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# Monitoring of Total Petroleum Hydrocarbon Degradation in Stagnant Water Media with the Application of First Order Kinetics

Ukpaka CP, Okirie FU

**ABSTRACT**

Prediction of total petroleum hydrocarbon was monitored in a stagnant water media of salt and fresh was examine with the application of first order kinetics. The research demonstrates the factors that influence the total petroleum hydrocarbon degradation rate to the physicochemical parameters as well as the microbial activity. However, the degradation rate of the TPH was more in salt water than fresh water medium as revealed in this investigation. The developed model was resolved using the necessary boundary conditions and the result obtained demonstrates the relationship between the  $\ln(CTPH)$  with variation in time. This research has revealed that first order kinetics is a good tools for monitoring, predicting and simulation of total petroleum hydrocarbon degradation in stagnant water environment under the influence of sedimentation process.

**Key words:** Monitoring, total petroleum hydrocarbon, degradation, stagnant, water media, application, first order kinetics

**1. INTRODUCTION**

The degradation of petroleum hydrocarbon has been investigated in a moving stream as well as stagnant water environment and the effect of dispersion [1-4]. However, the effect of physicochemical properties on the degradation of petroleum hydrocarbon in water environment has been studied and their obtained results revealed that the parameters of the physical and chemical characteristics influence the microbial growth by inhibiting the active site of the organism [5-8].

Temperature, oxygen, contaminants concentration and other parameters has been be considered as a contributing factors to microbial growth and decline in the organism, especially when the environment is not favourable for microbes to catalyse the reaction [9-10].

The dispersion of the petroleum hydrocarbon in a stagnant water environment is controlled with force of gravity and disturbers, which may be

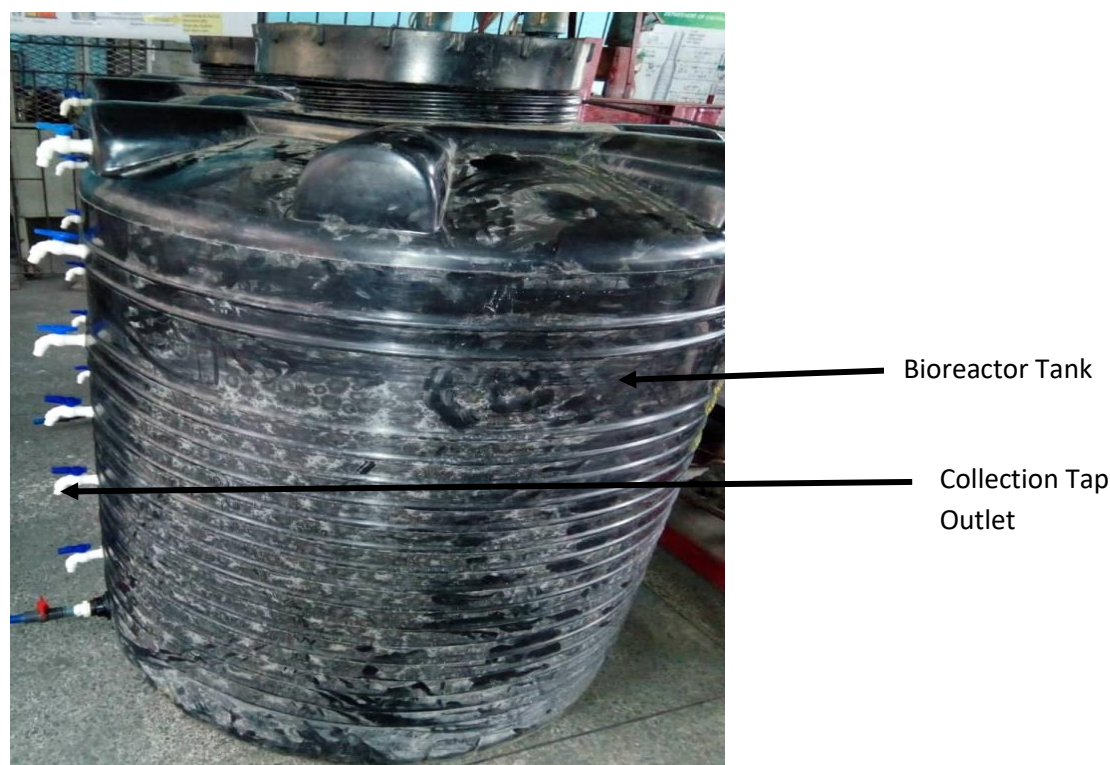
attributed the environment factors as revealed by different research groups [11-15]. The action of the microbes also enhance dispersion of contaminants in stagnant and moving stream as well as mitigate the contaminants concentration [16-17]. The degradation of the total petroleum hydrocarbon in this case was monitored as function of depth dependent and the variation in concentration reveals the integrity of the role of dispersion as related to concentration variation [18-21].

Water is a major substance use for domestic and industrial utilization and alteration of concentration, indeed, is a matter of interest as allot of researchers has demonstrated the effect of petroleum hydrocarbon on water environment [22-24]. The underground water is found to have been polluted due to dispersion of these contaminants from one level of the aquifer to the other [25-27]. Indeed, the investigation on the underground water contamination has revealed that the mechanism of crude oil dispersion and spreading has thoroughly influence the quality of the underground water not meeting the standard for domestic and industrial utilization without adequate treatment before utilization in most cases [28-31]. On this note, there is need to monitor the total petroleum hydrocarbon degradation in a stagnant water medium relation to time dependent.

## 2. MATERIALS AND METHODS

### Modelling of Total Petroleum Hydrocarbon Content

The degradation of the crude oil content, which is here expressed in terms of total petroleum hydrocarbon (TPH), is evaluated through mathematical models. First, the concentration of TPH degradation along the water surface (horizontal transport) and down the column (vertical transport), after it has been discharged onto the surface of the stagnant water, was studied using a two dimensional advection-diffusion transport model. Secondly, the settling rate of the crude oil content from the surface down the bottom of the water system was studied through a sedimentation model. Figure 1 shows a cylindrical control volume representing the water tank polluted by the crude oil sample. The crude oil diffusion in vertical (axial) and horizontal (radial) components is depicted in the hypothetical diagram.



**Figure 1: Experimental Set-Up of Cylindrical Batch Reactor**

The principle of mass balance was used to model the degradation process of TPH in stagnant water system. This general mass balance equation is given in equation (1).

$$\left\{ \begin{array}{c} \text{Mass flux} \\ \text{of} \\ \text{Component} \\ \text{into system} \end{array} \right\} = \left\{ \begin{array}{c} \text{Mass flux of} \\ \text{Component} \\ \text{from system} \end{array} \right\} + \left\{ \begin{array}{c} \text{Rate of} \\ \text{degradation} \\ \text{due to} \\ \text{reaction} \end{array} \right\} + \left\{ \begin{array}{c} \text{Rate of} \\ \text{accumulation} \\ \text{of mass} \\ \text{within system} \end{array} \right\} \quad (1)$$

$$\text{The mass flux into system} = \left( -D_r \frac{\partial C_i}{\partial r} + v_r C_i \right) + \left( -D_z \frac{\partial C_i}{\partial z} + v_z C_i \right) \quad (2)$$

The mass flux out of boundary

$$= -D_r \frac{\partial C_i}{\partial r} + v_r C + \frac{\partial}{\partial r} \left( -D_r \frac{\partial C_i}{\partial r} + v_r C_i \right) - D_z \frac{\partial C_i}{\partial z} + v_z C_i + \frac{\partial}{\partial z} \left( -D_z \frac{\partial C_i}{\partial z} + v_z C_i \right) \quad (3)$$

$$\text{Rate of TPH degradation} = R_i \quad (4)$$

$$\text{The rate of TPH accumulation} = \frac{dC_i}{dt} \quad (5)$$

Substituting equation (2) through (5) into (1) gives:

$$= -\frac{\partial}{\partial r} \left( -D_r \frac{\partial C_i}{\partial r} + v_r C_i \right) - \frac{\partial}{\partial z} \left( -D_z \frac{\partial C_i}{\partial z} + v_z C_i \right) - R_i = \frac{\partial C_i}{\partial t}$$

$$\frac{\partial C}{\partial t} = -v_r \frac{\partial C}{\partial r} - v_z \frac{\partial C}{\partial z} + D_r \frac{\partial^2 C}{\partial r^2} + D_z \frac{\partial^2 C}{\partial z^2} - R_i \quad (6)$$

Equation (6) is the model for dispersion of TPH concentration in stagnant water system.

#### Solution to the Model

The solution to the model can be obtained by either numerical or analytical techniques. In this study, the analytical method was used by adopting the transformation technique. Firstly, equation (6) is simplified by using the substitution method adopted. This is stated as:

$$X = z + r \sqrt{\left( \frac{D_r}{D_z} \right)} \quad (7)$$

By this substitution, the concentration,  $C$  is now a function of  $X$ . Hence, we have as follows:

$$\frac{\partial C}{\partial r} = \frac{\partial C}{\partial X} \cdot \frac{\partial X}{\partial r} \quad (8)$$

The direct differentiation of equation (7) with respect to  $r$  gives as follows:

$$\frac{\partial X}{\partial r} = \sqrt{\frac{D_r}{D_z}} \quad (9)$$

Now, combining equations (8) and (9) gives:

$$\frac{\partial C}{\partial r} = \sqrt{\frac{D_r}{D_z}} \frac{\partial C}{\partial X} \quad (10)$$

Similarly, if  $C$  is also a function of  $z$ , then, we have as follows:

$$\frac{\partial C}{\partial z} = \frac{\partial C}{\partial X} \cdot \frac{\partial X}{\partial z} \quad (11)$$

Again, the direct differentiation of equation (7) with respect to  $z$ , gives:

$$\frac{\partial x}{\partial z} = 1 \quad (12)$$

Hence, equation (11) becomes:

$$\frac{\partial C}{\partial z} = \frac{\partial C}{\partial X} \quad (13)$$

Further differentiation of equation (10) with respect to  $r$ , gives:

$$\frac{\partial^2 C}{\partial r^2} = \sqrt{\frac{D_z}{D_r}} \frac{\partial^2 C}{\partial X^2} \quad (14)$$

Also, further differentiation of equation (13) with respect to  $r$ , gives:

$$\frac{\partial^2 C}{\partial z^2} = \frac{\partial^2 C}{\partial X^2} \quad (15)$$

By substituting equations (10), (13), (14) and (15) into equation (6) gives as follows:

$$\frac{\partial C}{\partial t} = -v_r \sqrt{\frac{D_r}{D_z}} \frac{\partial C}{\partial X} - v_z \frac{\partial C}{\partial X} + D_r \sqrt{\frac{D_r}{D_z}} \frac{\partial^2 C}{\partial X^2} + D_z \frac{\partial^2 C}{\partial X^2} - R_i \quad (16)$$

Simplification of equation (16) gives:

$$\frac{\partial C}{\partial t} = \left( D_r \sqrt{\frac{D_r}{D_z}} + D_z \right) \frac{\partial^2 C}{\partial X^2} - \left( v_r \sqrt{\frac{D_r}{D_z}} + v_z \right) \frac{\partial C}{\partial X} + -R_i \quad (17)$$

Equation (17) is further simplified as:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial X^2} - v \frac{\partial C}{\partial X} + -R_i(C) \quad (18)$$

where:

$$D = \left( D_r \sqrt{\frac{D_r}{D_z}} + D_z \right)$$

$$v = v_r \sqrt{\frac{D_r}{D_z}} + v_z$$

Boundary conditions

$$t = 0 \quad 0 < X < \infty ; \quad C = 0 \quad (19)$$

$$t > 0 \quad X = \infty ; \quad C = 0 \quad (20)$$

$$t > 0 \quad X = 0 ; \quad C = C_o \quad (21)$$

Again, let  $\lambda$  be a function of  $X$  and  $t$

i.e  $\lambda(X, t)$ , and according to literature (Yadaw and Jaiswal, 2012; Jha *et al.*, 2012),  $\lambda$  and  $C$  are related by the transformation,

which is expressed as:

$$C(X, t) = \lambda(X, t) \exp \left\{ \frac{v}{2D} X - \left( \frac{v^2}{4D} + R_i(C) \right) t \right\} \quad (22)$$

Taking the Laplace transform of equation (14) gives:

$$\begin{aligned} s\bar{C} - C_{(o)} &= D \frac{\partial^2 \bar{C}}{\partial X^2} - V \frac{\partial \bar{C}}{\partial X} - \bar{R}_i(C) \\ \therefore s\bar{C} &= D \frac{\partial^2 \bar{C}}{\partial X^2} - V \frac{\partial \bar{C}}{\partial X} - \bar{R}_i(C) \end{aligned} \quad (23)$$

Applying the boundary condition at  $X = 0$  and  $t = 0$ , equation (22) becomes

$$C(0, 0) = \lambda(0, 0) \quad (24)$$

Thus, the Laplace transform of equation (24) gives

$$\bar{C} = \bar{\lambda} \quad (25)$$

Therefore, equation (23) can be written as

$$S\bar{\lambda} - D \frac{\partial^2 \bar{\lambda}}{\partial X^2} - V \frac{\partial \bar{\lambda}}{\partial X} - \bar{R}_i(C) \quad (26)$$

Again, when  $t = 0$  and  $\frac{\partial \lambda}{\partial X} = 0$ , equation (26) becomes

$$S\bar{\lambda} = D \frac{d^2 \bar{\lambda}}{dX^2} \quad (27)$$

It was notice that equation (27) is of the general form:

$$\frac{d^2 y}{dx^2} - n^2 y = 0 \quad (28)$$

And solution is

$$A e^{nx} + B e^{-nx} \quad (29)$$

Thus, the solution to equation (29) is

$$\bar{\lambda} = A e^{\sqrt{s/D} X} + B e^{-\sqrt{s/D} X} \quad (30)$$

Applying the boundary conditions:

When  $X = \infty$ ,  $\lambda = 0 \therefore \bar{\lambda} = 0$  and  $A = 0$

When  $X = 0$  and  $t > 0$ , we have from equation (22)

$$C_o = \lambda e^{-\left(\frac{v^2}{4D} + R_i(C)\right)t}$$

$$\therefore \lambda = C_o e^{\left(\frac{v^2}{4D} + R_i(C)\right)t} \quad (31)$$

The Laplace transform of equation (31) is

$$\bar{\lambda} = \frac{C_o}{s - \left(\frac{v^2}{4D} + R_i(C)\right)} \quad (32)$$

From equation (30), if  $A = 0$

$$\bar{\lambda} = B e^{-\sqrt{s/D} X}$$

and at  $X = 0$

$$B = \frac{C_o}{s - \left(\frac{v^2}{4D} + R_i(C)\right)} \quad (33)$$

Substituting  $B$  and  $A$  into equation (22) gives

$$\bar{\lambda}(X, s) = \frac{C_o}{s - \left( \frac{v^2}{4D} + R_i(C) \right)} e^{-\sqrt{s/D}X} \quad (34)$$

To transform the Laplace domain to the time domain, the inverse Laplace of equation (34) was taken to give

$$L^{-1}[\bar{\lambda}(X, s)] = L^{-1} \left[ \frac{C_o}{s - \left( \frac{v^2}{4D} + R_i(C) \right)} e^{-\sqrt{s/D}X} \right]$$

From Laplace table and further simplifications, we have

$$\lambda(X, t) = \frac{C_o}{2} \left\{ \operatorname{erfc} \left[ \frac{X - (v + R_i(C))t}{2\sqrt{Dt}} \right] + \operatorname{erfc} \left[ \frac{X + (v + R_i(C))t}{2\sqrt{Dt}} \right] \right\} \quad (35)$$

Substituting equation (35) into (22) gives:

$$C(X, t) = \frac{1}{2} C_o \left\{ \operatorname{erfc} \left[ \frac{X - (v + R_i(C))t}{2\sqrt{Dt}} \right] + \operatorname{erfc} \left[ \frac{X + (v + R_i(C))t}{2\sqrt{Dt}} \right] \right\} \exp \left[ \frac{v}{2D} X - \left( \frac{v^2}{4D} + R_i(C) \right) t \right] \quad (36)$$

The rate term is substituted into equation (36), which in this case, the first order degradation rate and Monod equations were tested to evaluate the TPH concentration in the stagnant water system.

### Basic Assumptions

1. The velocity term in x-direction (horizontal direction) is negligible. Hence, the dispersion of TPH in this direction is only due to diffusion.
2. In the vertical direction, the dispersion of crude oil is by convective and diffusion. Hence, both velocity and diffusion are considered.
3. The stream flow pattern is not altered by the presence of multiple contaminants in the water system.

### Boundary Conditions

The following boundary conditions are applied.

$$\begin{aligned} t = 0, \quad z = 0 \quad x = 0 \quad C_{TPH} &= C_{TPH(o)} \\ t > 0, \quad z > 0 \quad x > 0 \quad C_{TPH} &= C_{TPH(t)} \\ t = \infty \quad z = 0 \quad x = 0 \quad C_{TPH} &= 0 \end{aligned}$$

### Rate Expression

#### First order Degradation Rate

The first order TPH degradation rate is expressed as:

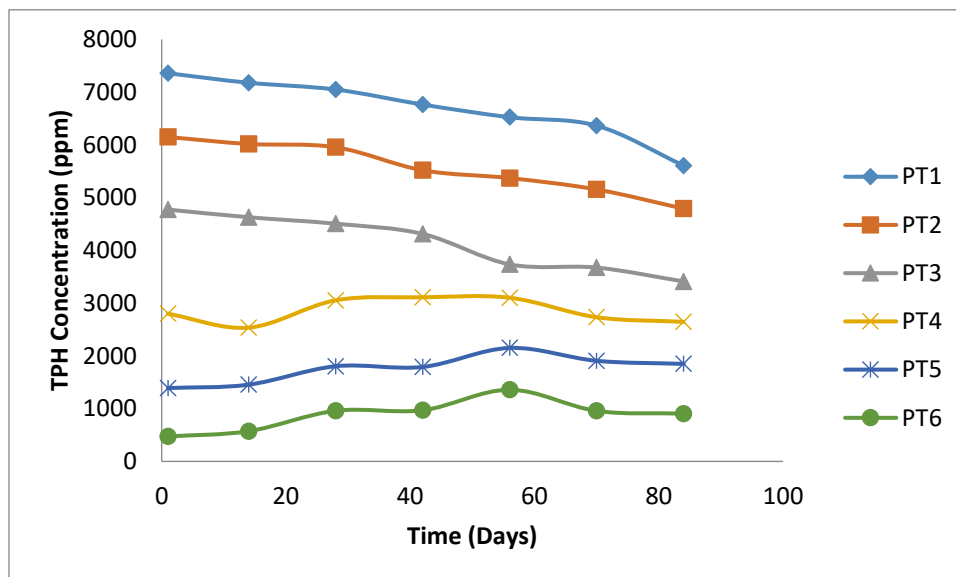
$$R_i(C) = k_d C_{TPH} \quad (37)$$

## 3. RESULTS AND DISCUSSION

The results obtained from this investigation are demonstrated in Figures as revealed below.

### Periodic Dispersion of TPH in Polluted Water

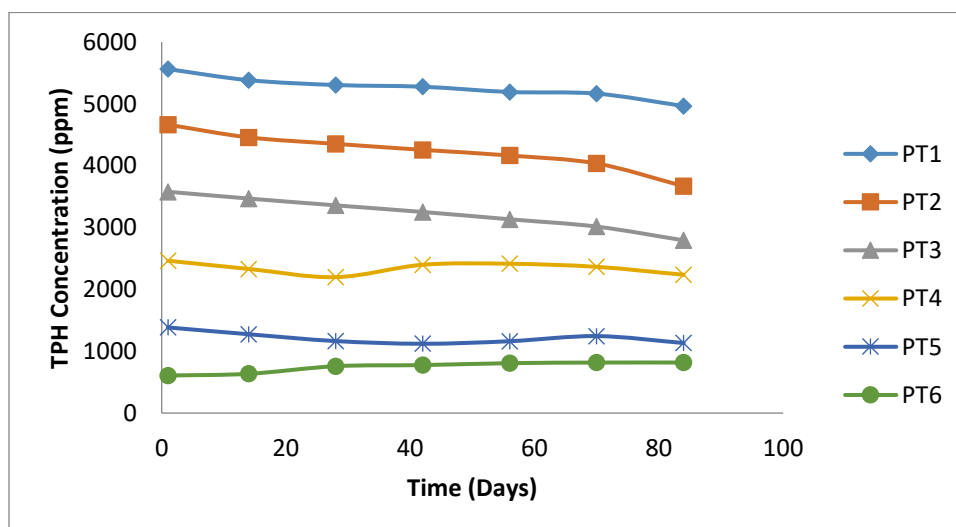
The TPH concentration in the polluted stagnant fresh and salt waters were analysed along the depth of the tank at intervals of 25 cm from the water surface, where the crude oil content was discharged, down the bottom of the tank on biweekly basis. The representative water samples collected from the prepared sample collection points were analysed via gas chromatograph to determine the concentration of TPH dispersed within the stagnant water. The results for this analysis are shown in Figure 2 for fresh water and Figure 3 for salt water.



**Figure 2: TPH Concentration versus Time at Different Depths for Fresh Water**

Pt 1 (0m), pt 2 (0.25m), pt 3 (0.5m), pt 4 (0.75), pt 5 (1m) and pt 6 (1.25).

Figure 2 showed the profiles of Total Petroleum Hydrocarbon (TPH) content recorded in polluted fresh water media with time at various depths. The specific concentration of TPH in the stagnant fresh water decreased with increase in time for sample points 1, 2 and 3 (PT1, PT2 and PT3), but increased with increase in time up to 42 days for sample point 4 (PT4) and up to 56 days for sample points 5 and 6 (PT5 and PT6). At sample point 1 (PT1), TPH decrease in fresh water ranged from 7358.18 to 5608.07ppm as time increased from 1 to 84 days. Also, TPH decreased from 6149.74 to 4790.93ppm as time increased from 1 to 84 days in PT2, while at PT3, TPH increased from 4773.79 to 3411.76ppm. However, at PT4 increased from 2799.72 to 3109.94ppm from 1 to 42 days and then, decreased to 2644.84ppm at day 84. Similarly, at PT5 and PT6, TPH increased from 1390.29 to 2158.88ppm and 472.32 to 2158.46ppm at day 56 and before decreasing again to 1848.39ppm and 905.68ppm respectively at day 84.



**Figure 3: TPH Concentration versus Time at Different Depths for Salt Water**

Pt 1 (0m), pt 2 (0.25m), pt 3 (0.5m), pt 4 (0.75), pt 5 (1m) and pt 6 (1.25).



Ordinarily, crude is supposed to float in water surface due to the fact that its density is less than that of water, but this was not so as high concentrations were recorded at the base of the water tank. These high TPH concentrations recorded at the base of the tank can be attributed of attachment of crude oil molecules to crude oil sediments. These sediments dragged the attached oil molecules down the base of the tank. It was also noticed that the concentration of crude over the 84 days analysis was still high, implying that the rate of degradation was low. This may be due to the fact that bacteria growth in the water media was slow, and this was apparent in the level of nutrients (nitrates) recorded during the physicochemical analysis, which was very low in the water system.

Figure 3 showed the profiles of Total Petroleum Hydrocarbon (TPH) content in polluted salt water media with time at various depths. Again, the specific concentration of TPH in the stagnant fresh water decreased with increase in time for sample points 1, 2 and 3 (PT1, PT2 and PT3), but increased with increase in time at some points and thereafter, decreased again. The decrease in TPH in salt water ranged from 5561.94 to 4965.14ppm as time increased from day 1 to day 84. Also, TPH decreased from 4663.62 to 3668.82ppm as time increased from 1 to 84 days in PT2, while at PT3, TPH increased from 3578.67 to 3029.82ppm. However, at PT4, PT5, and PT6, TPH concentrations ranged between 2463.84ppm and 2236.5ppm, 1384.92ppm and 1233.10ppm and 607.14ppm and 817.39mg/l respectively, as time increased from day 1 to day 84.

#### TPH Prediction in Polluted Stagnant Water

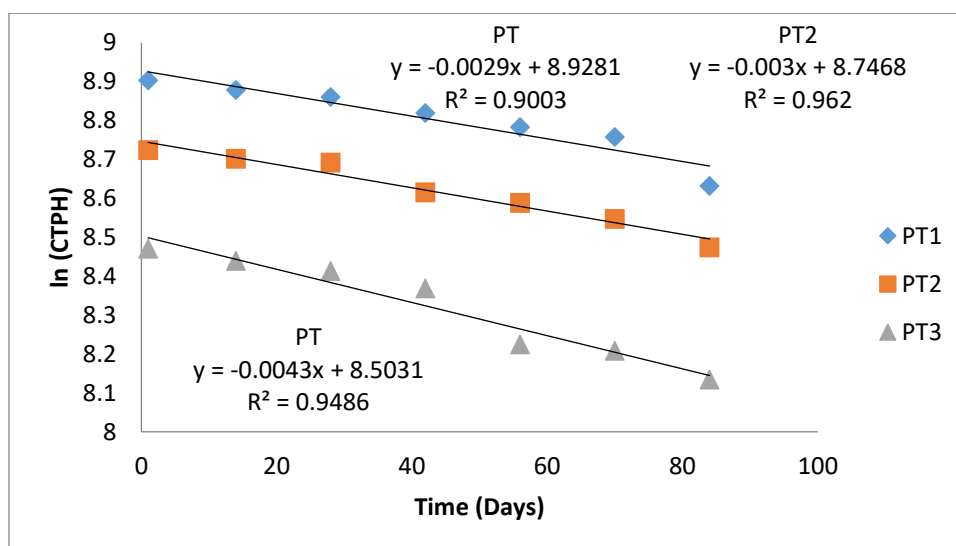


Figure 4: First Order Rate Kinetic Plots for Determination of Rate Constant used Fresh Water

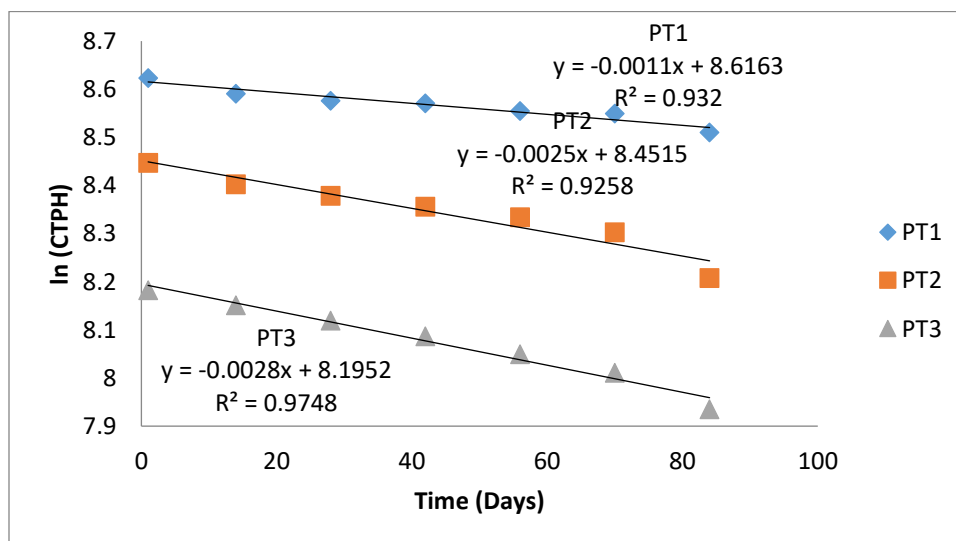


Figure 5: First Order Rate Kinetic Plots for Determination of Rate Constant used for Salt Water

The dispersion of TPH in both water media was studied via transport model incorporating diffusion and convective terms. The TPH concentration in vertical and horizontal dimension was monitored through this model. Thus, in the x-direction, which also means the horizontal direction, the transport or spreading of TPH in the water tank was assumed to be controlled by diffusion only, while in the vertical direction the TPH transport was assumed to be control by both diffusion and convection due to gravity. Therefore, the velocity in which the TPH influenced to move downward to the base of the tank was taken into consideration.

In literature, the rate of TPH degradation was mostly studied by first order degradation rate kinetics, especially in soil media. The rate term in the model was expressed using the first order degradation rate kinetics. The rate constants in the models were determined from the plots of experimental data and shown in Figures 4 and 5. The predicted TPH along the vertical and horizontal directions by the incorporated first order rate kinetics and Monod models was simulated and compare. The values of predicted TPH in the vertical direction were extracted at specified time and compare with experimental values. This was to determine the model that best fitted the TPH concentration measured from the experiment. In the horizontal direction, the model were not compared with TPH measured from the experiment since during experiment, the concentration of TPH was not determined due constrains involved in the process.

#### 4. CONCLUSION

**The research reveals the following:**

1. Degradation of TPH was experience in all sampling points of PT1, PT2 and PT3 and rapid degradation in point PT3 > PT2 > PT1 revealing the presence of highly an anaerobic organisms
2. The degradation of the TPH demonstrates the trend revealing the acceptability of the first order kinetics in predicting the process
3. The degradation of the TPH was controlled by the violation of the physicochemical of the process
4. In comparison it is revealed that TPH degradation was faster in salt medium than fresh water medium
5. This variation can be integrated the favorable condition of the microorganisms to catalyzes the bio-reaction process.

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#### Conflicts of interests

The authors declare that there are no conflicts of interests.

#### Data and materials availability

All data associated with this study are present in the paper.

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