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Process formulation and utilization of fertilizer admixture on the reduction of petroleum hydrocarbon polluted soil leachate

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ABSTRACT

Petroleum hydrocarbon leachate alters the physiochemical properties of ground water especially in areas with a higher water table. Failure to mitigate hydrocarbon polluted soils with the right fertilizer leads to ground water pollution. In this research, weathered crude oil was used as the soil polluter, and the biostimulants of pig manure (PM), pig manure biochar (PMB), inorganic fertilizer (IF) and their admixtures were used to examine their soil treatment capabilities. The effects of these biostimulants on pH, nitrate, phosphate and total petroleum hydrocarbon (TPH) levels in petroleum hydrocarbon polluted soil leachate were also investigated. The results obtained in this study indicate that as the percentage of petroleum hydrocarbon contamination increases, the pH, nitrate and phosphate contents of the soil leachate decreases. The results also revealed that there was a slight reduction in the levels of pH, nitrates, and phosphates as the incubating contact time increased. Furthermore, combined amendments, particularly PM+PMB application showed a remarkable reduction in the residual (<95%) TPH leachate levels. Therefore, this study has demonstrated that the biostimulant application of PM+PMB combinations is eco-friendly and more effective in the reduction of TPH, nitrates and phosphates in petroleum hydrocarbon soil leachate for agricultural and ground water security.

Keywords: petroleum hydrocarbon; leachate; pig manure; biochar

1. INTRODUCTION

Hydrocarbon soil contamination resulting from oil exploration is an issue in the Niger Delta region of Nigeria. The region is known for its increasing oil spills because of many years of oil and gas exploration, failures of oil installations, vandalism and natural seepage (Ogheneoruese Onoharigho et al., 2022). Low molecular weight hydrocarbon compounds are highly mobile in the environment, while high molecular weight tend to bind strongly to soil particles near the source or remain entrapped within an organic phase. There are many oil spill sites in Niger Delta and not one spill site has been adequately cleaned up. In some of the sites, pollution has reached ground water (Nuhu et al., 2021). Remediating these sites with eco-friendly fertilizer would provide more land for arable farming, portable water and a generally healthier environment (Sayed et al., 2021). Oil contamination from crude oil causes soil toxicity and contaminates ground water, particularly in low water table areas (Li et al., 2021). Pollution of soil and water is a general concern around the globe because of many sources, such as oil spillages, hydrocarbon storage tanks and the illegal refining of crude oil. Accidents from oil tankers and oil from waste pits also cause ground water pollution (Adeola et al., 2022). Kalhor et al. (2019) asserted that soil acts as route for contamination to ground water aquifers. Hydrocarbon leaching is the downward transport of dissolved hydrocarbons in the soil profile with percolated water. Such leached hydrocarbons and fertilizers contribute to ground water pollution (Pérez-Lucas et al., 2019). Leaching is faster in humid areas than in dry regions. The factors affecting petroleum hydrocarbon (PHC) ground water contamination are the depth of ground water, the particle size distribution of soil, and the level of hydrocarbons present in soil.

Managing hydrocarbons that have been spilled into the environment requires detailed knowledge on how they interact with soil water media (Ali et al., 2020). A major problem in groundwater is the cleaning of hydrophobic organic compounds, which are also dangerous to human health (Al-Hashimi et al., 2021). Improper disposal of petroleum products (e.g., jet fuel, refinery wastes, diesel, and used lubricating oil) and volatile organic solvents are recognized as two of the most widespread causes of groundwater contamination by chemical compounds. Immiscible liquids enter the ground water ground due to gravitational and capillary forces (Leharne, 2019).

PHCs can be held in voids in the soil, in the form of residual saturation, and can lead to long-term pollution of groundwater through the action of rainwater, if not remediated within a short time. Groundwater is easily contaminated if not protected by a low permeability layer such as clay and soil organic matter.

Natural oil spillages may leach from the soil for a longer period of time and thus become a continuous source of soil and groundwater pollution. The use of surfactants can reduce the mobility of hydrocarbon contaminants in soil–water systems by adsorbent hydrocarbons adhere to the surface of the surfactant (Saint-Fort, 2022). Many researchers have used fertilizer, poultry manure, and biochar for PHC remediation and nutrient enhancement without assessing the consequences on ground water. Therefore, this study examines the effect of inorganic fertilizer (IF), pig manure (PM), and their admixtures on total petroleum hydrocarbon (TPH), nitrates, and phosphates in PCH-polluted soil leachate. IFs are chemical fertilizers that are made up of a variety of chemical formulations to suit different uses (Kakar et al., 2020). Most of them are made of phosphorus, potassium, and nitrogen, which are elements needed by crops (Jiaying et al., 2022). IF only provides short term nutrient enhancement and remediation, and also induces secondary contamination of soil and ground water (Verma, 2022). Organic fertilizers are prepared from poultry droppings, animal dung, or vegetable waste. They add organic nutrients to soil, improve soil organic matter, increase water holding capacity, reduce soil crusting problems, reduce erosion from wind and water, and consistently release nutrients for crops and microorganism utilization. Another fertilizer is biochar. It is produced through the pyrolysis of biomass or carbonization at varying temperatures and times, depending on the intended use.

Biochar can be prepared to address agricultural and environmental needs by adding additives or undergoing treatments. It absorbs water in its cavities which assists plants to cope during drought (Ahluwalia et al., 2021). It also serves as a sink for microorganisms to grow. The pore volume of biochar is affected by the way it is prepared (Oni et al., 2019). Biochar has a higher adsorption affinity for many organic and inorganic contaminants compared to other forms of soil organic fertilizer (Ambaye et al., 2021). It detoxifies soil water by adsorbing compounds that inhibit microbial growth.

2. MATERIALS AND METHODS

2.1 Sample collection

The unpolluted soil (US) used for this study was obtained from farmland in Mosogar, Delta state (5.8790° N, 5.7334° E). The soil was air dried, and the debris and roots were removed. The dried soil was crushed in a porcelain mortar and filtered through a 2-mm sieve. Samples were stored in polyethylene bags that had been cleaned with chromic acid, and were labeled before analysis.

2.2 Fertilizer and poultry manure

PM and inorganic fertilizer (IF) were collected from a Songhai integrated farm in Amukpe, Sapele and the Ministry of Agriculture, Zonal Headquarters, Sapele respectively. The PM and the IF were crushed in a porcelain mortar and passed through a 2-mm sieve. Samples were preserved in polythene bags and labelled prior to analysis.

2.3 Biochar production

The PM were dried and carbonized in an oven at 350 °C. The carbonized biochar was allowed to cool at room temperature overnight (Amalina et al., 2022). The biochar was crushed into small granules and then passed through a 2-mm sieve for proper preservation.

2.4 Treatment of crude oil contaminated soils

Crude oil was obtained from Seplat Development Oil Company drilling sites in Sapele. Crude oil was weathered for 14 days (Lofthus et al., 2020), then 3 kg of soil were spiked with the crude oil at varying concentrations (0%, 2%, 4%, and 6%) and thoroughly homogenized.

The contaminated soil samples were left for 21 days as a stabilization period before the addition of the biostimulants. During this period, the polluted soil was maintained at 50% water holding capacity.

2.5 Soil amendment treatment and design

After 21 days treatment PM, PMB and IF were applied to the soil individually and in different ratios. The weight of soil to PM and PMB was 120 g per 3,000 g respectively. The PM/PMB mixture was 120 g (60g of PM and 60g of PMB) to 3,000 g soil, and that of PM+PMB+IF was also 120 g (40 g of PM, 40g of PMB and 40g of IF) to 3,000g. The amended soils were mixed thoroughly, watered with deionized water and maintained at 50% water holding capacity for an 84-day incubation period allowing proper aeration. Periodic sampling of soil from each container was done at 3-week intervals.

2.6 Leachate processing

Air-dried soil (100 g) was packed into a soil column (25 cm in length and 3 cm inner diameter) that was previously packed with cotton wool. The surface of the column was covered with filter paper. Deionized water was then added to the soil column using a dropping burette (De Gisi et al., 2016). The leachate was thereafter collected after 48 h.

2.7 Determination of physico-chemical properties of soil and total petroleum hydrocarbon (TPH)

Black method (Bahadori and Tofighi, 2016) was used for pH determination. The concentration of phosphorus was determined using the method of Wieczorek et al. (2022), the nitrogen content was determined following Tessier's (2018) method, and the level of soil nutrients (phosphorus, sodium, calcium, magnesium and potassium) were determined following standard methods (Hailu et al., 2015). The methods of Berkessa et al. (2019) were used for nitrate and phosphate determination, and the level of TPH was determined using gas chromatography with a flame ionization detector (Simeon et al., 2020).

2.8 Statistical analysis

Anova was used to determine the statistical significance (SPSS Version, 24) between different treatments and times. The probability was set at 0.05 level of significance.

3. RESULTS AND DISCUSSION

The physiochemical properties US, PM and PMB are shown in Table 1. Soil texture of US revealed that the US soil was sandy loam. The soil composition contained mostly sand (74.45%), followed by clay (23.39%) and silt (2.16%).

The pH of US is within the range of agricultural soil (Neina, 2019) (Table 1). The water holding capacity (WHC) of the US is slightly low. This might be attributed to low soil organic matter (SOM). Soil with low WHC is prone to IF leaching (Kuo et al., 2020).

The measured essential nutrients (N, P, Ca, Mg, K, Na, C), pH and WHC of PM and PMB (Table 1) showed that single or co-application for PHC soil amendment will be a better biostimulant for PHC polluted soil leachate reduction.

Table 1. Physiochemical properties of US, PM and PMB

Parameter	US	PM	PMB
pH	5.72±0.05	7.82±0.05	8.87±0.05
P (%)	1.03±0.02	12.43±0.14	9.89±0.07
N (%)	1.00±0.01	23.06±0.56	9.69±0.13
Na (mg/kg)	0.99±0.01	184.23±2.76	23.06±1.31
K (mg/kg)	0.85±0.02	1,334.95±6.72	1,339.65±17.02
Mg (mg/kg)	0.97±0.00	438.70±4.66	519.27±2.66
Ca (mg/kg)	0.66±0.00	25.53±0.22	27.09±0.24
% Water holding capacity	0.36±0.01	0.74±0.01	0.85±0.01

3.1 Level of pH, nitrate and phosphate in PHC leachate

In this context, leachate is any liquid that is in the course of passing through soil and it extracts some of the material through which it passes. Leaching is highly dependent upon structure with water infiltration rates and low nutrient retention capacities.

Sandy soils and well-structure ferralitic soils with low organic matter content are particularly conducive to leaching (Pareek, 2017). Results in Table 2 shows that pH reduces as the dosage of PHC contamination increases. However, the pH of leachate significantly increases after treatment with biostimulants. This might be due to releases of free bases by the biostimulants (Bartucca et al., 2022). In addition, there was a decrease in level of pH as the period of remediation increased from 21 days to 84 days. This observation was likely due to the utilization of free bases by the biodegrading microbes of the amended soil. PMB had the highest pH follow by the PM+PMB combination. The level of nitrates (Table 3) and phosphates (Table 4) in the leachate indicates that IF had the highest pH while PM+PMB had the lowest, followed by PMB. Though the concentration of the nitrate and phosphate decreased as the period of incubation/contact time increased, this might be due to the utilization of nitrates and phosphates by the TPH degrading microbes. The reduction of nitrate leaching by PMB and the PM+PMB combination included absorption of nitrates into PMB and PM+PMB, and enhanced immobilization and denitrification of nitrogen (Grzyb et al., 2021).Other factors that facilitate the reduction of nitrates in PMB and PM+PMB may be the presence of anaerobic conditions and organic carbon as an electron donor, thus facilitating denitrification (Kamp et al., 2015). The use of animal manure singly and wood manure biochar to reduce leaching of nitrates has been earlier documented (Vamvuka and Raftogianni, 2021). In the same vein, the decrease in phosphate concentration by PMB and PM+PMB compared with the control is attributed to adsorption of ortho-phosphate and organic phosphorus compounds by the PMB and PM+PMB. Similar results were also observed by (Afridi et al., 2019) in tillage amended biochar and manure soil. The significant concentrations of nitrates and phosphates in IF and IF combinations can pollute ground water, especially when the amendment is higher.

3.2 Level of TPH in petroleum hydrocarbon polluted soil leachate

The cumulative concentration of PHC in leachate is referred to as TPH leachate (TPHL). In this present study, the results of the biostimulants (PM, PMB, IF, PM+IF, PM+ PMB, PM+PMB+IF) on TPHL (Table 5) indicated that as the percentage of PHC contamination of soil increases the rate of TPHL also increases.

The results suggested that the TPH concentration in the leachate depended on the PHC concentration in the soil. However, there was a significant difference (p<0.05) between levels of TPHL contamination. In addition the level of TPH in leachate was low when compared to soil. This indicates that some proportion of the TPH must have been bound to the soil particles. Poor solubility of PHC with water must have also be responsible for this observation. These results are in agreement with Zabbey and Olsson (2017), who reported that less than 5% of crude oil will dissolve in water. Biostimulant treatment of PHC reduced the rate of TPHL significantly (p<0.05). Moreover, soil TPH leachate decreased to 0 in the PM+PMB amended soil with an 84-day incubation period for 2% soil contamination representing 100% remediation (Table 5). Over 96% remediation was also noted for treated soil with PMB+IF, PM+PMB+F and PMB+F. This suggests that the remediation of TPHL depends on the nature of the biostimulant, time and the concentration of PHC. The remediation of TPHL by the biostimulants follows the following trend: PM+PMB>PMB+PM+IF>PMB+IF PM+IF>PMB>IF>PM. The effectiveness of PM+PMB on TPHL might be due to the synergetic effects of PM and PMB which aid the activity of bacterial degrading TPH. Sorption properties of biochar could also be a contributor towards the effectiveness of PM+PMB.

Table 2. Comparison of mean pH content of soil leachate contaminated with 2%, 4%, and 6% PHC amended with biostimulants after 21 day, 42 day, 63 day, and 84 day remediation periods

		Remediation Period (days)			
		21	**42**	**63**	**84**
% Contamination	Biostimulant				
2%	Control	5.41	5.40	5.35	5.34
	PM	6.54	6.37	6.28	6.26
	PMB	7.23	7.14	7.03	6.94
	IF	6.14	6.05	5.98	5.49
	PM+IF	6.29	6.17	6.03	5.94
	PMB+IF	6.36	6.31	6.26	6.23
	PM+PMB	6.74	6.65	6.58	6.56
	PM+PMB+IF	6.34	6.29	6.22	6.17
4%	Control	5.39	5.37	5.30	5.21
	PM	6.17	6.13	6.10	5.90
	PMB	6.58	6.49	6.37	6.15
	IF	5.83	5.72	5.41	5.35
	PM+IF	5.89	5.81	5.63	5.33
	PMB+IF	6.17	6.15	6.03	5.81
	PM+PMB	6.34	6.30	6.17	6.04
	PM+PMB+IF	6.10	5.90	5.81	5.70
6%	Control	5.31	5.28	5.25	5.20
	PM	6.10	6.01	5.83	5.79
	PMB	6.41	6.23	6.21	6.08
	IF	5.71	5.54	5.38	5.31
	PM+IF	5.69	5.38	5.26	5.14
	PMB+IF	5.94	5.49	5.26	5.20
	PM+PMB	6.20	6.09	6.07	5.95
	PM+PMB+IF	6.03	5.66	5.61	5.47

Table 3. Comparison of mean nitrate (mg/L) content of soil leachate contaminated with 2%, 4%, and 6% PHC amended with biostimulants after 21 day, 42 day, 63 day, and 84 day remediation periods

		Remediation period (days)			
		21	**42**	**63**	**84**
% Contamination	Biostimulant				
2%	Control	0.36	0.34	0.33	0.30
	PM	2.18	2.17	2.16	2.11
	PMB	2.25	2.24	2.23	2.17
	IF	93.00	89.39	83.75	71.48
	PM+IF	93.87	87.36	58.48	33.21
	PMB+IF	59.21	53.43	52.85	49.39
	PM+PMB	2.34	2.30	2.12	2.10
	PM+PMB+IF	20.80	20.21	18.77	12.27
4%	Control	0.34	0.33	0.32	0.29
	PM	2.17	2.16	2.15	2.09
	PMB	2.16	2.14	2.12	1.59
	IF	58.48	54.87	49.82	39.71
	PM+IF	49.10	46.93	39.71	37.18
	PMB+IF	47.65	40.43	38.27	33.21
	PM+PMB	2.31	2.17	2.11	1.81
	PM+PMB+IF	20.22	20.10	18.61	12.25
6%	Control	0.32	0.30	0.29	0.28
	PM	2.16	2.15	2.14	2.00
	PMB	2.14	2.12	2.10	1.12
	IF	49.82	41.16	38.99	35.51
	PM+IF	40.72	37.18	29.68	26.72
	PMB+IF	39.57	36.10	31.77	28.89
	PM+PMB	2.24	2.15	2.10	1.20
	PM+PMB+IF	20.20	20.15	18.50	12.41

Table 4. Comparison of mean phosphate (mg/L) content of soil leachate contaminated with 2%, 4%, and 6% PHC amended with biostimulants after 21 day, 42 day, 63 day, and 84 day remediation periods

		Remediation period (days)			
		21	**42**	**63**	**84**
% Contamination	Biostimulant				
2%	Control	0.50	0.48	0.47	0.41
	PM	2.16	2.13	2.10	2.02
	PMB	2.15	2.13	2.11	2.05
	IF	57.27	51.77	48.11	42.51
	PM+IF	42.41	34.95	25.56	18.61
	PMB+IF	40.55	35.05	32.95	30.00
	PM+PMB	2.16	2.11	2.10	1.88
	PM+PMB+IF	31.61	32.05	30.00	29.01
4%	Control	0.41	0.40	0.39	0.35
	PM	2.11	2.10	2.09	2.00
	PMB	2.10	2.06	2.00	1.97
	IF	56.31	51.55	46.11	40.65
	PM+IF	41.55	32.75	22.50	17.40
	PMB+IF	39.90	36.50	30.25	29.58
	PM+PMB	2.12	2.10	2.09	1.08
	PM+PMB+IF	30.78	30.50	28.02	25.45
6%	Control	0.40	0.39	0.37	0.31
	PM	2.10	1.61	1.56	1.55
	PMB	2.08	1.61	1.58	1.57
	IF	47.75	38.11	28.14	22.25
	PM+IF	20.01	19.90	19.81	15.50
	PMB+IF	20.02	19.25	18.99	18.75
	PM+PMB	2.10	2.07	2.01	1.02
	PM+PMB+IF	18.25	17.21	17.15	17.00

Table 5. TPH (μg/ L) Content of soil leachate contaminated with 2%, 4% and 6% PHC amended with biostimulant after 21 day, 42 day, 63 day, and 84 day remediation periods

		Remediation period (days)			
		21	**42**	**63**	**84**
% Contamination	Biostimulant				
2%	Control	337,751.17±90.26	321,446.77±0.41	305,397.54±20.39	2412.71±20.81
	PM	239,580.16±40.53	217,952.77±13.66	168,037.41±30.41	2,188.34±0.51
	PMB	178,240.04±17.57	162,296.76±24.80	122,189.60±30.41	1,161.327±7.14
	IF	148,224.00±62.41	137,986.83±16.44	101,638.72±23.65	282.96±0.83
	PM+IF	100,975.96±54.11	94,091.93±17.34	69,797±24.33	291.22±0.83
	PMB+IF	91,850.38±50.24	85,240.54±19.70	36,368.03±25.47	262.96±2.78
	PM+PMB	29,169.37±51.59	24,940.84±17.23	18,554.43±22.13	0
	PM+PMB+IF	76,820.55±53.92	69,690.84±17.23	52,740.45±24.16	12.77±0.29
4%	Control	421,485.69±51.04	400,436.34±21.38	376,647.86±30.04	353,056.13±22.43
	PM	349,844.42±17.88	231,939.12±21.89	216,966.19±22.30	1,775.09±23.53
	PMB	222,072.63±19.85	204,878.77±14.52	169,312.47±23.55	1,162.21±12.10
	IF	203,114.20±20.44	184,303.87±64.79	15,324.35±26.72	1,076.92±16.21
	PM+IF	193,778.08±23.77	178,009.44±14.80	132,157.93±20.62	821.19±3.52
	PMB+IF	151,788.27±21	127,175.59±20.13	92,147.43±48.56	523.38±8.20
	PM+PMB	93328.90±21.10	55,565.47±18.68	41,874.37±22.31	110.51±2.09
	PM+PMB+IF	102,757.85±20.18	95,627.05±20.60	71,288.03±22.43	265.18±5.20
6%	Control	524,118.26±40.73	503,345.84±38.68	488,312.37±52.82	467,308.14±50.16
	PM	425,986.85±41.37	318,492.27±39.80	225,498.51±26.63	129,259.83±24.89
	PMB	361,855.03±40.07	264,112.79±32.65	215,233.30±25.19	108,124.13±37.25
	IF	274,248.19±54.11	242,807.77±36.17	211,700.95±28.11	105,257.79±35.04
	PM+IF	254,948.21±34.11	235,876.60±33.22	190,460.80±26.74	70,424.41±26.00
	PMB+IF	158,780.03±32.12	148,143.56±36.26	124,820.71±13.94	60,235.79±10.42
	PM+PMB	104,588.31±36.98	78,463.30±28.71	62,377.40±16.97	15,454.10±28.06
	PM+PMB+IF	151,937.34±39.65	123,304.10±36.26	98,060.94±13.77	16,220.10±39.00

4. CONCLUSION

The results of this study indicate that the unpolluted soil was sandy loam. The results also revealed that PM and PMB contain relative high essential-nutrient and water-holding capacities. Furthermore, PM+PMB were more effective in the reduction of nitrates and phosphates than IF and its admixtures. In the same vein, the admixtures of PM+PMB revealed a higher reduction of residual TPH in soil leachate. Therefore, the PM+PMB combination decreased nitrate, phosphate and TPH leachate for ground water security.

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