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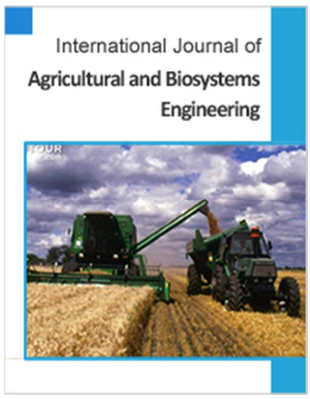

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Simulation of Mathematical Model for Petroleum Spill Absorption from Contaminated Soil Remediated with Goat Droppings

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Abstract
With the frequent reports of oil spillages in the Niger Delta area, there is need to seek for a cost effective method for remediation of crude oil impacted soils. In this research work a mathematical model for petroleum spill absorption from contaminated soil remediation with goat droppings was developed. The field test was carried out on the effect of nutrient sources NPK Fertilizer and goat droppings of 1, 5, 10, 20 and 50g added to crude oil contaminated soil in 20 polyethylene bags at random design. The treatment samples were tested at 2 weeks interval for 10 weeks. It was observed that the application of goat droppings was able to ameliorate the levels of total hydrocarbons form 7568mg/g to 439mg/kg as against 500mg/kg produced by NPK fertilizer which indicates a better remediation in goat dropping application. The model developed can be used to generate the level of reduction of the Total Hydrocarbon with respect to time. The predicted residual concentration of THC was compared with the measured values which show a very strong correlation 0.932 and 0.937 for F1 and F2 respectively at 50g. The rate of degradation was computed as follows; 0.016 and 0.054 $days^{-1}$ for F1 and F2 at 50g. The study revealed that goat droppings can degrade THC faster than NPK fertilizer.

1. Introduction

The natural environment may be altered or even exterminated by man through a series of agencies and effects. One of the major agencies that have caused habitat destruction and modification i.e. ecosystem alterations is industrialization and technology, through air, water, and land pollution. In the petroleum industry for example, pollution of the environment may occur through effluent discharges and accidental oil spillages. Environmental studies in Nigerian reveal that the development and production processes in the oil industry require an urgent need to plan, protect, and prudently utilize environmental resources for a better environment for man. These studies indicate that subtle changes occur in the Nigeria aquatic and terrestrial ecosystems due to the activities of the oil industry [17]. Most of the environmental changes occur from the release of crude oil into the environment. There were about 5334 reported cases of crude oil spillages between 1976 and 1977, with over 2.8m barrels of crude oil released into the environment [15]. [9] Reported that about 400,000 barrels of crude oil was released into the sea of Bayelsa State in the Texaco's Funiwa-5 well blow-out in 1980. About 40,

0000 barrels of crude oil was also released into the sea on January 12 1998 from Mobil producing Nigeria Unlimited Idaho oil spill which occurred near Akwa Ibom State, Nigeria due to a burst on corroded oil pipeline conveying crude oil from Idaho oil field to Qua Iboe Terminal at Mkpanak. When crude oil is spilled on land, the greasy fraction permeates slowly into the soil and is slowly biodegraded, while the light hydrocarbon fractions evaporate. Some that does not permeate the soil become thicker like tar. All, though, have toxic ingredients. Among the most troublesome are the high molecular weight compounds, especially the polycyclic aromatic hydrocarbon (PAH's) that include many known carcinogens which can combine with common environmental materials to form other carcinogens [8]. The release of crude oil into the environment may therefore result in habitat fragmentation, destruction, and disruption of ecosystems, intelligence in natural biogeochemical cycles, and the loss of plants and animal. [9] reported that the oil spill incident of 1970 at Ejamah-Ebubu near Eleme. Rivers state rendered farmlands and streams devastated. The NNPC spillage of 1982 at Abudu and Owa in Delta State also rendered the soil totally oil logged, whereas economic crops were scorched to death. The pollutants and toxicants in crude oil may cause deaths of plants and animals, disrupt biochemical pathways, metabolites and enzyme systems in all the cells and tissues of organisms.

The prevalent method adopted in curbing oil-spilled on land is by scooping off the polluted top soil and replacing it with another layer of fertile soil. But this approach is expensive and labor-intensive compared to the economical use of fertilizers to restoring fertility of such crude oil-polluted soils [19]. Hence, the researcher's interest in comparing the remediation effects of two kinds of fertilizers (Organic and Inorganic) on oil-polluted soil.

Prior to the period of oil boom, the Niger Deltans were predominantly farmers. They provide the nation with most of the agricultural produce like fishes, palm oil, to mention but a few, but now they import these farm produce. This is due to reduced agricultural activities in the area occasioned by the quest for white collar jobs that are non-existence and oil pillages on their waters and farm lands. It is also an obvious fact that in event of spillages, the communities that are affected go after compensation that the oil company concerned will pay rather than the cleanup of the mess. Consequently, a lot of spilled sites are left unattended to, thus reducing the available fishing, farming, and even building spaces. Again, there are cases of remediation attempts by the oil companies but most of them were inconclusive and abandoned.

The effectiveness of various strategies for oil spill response depends on the physical and chemical properties of the spilled oil in relation to the environment [2]. A larger member of different types of crude oils and refined producers are produced and transported in Nigeria. Petroleum products (crude oil and fuels) vary in their volatility, flammability, water solubility, tendency to emulsify viscosity and density. Some products are light, volatile, non-viscous, and less flammable (heavy fuel oil). Spill response differ depending on the oil type and its distinct characteristics [15]. Crude oil pollution adversely affects the soil ecosystem through adsorption to soil particles, provision of an excess carbon that might be unavailable for microbial use and an induction of a limitation in soil nitrogen and phosphorus [5].

However, the main purpose of this study is to determine the effectiveness of remediation of contaminated soil using goat droppings and compare its results with inorganic fertilizer, also to develop a mathematical model that can be used to predict the rate of degradation of THC and make recommendations based on findings.

2. Materials and Methods

2.1. Site Selection

The experimental site is located in a small village called Afam-Nta in Ndoki community with is in Oyibo Local Government area of Rivers State. It is Oil well head site which blew on Saturday 15 May, 2004. Ndoki has a population of about 100,000 people whose major occupation is farming. Majority of the populace there are foreigners working at the Afam power station. The coastal plain sand geological formation where the area is situated is characterized by sand and clay deposits. The topsoil is usually sandy loam and the vegetative cover is the tropical rainforest. The volume of rainfall provides great amount of surface run-offs rivulets and occasional streams which may carry substance like crude oil to nearby lands and rivers.

The equipment used in this work include shovel polyethene, black bag, hoe, rain gauge, distilled water, glass beaker, Orion research pH meter model 407A, organic and inorganic fertilizer, bean seed, maize seed, weighing balance, heater, sieve, desiccators. A ruler was used to measure the depth of the subsoil to about 15-30cm. The remediation study took place within 4 months. The soil was divided into 12polyethene bags as presented in Table 1.

2.2. Analysis of Soil Sample

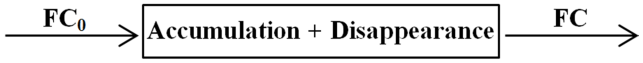
The soil pH and moisture content were measured according to [3]. Organic carbon was determined by Wakley-Black chromic acid digestion method [3]. The organic matter content of the soil was determined indirectly [3]. The phosphate content of the soil was determined according to the American Standards of Testing Materials (ASTM) method. Sulphate content of the soil was determined according to the ASTM method. The total hydrocarbon content of the soil was determined according to the ASTM D3921 standard with slight modification to suit laboratory conditions.

2.3. Mathematical Modeling

The interaction between the pollutant and micro-biota can result in the transformation of parent compounds to toxic

metabolites which can lead to abortive pathways [13, 10, 12], while adsorbents like clay and organic matter, which are site specific can decrease the bioavailability and therefore a lower risk for higher organisms (reduction in toxicity) and lower biodegradation efficiency as contaminants are tightly bound to the soil matrix [10, 21] The interaction between the pollutant and soil components of land-farm bioremediation can hamper the efficiency of land-farming for agriculture, the knowledge that has been generated during the last decades, which addresses these limitations [20, 11] has made it possible for the treatment of petroleum products in an environmentally safe manner.

In this study, the model for computing the rate degradation of concentration of Total Hydrocarbon which is represented by C is given by: Constant stirred tank reactor equation [1].



According to mass balance,

$$\begin{pmatrix} Input\ of\ Total \\ hydrocarbon\ to\ soil \end{pmatrix} = \begin{pmatrix} Output \\ rate \end{pmatrix} + \begin{pmatrix} Disappearance\ due\ to \\ biochemical\ reaction \end{pmatrix} + \begin{pmatrix} Accumulation \\ rate \end{pmatrix} \quad (1)$$

$$FC_0 = FC + R_S V + V\frac{dc}{dt} \quad (2)$$

Dividing all through of equation (2) by V

$$\frac{FC_0}{V} = \frac{FC}{V} + R_S + \frac{dc}{dt} \quad (3)$$

$$\frac{F_0}{V}C_0 = \frac{FC}{V} - R_S + \frac{dc}{dt} \quad (4)$$

$$\frac{dc}{dt} = \frac{F_0}{V}(C_0 - C) - R_S \quad (5)$$

$$\frac{dc}{dt} = -R_S \quad (6)$$

$$R_S = K_S C \quad (7)$$

$$\frac{dc}{dt} = -K_S C \quad (8)$$

Eq. 8 is the mathematical model representing the process the above model can be solved using separation of variable.

$$\frac{dc}{dt}.\frac{dc}{C} = K_S C.\frac{dc}{C} \quad (9)$$

Integrating both sides of equation (9)

$$\int_{C_O}^{C}\frac{dC}{C} = -K_S\int_0^t dt$$

$$In\ \frac{C}{C_0} = -K_s t \quad (10)$$

Taking exponential on both sides of equation

$$C_t = C_0 e^{-Kt} \quad (11)$$

The above equation (11) shows how the petroleum contaminant reduces in concentration [1]

3. Results and Discussion

Results of investigation as shown below indicate soil characteristics and outcome of the various treatment employed viz. physical and biological treatments.

3.1. Initial Condition of Soil Sample Prior to Remediation

Examination of composite contaminated soils revealed high concentrations of total hydrocarbon (7568 mg/kg). On the other hand, the subsoil of the control sample recorded 1mg/kg, 0.034mg/kg, 0.90mg/kg, and 3.93mg/kg for Total hydrocarbon. The pH of uncontaminated and contaminated soil samples was 5.42 and 6.33 respectively indication slightly acidic condition. The pH of uncontaminated and contaminates soil samples were 5.42 and 6.33 respectively indicating slightly acidic condition as shown in table 2.

3.2. Model Simulation

$$\ln(C_t) = \ln(C_o e^{-kt}) \quad (12)$$

$$\ln(C_t) = C_o(-kt) \quad (13)$$

Determination of the rate constant for the Effect of Goat dropping onTHC Remediation from Eq. 13.

3.3. Model Validation

In this study, a model which simulates the biodegradation of organic contaminants using two types of fertilizers was developed. The rate constants were computed as [k_s] in days^{-1} according to the weight of the fertilizer applied (see table 7). There is a strong and positive correlation between predicted (mathematical model) and measured values THC concentration; 0.932 and 0.937 for F1 and F2 as shown in figure 6. Therefore, the model is valid and can be used for prediction of bioremediation of THC polluted soil using goat droppings. However, 1g, 20g, and 50g weight of goat droppings showed high rate of remediation as indicated in figure 1 and table 8.

The results obtained from numerical simulation of the mathematical model using MATLAB 2013a as shown in tables 8 and 9, 50grams of fertilizer nutrient added to contaminated soil brought about the highest reduction of the total hydrocarbon concentration from 7568mg/kg to 500mg/kg unlike the 1g application which shows that the greater the application the better the remediation because the nitrogen supply from NPK fertilizer, the microorganisms that break down the substrate needed it for speedy remediation.

The rate of biodegradation of THC showed significant variations at different levels of fertilizer applied i.e. F1 and F2 as shown in figure 5. However, 50g of the remediation agent showed a better result (see figure 5). From the rate constants, it shows that F2, goat droppings (organic fertilizer) can degrade THC faster than F1 (inorganic fertilizer) (table 7).

The pH values for the original soil samples were 5.42 and 6.33 for control uncontaminated samples respectively, which indicate that both soils are acidic. On application of the nutrients, there were some changes in the pH value at various level of nutrient application; the values fluctuated between 7.91 and 4.84, [see tables 3 and 4]. It was observed that the pH values for NPK fertilizer levels of 5g and 10g of soil remained within the neutral pH range whereas goat dropping levels of 20g and 50g of soil remained within the neutral pH throughout the study duration. The pH Values observed in 5g and 10g soil of NPK fertilizer and 20g and 50g of soil of goat dropping fall within the optimum pH for rapid decomposition of waste which is usually in the range of 6.5 to 8.5 [8].

Moisture content values of 1.55 and 3.077 were observed for control and contaminated samples respectively before remediation was initiated. The reason for the difference is not farfetched; it was due to the saturation of the contaminated site with crude oil. After remediation with 1g each of the nutrients, there was an increment in the moisture content in both cases i.e. 3.76 and 4.60 for NPK fertilizer and goat droppings respectively. Both values are higher than the initial value of 3.077. However, the goat dropping has a higher impact on the moisture content than the fertilizer.

Nitrate concentration in both control and contaminated soils were 4.034mg/kg and 0.041 mg/kg respectively. After 10 weeks of remediation with 2 tonnes/ha each of the nutrients, there was an increase in the values to 148mg/kg (fertilizer) and 133 mg/kg (goat droppings). Increment to fertilizer application was higher than that of goat dropping, because of the presence of nitrogen in the inorganic fertilizer composition [1]. At the end of 10 weeks of treatment with 1g each of nutrients, there was increase in the values to 50mg/kg (fertilizer) and 30mg/kg (goat droppings). Fertilizer has a higher effect than goat dropping application of fertilizer the different levels recorded a percentage reduction in THC concentration as follows: 1g (86.70%), 5g (8.86%), 10g (87.32%), 20g (82.82%), 50g (84.54%) whereas that of goat dropping is as follows: 1g (84.69%), 5g (12.95%). The difference in the level of THC degradation can be attributed to the fact that fertilizer readily provided Nitrogen, phosphorus and potassium to the inherent microbial population unlike the goat dropping which does not readily release these same minerals until after mineralization of the goat droppings.

At the end of the study, fertilizer recorded the following percentage in THC concentration: 2tonnes/ha (91.62%), 10tonnes/ha (91.94%), 20tonnes/ha (92.73%), 20g (91.53%), 50g (93.39%) while that of goat dropping is as follows: 1g (94.20%), 1g (93.39%), 10g (92.07%), 20g (95.38%), 50g (94.71%). It was observed that, goat dropping recorded higher percentage reduction in THC concentration than NPK fertilizer (inorganic fertilizer) [see tables 8 and 9]. This could probably be attributed to the fact that the NPK applications ability to readily release nitrogen, phosphate, and potassium exposed the elements to leaching effect. Thereby depriving the microorganisms of the nutrients they require functioning effectively in the degradation of the hydrocarbon in the soil.

3.4. Seed Germination and Plant Growth

There was seed germination in both controls A (i.e. contaminated soil un-amended with nutrients) and control B (i.e. uncontaminated soil un-amended with nutrients), crude oil affect germination and plant growth in diverse ways.

It may directly kill the plant or inhibit continued growth of already established vegetation. Growth may be indirectly affected through the creation of nutrient-deficient conditions by microbial immobilization [17]. At high levels of pollution, the seeds are destroyed while at lower levels, germination is retarded [19]. Therefore, the rate of germination has an inverse relationship with the dose of oil in contamination. Brim [6] reported that the poor levels of seed germination is due to poor soil wet ability, aeration and toxic effect of oil. The use of NPK fertilizer (20:10:10) in the pre planting remediation of crude oil-polluted soil inhibited germination of both beans and maize seeds. On the other hand, there was germination in both contaminated and uncontaminated soils amended with goal droppings.

The use of goat dropping provided some benefit to the seeds and seedlings. This is hinged on its moderate solubility, balanced macro and micro-nutrients, gradual but steady release of these nutrient into soil, its tendency of increasing the humus level, its non-toxicity to the soil biota, and finally, its slight alkaline pH [7]. On the other hand, NPK fertilizer did not encourage the germination of seeds beyond 1g level which is primarily due to over-nitration of the soil, its high solubility, water-logging capacity, and its toxic nature to the soil biota. Moreover, the water-logging effect observed in the use of NPK fertilizer is a contributory factor to the observed inhi8bited growth of plants, because of its potentials in displacing available soil-air-which is otherwise, important to the planted seeds and soil microbes [14].

However, beans seed was used in the study because of the bulky nature of maize seedlings. In terms of plant growth variables, leaf length, leaf height etc. control B (uncontaminated soil) showed a better growth variable than control A, (contaminated soil). On application of 1g of the nutrients, the inorganic fertilizer showed better growth variable on uncontaminated soil than goat dropping whereas goat dropping showed better growth variables on contaminated soil than the inorganic fertilizer. (See figure 5). The highest plant height was recorded in uncontaminated soil when 50g of goat dropping was applied whereas the highest plant height was recorded in contaminated soil when 20g of goat droppings was applied [7].

4. Conclusion

Generally, remediation with fertilizers (organic or inorganic) is advantageous in replenishing the lost nutrients of the polluted soils. Inorganic fertilizer releases more of the element at a faster rate than its organic counterpart, but the inorganic fertilizer is able to sustain its nutrient for a longer period. Integration of the results showed that organic fertilizer (goat dropping) is preferable in the pre-planting

remediation of crude oil polluted soil. This is mainly because, its actions in the soil does not have toxic or negative effects on the ecological flora and fauna both in the short or long run. On the other hand, inorganic fertilizer remediated soil can only be cultivated three months after the remediation has been initiated. The results obtained from Numerical Simulation of the Mathematical Model showed that 50grams of fertilizer nutrient added to contaminated soil brought about the highest reduction of the total hydrocarbon concentration from 7568mg/kg to 500mg/kg unlike the 1g application. This shows that the greater the application the better the remediation because of the nitrogen supply from NPK fertilizer, the microorganisms that break down the substrate needed it for speedy remediation. Therefore, the model can be used to predict the concentration of TPH at any time.

Table 1. *Description of sample.*

Sample Label	Description
OF_1	1.0g organic fertilizer/1kg of contaminated soil
OF_2	5.0g organic fertilizer/1kg of contaminated soil
OF_3	10.0g organic fertilizer/1kg of contaminated soil
OF_4	20.0g organic fertilizer/1kg of contaminated soil
OF_5	50.0g organic fertilizer/1kg of contaminated soil
IF_1	1.0g inorganic fertilizer/1kg of contaminated soil
IF_2	5.0g inorganic fertilizer/1kg of contaminated soil
IF_3	10.0g inorganic fertilizer/1kg of contaminated soil
IF_4	20.0g inorganic fertilizer/1kg of contaminated soil
IF_5	50.0g inorganic fertilizer/1kg of contaminated soil

Table 2. *Characteristics of Contaminated Soil before nutrient application.*

Parameter	Control Sample	Contaminated Sample
Ph	5.42	6.33
Moisture content (%)	1.55	3.077
Total Hydrocarbon (mg/kg)	<1	7568
Organic matter (%)	0.1176	0.1799
Sodium absorption ration (SAR)	0.87	0.90

Table 3. *Result of the effect of fertilizer (F_1) on pH of the contaminated soil sample.*

Time (weeks)	1g	5g	10g	20g	50g
2	6.99	7.81	7.78	7.07	7.17
4	7.01	7.76	7.72	6.97	7.01
6	6.89	7.82	7.75	7.17	6.90
8	7.03	7.90	7.750	6.89	7.21
10	4.84	7.50	7.50	6.66	6.29

Table 4. *Result of the effect of Goat dropping (F_2) on pH of the contaminated soil sample.*

Time (weeks)	1g	5g	10g	20g	50g
2	7.94	6.67	6.80	7.22	7.82
4	7.60	6.60	6.90	7.01	7.60
6	7.80	6.80	6.81	6.90	7.71
8	7.91	6.70	6.85	7.20	7.50
10	5.88	6.70	6.80	7.05	7.68

Table 5. *Result of the effect of fertilizer (F_1) on THC of the contaminated soil sample.*

Time (weeks)	1g	5g	10g	20g	50g
2	1006	1070	960	1300	1170
4	980	960	870	1100	920
6	840	800	746	970	790
8	710	760	610	890	650
10	634	610	550	641	500

Table 6. *Result of the effect of Goat dropping (F_2) on THC of the contaminated soil sample.*

Time (weeks)	1g	5g	10g	20g	50g
2	1158	1390	1631	2676	6588
4	980	1060	1240	1480	4260
6	670	870	1010	1000	1550
8	510	770	910	600	800
10	634	610	550	641	500

Table 7. *Rate of biodegradation constants.*

Weight of fertilizer (g)	K_s ($days^{-1}$)	
	F1	F2
1	0.010	0.015
5	0.011	0.015
10	0.012	0.019
20	0.012	0.030
50	0.016	0.054

Table 8. *The Residual Concentration of THC after remediation with F1.*

Time (weeks)	Residual Concentration of THC ($mg.kg^{-1}$).				
	1g	5g	10g	20g	50g
0	7568.00	7568.00	7568.00	7568.00	7568.00
1	6793.25	6793.25	6624.86	6114.90	5186.34
2	6097.80	6097.80	5799.25	4940.81	3554.20
3	5473.56	5473.56	5076.53	3992.15	2435.69
4	4913.22	4913.22	4443.88	3225.63	1669.17
5	4410.24	4410.24	3890.08	2606.29	1143.88
6	3958.75	3958.75	3405.29	2105.87	783.90
7	3553.49	3553.49	2980.91	1701.53	537.21
8	3189.71	3189.71	2609.42	1374.83	368.15
9	2863.17	2863.17	2284.23	1110.85	252.29
10	2570.06	2570.06	1999.56	897.56	172.89

Table 9. *The Residual Concentration of THC after remediation with F2.*

Time (weeks)	Residual Concentration of THC ($mg.kg^{-1}$)				
	1g	5g	10g	20g	50g
0	7568.00	7568.00	7568.00	7568.00	7568.00
1	5622.80	7271.30	7418.10	5592.00	5623.40
2	4177.60	6986.10	7271.30	4131.90	4178.40
3	3103.80	6712.20	7127.30	3053.00	3104.70
4	2306.00	6449.00	6986.10	2255.90	2307.00
5	1713.30	6196.20	6847.80	1666.80	1714.20
6	1272.90	5953.20	6712.20	1231.60	1273.70
7	945.76	5719.80	6579.30	910.04	946.42
8	702.67	5495.50	6449.00	672.42	703.23
9	522.06	5280.00	6321.30	496.85	522.53
10	387.88	5073.00	6196.20	367.12	388.26

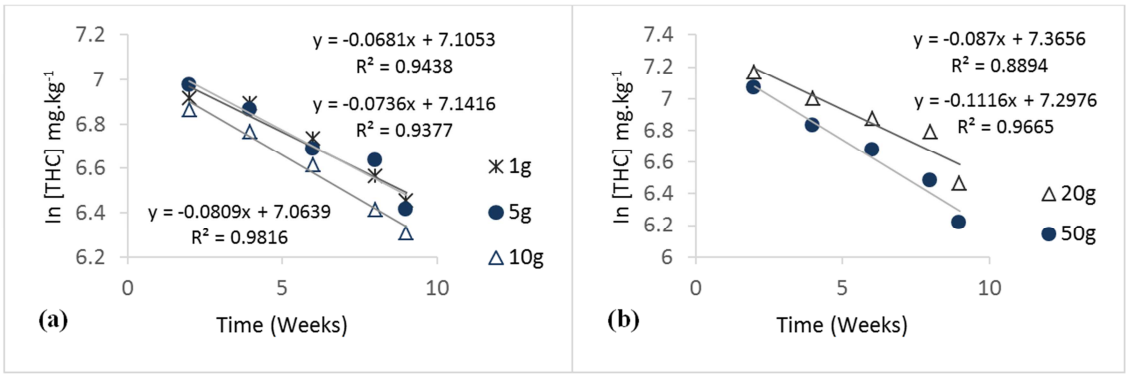


Figure 1. *(a) and (b): Graph In [THC] vs Time to determine the reaction rate constant for F1.*

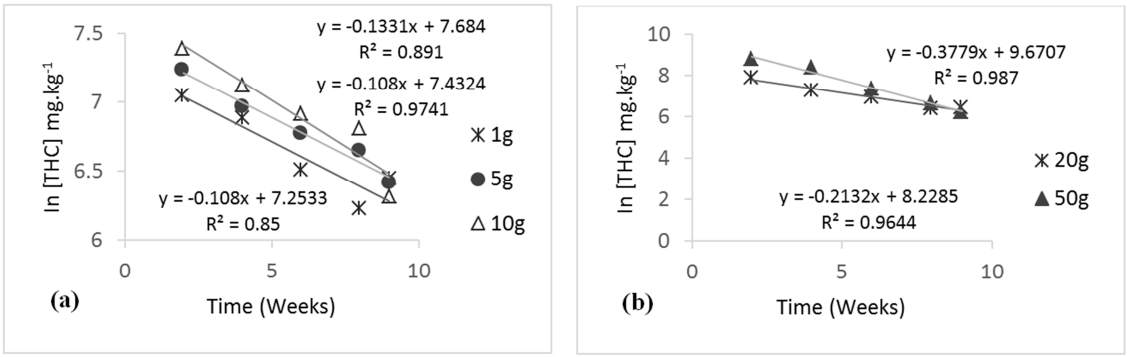


Figure 2. *(a) and (b): Graph In [THC] vs Time to determine the reaction rate constant for F2.*

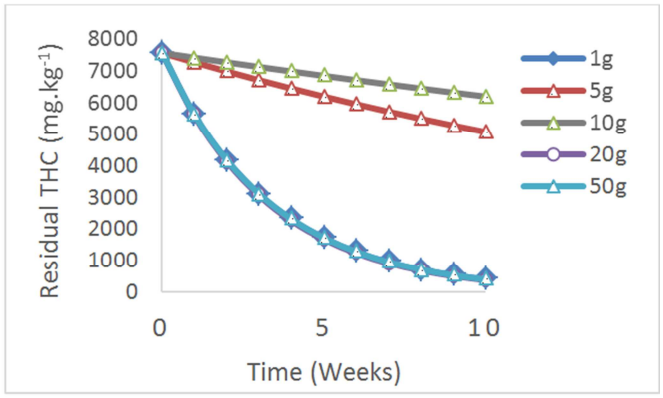


Figure 3. *Residual THC after 10 weeks remediation with F2 from numerical simulation.*

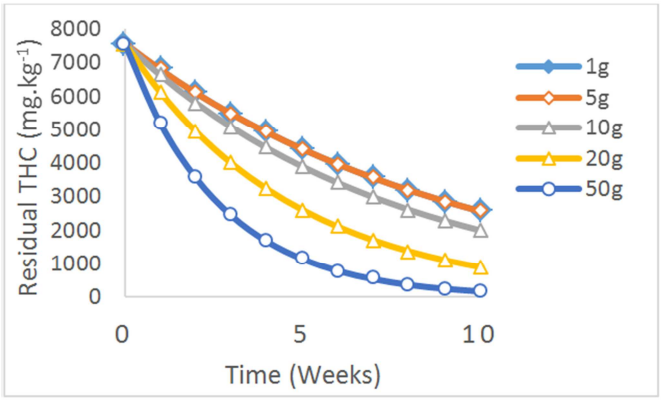


Figure 4. *Residual THC after 10 weeks remediation with F1 from numerical simulation.*

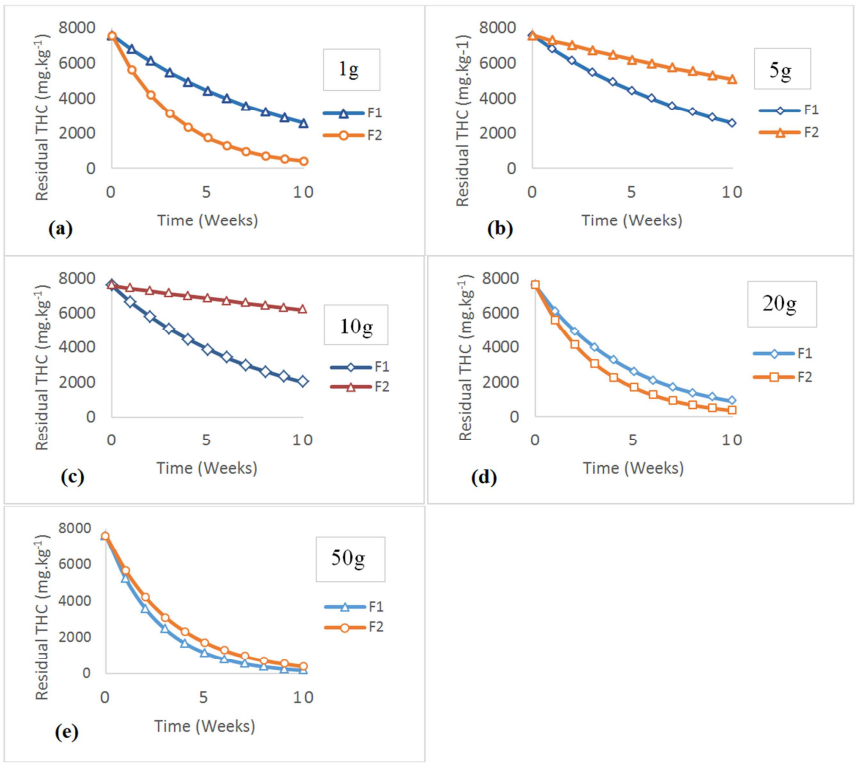


Figure 5. *(a), (b), (c), (d) and (e): The effect of F1 and F2 in THC remediation.*

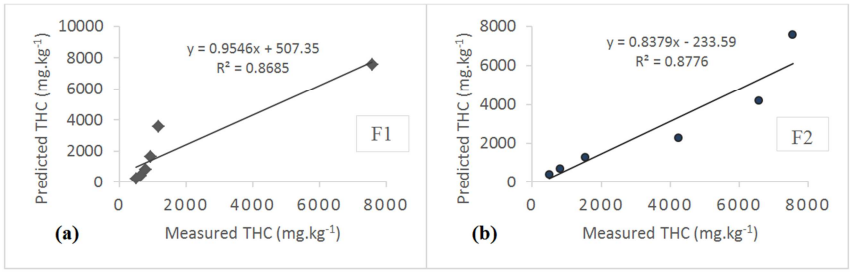


Figure 6. *(a) and (b): The accuracy of presented model for predicted THC against measured values at 50g.*

Nomenclature

Symbols	
R_sV	rate of disappearance due to biochemical reaction
B	blank reading
C	final concentration of Total Hydrocarbon
C_0	initial concentration of Total Hydrocarbon
FC	output of total Hydrocarbon from the soil
FC_0	input of Total Hydrocarbon to the soil
K	rate of degradation of Total Hydrocarbon
N	concentration or normality of $K_2Cr_2O_7$
NPK	nitrogen, phosphorus and potassium [fertilizer]
T	volume of titre (ml) of $K_2Cr_2O_7$
T	time [weeks]
THC	total hydrocarbon content [mg/g]
W	moist content of soil
W_1	weight container plus moist soil [g]
W_2	weight of container plus oven – dried soil [g]
W_c	weight of container [g]
$V\frac{dc}{dt}$	rate of accumulation
US	uncontaminated soil
CS	contaminated soil
F1	NPK fertilizer (inorganic fertilizer)
F2	goat droppings (organic fertilizer)

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