of flow stations including numerous pipelines render the area highly vulnerable to oil spills.

2.2 Collection of Samples

Samples were gotten from Abonnema Wharf River from three different points for this project and the samples were used for the purpose of the characterization of aqueous petroleum hydrocarbon pollution in the above river. The water sample of 100mL was used for each coagulant dosage. The analytical test procedures used to investigate the surface water petroleum contamination was total petroleum hydrocarbons (TPHs) determinations.

The TPHs was classified into surrogated fractions (C8 – C10, C11 – C28 and C29 – C40) characterised by similar adverse health effects, toxicological information, chemical and physical properties; in accordance to the guidelines developed by the Agency for Toxic substances and Disease Registry [12], Environmental Protection Agency (EPA) [20], TPH Criteria working Group (TPHCWG) [21], U.S Department of

Environmental Quality (DEQ), [22], and American Petroleum Institute (API) for ease of quantification and interpretation.

The representative extract (sample) of total petroleum hydrocarbons was analytically identified and quantified using Gas chromatography mass spectrometry detection (GC-MSD) Agilent Technologies 7890A in adherence to the standard analytical method of U.S EPA 8270;625. The instruments setting and operational conditions were done in accordance with the manufacturers specifications.

Chemicals: Sodium sulphate anhydrous salt (Na₂SO₄), Dichloromethane, O-terphenyl.

Equipment: The equipment used is called ‘Gas chromatography’.

2.3 Experimental Procedures

2.3.1 Parameters Analyzed:

- i. Total Petroleum Hydrocarbon (TPH)

Total Petroleum Hydrocarbon (TPH):

- i. 100ml of water was weighed
- ii. Sample was dried with sodium sulfate anhydrous salt (Na₂SO₄).
- iii. Dichloromethane solvent was added (30ml) and spiked with O-Terphenyl standard (25µl)
- iv. Shacked for 30 minutes at 120 revolutions per minute.
- v. Poured into separatory funnel and allowed to stand for 30 minutes.
- vi. The mixture was separated and the solvent injected into the GC.
- vii. Packed into the GC vials
- viii. Final analysis done using GC-FID for both the Total Petroleum Hydrocarbon (TPH) Final analysis done using GC-FID for both the Total Petroleum Hydrocarbon (TPH).

2.4 Modelling of Total Petroleum Hydrocarbon contamination

The Michaelis-Menten Equation was employed in modeling the Total Petroleum Hydrocarbon (TPH) concentration contamination [23].

Basically,

$$V = \frac{\text{Change in hydrocarbon concentration (ppm)}}{\text{Change in time period}} \quad (1)$$

Michaelis-Menten Equation is given by:

$$V = \frac{V_{\max} [S]}{k_s + [S]} \quad (2)$$

[S] is the substrate concentration (ppm), k_s is the dissociation constant. V_{max} is the maximum specific rate or velocity of substrate. V is the velocity of the substrate at any time.

The linearized form of the Michaelis-Menten Equation can be rewritten as:

$$\frac{1}{V} = \frac{K_s}{V_{\max}} \cdot \frac{1}{[S]} + \frac{1}{V_{\max}} \quad (3)$$

Where $\frac{K_s}{V_{\max}}$ represents the slope of the graph and $\frac{1}{V_{\max}}$ represents the intercept on the vertical axis.

$V_{\max} = \frac{1}{\text{intervept}}$ and $K_s = \text{slope} * V_{\max}$ from the graph.

2.4.1 MATLAB program for Total Petroleum Hydrocarbon contamination

```
clear all;
clc;
syms S; syms V;
n=input('Enter number of observations:');
C8=zeros(n,1);
C12=zeros(n,1);
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C16=zeros(n,1);
C25=zeros(n,1);
S8=zeros(n-1,1);
S12=zeros(n-1,1);
S16=zeros(n-1,1);
S25=zeros(n-1,1);
V8=zeros(n-1,1);
V12=zeros(n-1,1);
V16=zeros(n-1,1);
V25=zeros(n-1,1);
x1=zeros(n-1,1);x2=zeros(n-1,1);x3=zeros(n-1,1);
x4=zeros(n-1,1);
y1=zeros(n-1,1);y2=zeros(n-1,1);y3=zeros(n-1,1);
y4=zeros(n-1,1);
for i=1:1:n
    TEST=i
    C8(i)=input('Enter the concentration of C8:');
    C12(i)=input('Enter the concentration of C12:');
    C16(i)=input('Enter the concentration of C16:');
    C25(i)=input('Enter the concentration of C25:');
end
for i=1:1:n-1
    S8(i)=C8(i+1);
    x1(i)=1/S8(i);
    V8(i)=C8(i+1)-C8(i);
    S12(i)=C12(i+1);
    x2(i)=1/S12(i);
    V12(i)=C12(i+1)-C12(i);
    S16(i)=C16(i+1);
    x3(i)=1/S16(i);
    V16(i)=C16(i+1)-C16(i);
    S25(i)=C25(i+1);
    x4(i)=1/S25(i);
    V25(i)=C25(i+1)-C25(i);
end
table(S8,V8)
table(S12,V12)
table(S16,V16)
table(S25,V25)
subplot(4,1,1)
grid off;
P1=polyfit(1/S8,1/V8,1);
slope=P1(1)
intercept=P1(2)
Vmax=1/intercept
Ks=slope*Vmax
V=Vmax*S/(Ks+S) %Michaelis-Menten Model
hold on;
x1;
y1=x1*P1(1)+P1(2);
table(x1,y1)
plot(x1,y1,'b-*')
xlabel('x1 (per ppm)')
ylabel('y1 (time period per ppm)')
title('C8 @ Total Petroleum Hydrocarbon (TPH)')
legend('C8 substrate-TPH specific rate')
subplot(4,1,2)
P2=polyfit(1/S12,1/V12,1);
slope=P2(1)

```

```

intercept=P2(2)
Vmax=1/intercept
Ks=slope*Vmax
V=Vmax*S/(Ks+S) %Michaelis-Menten Model
hold on;
x2;
y2=x2*P2(1)+P2(2);
table(x2,y2)
plot(x2,y2,'m-*)
xlabel('x2 (per ppm)')
ylabel('y2 (time period per ppm)')
title('C12 @ Total Petroleum Hydrocarbon (TPH)')
legend('C12 substrate-TPH specific rate')
subplot(4,1,3)
P3=polyfit(1/S16,1/V16,1);
slope=P3(1)
intercept=P3(2)
Vmax=1/intercept
Ks=slope*Vmax
V=Vmax*S/(Ks+S) %Michaelis-Menten Model
hold on;
x3;
y3=x3*P3(1)+P3(2);
table(x3,y3)
plot(x3,y3,'k-*)
xlabel('x3 (per ppm)')
ylabel('y3 (time period per ppm)')
title('C16 @ Total Petroleum Hydrocarbon (TPH)')
legend('C16 substrate-TPH specific rate')
subplot(4,1,4)
P4=polyfit(1/S25,1/V25,1);
slope=P4(1)
intercept=P4(2)
Vmax=1/intercept
Ks=slope*Vmax
V=Vmax*S/(Ks+S) %Michaelis-Menten Model
hold on;
x4;
y4=x4*P4(1)+P4(2);
table(x4,y4)
plot(x4,y4,'r-*)
xlabel('x4 (per ppm)')
ylabel('y4 (time period per ppm)')
title('C25 @ Total Petroleum Hydrocarbon (TPH)')
legend('C25 substrate-TPH specific rate')

```

III. RESULTS AND DISCUSSION

3.1 Presentation of Results

The data of TPHs and Summary of concentration (ppm) of TPH detected in the surface water are shown in Tables 1 and 2 respectively. The values of the TPHs in Table 2 were identified and quantified by classification of surrogated standard fractions (C8 – C10, C11 – C28 and C29 – C40) that were characterised by similar adverse health effects, toxicological information, chemical and physical properties; in accordance to the guidelines developed by the (ATSDR), (EPA), (TPHCWG), (DEQ) and (API).

Table 1: Summary of Concentration (ppm) of TPH Detected

S/N	Group Name	Concentration (ppm)					
		Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
1	C8	93.4	87.3	78.2	74.3	66.1	52.7
2	C9	-	46.1	39.7	28.6	-	-
3	C11	117.6	108.4	96.3	81.2	74.9	63.2

4	C12	142.1	135.7	116.8	102.5	99.2	95.5
5	C13	-	46.4	-	31.6	28.5	-
6	C14	226.9	207.8	-	134.1	-	113.4
7	C16	205.8	178.3	136.2	112.2	75.4	48.7
8	C18	151.2	-	124.6	93.8	88.8	77.1
9	Ph	436.7	351.9	-	136.2	101.6	98.7
10	C20	49.5	36.0	23.9	-	18.2	-
11	C21	71.2	63.6	-	42.4	-	29.2
12	C22	84.8	82.8	81.3	79.5	83.8	52.6
13	C25	217.2	189.4	119.7	81.3	51.9	37.6

Table 2: Total Petroleum Hydrocarbons (TPHs) Concentrations for Raw Petroleum contaminated surface water

Components	Categories 1TPHCWG 2USEPA/ 3DEQ/ 4API	Concentrations (mg/l)
C8 - C10	+GRO	1397.53
C11 - C28	++DRO	102305.63
C29 - C40	+++EDRO	1059.25
C8 - C40	TPH	104762.42
DPR/EGASPIN (MCL)	TPH	10.00

3.2 Results of MATLAB Program Model

The reciprocal of specific rate, $\frac{1}{V}$ was plotted against the reciprocal of substrate concentration of hydrocarbon,

$\frac{1}{[S]}$ in MATLAB for C8, C12, C16 and C25. The results are displayed graphically below:

The graphical representation of C8 hydrocarbon gives the following results:
slope=1.6287;

intercept=-0.0187

Vmax=-53.6000; Ks=-87.3000

Michaelis-Menten equation for C8 via MATLAB is: $V = -(268*S)/(5*(S - 873/10))$

The graphical representation of C12 hydrocarbon gives the following results:
slope = 1.7950; intercept = -0.0132

Vmax = -75.6000; Ks = -135.7000

Michaelis-Menten equation for C12 via MATLAB is: $V = -(378*S)/(5*(S-1357/10))$

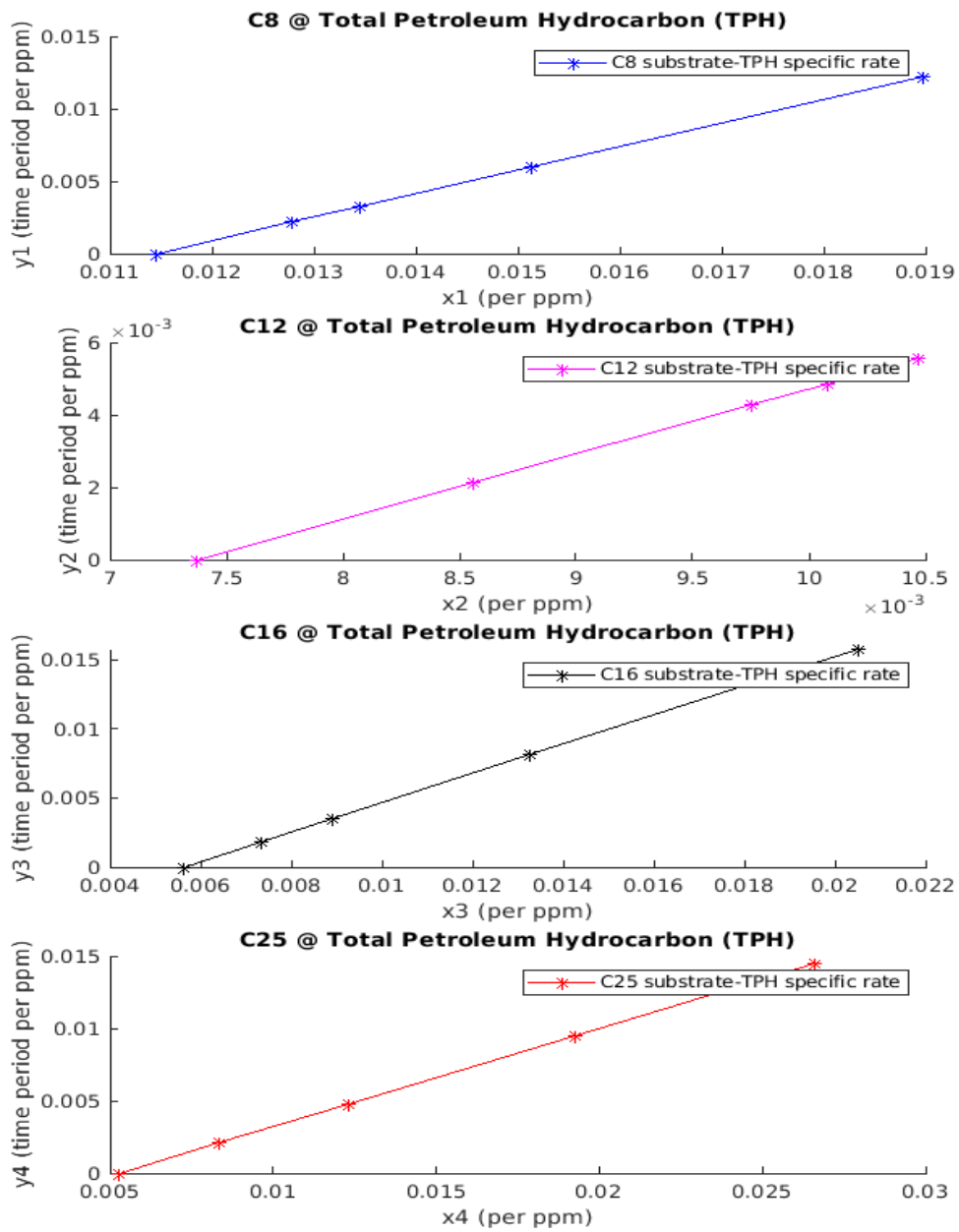


Figure 1: TPH graphical representation for C8, C12, C16, and C25 contaminations

The graphical representation of C16 hydrocarbon gives the following results:

slope = 1.6287; intercept = -0.0187

Vmax = -53.6000; Ks = -87.3000

Michaelis-Menten equation for C16 via MATLAB is: $V = -(268*S)/(5*(S - 873/10))$

The graphical representation of C25 hydrocarbon gives the following results:

slope=0.6793;

intercept=-0.0036

Vmax=-278.8000;

Ks=-189.4000

Michaelis-Menten equation for C25 via MATLAB is: $V = -(1394 * S) / (5 * (S - 947/5))$

IV. CONCLUSION

The elevated levels of Total Petroleum Hydrocarbons (TPHs) from Abonnema wharf have provided evidences of severe petroleum contamination as compared with the international certified relevant regulatory agencies standards. The levels are high enough to cause public health concerns, since residents may ingest, inhale or directly exposed to this contaminants by skin contacts. Also livestock and plants can bio accumulate these contaminants and magnify them along the food chain leading to man. On a regular basis, analytical techniques for TPH in water are being developed and diverse factors are responsible for this tests carried out. Top on this list of factors is the increasingly persistent demand for more profitable and simple environmental diagnostic tools capable of generating reliable data on a time scale. From this study, it is obvious that there is an analytical technique that is problem-free although, some are less problematic than others. This is why efforts have been made in this study to extract as much relevant information from the open access literature as possible on the application and analytical techniques in general, in the detection of petroleum contamination in water. From this study, Total concentration ranged from 9.5276 ppm to 223.8895 ppm. The Michaelis-Menten model from the MATLAB program shows that the petroleum contamination transition levels are linear and have positive trends for C8, C12, C16 and C25 hydrocarbons.

4.1 Recommendations

Many analytical methods are available for the TPH measurement in the environmental samples. Some measure TPH on the basis of the aliphatic or aromatic fraction alone while few others combine the two fractions for the measurement. The results taken using different methods are therefore in most cases incomparable owing to their different background. However, off-site laboratory equipment is more preferred for the analytical determination of the analyses than the portable field devices because it generates good data which are much more reliable. The TPH concentrations were found to be extremely higher in the spillage sites and greatly industrialized areas. Therefore, it is highly recommended that human health and ecological risk assessments be carried out as a follow up of this study. This will provide the additional relevant information needed for an effective clean-up of the Abonnema wharf. Hence, there is a greater need for compliance with environmental laws as regards the oil exploration and transportation, as well as waste discharge into the marine environment in order to ensure safety of aquatic and human lives. This paper has helped in understanding Total Petroleum Hydrocarbon (TPH) to Human Health, Agriculture and Aquatic Life.

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