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The Removal of Total Petroleum Hydrocarbon in Soil Polluted by Crude Oil-Spillage using Response-Surface Technique and Artificial Neutral Network

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Abstract: Optimization of environmental process parameters not only increases the efficiency, but also the quality of the process without increasing the cost to a great extent. In the present study, Response Surface Methodology (RSM) and Artificial Neural Network (ANN) were employed to predict the %TPH removal in petroleum polluted soil. The aim is to identify the optimal conditions for TPH removal in petroleum polluted soil remediated with mushroom substrate. In RSM model, two quadratic equations of percentage TPH removal were derived from experimental data. The analysis of variance (ANOVA) was also computed. In ANN model, 80% data were used to train and 20% data were employed for testing.

Error analysis conducted shows that RSM has more errors than ANN. The accuracy of the RSM and ANN model was found to be ≤96%. The ANN models exhibit an error of ~6% MAE for testing data. The regression coefficient was found to be greater than 95.0%. However, ANN predicted values have been found to be very close to experimental values compare to RSM predicted values, hence ANN model demonstrated a higher accuracy.

Keywords: Optimization; bioremediation; polluted soil; mushroom substrate; oil spillage; hydrocarbon.

1 Introduction

Hydrocarbons inhibit the germination of seeds because of insufficient soil evaporation and water in the soil leading to loss in viability (Rowell, 1977; Udo and Oputa 1984). Hydrocarbons in the soil injures or kills the seed embryo when they come in contact and enters the seed through the micropyle end of the seed, leading to germination failure (Atunuya, 1987).

Even if the seed eventually germinates, presence of Hydrocarbons in the soil results in growth retardation, wilting of the plant, defoliation of leaves and chlorosis (Hitivani and Mecs, 2003)

Therefore, adequate restoration of total petroleum hydrocarbon (TPH) in contaminated soil depends on the nature and activity of remediating microbes. Whereas low molecular weights TPH is usually readily degraded, high molecular weights TPH resist extensive bacteria degradation in soil and sediment media (Aust 2003; Mueller, 1998). In addition, TPH is considered the most acute toxic component of petroleum products, and is associated with chronic and carcinogenic effects (Anderson, 1980).

Bioremediation, which is the enhancement of natural biological degradation processes, has been proposed as cost-effective technology for removing contaminants from oil-spills in soils (Mulligan and Galver, 2003). Hydrocarbon-degrading microbes have been studied for its effectiveness in microbial oil degradation in soils (Perelo, 2010). The activities of microbes naturally present in the soil can further improved upon using bioremediation techniques through increased aeration of the polluted area or by nutrient (Substrate) additions (Agrodok, 2005. The aim of this technique is to remove pollutant from the natural environment or to reduce the pollutant to a less harmful product using indigenous microbiological community of the contaminated environment. Many researchers have adopted bioaugmentation or biostimulation (physical or chemical) degradation to clean up or remediate hydrocarbon-contaminated soil (Gibson and Parales, 2000).

However, these methods in some cases may leave behind traces of compounds that are more harmful to the environment than the initial parent compounds. The adoption Biological breakdown of contaminants has offered the most interesting environmentally friendly alternative for remediating TPH in crude oil contaminated soil because of its capacity to utilize indigenous microorganisms within the soil environment to break down the TPH into innocuous constituents (Lan et al., 2003).

The harm caused by TPH to the natural environment due to the activities of the oil industry in Nigeria has had enormous impact on the quality of soil, water, and air resulting in huge Agro- economic losses and

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lower life expectancy (Sample et al., 2000). TPH persistent nature and tendency to spread into the soil, surface, and groundwater has made its removal a national issue in Nigeria. To buttress the danger of TPH, to the environment, the federal government of Nigeria recently flagged off a multimillion dollars cleaning of oil producing Ogoni land after much protest and death by the environmental activist in Nigeria. Although the major generator of TPH in the Nigerian Niger Delta is the activities of the petroleum industry but largely the increasing number of vehicles and electric generators in Nigeria has increased the use of lubrication and transmission oil, which has constituted another source of TPH in the environment.

Mushroom substrate has shown to resist contaminated soil due to secretion of manganese dependent peroxidase, and Lucases, therefore could be a good bioremediation of the contaminated soil (Aust and Swaner, 2003; Mansur and Arias, 2005; Barr and Aust, 2010). Mushroom exhibits different optimum growth ability on different kinds of contaminant present in the soil (Emuh, 2009).

This shows the resilience of different mushroom species to breakdown and adsorbs or mineralizes particular contaminants (Oudot 1990, Stemets 2005). Several mushroom substrates developed on different parent material has been applied in soil remediation. Lan et al. (2003), reported the use of mushroom grown on compost to degrade TPH in crude oil contaminated soil. Thomas and Becker (1999) reported that oyster mushroom mineralized and metabolized 97% of oil after 8 weeks of incubation. Eggen and Sasek (2003) reported that oyster mushrooms compost significantly and effectively reduced toxin in polluted soil. Similarly, Ruma et al., (2007) and Sasek (2003) reported the use of mushroom to degrade hydrocarbons and its by-product while Gadd (2001), Kondo and Sasek (2003) and reported the use of mushroom to degrade Agro- chemicals. Mushroom are particularly proficient in breaking down many recalcitrant compounds, long chain molecules, and harmful toxins to less but simpler chains (Stamets, 2005). Mushroom shows rugged abilities to transfer recalcitrant pollutants and degrades a wide range of structurally diverse poisonous environmental pollutants (Mandel et al., 1998; Lang, 2003, Reddy and Quinn, 1999). Their extra cellular capacity, help them to degrade non-soluble poisonous compounds and non-polar compounds and utilize them as food (Levin et al., 2003; Barr and Aust, 2010).

Hamman, (2004) reported that enzymes of fungi degraded several Polycyclic aromatic hydrocarbons (PAH) after 50 days of incubation to 1 -7%. Levin et al. (2003) reported that Trametestrogil metabolized and degraded 90-97% highly concentrated nitrobenzene and enuthrencenene.

Although the concept of using mushroom substrate in treating TPH contaminated soil has been carried out in some countries and has been proven efficient, TPH treatment with mushroom substrates can still be considered as an evolving ex situ biotreatment for TPH contaminated soil especially in Nigeria. Knowledge gap still exist on optimum condition for TPH bioremediation and the properties a given mushroom substrate should exhibit for fast removal of TPH. The success of any bioremdiation strategy depends on environmental variables, which are easier to provide, but optimization of these variables is key to avoid inhibition of microbial activity that affects removal efficiency (Philips, 2005). According to Schmidt (2008), there is scarcity of biological and biochemical models for in-depth description of bioremediation process, therefore statistical tools present a veritable option to obtain relevant information for process optimization and has been applied in different research areas (Montgomery, 2008).

The major aim of this work therefore, is to study the effect of mushroom substrates on TPH on the optimization of soil bioremediation process using Response surface method and artificial neural network. Response surface methodology (RSM) is the application of design expert software in modeling and optimization. However, the benefits of design expert over other software include asset in statistical quality control, expressive and inferential statistics, statistical process control, reliability, gage repeatability and reproducibility studies, process ability with an enhanced graphing output (Jahirul et al., 2014). On the other hand, Artificial neural network (ANN) is an educative based on assessment method, which shows a nonlinear relationship with joining factors and product formation thru reiteration of outcomes acquired from experimental design (Yan, 2010). ANN has showed to be enhanced design software in terms of data fitting and prediction (Bourquin et al., 1998; 1998; Ghobadiana et al., 2008; Sulaiman et al., 2010; Adepoju et al., 2018). These software's and others have been applied either in single or combination form to model and optimize the experimental factors so as to determine the optimum yields (Canakci et al., Yuste and Dorado, 2006; Najafi et al., 2007; Shchinas et al., Canakci et al., 2009; Shiyakumara et al., 2011; Cay et al., 2012; Moradi et al., 2013; Adepoju et al., 2018).

2 Materials and Method

2.1 Description of Study Area

This study was carried out at the research farm of the Niger Delta University, Amassoma, Bayelsa State in Niger Delta region, Nigeria. The region is within the tropical rain-forest zone of Nigeria with an ambient

environment having a mean annual rainfall of 2400 mm; a mean monthly relative humidity of 85%, a mean daily minimum temperature of about 230C and a mean daily maximum in temperature of 31.50C. The soil is normally moisture laden due to the high annual rainfall, which results in surface run-offs, rivulets and streams, which may convey substances like crude oil to contaminate nearby land and rivers (Fasidi, 2005).

The crude oil used for this experiment was obtained from Nigeria National Petroleum Company (NNPC) in Port Harcourt, Rivers State. The mushrooms used were bought from Santannah market in Benin City Nigeria.

2.2 Experimental Design and Analysis

The soil was divided into six treatment sample cells in six different containers (bucket). The different samples were coded as 1,6. Cell 1, the control, did not receive any treatment, whereas cells 2,6 were earmarked to receive (900-1000g), (750-800g) and (700-800g) of saprophytic, parasitic and symbiotic mushrooms respectively, during the remediation period. The weight of pollutant is 30.4mg in 5000g weight of soil, and 0.75 litres of water was added to the soil during the remediation process. Soil Treatment

The crude oil was added to each treatment cell. The cells were left undisturbed for three days, at the end of which the treatment options were then applied. The three-day period allows degradation to commence and follows the work of Fasidi (2005). Soil sample were collected using a 22 cm hand-held soil auger capable of obtaining uniform cores of equal volume at predetermined depths bulked together (composite soil samples) and put in sample bottles labeled 1,…6. Different types of mushroom were applied by broadcasting to the relevant cell. Various quantities of mushroom applied to the different cells. Then 1000g, 900g, 800g of the mushroom substrate was applied once in ten (10) weeks to cells 1,6, respectively. These quantities of mushroom supplied nitrogen to the cells for the ten (10) weeks remediation period.

2.2.1 Analysis of Total Petroleum Hydrocarbon

Three days after pollution, 10g of each sample was taken and put into sample bottles labeled 1, 6. 80ml of chloroform was measured and added to each sample and the sample was tightly closed and thoroughly shaken for proper mixing of contents. The mixtures in the bottles were left to stand for 2 days to allow for complete extraction of the crude oil by the chloroform. On the 4th day, each of the samples was decanted; the clear liquid was transferred to fresh sample bottles and the volume made up to 60ml using chloroform. The UV-VIS spectrophotometer was standardized using chloroform for the blank, with wavelength set at 290nm. The absorbance of sample was measured immediately after completion of the last step and the digital readout of the instrument recorded.

2.2.2 Soil pH and Electrical Conductivity (EC)

The hydrogen ions concentrations of the soil samples were determined using the pH electrical conductivity meter (pH meter). To achieve this, 10g homogenized soil sample (pounded in a soil mortar and sieved through a 2mm sieve was weighed and put in a pH cup and the addition of 25ml of deionized water followed suit. This then resulted into soil: water concentration of 1:2. The mixture was stirred for 1 hour and reading was taken. The pH meter was already calibrated using a buffer solution of 4 and 7. The readings were then taken by inserting the probe of the pH/EC meter into solution (soil solution). The EC of the soil samples were measured in micro Siemens/cm (µs/cm). The probe of the electrode was washed after each reading for accurate results and to avoid cross-contamination.

2.2.3 Moisture Content (MC)

This was determined using the oven drying method. In this method, 20g of wet soil (W1) were put into an aluminum foil and placed in an oven to dry at 1050C. After 24 hours, the soil samples in the oven were removed and reweighed. The dry weight, therefore, become an index for determining the moisture content of the soil sample. The final weight (W2) of each sample is recorded using an electronic weighing balance.

2.2.4 Total Organic Content (TOC)

To determine the TOC, 250mg of air-dried soil sample were taken in 250ml conical flasks, and 5ml of potassium dichromate solution was added. Hence, 10ml of concentrated sulfuric acid was added gradually and the contents were allowed to incubate for 30 minutes at room temperature. Then 100ml of de-ionized water, 5 ml of concentrated phosphoric acid, 0.1g of dry sodium fluoride, and 0.5 ml of diphenylamine indictor were added sequentially. The contents of the flask were titrated against 0.5M ferrous ammonium sulfate. The end point was noticed as dull green through turbid blue to brilliant green. Distilled water blank was run simultaneously, and the TOC was calculated as described by Hooda and Kaur (1999).

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2.3 Response Surface Methodology (RSM)

The experiment was designed using Response Surface Methodology, and after performing 17 experimental runs of the Box-Behnken design (BBD) and one control. The three different independent variables were investigated at three levels of coded low level of -1, medium coded level of 0 and high coded level of +1 with 17 standard runs of experiment as presented in Table 1 and 2.

Table 1. Experimental range and the levels of the variables

Factors	**Low level (-1)**	**Medium level (0)**	**High level (+1)**
Parasitic mushroom (A) g	750	775	800
Saprophytic mushroom (B) g	900	950	1000
Symbiotic mushroom (C) g	700	750	800

Table 2. Full-factorial design at three levels for the independent variables

	Factor 1		Factor 2		Factor 3	
Runs	**A: Parasitic [g]**		**B: Saprophytic [g]**		**C: Symbiotic [g]**	
	Coded	Uncoded	Coded	Uncoded	Coded	Uncoded
1	0	775	0	950	0	750
2	+1	800	0	950	+1	800
3	0	775	0	950	0	750
4	+1	800	+1	1000	0	750
5	0	775	+1	1000	+1	800
6	+1	800	-1	900	0	750
7	0	775	0	950	0	750
8	0	775	-1	900	+1	800
9	+1	800	0	950	-1	700
10	0	775	0	950	0	750
11	0	775	0	950	0	750
12	0	775	-1	900	-1	700
13	-1	750	0	950	-1	700
14	-1	750	0	950	+1	800
15	-1	750	+1	1000	0	750
16	-1	750	-1	900	0	750
17	0	775	+1	1000	-1	700
18	-	-	-	-	-	-
Control						

2.4 Analysis of Total Petroleum Hydrocarbon

The percentage removal of total petroleum hydrocarbon was determined using the equation below (Olatunji et al., 2018)

$$\%TPH = \frac{TPH_I - TPH_F}{TPH_I} \times 100 \quad (1)$$

where,
TPH_I = the initial amount of TPH in the soil
TPH_F = the residual amount of TPH after remediation

3 Results and Discussion

The initial screening of TPH for all samples showed almost equal value of concentration, because each sample was contaminated with the same amount of crude oil. Each sample showed reduction in TPH, including sample 1, to which no mushroom substrates was added. The soil characteristics that were used as indicators of the levels of pollution and remediation, before and after the crude- oil contamination, as well as remediation process. The particle size analyses of the soil before treatment showed that the soil texture is silty clay (see Table 3). The soil parameters from the initial assessment indicate that the soil to remediate is acidic with a mean pH value of 4.65 and the moisture content was found to be 14% which is not suitable environmental condition

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for bioremediation (Achwendinger, 2010)

3.1 Soil Parameters

Table 3 showed the values for the soil parameters that were tested.

Table 3. Soil Parameters for the TPH polluted soil.

Parameters	Values
Soil pH	4.65
Moisture content (%)	14.0
Total organic content (%)	0.18
Electrical conductivity (/)	29.0

3.2 Modeling and Statistical Analysis

The results of the statistical experiments were analyzed with reverence to the actual design matrix. The regression equation showed that TPH degradation rate was an experimental function of test variables in uncoded units (actual values). The RSM design was carried out in design expert version 10.

Table 4. Experimental results, predicted and residual values by RSM and ANN for percentage TPH removal

	Percentage TPH removal			**Residual**	
Run	**Experimental values (%)**	**Predicted values RSM (%)**	**Predicted values ANN (%)**	**RSM**	**ANN**
1	46.3	45.7	45.5	0.6	0.8
2	47.6	47.2	47.6	0.4	0.0
3	46.2	45.7	45.5	0.5	0.7
4	47.9	48.3	47.9	-0.4	0.0
5	46.6	46.6	46.6	0.0	0.0
6	47.8	47.6	47.8	0.2	0.0
7	45.9	45.7	45.5	0.2	0.4
8	45.8	46.4	45.8	-0.6	0.0
9	46.6	46.9	46.9	-0.3	-0.3
10	45.2	45.7	45.5	-0.5	-0.3
11	45.1	45.7	45.5	-0.6	-0.4
12	45.6	45.6	45.6	0.0	0.0
13	46.9	47.3	46.9	-0.4	0.0
14	47.0	46.8	47.0	0.2	0.0
15	48.4	48.6	48.8	-0.2	0.0
16	47.5	47.1	47.5	0.4	0.0
17	48.2	47.6	48.2	0.6	0.0
18	21.15	-	-	-	-
Control					

3.2.1 Second Order Polynomial Regression Model and Statistical Analysis

The experimental data were fitted to a second order polynomial regression model containing 3 linear, 3 quadratic and 3 interaction terms (Montgomery, 2008) using the same experimental design software to derive the Regression equation for TPH removal from polluted soil as stated in Eq3. The significance of each coefficient in the equation was determined by F-test and P- values. F-test showed that all the factors and interactions considered in the experimental design are statistically significant i.e. $P < 0.05$, at 95% confidence level. The regression equation obtained after analysis of variance gave the level of TPH removal as a function of the different bio-stimulation variables: Parasitic mushroom, Saprophytic, and Symbiotic mushroom.

The response (Y) generated in coded factor is given as:

$$Y = 45.74 + 0.013A + 0.55B - 0.038C - 0.20AB + 0.22AC - 0.45BC + 1.32A^2 + 0.84B^2 – 0.032C^2 \quad (2a)$$

where, Y = TPH removal (%)

A = Parasitic mushroom, B = Saprophyic mushroom and C = Symbiotic mushroom

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The equation in terms of coded factors can be used to make predictions about the response for given levels of each factor. By default, the high levels of the factors are coded as +1 and the low levels of the factors are coded as -1. The coded equation is useful for identifying the relative impact of the factors by comparing the factor coefficients.

Final equation in terms of actual factors:

Y = 1457 - 3.25A - 0.37B + 0.05C - 1.6e - 004AB + 1.8e - 004AC - 1.8e - 004BC + 2.1e - 003A2 + 3.37e - 004B2 - 1.3e - 005C2 (2b)

The equation in terms of actual factors can be used to make predictions about the response for given levels of each factor. Here, the levels should be specified in the original units for each factor. The result from Table 4 showed that on day 28, TPH content had decreased in all the soil microcosms. In control, natural Bio-attenuation removed 21.15% of petroleum hydrocarbons from the soil. It was observed that the respective reduction in petroleum hydrocarbon (TPH) of soil microcosms with amendments was much higher when compared to control in the same period in Table 4. This observation showed that the addition of bio-stimulants increased the rate of TPH degradation in the soil. This is in agreement with the report of Agarry and Ogunleye (2012a) that an increase spent engine oil biodegradation with the addition of bio-stimulants such as NPK, Tween 80 and Pig Manure as supplements and Olawale et al., (2015) who optimized diesel polluted soil with the following bio-stimulants, Tween 80, Poultry droppings and Hydrogen perioxide. According to Olawale et al. (2015), Mohajeri, et al., (2010) reported that one of the major factors limiting degradation of hydrocarbons is their low availability to the microbial cells.

Table 5. Analysis of variance (ANOVA) for the quadratic response surface model fitting to the biodegradation data of TPH

Source	SS	DF	MS	F-Value	P-Value	Remarks
Model	14.46	9	1.61	4.04	0.0396	Significant
A	1.25e-003	1	1.25e-003	3.142e-003	0.9569	Not Significant
B	2.42	1	2.42	6.08	0.0431	Significant
C	0.011	1	0.011	0.028	0.8712	Not Significant
AB	0.16	1	0.16	0.40	0.5461	Not Significant
AC	0.20	1	0.20	0.51	0.4986	Not Significant
BC	0.81	1	0.81	2.04	0.1966	Not Significant
A2	7.31	1	7.31	18.37	0.0036	Significant
B2	2.99	1	2.99	7.51	0.0289	Significant
C2	4.45e-003	1	4.45e-003	0.11	0.9188	Not Significant
Residual	2.78	7	0.40			
Lack for fit	1.53	3	0.51	1.63	0.3163	Not Significant
Pure error	1.25	4	0.31			
Correlation	17.24	16				
Total						

The Model F-value of 4.04 implies the model is significant. There is only a 3.96% chance that an F-value this large could occur due to noise. Values of Prob > F less than 0.05 indicate model terms are significant. In this case B, A^2, $B^2$are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve your model. The Lack of Fit F-value of 1.63 implies the Lack of Fit is not significant relative to the pure error. Non-significant lack of fit is good.

A higher F-value signifies a well-fitting of the RSM model to the experimental data (Panwal et al., 2011). Datta and Kumar (2012) reported that an F-value along with low p-value indicates a high significance of the regression model. Nevertheless, the p-value should be lower than 0.05 for the model to be statistically significant (Patel et al., 2011). Based on the research reports, the regression model found in Table 5 was highly significant, as it is evident by the F-value 4.04 and the low p-value 0.0396, respectively. Standard deviation = 0.63, C. V (%) = 1.35, R-Squared = 0.8385, Mean = 46.74, Adjusted R-Squared = 0.6309, Predicted R-Squared = -0.5356, Adequate Precision = 6.305. A negative Predicted R-Squared implies that the overall mean may be a better predictor of your response than the current model. A ratio greater than 4 is desirable. The ratio of 6.305 indicates an adequate signal. This model can be used to navigate the design space.

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Table 6. Coefficient of the model for TPH biodegradation.

Factor	Coefficient	DF	Standard Error	95% Low	CI 95% High	CI VIF
Intercept	45.74	1	0.28	45.07	46.41	1.00
A	0.013	1	0.22	-0.51	0.54	1.00
B	0.55	1	0.22	0.023	1.08	1.00
C	-0.038	1	0.22	-0.56	0.49	1.00
AB	-0.20	1	0.32	-0.95	0.55	1.00
AC	0.22	1	0.32	-0.52	0.97	1.00
BC	-0.45	1	0.32	-1.20	0.30	1.00
A^2	1.32	1	0.31	0.56	2.04	1.01
B^2	0.84	1	0.31	0.12	1.57	1.01
C^2	-0.032	1	0.31	-0.76	0.69	1.01

The ideal VIF value is 1.0. VIFs above 10 are cause for concern. VIFs above 100 are cause for alarm, indicating coefficients are poorly estimated due to multi-collinearity. Figure 1 showed the studentized residuals and normal percent probability plot. Residual showed the difference between the observed value of a response measurement and the value that is fitted under the theorized model. Small residual values indicated that model prediction is accurate.

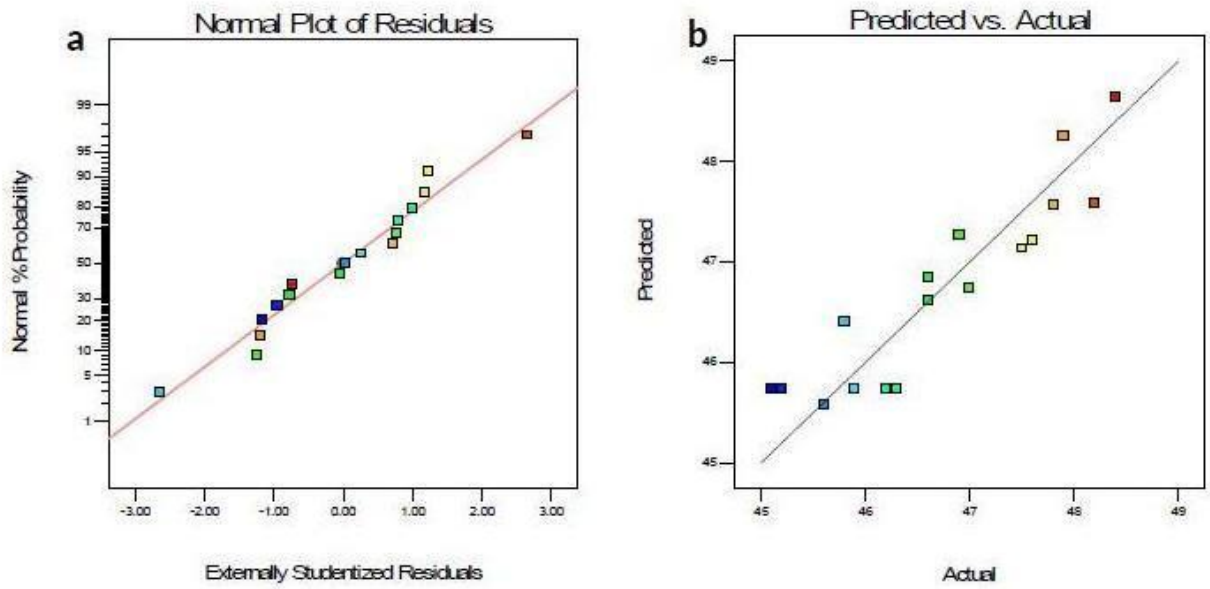

Figure 1. (a) Normal plot of residuals plot of soil TPH bioremediation (b) Predicted versus actual plot of soil TPH bioremediation.

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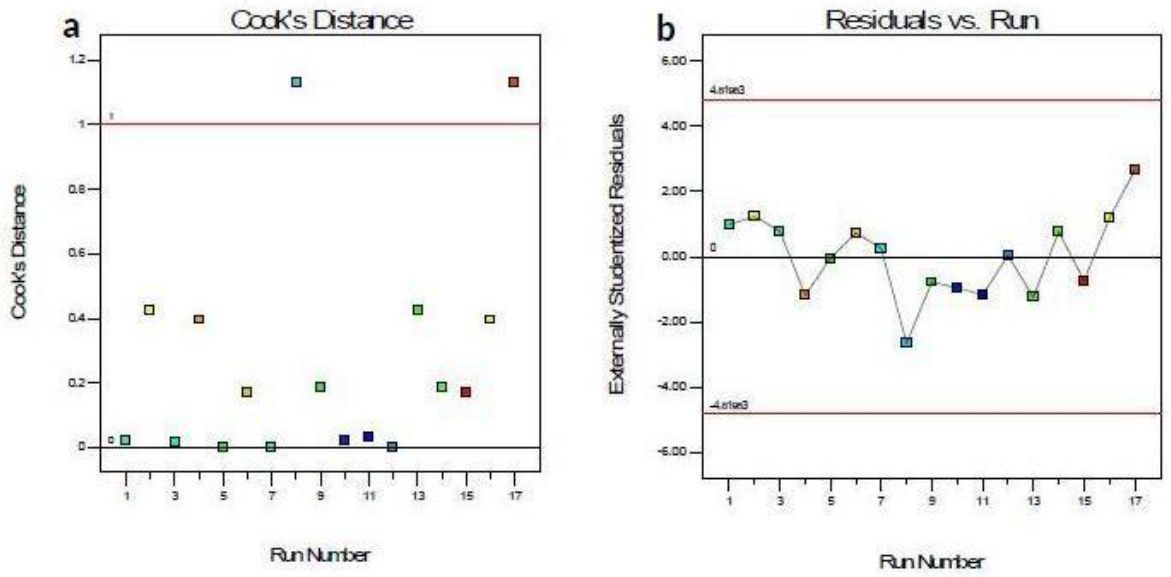

Figure 2. (a) Cook's distance plot of soil TPH bioremediation (b) Residuals versus experimental runs

3.2.2 Interaction Among Factors That Influence TPH Removal

The graphical representation of the response shown in Figures 3– 5 showed the effect of Parasitic mushroom (A), Saprophytic mushroom (B) and Symbiotic mushroom (C) on removal of TPH. The effect of the interaction of poultry droppings and Parasitic mushroom on TPH bioremediation is illustrated in Figure 3. It was observed in this study that; higher rate of TPH removal was attained with higher amendment Parasitic mushroom and relatively high amount of Saprophytic mushroom. The maximum degradation yield of TPH (48.4%) was obtained with 775g of Parasitic mushroom and 950g of Saprophytic mushroom at a fixed amount of Symbiotic mushroom of 774.324g. This was because of better bioavailability of substrate for the inherent microorganisms. Figure 4 shows the 3D response surface plot of the interaction effect between Parasitic mushroom and Symbiotic mushroom.

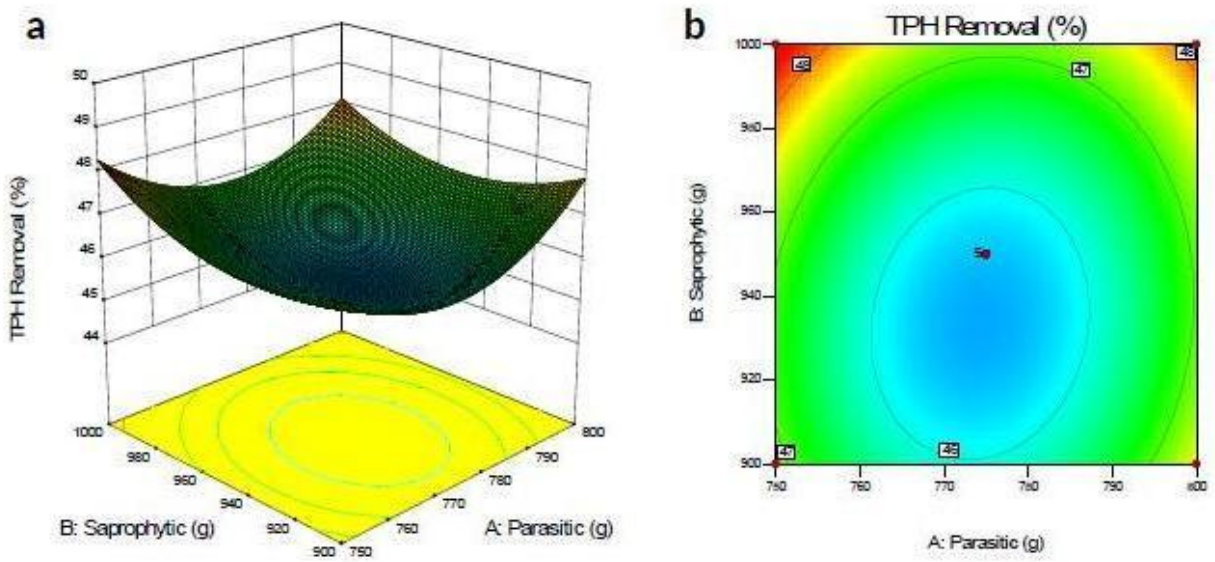

Figure 3. Response surface 3D and contour plots indicating interaction effects of factors Parasitic and Saprophytic mushroom

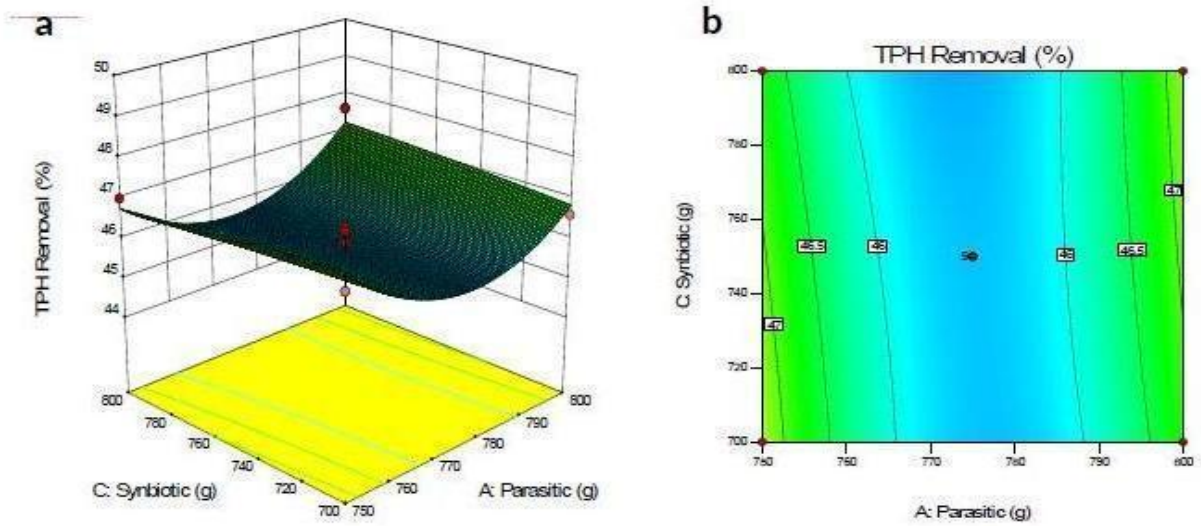

Figure 4. Response surface 3D and contour plots indicating interaction effects of factors Parasitic and Symbiotic mushroom

The plot demonstrated that both Parasitic and Symbiotic mushroom have interaction performance at optimum weight. A higher percent TPH removal was obtained at a higher amount of symbiotic mushroom with relatively high amount Parasitic mushroom and a fixed amount of Saprophytic mushroom, 950g. This three dimensional plot explained that both Parasitic and Symbiotic have individual impact on TPH removal as the individual coefficient of both Parasitic and Symbiotic is positive and their interaction effect is positive. Figure 5 showed the response surface 3D plot of the effect of interaction between Saprophytic and Symbiotic mushroom weights. Higher rate of TPH removal was observed with increase in Saprophytic and Symbiotic mushroom weights S due to positive interaction effect. Due to dominating interaction effects of Saprophytic mushroom, higher levels of this variable gave higher yields of TPH removal.

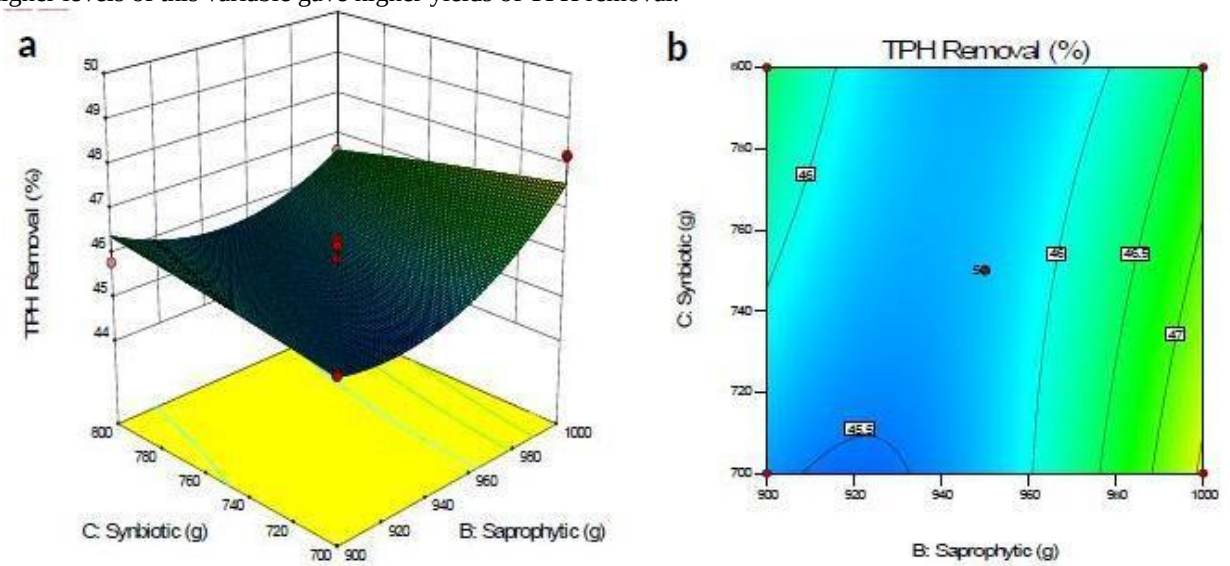

Figure 5. Response surface 3D and contour plots indicating interaction effects of Saprophytic mushroom and Symbiotic mushroom.

From the plots in Figures 3-5, it showed that each of the three variables used in the present study has its distinct effect on TPH removal by the inherent microbial populations in the soil. Gradual increase in Parasitic mushroom, Saprophytic and Symbiotic mushroom mass from low level (coded value –1) to a higher level (coded value +1) resulted in both increase and decrease of TPH degradation.

3.2.3 Artificial neural network (ANN)

Artificial neural network (ANN) work and react like the human brain and nerve systems that are known for their extreme ability to learn and arrange data (Prakash et al., 2008; Shojaeimehr et al., 2014). ANNs consist of an input and an output layer and one or more hidden layers. In input and hidden layers, each neuron receives input values. Neurons transfer input values to next layer that the strength of these connections determined by weights (Khataee and Khani, 2009; Shojaeimehr et al., 2014). In the present study, neural network is used for prediction of percentage removal of TPH in petroleum polluted soil. The best ANN chosen in the present work was a cascade-forward back prop type of network with training as TRAINLM, adaption learning function, LEARNGDM, performance function, MSE, number of layers, 2 with 8 neurons and TRANSIG transfer function. Figure 6 describes the ANN architecture showing the optimal design. A regression analysis between ANN outputs and the experimental data was carried out. This ANN model indicated a precise and effective prediction of the experimental data with a correlation coefficient (??) of 0.94874, 0.97651, 1.000 and 0.95688 for training, validation, testing and all data, respectively (see figure 7).

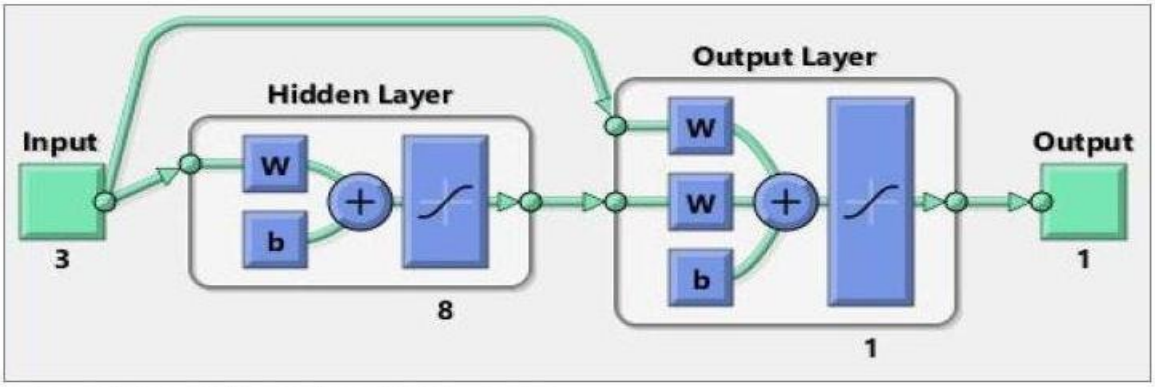

Figure 6. Optimal architecture of ANN model

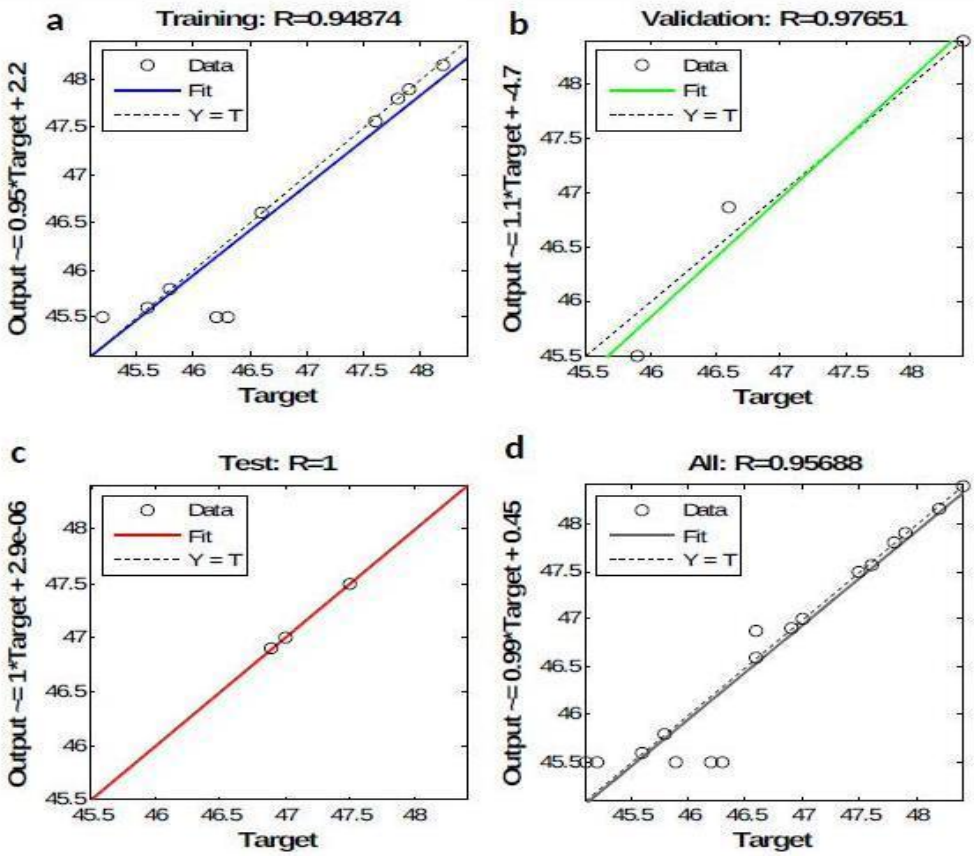

Figure 7. Network model with training, validation, test and all prediction set

Figure 7 shows the regression plots for the output with respect to training, validation, and test data. The output tracks the targets very well, and the R-value is 0.95688. In this case, the network response is satisfactory.

3.3 Optimization and Validation

Numerical optimization technique based on desirability function was used to determine the workable optimum conditions for the TPH bioremediation process. In order to provide an ideal case for biodegradation, the goal for Parasitic, Saprophytic and Symbiotic mushroom was set in range based upon the requirements of the TPH bioremediation and TPH removal was set on maximize. The predicted optimum (uncoded) values of Parasitic mushroom, Saprophytic and Symbiotic mushroom were found to be: 750g, 1000g and 700, respectively, to achieve 49.3175% maximum diesel oil removal; while desirability for the predicted optimum values was 0.821 from RSM optimization. ANN optimization was carried out in MATLAB following the predicted equations as shown in figure 7. Errors between predicted and actual values were calculated according to Eq. (4) (Mohajeri et al., 2010; Agarry and Ogunleye, 2012a; Agarry 2018)

$$\%\text{Error} = \frac{\text{Experimental value} - \text{Predicted value}}{\text{Experimental value}} \times 100$$

The error computation showed a reasonable accuracy in ANN prediction when compared with RSM (Figure 8a). The relationship between the variables and the response can be represented on graphical drawing, placing the experimental levels of each variable on the one side, and the type of interactions between the test variables, on the other, which allows deducing the optimum conditions. Figure 8b therefore shows a plot predicted against the experimental values for both RSM and ANN. The perfect straight line obtained indicated that the models factor and the predicted values are in agreement with each other.

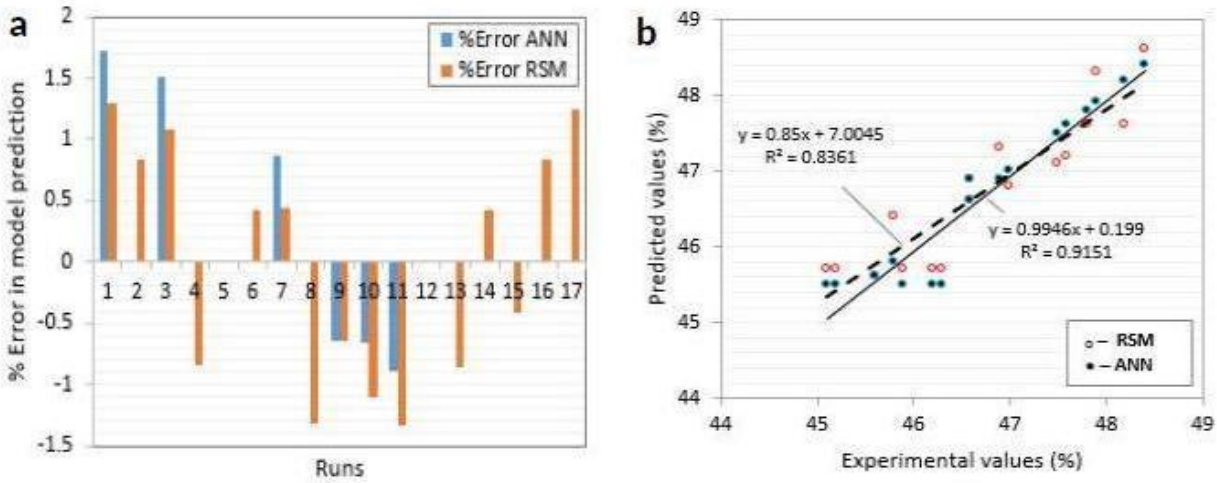

Figure 8. (a) Percentage error between predicted values versus no. of experimental runs (b) Correlation between experimental and predicted values of %TPH removal

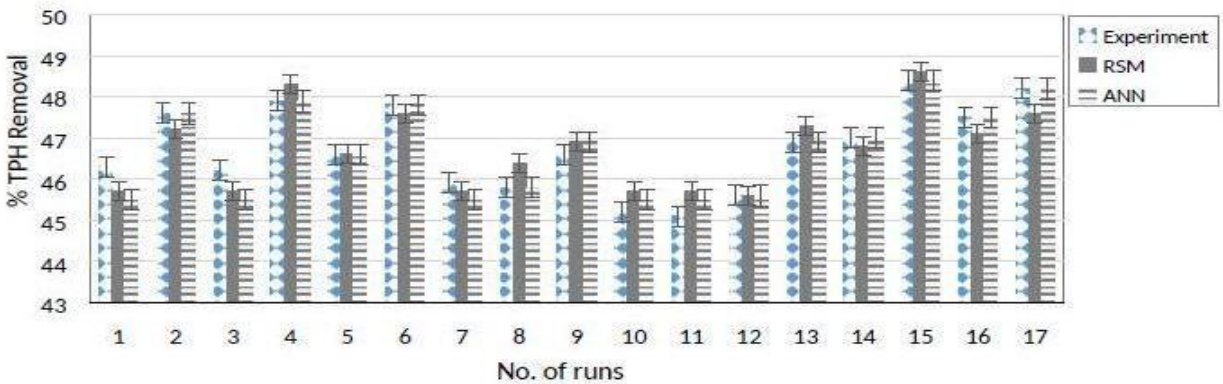

Figure 9. Comparison between experiment and predicted values

Coefficient of correlation, R2 = 0.8361 and 0.9151 for RSM and ANN respectively by comparing experimental and predicted %TPH removal. It shows that ANN has a higher correlation value than RSM (see figure 8b). Therefore, ANN is proven to be a better tool for optimization than RSM. This in agreement with Sushovan et al. (2018) on application of RSM and ANN for optimization and modeling of biosorption of chromium (VI) using cyanobacterial biomass. Similiarly, Tahereh et al. (2014) reported high acuracy in ANN compared to RSM having correlation coefficients of 0.962 and 0.941 for ANN and RSM respectively. Also, in the study of Hargovind and Narendranath (2018) ANN predictor gives more accurate values compare to RSM predicted values. However, comparing experimental values versus the predicted shows that ANN prediction is closer to the experimental values of %TPH removal than RSM prediction (see figure 9).

Conclusion

This study investigated the bioremediation of TPH polluted soil and its optimization using Response Surface Methodology. The petroleum contaminated soil was incubated for 28days bioremediation period and statistical analysis was carried out. The predicted optimum parameters were Parasitic mushroom: 750g, Saprophytic mushroom: 1000g and Symbiotic mushroom: 700g. The optimal TPH removal was found to be 49.3175%. At this optimum condition, it can be concluded that bioremediation resulted in petroleum hydrocarbon degradation.

Finally, an ANN model has been developed for TPH degradation based on part of the experimental data, and the model has sufficient generalization ability as evident from prediction of the unseen experimental data with reasonable accuracy.

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