THE INTERNATIONAL JOURNAL OF
SCIENCE & TECHNOLEDGE

Mathematical Modelling of the Remediation of Total Petroleum Hydrocarbon in Soil Samples Case Study: Shell Spill Site

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Abstract:
An oil spill is the release of a liquidpetroleumhydrocarbon into the environment, especially marine areas, due to human activity, and is a form of pollution. Environmental pollution is a common hazard in the Niger Delta Region. It is largely due to crude oil exploration and exploitation in the area. This work covered model of variation of total petroleum hydrocarbon (TPH) parameters with no deep consideration and performance of the experimental analysis of the parameters. Data from analyses done on different soil samples gotten from Shell Petroleum spill site were used in the modelling so as to validate mathematically, the field data. It also improved some statistical analysis of soil parameters by overcoming statistical limitations in the correlation of parameters. The study also confirmed and evaluated the possibility that a smaller group of soil sample parameter provide sufficient information for soil remediation assessment. The models for the amount of total petroleum hydrocarbon in the four samples were-0.005x^2+ 1.747x+2.581, 7.964e 0.064x, 7.266e0.068x and 10.56e 0.069x, respectively, where x is the retention time of TPH in the soil samples.

Keywords: *Oil spill, modeling, remediation*

1. Introduction

The Major source of energy for industry and daily life are products from Petroleum-based., during the exploration, production, refining, transport, and storage of petroleum and petroleum products accidental spills and leakages occur regularly. Poor miscibility of crude oil accounts for accumulation of free oil on the surface of ground water and this may migrate laterally over a wide distance to pollute other zones very far away from the point of pollution. Industrial and municipal discharges as well as urban run-offs, atmospheric deposition and natural seeps also account for petroleum hydrocarbon pollution of the environment.The contamination of soil by these crude oil and petroleum products has become a serious problem that represents a global concern for the potential consequences on ecosystem and human health (Onwurah et al., 2007).

Crude oil spills have caused great negative impact on food productivity and affect plants by creating conditions which make essential nutrients like nitrogen and oxygen needed for plant growth unavailable to them (Adam and others, 2002).Toxic components in oil may exert their effects on man through inhibition of protein synthesis, nerve synapse function, and disruption in membrane transport system and damage to plasma membrane (Prescott, et al., 1996). Crude oil hydrocarbons can affect genetic integrity of many organisms, resulting in carcinogenesis, mutagenesis and impairment of reproductive capacity (Short and Heintz, 1997). The risk of drinking water contaminated by crude oil can be extrapolated from its effect on rats that developed haemorrhagic tendencies after exposure to water soluble components of crude oil (Onwurah, 2002). Volatile components of crude oil after a spill have been implicated in the aggravation of asthma, bronchitis and accelerating aging of the lungs (Kaladumo, 1996). Other possible health effects of oil spill can be extrapolated from rats exposed to contaminated sites and these include increased liver, kidney and spleen weights as well as lipid per-oxidation and protein oxidation (Anozie and Onwurah, 2001).

Among petroleum products, diesel oil is a complex mixture of alkanes and aromatic compounds that are frequently reported as soil contaminants leaking from storage tanks and pipelines or released in accidental spills (Gallego et al., 2001). The scale of hazards imposed on the natural environment depends on the surface of the area contaminated by the petroleum products, their chemical composition, and the depth at which pollutants occur (Wolicka et al., 2009).

The technology commonly used for soil remediation includes mechanical, burying, evaporation, dispersion, and washing. However, these technologies are expensive and can lead to incomplete decomposition of contaminants (Das and Chandra, 2011). For this reason an increasing attention has been directed toward the research of new strategies and environmental-friendly technologies to be applied for the remediation of soil contaminated by petroleum hydrocarbons. Therefore, apart from the environmental problem caused by oil pollution, the agronomic and economic aspects are significant (Jobson et al., 1974; Kuhn et al., 1998).Cleanup or remediation and recovery from an oil spill is difficult and depends upon many factors, including the type of oil spilled, the temperature of the water (affecting evaporation and biodegradation), and the types of shorelines and beaches involved. Methods for cleaning up include bioremediation (use of microorganismsor biological agents to break down or remove oil), phytoremediation (the use of living green plants for the removal of contaminants and metals from soil), gas chromatography, infrared spectroscopy, controlled burning, dredging and the use of dispersants.

Total petroleum hydrocarbon (TPH) contamination is recognized as a serious threat to environmental ecosystems. TPHs are a complex mixture of chemical substances such as alkanes, aromatics and asphaltene fractions and are very toxic to living organisms. Phytoremediation has been proposed as a cost effective, non-intrusive, and environmental friendly technology for the restoration of soils contaminated with TPH.In order for remediation of TPH in soil samples to be carried out, accurate knowledge of the physico-chemical parameters of the soil must be known. These include soil pH, soil moisture levels, soil texture, electrical conductivity, salinity, soil bulk density and soil bulk density.

Bioremediation Kinetics Kinetic analysis is a key factor for understanding biodegradation process, bioremediation speed measurement and development of efficient clean up for a crude oil contaminated environment. The information on the kinetics of soil bioremediation is of great importance because it characterizes the concentration of the contaminant remaining at any time and permit prediction of the level likely to be present at some future time. Biodegradability of crude oil is usually explained by first order kinetics (Pala et al., 2006; Agarry et al., 2010b; Zahed et al., 2011) and this is given as in Eq. (1):

$$C_t = C_o e^{-kt} \quad (1)$$

Where Co is the initial TPH content in soil (mg/kg), Ct is the residual TPH content in soil at time t, (mg/kg), k is the biodegradation rate constant (day^{-1}) and t is time (day). Plotting the logarithm of TPH concentration versus time presents appropriate information about the biodegradation rate.

Biodegradation half-lives are needed for many applications such as chemical screening (Aroson et al., 2006), environmental fate modelling (Sinkkonen and Paasivirta, 2000) and describing the transformation of pollutants (Dimitrov et al., 2007; Matthies and Klasmeier, 2008). Biodegradation half-life times ($t_{1/2}$) are calculated by Eq. (2) (Yeung et al., 1997; Zahed et al., 2011; Agarry et al., 2013):

$$t½ = \frac{\text{In}2}{k} \quad (2)$$

Where k is the biodegradation rate constant (day^{-1}). The half life model is based on the assumption that the biodegradation rate of hydrocarbons positively correlated with the hydrocarbon pool size in soil (Yeung et al., 1997).

2. Methodology

2.1. Model Conceptualization

The model conceptualizes the stretch of Total petroleum hydrocarbon determination data studied in four (4) stations; 28" Nkporu-Bomu TNP at Elelenwo community, 36" Rumuekpe-Nkporu D/L at Mgodo, Aluu Community, 36" Nkpoku-Bomu F/L at Atali Community and 6" Obigbo North-Komkom D/L at Komkom Community.

The parameters: retention time of each hydrocarbon component in each sample (Rettime) and the Amount of each component in each sample (Amount) from the four stations for the modelling is tabulated below.

Sample 1: 28" Nkporu-Bomu TNP at Elelenwo community.
Sample 2: 36" Rumuekpe-Nkporu D/L at Mgodo, Aluu Community.
Sample 3: 36" Nkpoku-Bomu F/L at Atali Community
Sample 4: 6" Obigbo North-Komkom D/L at Komkom Community.

Name Group	Rettime (Minutes)	Sample 1 Amount(Ppm)	Rettime (Minutes)	Sample 2 Amount(Ppm)
C8	3.133	-	3.133	-
C9	4.614	289.98689	4.567	-
C10	6.701	103.38001	6.725	754.09696
C11	8.284	33.50863	8.349	833.50255
C12	9.767	49.66969	9.689	353.92340
C13	11.181	145.97399	11.144	1749.45425
C14	12.471	249.89316	12.511	2121.24674
C15	13.744	28.12555	13.789	673.27476
C16	15.533	539.76027	15.518	288.87757
Pristane	18.303	631.58647	18.151	560.37833
C17	18.464	631.58647	18.504	517.62852
Phytane	21.012	967.11536	21.026	950.63349

Name Group	Rettime (Minutes)	Sample 1 Amount(Ppm)	Rettime (Minutes)	Sample 2 Amount(Ppm)
C18	21.649	3116.63331	21.738	4356.74682
C19	23.535	1064.23253	23.575	958.73220
C20	25.827	1477.94817	25.838	916.66781
C21	27.304	886.27984	27.934	625.49942
C22	29.581	978.02756	29.631	534.21846
C23	31.478	606.16183	31.465	420.08992
C24	33.046	493.72997	33.059	498.56029
C25	34.599	1025.97633	34.576	744.82557
C26	36.473	1717.81572	36.141	441.73631
C27	37.582	856.67228	37.583	611.92853
C28	39.055	391.16185	39.036	321.67251
C29	40.472	1610.94366	40.470	473.00190
C30	41.740	82.46457	41.709	58.47198
C31	43.006	48.24004	43.011	47.93117
C32	44.185	56.82787	44.167	57.13137
C33	45.361	57.46829	45.361	57.76138
C34	46.546	57.91219	46.512	56.93223
C35	47.338	56.07453	47.862	54.30729
C36	49.500	58.23796	49.343	59.54750
C36	51.322	72.72885	51.313	70.87669
C38	52.322	92.31713	52.981	88.37785
C39	54.834	71.60886	54.871	62.75592
C40	57.053	124.74565	56.960	123.97062

Tables 1: Parameters of the Total Petroleum Hydrocarbon Samples Modelled

Name Group	Rettime (Minutes)	Sample 3 Amount(Ppm)	Rettime (Minutes)	Sample 4 Amount(Ppm)
C8	3.133	-	7.471	2.19424e-2
C9	4.567	-	10.060	1.77596e-3
C10	6.740	743.36064	12.610	-
C11	8.353	801.95294	13.241	-
C12	9.693	322.89550	14.969	-
C13	11.143	1667.47583	15.228	-
C14	12.509	2015.19639	17.403	-
C15	13.782	661.90927	17.774	-
C16	15.698	1156.41393	20.186	-
Pristane	18.141	694.34124	28.605	2.53288e-9
C17	18.494	543.06134	28.730	-
Phytane	21.036	941.04979	30.455	-
C18	21.737	4268.91426	30.634	-
C19	23.578	900.36777	32.220	-
C20	25.838	1474.50399	33.903	-
C21	27.923	510.67943	35.519	-
C22	29.638	527.03252	37.067	-
C23	31.480	922.73104	38.554	-
C24	33.053	386.75914	39.983	-
C25	43.725	285.33151	41.358	-
C26	36.235	380.11716	42.682	-
C27	37.591	301.02766	43.962	-
C28	39.104	37.79009	45.203	-
C29	40.463	60.08606	45.647	-
C30	41.726	69.71040	46.396	-
C31	42.986	47.47585	47.560	-
C32	44.202	49.32558		

Name Group	Rettime (Minutes)	Sample 3 Amount(Ppm)	Rettime (Minutes)	Sample 4 Amount(Ppm)
C33	45.380	55.43551		
C34	46.522	54.06229		
C35	47.871	55.17828		
C36	49.401	63.27888		
C36	51.293	72.19585		
C38	52.984	94.03612		
C39	54.838	73.21136		
C40	57.003	122.46252		

Tables 2: Parameters of the Total Petroleum Hydrocarbon Samples Modelled

Statistical analysis of Total Petroleum hydrocarbon (TPC) data maybe very broadly classified into two groups:

- Descriptive statistics
- Inferential statistics

These classifications suggest that initially the task is to provide an accurate and reliable statistical description of the data set. Following that, it may be possible to give some account of how well the data set and/or its interactions conforms to some theories as to its origin, and so possibly identify potential means of modification. Therefore the data obtained was analyzed using method of least square which would fit two of the parameters by linear, quadratic, exponential, and logarithmic regression models so as to determine the best fit for the pair of the data set.
The simplest type of approximating curve is a straight line which can be represented by the equation.
$Y = a_o + a_1x$... ... (3)
Finding the estimates of a_o and a_1 such that the line forms a good fit to the data, method of least square was applied.
The "goodness of fit" of the given data set is provided by the sum of the deviations. That is,
$p_1{}^2 + p_2{}^2 +$.. $p_n{}^2$, which must be minimum. Thus:
SUM = $\sum_{i=1}^{n} p_n{}^2$ = minimum... (4)
Mathematically, equation (4) is described as
sum = s = $\sum_{i=1}^{n}(p_1{}^2)$
$\sum_{i=1}^{n}[y_i - (a_0 + a_1x_i)]^2$... (5)
Minimizing equation (5) to obtain a_o and $a_1$by taking the partial derivatives of S with respect to a_o and a_1, and setting them equal to zero respectively,
$\frac{ds}{da_o} = \sum 2(y_i\, a_o + a_1x_1)(-x_i) = 0$....................................... (6)
$\frac{ds}{da_1} = \sum 2(y_i - a_o - a_ix_i)(-x_i) = 0$ .... (7)
Rearranging equation (6) and (7) gives:
$na_o + a_i \sum x_i = \sum y_i$ .... (8)
$a_o \sum x_i + a_1 \sum x_i{}^2 = \sum x_i\, y_i$ ....(9)
Solving the above equations simultaneously by matrix techniques, $\begin{bmatrix} n & \sum x_i \\ \sum x_i & \sum x_i{}^2 \end{bmatrix}\begin{pmatrix} a_o \\ a_1 \end{pmatrix} = \begin{pmatrix} \sum y_i \\ \sum x_iy_i \end{pmatrix}$ ....(10)
Applying Gaussian Elimination to equation (10), divide row 1 by n $\begin{pmatrix} 1 & \frac{\sum x_i}{n} \\ \sum x_i & \sum x_i{}^2 \end{pmatrix}\begin{pmatrix} a_o \\ a_1 \end{pmatrix} = \begin{pmatrix} \sum\frac{y_i}{n} \\ x_iy_i \end{pmatrix}$
Multiply row 1 by -$\sum$ x, and add same to row 2,
$$\begin{bmatrix} 1 & \frac{\sum x_i}{n} \\ 0 & \sum x_i - \frac{(\sum x_i)^2}{n} \end{bmatrix}\begin{pmatrix} a_o \\ a_1 \end{pmatrix} = \begin{pmatrix} \frac{\sum x_i}{n} \\ \sum x_iy_i - \frac{\sum x_i \sum y_i}{n} \end{pmatrix} \quad (11)$$
Performing backward substitution on equation (11):
$$\left[\sum x_i{}^2 - \frac{\left(\sum x_i{}^2\right)}{n}\right] a_i = \sum x_i\, y_i - \sum x_i \sum y_i.$$
$$a_1 = \frac{\sum x_iy_i - \sum x_iy_i}{\sum xi^2 - \frac{(xi^2)}{n}}.$$
$$a_1 = \frac{n\sum x_i \sum y_i - \sum x_i \sum y_i}{n\sum x_i{}^2 - (\sum x_i)^2} \quad (12)$$
From equation (8), making a_o the subject of the formular result to:
$$a_o = \frac{\sum y_i - a_i \sum x_i}{n} \quad (13)$$
Equations (12) and (13) are used to find the constants parameters in linear regression of two set of the variables.
Now consider a polynomial of the form:
$y = a_o + a_1x + a_2x^2$ .... (14)

Given n set of measurements; (y_1, x_1), (y_2, x_2), (y_3, x_3) .... (y_n, x_n), The least square estimates of a_o, a_1, and a_2 are obtained in a similar way to that previously presented for linear regression. The sum of squared deviations of the observed values of y from the predicted values is given by:

$S = \sum(y = a_o - a_i x - a_2 x^2)^2$.. (15)

Minimizing equation by setting its partial derivatives with respect to a_1, a_2, and a_3 and equate to zero to obtain the following three simultaneous equations,

$\sum y = na_o + a_1 \sum x + a_2 \sum x^2$.

$\sum xy = a_o \sum x + a_1 \sum x^2 + a_2 \sum x^3$.

$\sum x^2 y = a_o \sum x^2 + a_1 \sum x^3 + a_2 \sum x^3$.................................... (16)

Solving equation (16) above for a_o, a_1 and a_2 gives a quadratic regression model of two set of variables. For goodness of fit,

$r^2 = \frac{\sum(y_{east} - \ddot{y})^2}{\sum(y - \ddot{y})^2}$. where y_{est} = estimated value of y from a regression equation.

$y = \frac{\text{observed}}{\text{actual}}$ value of independent variables.

$\ddot{y}$ = mean of independent variables.

Taking:

$y_{east} = a_o + a_1 x$.

$\ddot{y} = \frac{\sum y}{n}$ and,

n = number of observations,

Then for a linear model,

$$r^2 = \frac{\sum\left(a_o + a_1 x - \frac{(\sum y)^2}{n}\right)}{\sum\left(y - \frac{(\sum y)^2}{n}\right)} \quad (17)$$

Multiplying both the numerator and the denominator by y and summing the individual terms which takes care of the squared bracket, results as:

$$r^2 = \frac{a_o \sum y + a_1 \sum xy - \frac{(\sum y)^2}{n}}{\sum y^2 - \frac{(\sum y)^2}{n}} \quad (18)$$

similarly, for a quadratic regression model,

$$r^2 = \frac{a_o \sum y + a_1 \sum xy + a_2 \sum x^2 y - \frac{(\sum y)^2}{n}}{\sum y^2 - \frac{\sum y^2}{n}} \quad (19)$$

Equation (17) and (18) are used to determine the coefficients of regression of two set of parameter variables. Owing to computational and approximation errors, Microsoft excel was applied to obtain the graph of the four set of the parameters by plotting Amount against Retention time using Linear, quadratic, logarithmic and exponential curves with the corresponding regression models and coefficient of correlation determined.

3. Results and Discussion

The results for each sample analysis on the total petroleum hydrocarbon data are presented in Table 1 and Table 2 respectively. The measured data characteristics of the different hydrocarbon samples indicated that:

- The Retention time which is the time it takes for each component of the Total petroleum hydrocarbon to be detected in each sample from C8-C40 increases with time.
- The concentration of the hydrocarbon in each sample varied accordingly and also varies with the retention time.

The Total petroleum hydrocarbon parameters given in Table 1 were subjected to statistical mathematical model using data from sampling test at the stations. The regression relationships were made between Amount (ppm) and Retention time (min).

Model Type	Sample 1	R^2
Linear	1.559x + 3.707	0.992
Polynomial*	$-0.005x^2 + 1.747 + 2.581$	0.998
Exponential	$8.650e^{0.063x}$	0.900
Logarithmic	16.69ln(x)-12.43	0.853
	Sample 2	
Linear	1.561x+2.139	0.997
Polynomial*	$-0.006x^2 + 0.35x + 2.47$	0.998
Exponential	$7.964e^{0.064x}$	0.901
Logarithmic	19.41ln(x)-21.85	0.899
	Sample 3	
Linear*	1.570x + 2.246	0.986

Model Type	Sample 1	R^2
Polynomial	$-0.009x^2+1.889x+0.325$	0.849
Exponential	$7.266e^{0.068x}$	0.868
Logarithmic	17.30In(x) -15.04	0.785
	Sample 4	
Linear	1.788x+ 6.373	0.978
Polynomial*	$0.017x^2 + 2.198+ 4.732$	0.981
Exponential	$10.56e^{0.069x}$	0.907
Logarithm	14.43In(x) – 4.107	0.880

Table 3: Summary of the Different Models
(Implies Best Fit Model)*

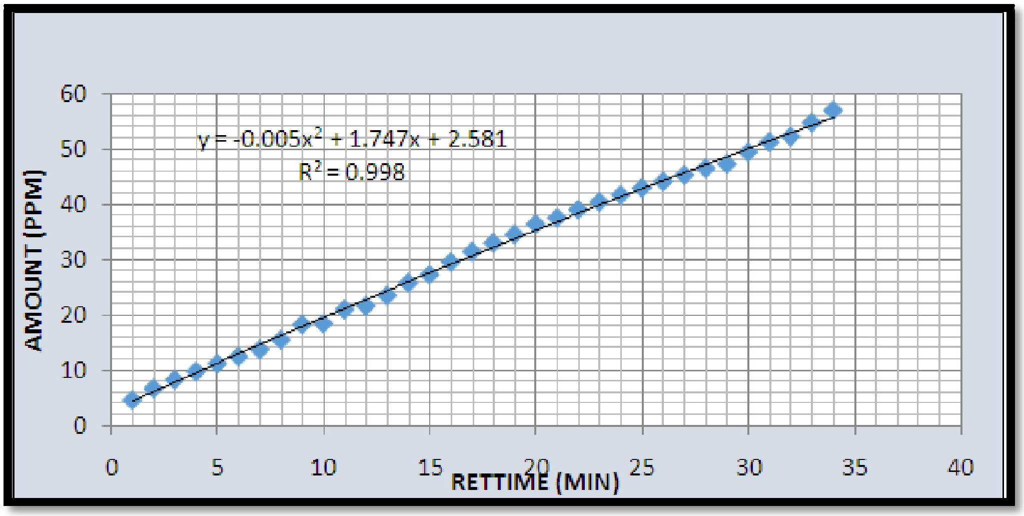

Figure 1: Plot of Amount versus Rettime for Sample 1

Comparison of the different models of Amount against time for Sample 1 shows that the best fit for the two parameters is the polynomial model (quadratic). Therefore the Amount against time of the different hydrocarbon samples of the area was related by:

Amount = $-0.005x^2 + 1.747x + 2.581$... (20)

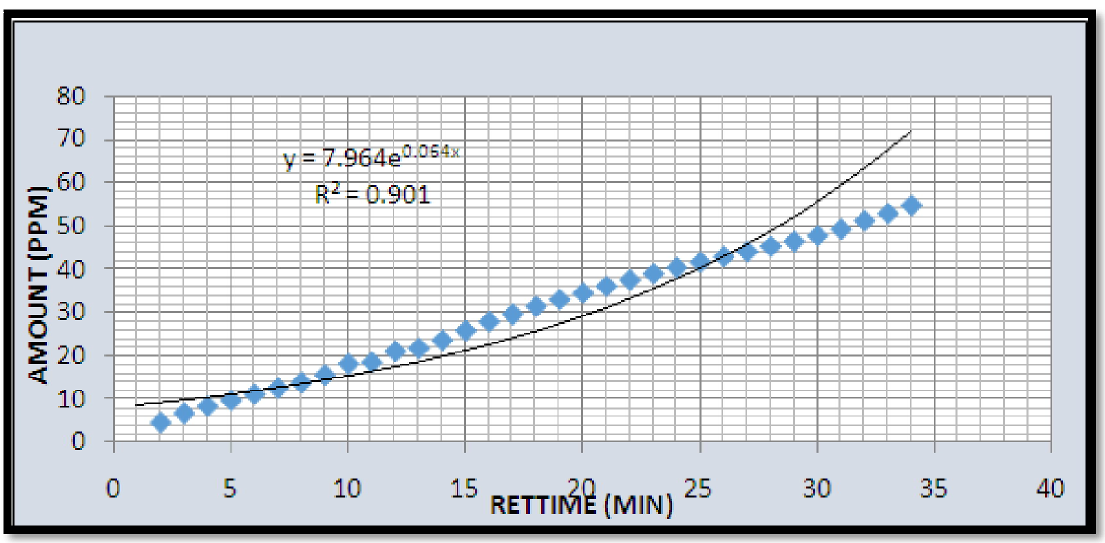

Figure 2: Plot of Amount versus Rettime for Sample 2

Comparison of the different models for Amount against time gave exponential model as the best fit: Thus,
Amount=$7.964e^{0.064x}$ .... (21)

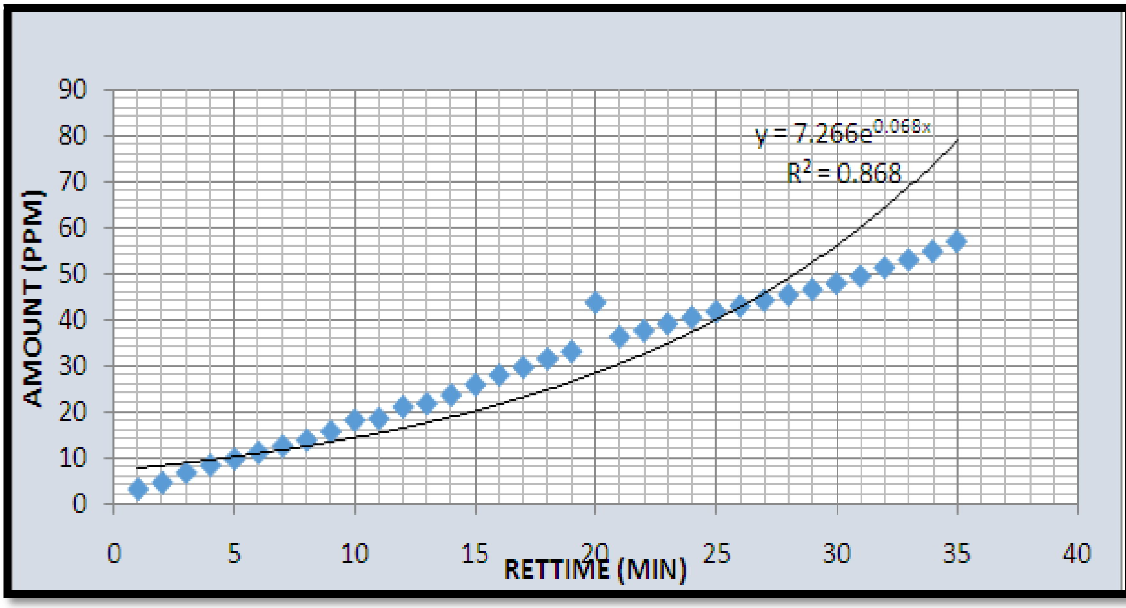

Figure 3: Plot of Amount versus Rettime for Sample 3

By comparison, the exponential model best fit Amount against time Thus:
Amount = $7.266e^{0.068x}$……………………………………………. (22)

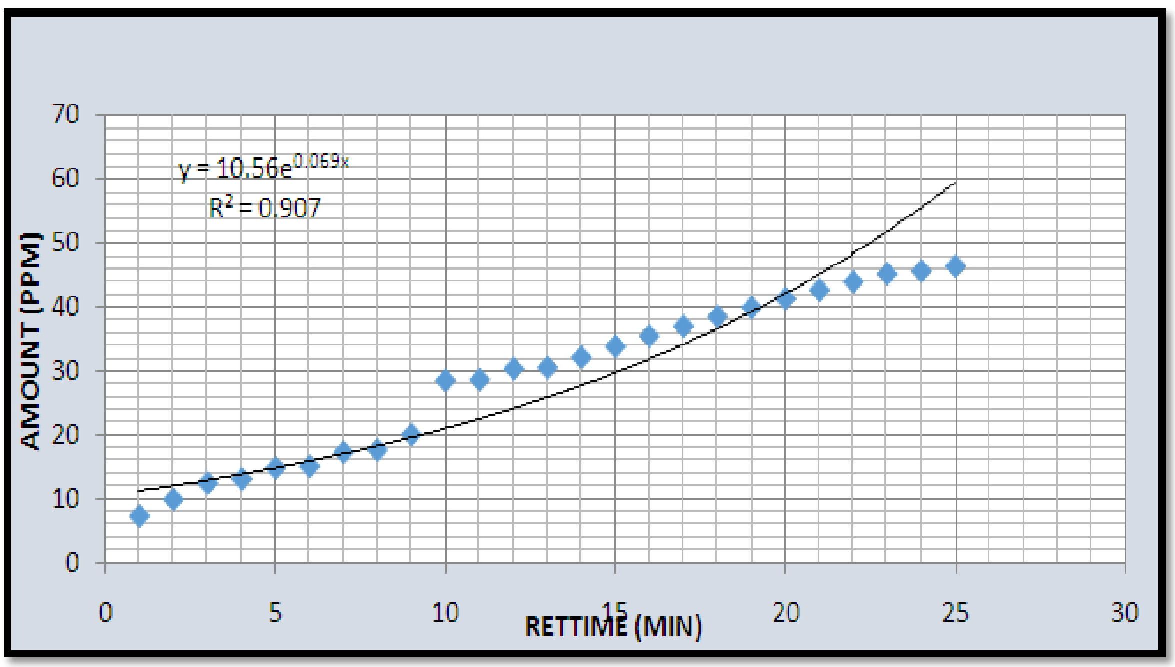

Figure 4: Plot of Amount versus Rettime for Sample 4

By comparison, the exponential model best fit Amount against time Thus.
Amount =$10.56e^{0.069x}$……………………………………………. (23)

3.1. The Test of the Models

The validity of the predicted model is verified by analysis of variance (ANOVA). The second-order model quality is assessed by the correlation factor (R^2) and the result analysis is carried out using regression model value (with 78-99% confidence level). Finally, the optimal values of tested variables are obtained by analyzing the surface curves and regression equation optimization. Standard shows that some of the activities such as dumping of refuse, defecation, and oil spillage (from pipeline) rendered the four sample sites unfit except they are treated.

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