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Research article

Evaluation of Organic Manures Effects on Crude Oil Polluted Soil

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Abstract

Artificial pollution experiment studies were carried out on the bioremediation of crude oil polluted soil using poultry droppings (PD), cow dung (CD) and urban waste (UW) in singly and equally mixed proportions. The natural soil sample was collected at random at the back of School of Postgraduate Studies building within the Federal University of Technology, Akure and was artificially spiked with 50ml Forcados Crude Oil per 500g soil samples, biostimulated by 100g of PD, CD and UW in singly and equally mixed proportions per experimental pot and studied. Natural soil, artificially polluted soil and bioremediated soil samples were all characterized for pH, K^+, Na^+, Ca^{2+}, Mg^{2+}, CEC, moisture, WHC, porosity, electrical conductivity, total organic carbon, organic matter, total nitrogen, phosphorus and soil particle size analysis using standard analytical methods to determine the effects of crude oil pollution on these properties. Results revealed the improvement in soil minerals and nutrients in organic manures biostimulated soils in the sequence of PD + PS > CD + PS > PD + CD + PS > UW + PS > PD + CD + UW + PS > PD + UW + PS > CD + UW + PS. Total petroleum hydrocarbon (TPH) was determined by measuring the amount of TPH left in the crude oil polluted and organic manures biostimulated soils at 7 days interval for 42 days of crude oil treatments and organic manures biostimulation. Results showed remarkable reduction of TPH in organic manures biostimulated soils samples in the sequence of PD + PS (16.25mg/kg and 90%) > CD + PS (18.26mg/kg and 88.88%) > PD + CD + PS (50.13mg/kg and 69.48%) > UW + PS (62.25mg/kg and 62.10%) > PD + CD + UW + PS (65.53mg/kg and 60.10%) > PD + UW + PS (68.88mg/kg and 58.06%) > CD + UW + PS (71.13mg/kg and 56.69%) till the 42nd day of the study which revealed better effectiveness of singly organic manures to mixed organic manures in applications to biodegrade petroleum hydrocarbons in the crude oil polluted soil with improved soil minerals and nutrients.

Keywords: Biostimulation, Biodegradation, Crude oil, Total petroleum hydrocarbons (TPH)

Introduction

Incessant crude oil pollution in oil rich areas of Niger delta of Nigeria is gradually becoming worrisome due to its direct socioeconomic challenges, confronting the people of the region. In Nigeria, a number of oil spill incidents have been recorded and documented which include Chevron's oil spill in Ilaje, Ondo state; Shell's oil spill in Barrale, Ogoniland, Rivers state and Shell's oil spill in Gana and Urhobo community in Delta state among others [1]. Soil, among the receiving end of the crude oil pollution is called for critical attention since majority of the affected people are arable farmers that depend solely on soil for their agricultural activities. The unsatisfactory effects of the crude oil pollution on soil chemistry and biota have been reported by many researchers. Meanwhile, the use of fertilizers have been noted to be effective in remediating crude oil polluted soil but not the use of inorganic fertilizers that have not been accommodating as it is allied with increased soil acidity and nutrient imbalance and further continuous use of fertilizers creates potential polluting effect in the environment [2]. The availability, affordability and eco-friendly characteristic advantages of organic manures over inorganic fertilizers make them suitable for bioremediation of crude oil polluted soils. Biostimulation plays a direct role as a source of all necessary nutrients in available forms during mineralization of biostimulated soil, thereby improving both the physical and the biological properties of the soil [3]. Okolo *et al.* [4] investigated the impact of addition of poultry manure alone to enhance bioremediation process in crude oil contaminated soil, while Ibekwe *et al.* [5] studied the effect of organic nutrient on microbial utilization of hydrocarbons on crude oil contaminated soil. However, Lee *et al.*, [6], Ebere *et al.*, [7] and Rowland *et al.*, [8] have investigated the bioremediation of crude oil polluted soil using inorganic N.P.K fertilizer, poultry manure, cow manure, palm kernel ash and saw dust organic manures in singly and mixed proportion. Organic manures have been tested as biostimulating agents with varying degree of successes in bioremediation of crude oil polluted soils. Meanwhile, much have to be done and reported on biostimulation and biodegradation of crude oil polluted soil using a proper mixture proportion array of equally mixed organic manures to establish the potential of organic manures in biostimulation and biodegradation process. In this study, poultry droppings, cow dung and urban waste organic manures as biostimulating and biodegrading agents would be employed in singly and equally mixed proportions to evaluate the effects of organic manures on crude oil polluted soil.

Materials and Methods

Samples Collection:

Soil samples were collected at random from an unutilized land at the back of School of Postgraduate Studies building within the Federal University of Technology, Akure using a hand trowel at a depth of 0-20cm below soil surface, having no pollution history and devoid of petroleum hydrocarbons contamination. Crude oil was obtained from Shell Petroleum Development Company (SPDC) forcados terminal, Burutu, Delta state, Nigeria. Urban waste was collected at the general township dumping site along Irese road in Akure while Poultry droppings and Cow dung were collected from the Teaching and Research Farm (Livestock section) of the Federal University of Technology, Akure, Nigeria.

Sample Preparation, Artificial pollution and Treatment:

Soil was air dried for a period of seven days in a clean well ventilated laboratory and sieved by passing through a 2mm mesh sieve. 500g of soil was each measured into clean dry experimental pot and moistened with distilled water to ensure proper mixing with the crude oil. Artificial pollution of the soil samples was done by measuring 50ml of crude oil into the experimental pot containers containing 500g of soil each. The individual mixtures were thoroughly mixed to achieve a 10% artificial pollution. 10% spiking was adopted to achieve severe pollution because beyond 3% concentration [9], crude oil has been reported to be increasingly deleterious to soil biota [10]. The poultry droppings, cow dung and urban waste manures samples were sun dried for seven days after which they were grinded, thoroughly mixed, sieved through a 2mm sieve to achieve uniform particle size and stored in neat polythene bags for use. 100g of the poultry droppings, cow dung and urban waste manures samples was added to the experimental pots containing 500g of 50ml crude oil artificial polluted soil and thoroughly mixed in singly and equally mixed proportions to obtain homogeneity and to allow proper bioremediation for another 42 days with periodic watering. The experimental pots contained natural soil (NS); polluted soil (PS); poultry droppings + polluted soils (PD + PS); cow dung + polluted soils (CD + PS); urban waste + polluted soils (UW + PS); poultry droppings + cow dung + polluted soils (PD + CD + PS); poultry droppings + urban waste + polluted soils (PD + UW + PS); cow dung + urban waste + polluted soils (CD + UW + PS) and poultry droppings + cow dung + urban waste + polluted soils (PD + CD + UW + PS) for 42 days bioremediation study periods. This experimental design was a randomized complete block and duplicated.

Soil Characterization/Physicochemical Analysis:

Soil physicochemical characteristics such as soil particle size analysis, pH, K^{+}, Na^{+}, Ca^{2+}, Mg^{2+}, cations exchange capacity (CEC), moisture, water holding capacity (WHC), porosity, electrical conductivity, total organic carbon (OC), organic matter (OM), total nitrogen (N) and phosphorus (P) were determined before pollution, 7 days interval for 42 days after pollution and bioremediation process. Soil pH was determined electrometrically following the procedure outlined by Mylavarapus and Kennelley [11]. Particle size analysis was done using bouyoucos hydrometer method [12]. Soil minerals were determined by the method of Tel [13]. Total organic carbon and matter were determined by the wet dichromate acid oxidation method of Nelson and Sommers [14]. Soil water holding capacity and porosity were determined by the method of Michael [15]. Total Nitrogen was determined using the method of Radojevic and Bashkin [16]. Total Phosphorus was determined by Bray and Kurtz method [17]. Electrical conductivity was carried out as described by Chopra and Kanzer [18]. Soil moisture was determined using the method of Michael [15].

Determination of Total petroleum Hydrocarbon (TPH):

1g each of the artificially polluted and biodegradated soil samples were dissolved in 10ml of hexane and shaken for ten minutes using a mechanical shaker. The solution was filtered using a whatman filter paper and the filtrate diluted by taking 1ml of the extract into 50ml of hexane [9]. Procedural blanks and standard solutions were prepared and included to ensure analytical quality control so as to ensure the accuracy and reproducibility of the results. The absorbance of this solution was read at 460nm with HACH DR/2010 Spectrophotometer using n-hexane as blank. Replicate analyses were carried out on the determination of TPH to yield a statistical mean which will be used to determine trueness and also standard deviation of the mean to measure precision [19] [20]. Total petroleum hydrocarbon (TPH) was determined at 7 days interval for 42 days.

Results and Discussion

Table 1 showed the nutrients and minerals analysis results of crude oil (CO), poultry droppings (PD), cow dung (CD) and urban waste (UW) samples used in this study. Crude oil polluted soil (control 2) was observed to have reduced in pH, conductivity, K^+, Na^+, Ca^{2+}, Mg^{2+}, cations exchange capacity (CEC), moisture, WHC and porosity as compared to the natural soil (control 1) as shown in Table 2. The observed reduction followed the submissions of Oyedele and Amoo [21] and Osuji and Nwoye [22]. The observed reduction in pH and conductivity could be as a result of increase in hydrophobicity of the soil condition which prevented air and water to penetrate the crude oil polluted soil that favoured the release of hydrogen ion (H^+) which made the soil to increase in acidic condition [21] and non-polar nature of the crude oil, bringing about reduced ionic movement in the soil [19] respectively. Soil K^+, Na^+, Ca^{2+}, Mg^{2+}, cations exchange capacity (CEC), moisture, WHC, and porosity reduction in crude oil polluted soil might be due to the immobilization of these soil nutrients and minerals by crude oil that made them to be trapped in organic phase of the crude oil polluted soil and unavailable to the soil in solution [21]. The organic carbon and organic matter increase in crude oil polluted soil as compared to the natural soils as revealed in Table 2 may be due to the added carbon substrate from the crude oil [23]. The increase in C/N ratio was an indication of stress caused by crude oil pollution to the soil.

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Table 1: Results of nutrients and minerals analysis of crude oil and organic manures

Parameters	Crude oil (CO)	Poultry droppings (PD)	Cow dung (CD)	Urban waste (UW)
pH	4.22 ± 0.100	6.94 ± 0.158	7.70 ± 0.000	5.3 ± 0.100
Organic Carbon (%)	97.05 ± 0.014	8.53 ± 0.016	13.45 ± 0.014	3.41 ± 0.000
Organic Matter (%)	-	14.71	23.19	5.88
Nitrogen (%)	0.10 ± 0.005	1.96 ± 0.000	1.29 ± 0.001	1.10 ± 0.001
Phosphorous (%)	0.06 ± 0.000	1.33 ± 0.014	0.56 ± 0.000	0.52 ± 0.000
K^+ [Cmol/kg]	0.07 ± 0.001	5.20 ± 0.010	2.12 ± 0.005	1.99 ± 0.001
Na^+ [Cmol/kg]	0.07 ± 0.005	1.29 ± 0.000	0.98 ± 0.014	0.84 ± 0.005
Ca^{2+} [Cmol/kg]	0.11 ± 0.014	1.05 ± 0.001	0.20 ± 0.000	0.20 ± 0.000
Mg^{2+} [Cmol/kg]	0.09 ± 0.010	2.90 ± 0.005	0.51 ± 0.005	0.48 ± 0.001
CEC [Cmol/kg]	0.34	10.44	3.81	3.51
C/N	970.5	4.35	10.43	3.10

Results = Mean values ± standard deviation

The results in Table 2 revealed the increase in pH, conductivity, organic carbon, organic matter, total nitrogen, phosphorus, K^+, Na^+, Ca^{2+}, Mg^{2+}, cations exchange capacity (CEC), moisture, WHC and porosity in organic manures biostimulated soils as compared to crude oil polluted soil (control 2) in the sequence of PD + PS > CD + PS > PD + CD + PS > UW + PS > PD + CD + UW + PS > PD + UW + PS > CD + UW + PS which may be due to the additional nutrients supplement and energy being supplied by the organic manures that microbially mineralized the biostimulated soils that enhanced the improvement of the soil properties. This is in line with the findings of Obasi *et al* [24] and Okoro *et al* [25]. The higher pH and conductivity values of PD + PS encouraged more soil microbes to thrive in order to mineralize organic manure biostimulated soil more than the other singly and equally mixed organic manures biostimulated soils in the sequence of CD + PS > PD + CD + PS > UW + PS > PD + CD + UW + PS > PD + UW + PS > CD + UW + PS [26]. The increase in organic carbon and organic matter in organic manures biostimulated soils as compared to crude oil polluted soil (control 2) is in line with the earlier reports of Mbah *et al.*, [27]. The increase may be due to the biohumification of organic materials of the organic manures [21] to influence the increase in microbial biomass and population for soil mineralization towards the enhancement of physicochemical properties of the organic manures biostimulated soils. The highest improved soil organic carbon, organic matter, total nitrogen, phosphorus, minerals, nutrients, moisture, WHC and porosity in PD + PS organic manure biostimulated soil was due to the high minerals and nutrients dissolvability, decomposability and absorbability of poultry droppings than other singly and equally mixed organic manures biostimulated soils [28]. The sharp reduction of C/N of organic manures biostimulated soils was also observed in Table 2 as compared to the crude oil polluted soil (control 2). This is in line with the submissions of Obasi *et al* [24]. It may be due to the microbial fixation of nitrogen from atmosphere into the organic manures biostimulated soils with the added nutrients from organic manures [21]. The reduction C/N of organic manures biostimulated soils was in the sequence of PD + PS > CD + PS > PD + CD + PS > UW + PS > PD + CD + UW + PS > PD + UW + PS > CD + UW + PS. The reduction sequence indicated that the singly organic manures biostimulated soil, PD + PS mostly encouraged

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microbial fixation of nitrogen from atmosphere into the organic manures biostimulated soils. However, the C/N value of crude oil polluted soil was higher than the organic manures biostimulated soils [24]. This may be due to the added carbon substrate from the crude oil that was responsible for the immobilization of available soil nitrogen to cause stress to the soil. Crude oil pollution did not affect the Particle size of the soil as shown in Table 2 that the sand (45%), clay (55%) and silt (50%) fractions respectively were all in the same range for the natural, artificial crude oil polluted soil and bioremediated soils. A classification of the soil based on the USDA textural class [29] shows that the soil is clay loam. Fig 1 and Fig 2 results showed remarkable reduction of total petroleum hydrocarbon (TPH) in mg/kg and % respectively in organic manures biostimulated soils as compared to crude oil polluted soil (114.25mg/kg) for the period of 42 days. Total petroleum hydrocarbon TPH content expectedly increased in concentration in crude oil polluted soil [30]. The TPH reduction results is in line with the findings of Lee *et al*., [6], Ebere *et al*., [7], Rowland *et al.*, [8], Obasi *et al.*, [24] and Okoro *et al.*, [25]. The highest TPH reduction was observed in PD + PS (16.25mg/kg and 90%) > CD + PS (18.26mg/kg and 88.88%) > PD + CD + PS (50.13mg/kg and 69.48%) > UW + PS (62.25mg/kg and 62.10%) > PD + CD + UW + PS (65.53mg/kg and 60.10%) > PD + UW + PS (68.88mg/kg and 58.06%) > CD + UW + PS (71.13mg/kg and 56.69%) till the 42^{nd} day of the study. The sequence of the TPH reduction in organic manures biostimulated soils followed the same soil physicochemical properties improvement sequence of organic manures biostimulated soils as observed in Table 2. Highly perceived dissolvability, decomposability and absorbability of poultry droppings to cow dung, urban waste and equally mixed organic manures biostimulated soils respectively in sequence of PD + PS > CD + PS > PD + CD + PS > UW + PS > PD + CD + UW + PS > PD + UW + PS > CD + UW + PS may be due to the increase in petroleum utilizing microbes population and biomass in organic manures [31] that utilized the crude oil for carbon and energy source to degrade crude oil in organic manures biostimulated soils [32]. However, the observed decrease in biodegradation of TPH in equally mixed organic manures biostimulated soils as compared to the singly applied organic manures in Fig 1 and Fig 2 may be due to the higher symbiotic relationships of the consortium of petroleum utilizing microbes in equally mixed organic manures than expected utilization of carbon and energy by petrophiles from the petroleum hydrocarbons for increase biomass and population that could enhance biodegradation of the petroleum hydrocarbons. Some of the products of biodegradation of petroleum hydrocarbons using organic manures are useful plants nutrients and beneficial to soil ecosystem [33].

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Table 2: Results of physicochemical properties of natural, artificial polluted and organic manures treated soils for 42 days

Parameters	Natural soils (control 1) (NS)	Polluted soils (control 2) (PS)	Poultry droppings + Polluted soils (PD + PS)	Cow dung + Polluted soils (CD + PS)	Poultry droppings + Cow dung + Polluted soils (PD + CD + PS)	Urban waste + Polluted soils (UW + PS)	Poultry droppings + Cow dung + Urban waste + Polluted soils (PD + CD + UW + PS)	Poultry droppings + Urban waste + Polluted soils (PD + UW + PS)	Cow dung + Urban waste + Polluted soils (CD + UW + PS)
pH	6.07 ± 0.141	4.68 ± 0.100	6.82 ± 0.000	6.78 ± 0.001	6.76 ± 0.050	6.53 ± 0.005	6.15 ± 0.000	6.13 ± 0.000	6.09 ± 0.016
Conductivity (µs/cm)	220.20 ± 0.014	169.77 ± 0.000	247.40 ± 0.000	245.96 ± 0.002	245.23 ± 0.000	236.88 ± 0.005	233.10 ± 0.002	222.37 ± 0.001	220.92 ± 0.000
Organic Carbon (%)	0.71 ± 0.014	1.62 ± 0.002	1.72 ± 0.000	1.70 ± 0.001	1.69 ± 0.000	1.60 ± 0.005	0.69 ± 0.000	0.65 ± 0.000	0.64 ± 0.001
Organic Matter (%)	0.95 ± 0.000	2.83 ± 0.005	2.97 ± 0.002	2.94 ± 0.016	2.91± 0.014	2.76 ± 0.000	1.19 ± 0.050	1.12 ± 0.001	1.10 ± 0.000
Nitrogen (%)	0.29 ± 0.002	0.22 ± 0.000	0.93± 0.001	0.66 ± 0.000	0.59 ± 0.002	0.55 ± 0.000	0.23 ± 0.000	0.21 ± 0.000	0.18 ± 0.001
Phosphorous (%)	24.32 ± 0.100	18.75 ± 0.158	33.31 ± 0.0160	30.98 ± 0.141	30.01 ± 0.100	26.61± 0.100	22.60 ± 0.158	22.56 ± 0.100	22.12 ± 0.160
K^+ [Cmol/kg]	0.30 ± 0.000	0.23 ± 0.002	0.44 ± 0.000	0.43 ± 0.002	0.42 ± 0.000	0.32 ± 0.001	0.31 ± 0.000	0.25 ± 0.000	0.19 ± 0.000
Na^+ [Cmol/kg]	0.24 ± 0.002	0.19 ± 0.000	0.33 ± 0.001	0.31 ± 0.000	0.31± 0.001	0.27 ± 0.000	0.23 ± 0.001	0.21 ± 0.002	0.20 ± 0.005
Ca^{2+} [Cmol/kg]	2.72 ± 0.010	2.10 ± 0.024	3.48 ± 0.000	3.47 ± 0.014	3.46 ± 0.010	2.64 ± 0.000	2.36 ± 0.002	2.29 ± 0.000	2.24 ± 0.001
Mg^{2+} [Cmol/kg]	2.31 ± 0.000	1.78 ± 0.001	2.35 ± 0.002	2.34 ± 0.000	2.32 ± 0.010	1.95 ± 0.001	1.08 ± 0.000	0.93 ± 0.005	0.87 ± 0.000
CEC [Cmol/kg]	5.57	4.30	6.60	6.55	6.51	5.18	3.98	3.68	3.50
C/N	2.45	7.36	1.85	2.58	2.86	2.91	3.00	3.10	3.56
Moisture (%)	10.42 ± 0.158	10.08 ± 0.100	11.54 ± 0.000	11.51 ± 0.240	11.25 ± 0.0141	11.12 ± 0.158	10.57 ± 0.160	10.46 ± 0.000	10.29 ± 0.141
WHC (%)	37.64 ± 0.100	36.93 ± 0.160	37.98 ± 0.158	37.83 ± 0.000	37.62 ± 0.100	37.59 ± 0.000	37.21 ± 0.000	37.08 ± 0.141	36.86 ± 0.158
Porosity (%)	46.71 ± 0.000	46.39 ± 0.141	47.83 ± 0.000	47.75 ± 0.158	46.91± 0.160	46. 75 ± 0.000	46.48 ± 0.141	46.30 ± 0.158	46.22 ± 0.100

Sand (%)	45	45	45	45	45	45	45	45	45
Clay (%)	55	55	55	55	55	55	55	55	55
Silt (%)	50	50	50	50	50	50	50	50	50
Textural class	Clay loam	Clay loam	Clay loam	Clay loam	Clay loam	Clay loam	Clay loam	Clay loam	Clay loam

Results = Mean values ± standard deviation

Fig 1: Concentration (mg/kg) decrease of total petroleum hydrocarbons (TPH) with time for 42 days of biostimulation of crude oil polluted soil

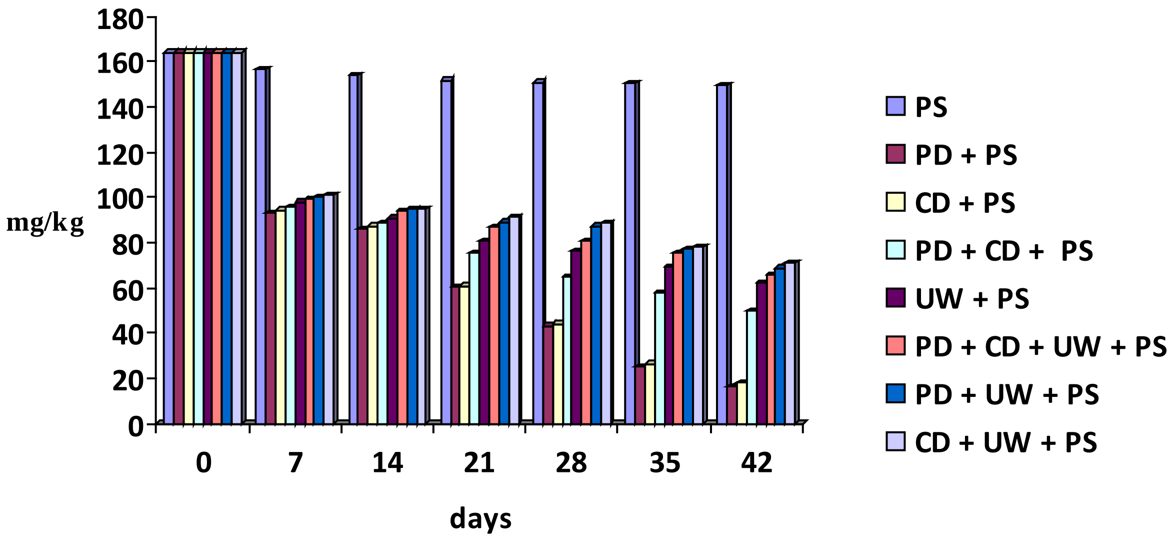
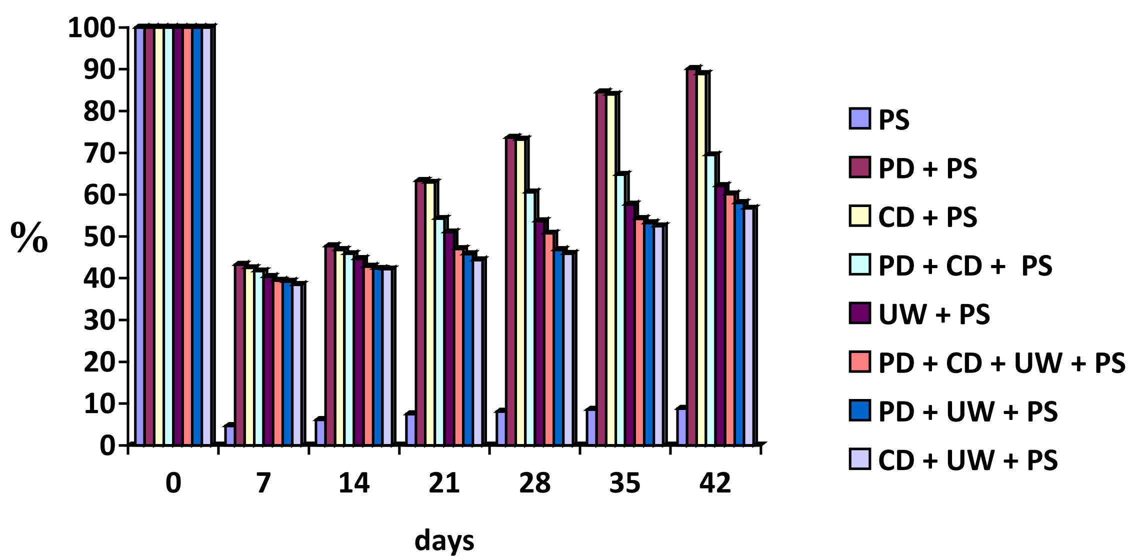


Fig 2: Percentage (%) decrease of total petroleum hydrocarbons (TPH) with time for 42 days of biostimulation of crude oil polluted soil



Conclusion

It was observed in this study that 10% artificial pollution of natural clay loam soil rendered the physicochemical properties of soil unsatisfactory. 100g of singly and equally mixed poultry droppings, cow dung and urban manures in proportions in the presence of 50ml of crude oil appreciably improved the physicochemical properties of 500g organic manures biostimulated soils in the remediation sequence of PD + PS > CD + PS > PD + CD + PS > UW + PS > PD + CD + UW + PS > PD + UW + PS > CD + UW + PS. These results have shown the effectiveness of poultry droppings, cow dung and urban manures at reducing / degrading crude oil polluted soils TPH in similar sequence of PD + PS (16.25mg/kg and 90%) > CD + PS (18.26mg/kg and 88.88%) > PD + CD + PS (50.13mg/kg and 69.48%) > UW + PS (62.25mg/kg and 62.10%) > PD + CD + UW + PS (65.53mg/kg and 60.10%) > PD + UW + PS (68.88mg/kg and 58.06%) > CD + UW + PS (71.13mg/kg and 56.69%) till the 42nd day of the study. Organic nutrients supplementation from these organic manures was discovered to be responsible for the enhancement of the biodegradation rate throughout the 42 days of research period. Biostimulation and biodegradation of crude oil polluted soil with poultry droppings, cow dung and urban manures in singly and equally mixed applications has been established to be highly effective towards the improvement of the minerals, nutrients and physicochemical properties of the biostimulated soil.

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