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Effects of Natural Attenuation on the Nutrient Stability of Simulated Oil Spill Contaminated Soils

Roberts Ashley Ubani [a], Osuji Leo Chigbu [a,b] and Onojake Mudiaga Chukunedum [a,b*]

[a] World Bank Africa Centre of Excellence in Oilfield Chemicals Research (ACE-CEFOR), University of Port Harcourt PMB 5323, Port Harcourt, Nigeria.
[b] Petroleum and Environmental Chemistry Research Group, Department of Pure and Industrial Chemistry, University of Port Harcourt, PMB 5323, Port Harcourt, Nigeria.

Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Natural attenuation of off-field laboratory-simulated crude oil-contaminated soils were investigated to determine the effects on the nutrient stability without organic inputs. Soil samples were collected from Elele and Isiokpo in the Ikwere local government area of Rivers State, Nigeria. Twenty kilograms of the soils were spiked with different milliliters of crude oil and the characteristics of the soils, such as pH, electrical conductivity (EC), total organic carbon (TOC), total organic matter (TOM), cation exchange capacity (CEC), total hydrocarbon content (THC), and total petroleum hydrocarbon (TPH), were determined immediately after contamination (CT) and six months after natural attenuation (NA) using standard analytical methods. Results obtained showed a decrease in

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**Corresponding author: E-mail: mudiaga.onojake@uniport.edu.ng;*

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the values of the physicochemical properties of the soils as shown: pH (6.11 to 5.60); TOC (4.49% to 3.19%); TOM (7.76% to 5.52%) for Elele soils, and for soil samples from Isiokpo pH (5.89 to 5.85); TOC (3.96% to 2.77%); TOM (6.84% to 4.79%). Natural attenuation decreased THC and TPH values by 14.32% and 12.53% in Elele soil and 12.37% and 9.75% in Isiokpo soils, respectively. The result of the study showed that natural attenuation, without any organic input, is effective in remediating petroleum hydrocarbon contents of oil-impacted soil and thus restoring soil physicochemical properties to arable form.

Keywords: Hydrocarbons; contamination; attenuation; remediation; nutrients.

1. INTRODUCTION

The utilization of natural processes to limit the spread of contamination from chemical spills and lower the quantity and concentration of pollutants at polluted locations is known as natural attenuation (Directive, 1999). Other names for it include bioattenuation, intrinsic bioremediation, and intrinsic remediation. In this instance, the pollutants remain on the property, and the place is left to be cleaned by naturally occurring processes. Biological degradation, volatilization, dispersion, dilution, radioactive decay, and sorption of the pollutant onto the soil's organic matter and clay minerals are examples of natural processes. It is mostly utilized for aquifer remediation once the source of contamination has been eliminated, though it can also be applied if the source remains or if certain hot spots have been eliminated.

Since there must be no risk to humans or the environment, long-term monitoring is absolutely required. Natural attenuation can take a long time and is influenced by hydrological changes. Humans and aquatic wildlife habitats must not be reached by the pollutant plume. Hydrocarbons including benzene, toluene, ethyl benzene, and xylene (BTEX) and, more recently, chlorinated hydrocarbons are the primary hydrocarbons for which natural attenuation is employed. Pesticides and inorganic compounds are among the other contaminants that may be remedied by natural attenuation (Mulligana & Yong, 2004).

The impact of hydrocarbon contamination, especially from crude oil spillage, is a major environmental concern owing to the enormous deleterious effects on humans and the entire ecosystem. Crude oil is frequently spilled on soil and water in the Niger Delta region, which is the hub of oil exploration activities in Nigeria (Abosede, 2013). Oil spillage can result in all forms of pollution to soil and water bodies, which poses serious health risks to habitats, vegetation, and animals. It has been observed that oil exploration and exploitation activities in the Niger Delta region over the past two decades have devastatingly impacted on the physical and social-economic environments of communities in the region, largely threatening the environment, livelihood, and subsistent peasant economy, hence, the basic survival of the people (Idam et al., 2023). Farmlands and rivers, which are the main economic sources of the people, have been rendered useless due to widespread spillage of hydrocarbon products. The degree of pollution and toxicity largely depend on several physical and chemical properties as well as the quantity of the crude oil (Okafor, 2023). Several varieties of toxic chemicals, such as petroleum hydrocarbons, are released into the environment through oil spillage (Udinyiwe & Aghedo, 2023). These toxic chemicals, which are poisonous to humans, have now been incorporated into the food chain through plant uptake.

One major issue of soil contamination by hydrocarbons is the severely adverse impact on soil physicochemical properties. The hydrocarbons when released penetrate soil capillaries and bind to soil particles within the interstitial spaces which may reduce soil aeration, water infiltration capacity, and increased bulk density of the soil with the overall result of the altering of the soil physicochemical characteristics and inhibition microbial activities (Ejairu & Okiotor, 2022). This has deleterious effect on the soil ecological and agricultural use of the soil as plant growth may be retarded (Osuji, 2002). Studies on the effects of hydrocarbon contamination of soil has generally been focused on the soil physicochemical and microbial properties, total hydrocarbon contents and effects on plant growth and yields (Wang et al., 2013; Abosede, 2013; Shehla et al., 2018; Udinyiwe & Aghedo, 2023).

In order to decontaminate and restore the nutrient integrity of soil, several remediation approaches are being deployed. Some environmental studies have focused on how to decontaminate the soils from crude petroleum contaminants in a more economical and effective ways (Hang Lv et al., 2018). Among the soil remediation methods, bioremediation by natural attenuation (NA) is the most common approach. Natural attenuation refers to the monitoring of natural processes in soil environments that act without any anthropogenic interventions to reduce concentration of contaminants and toxicity (Idam et al., 2023). Natural attenuation method of soil remediation relies on natural processes to control soil pollutants by turning the soil to promote microbial proliferation, aeration, nutrient, moisture and degradation (Okoye et al., 2019). The processes may include a variety of biological, physical, or chemical processes or combination of them, that under favorable conditions act without human intervention to reduce the mass, toxicity, mobility, volume, and concentration of contaminants in soil (Udinyiwe & Aghedo, 2023).

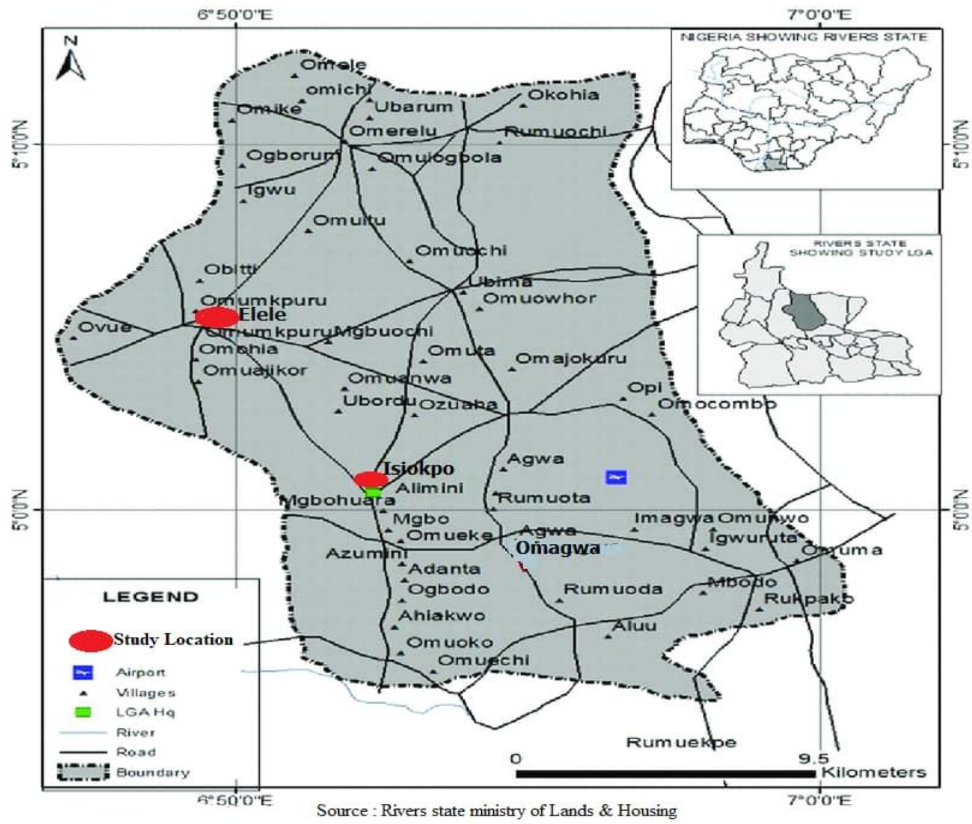
The process of rapid removal of pollutants by natural attenuation may be implemented with the introduction of organic nutrients and chemicals to stimulate indigenous microorganisms (biostimulation) and inoculation with exogenous microorganisms (bioaugmentation), this is referred to as enhanced natural attenuation (ENA) (Idam et al., 2023). Bioremediation by NA or ENA has proven to be more environmentally friendly without additional introduction of pollutants and less expensive because the method can be practiced with little or no experience and in a natural environment.

The aim of this research is to study the effectiveness of natural attenuation involving no external introduction of organic manure or soil tilling as an off-site remediation of crude oil impacted soil and the restoration of the soil physiochemical properties.

2. MATERIALS AND METHODS

2.1 Description of the Study Area

Soil samples for crude oil contamination were collected from Isiokpo and Elele, both in Ikwerre Local Government Area (LGA) of Rivers State (Fig. 1). Ikwerre LGA lies roughly within the coordinates of 6°50’0’E and 7°0’0’E latitude, and 5°0’0N and 5°100N longitude (Fig. 1).



Source : Rivers state ministry of Lands & Housing

Fig. 1. Map showing soil sample collection locations

2.2 Samples Collection and Analysis

Soil samples were collected randomly from an oil spill uncontaminated location at a depth of 0 –30 cm at different points using a hand augur from the two study locations. For representative sampling and homogeneity of samples, five composite soil samples comprising of 10 to 20 subsamples were obtained. The samples were placed in sampling bags, labelled accordingly and thereafter, taken to the laboratory for preparation and analysis. Plant debris and pebbles were removed from the soil samples. Plant debris and pebbles were removed from the soil samples. The composite samples from location 1 (Elele) were labelled E_1 to E_5 for soil obtained from Elele and I_1 to I_5 for that of Isiokpo.

2.3 Crude Oil Contamination

Two kilogram of each soil samples were weighed and used for the crude oil contamination. Different milligrams of bonny light crude soil sample was weighed and used for the contamination of 20kg of soil sample which were manually spiked with different concentrations of the crude oil and thoroughly mixed for homogeneity. The experimental design for the crude oil contamination is as follows:

Elele Samples (E)

E_1 = control (no contamination)
E_2 = 10 ml of crude oil contamination
E_3 = 20 ml of crude oil contamination
E_4 = 50 ml of crude oil contamination
E_5 = 100 ml of crude oil contamination

Isiokpo samples (I)

I_1 = control (no contamination)
I_2 = 10 ml of crude oil contamination
I_3 = 20 ml of crude oil contamination
I_4 = 50 ml of crude oil contamination
I_5 = 100 ml of crude oil contamination

Each prepared soil sample was divided into two equal parts and kept in separate polyethene bags. One part was used for the study of impact of crude oil contamination (CT) on soil physicochemical properties and hydrocarbon contents while the other part was for the study of the effectiveness of natural attenuation (NA) remediation. The samples were analysed for physicochemical properties such as soil pH, electrical conductivity (EC), total organic carbon (TOC), total organic matter (TOM), cation exchange capacity (CEC), total hydrocarbon content (THC) and total petroleum hydrocarbon (TPH). For the CT study, the analyses were commenced 24 hours after the contamination whereas for NA. For the study NA remediation, the samples were kept for six months thereafter the analyses.

2.4 Determination of Soil Physicochemical Properties

The soil physicochemical properties were determined following outlined standard procedures (Amadi et al., 2017; Idam et al., 2023). Soil pH and EC were measured using calibrated pH and conductivity meter in a suspension of 20 g of air-dried soil sample in distilled water (1:1 soil-water solution). The meter probes were inserted into the partly settled suspension and measured. CEC was determined by summation of the exchangeable cations with ammonium acetate buffer. TOC and TOM were determined by titrimetric procedures of Walkley-Black method (Adeyeni et al., 2016; Saikia et al., 2023).

2.5 Determination of Total Hydrocarbon Content and Total Petroleum Hydrocarbon

Two gram of soil sample was weighed into a clean extraction container, 20 ml of hexane was added, mixed thoroughly, and then allowed to settle. The mixture was carefully filtered into clean extraction bottle by means of filter paper in a Buchner funnel. The extract was then concentrated to 2 ml and thereafter transferred for separation. Separation was done using slurry of 2 g activated silica in 10 ml dichloromethane placed into the chromatographic column. 8 ml of the extracted eluant was collected into labelled glass vials for GC analysis. The concentration of THC and TPH in sample was measured at a particular chromatogram in units of mg/kg.

3. RESULTS AND DISCUSSION

3.1 Impact of Crude Oil Contamination on THC, TPH and Soil Physico-chemical Properties

The results of THC and TPH are presented in Tables 1 and 2 for soil collected from Elele and Isiokpo respectively. For the Elele soil, the THC and TPH values for the five soil conditions (E_1 to E_5) were 153.1, 243.5, 312.2, 1079 and 2513 mg/kg, and 71.08, 175.32, 231.01, 820.02, and 1862.19 mg/kg respectively. For the Isiokpo soil (I_1 to I_5) the THC, and TPH values were 124.2, 158.9, 358.0, 1104, and 2043 mg/kg, and 61.97, 119.17, 261.29, 783.78, and 1532.25 mg/kg respectively. The results showed that all CT soils had higher concentrations of THC and TPH. Significant increase was observed in the concentrations of the hydrocarbon with increase in quantity of crude oil spill. As shown in the Figs. (2 to 5), the concentrations of THC and TPH in the CT soils were significantly higher than those of the control soil. This revealed that crude oil contamination caused a significant increase in the THC, TPH and PAH contents of the contaminated soils in comparison with the control sample. Also, the results showed a significant dependent of THC and TPH on the volume of crude oil spillage, with the 100 ml having the highest level. The differences in THC and TPH contents between the Elele and Isiokpo soils clearly demonstrated the background natural activities of hydrocarbon materials in the soils. THC in the soil could increase carbon concentration, which may affect the equilibrium of nutrients available to plants.

The results of measured soil pH, electrical conductivity (EC), total organic carbon (TOC) total organic matter (TOM) and cation exchange capacity (CEC) are also given in Tables 1 and 2 for soils collected from Elele and Isiokpo respectively. The results showed a linear dependent reduction in the pH and EC of the Elele CT soil while that of Isiokpo soil showed no dependent on volume of crude oil contamination. When compared to the control soil, the result showed a significant decline in the soil pH and EC of the Elele CT soil unlike that of Isiokpo. The CT control soil of Elele registered a pH and EC value 6.84 and 88.0 μS/cm respectively, while that of Isiokpo had value of 6.09 and 68.0 μS/cm respectively. The CT Elele soil pH decreased from 6.34 to 5.84 in response to the quantity of crude oil contamination E_2 to E_5 respectively. This trend was not the case of the Isiokpo CT soil as the pH decreased from 7.02 to 4.61 for I_2 to I_4 and then had an increased value of 5.45 for I_5 contamination.

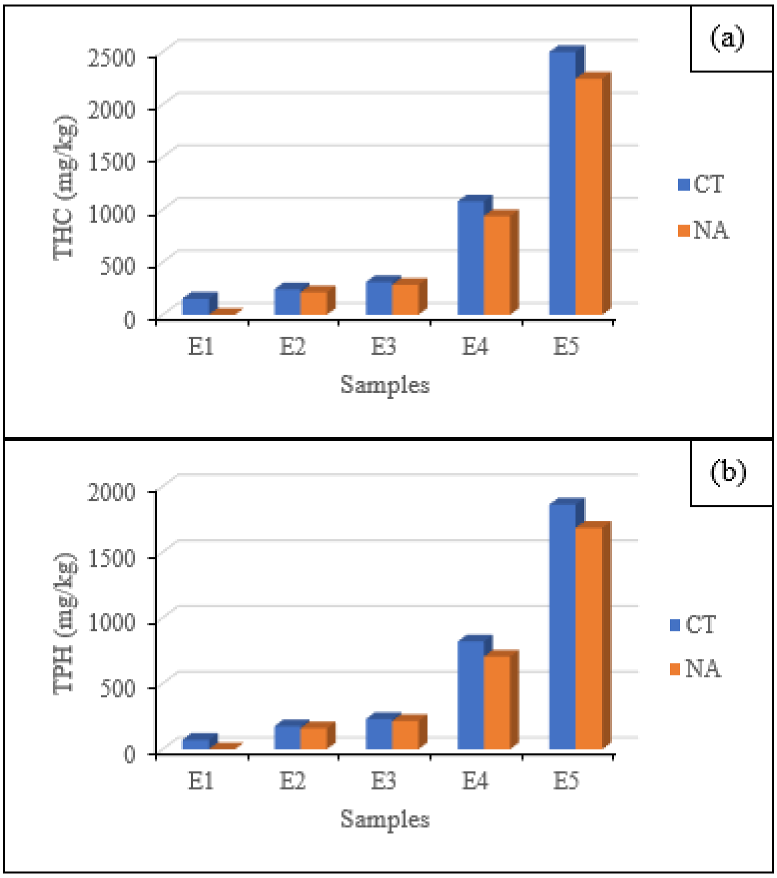


Fig. 2. Comparison of THC and TPH of contaminated and natural attenuation of elele soil

Table 1. Results of the physicochemical properties and the hydrocarbons of soil samples from Elele

Physicochemical properties	CT of Elele soil					NA of Elele soil				
	E_1	E_2	E_3	E_4	E_5	E_1	E_2	E_3	E_4	E_5
THC (mg/kg)	153.10	243.50	312.20	1079.00	2513.00	0.57	213.90	285.72	938.74	2245.90
TPH (mg/kg)	71.08	175.32	231.01	820.02	1862.19	0.43	160.43	214.30	704.06	1684.40
pH	6.84	6.34	5.57	5.95	5.84	6.56	5.55	5.62	5.24	5.05
EC (µS/cm)	88.00	53.00	78.00	34.00	29.00	153.00	140.0	106.0	72.0	75.0
TOC (%)	2.77	4.52	4.76	5.16	5.23	1.60	3.315	3.432	3.783	3.822
TOM (%)	4.79	7.82	8.23	8.90	9.04	2.77	5.732	5.934	6.541	6.608
CEC (meq/100g)	12.20	16.80	13.35	4.86	3.95	11.47	15.303	13.16	5.010	3.631

Table 2. Results of the physicochemical properties and the hydrocarbons of soil samples from Isiokpo

Physicochemical properties	CT of Isiokpo soil					NA of Isiokpo soil				
	I_1	I_2	I_3	I_4	I_5	I_1	I_2	I_3	I_4	I_5
THC (mg/kg)	124.20	158.90	358.00	1104.00	2043.00	4.47	149.41	281.61	931.66	1952.20
TPH (mg/kg)	61.97	119.17	261.29	783.78	1532.25	3.20	112.06	211.21	698.75	1464.20
pH	6.09	7.02	6.28	4.61	5.45	6.05	6.10	6.04	5.43	5.63
EC (µS/cm)	68.00	82.00	41.00	123.00	185.00	106.00	205.00	135.00	110.00	174.00
TOC (%)	0.98	3.59	4.80	5.19	5.23	0.55	2.42	3.51	3.63	3.74
TOM (%)	1.69	6.20	8.29	8.97	9.04	0.94	4.18	6.07	6.27	6.48
CEC (meq/100g)	5.28	14.47	9.82	3.59	6.54	6.13	13.25	8.20	4.14	7.33

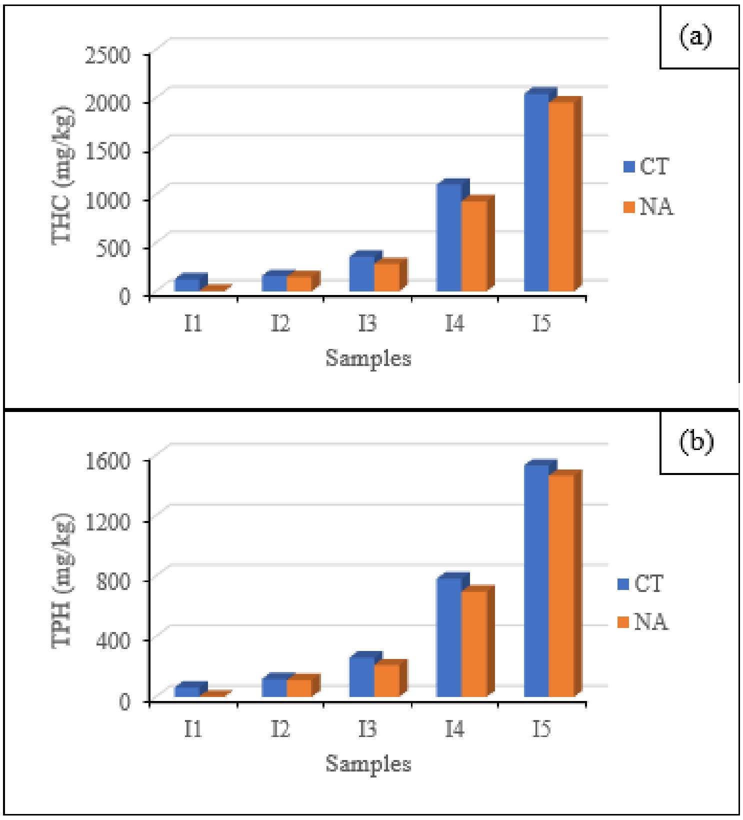


Fig. 3. Comparison of THC and TPH of contaminated and natural attenuation of Isiokpo soil

The EC of the control soil of Elele was higher than those of the contaminated soil, soil E_3 had the highest EC of 78.0 μS/cm among the contaminated soil. Except soil I_3, all other contaminated soil of Isiokpo had EC higher than the control soil with highest value of 185.0 μS/cm for I_5 contamination. Generally, the result shows that increasing contamination by crude oil reduced soil pH level to acidic nature (pH level < 6.0), similar to earlier reported studies (Kisic et al., 2009; Amadi et al., 2017). Soil pH and EC are vital properties of soil in ensuring crop yield as they safeguard soil fertility and micronutrients availability. Although there are mixed arguments on effect of crude oil contamination on soil pH and EC values. Some studies have reported increased pH of crude oil contaminated soil (Wang et al., 2013; Saikia et al., 2023; Okafor, 2023). Several authors have opined bioremediation strategies through microbial actions as the best stabilization option of hydrocarbon contaminated soil pH (Osuji and Nwoye, 2007; Obasi et al, 2013; Amadi et al., 2017). Previous studies in China oilfields also showed that oil pollution raised soil pH (Wang et al., 2010). The EC of the soil can be generally described as non-saline since the measured values were below 120 μS/cm prescribed by DPR (Ejairu and Okiotor, 2022), except that of I_4 and I_5. The result of EC of the CT soils in this study is far below values of 417.50 to 794.45 μS/cm reported in contaminated soil of Ovia North East local government area of Edo State (Udinyiwe and Aghedo, 2023).

Total Organic Carbon and Total Organic Matter are direct measure of organic richness (Adeyeni et al., 2016). The result of the TOC and TOM showed that the soils contain significant amount of organic matter. TOC and TOM values of all the CT soil of both locations were noted to be higher than the respective control soil. TOC and TOM values of 2.77 % and 4.79 % were measured in the Elele control soil, while in the CT soil the values increased steeply with quantity of crude oil contamination from 4.52 % to 5.23 % for TOC and 7.82 % to 9.04 % for TOM. In Isiokpo soil, the control soil recorded TOC and TOM values of 0.98 % and 1.69 % which also increased suddenly in the CT soils with increasing quantity of crude oil. Generally, the investigation showed that values of both organic carbon and matter indicated a linear dependent on the quantity of contaminating crude oil, implying that crude oil has a significant influence on organic matter content of soil. Soil TOC and TOM are generally due to biota degradation (Saikia et al., 2023), however, crude oil contamination may also have contributed to the organic content of soil. The increase of TOC in response to petroleum hydrocarbon in the soils indicated that crude oil contamination could affect soil chemical properties (Wang et. al., 2010). Soil organic carbon is a useful tool for monitoring assessing hydrocarbon contamination because it includes all weight fractions of THC and TPH (Schreier et al., 1999). In the present study, the noticed increase in the total organic carbon in the contaminated soils may be attributed to the hydrocarbon content present in crude oil (Saikia et al., 2023). The results of present study aligned with the findings of Saikia et al. (2023), Wang et al. (2010), Agbogidi et al. (2007) that crude oil contamination significantly increased soil organic contents which might be as a result of slow decomposition rate owing to poor soil aeration occasioned by the crude oil content. Organic carbon and matter are useful tools for monitoring hydrocarbon pollution in soil because organic carbon comprises all weight fractions of hydrocarbon (Wang et al., 2010, 2013). The noted response of the soil organic content to crude oil contamination, indicated that hydrocarbon residuals in soils may as well affect the soil chemical parameters thereby exerting adverse effects on soil ecosystems (Wang et al. 2010). High hydrocarbon concentration reduces plants growth, stem height and density, and above ground biomass of plant (Lin & Mendelssohn, 1996). Soil CEC is an indication of the soil capacity to retain and exchange cation elements. The decrease in the CEC at crude oil contamination volume of 50 and 100 ml implies that crude oil spillage at higher quantity may result to wholesome loss of soil cations with negative impact on the soil ability to retain necessary and essential nutrients needed for plant growth (Udinyiwe & Aghedo, 2023).

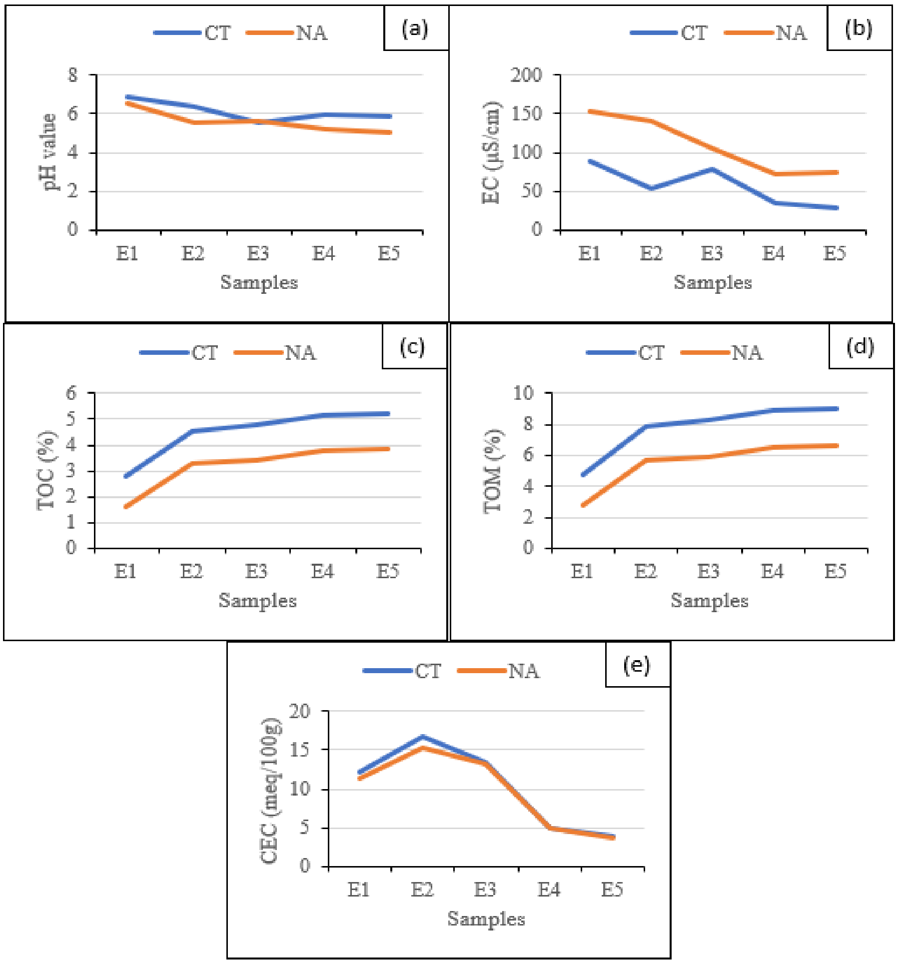


Fig. 4. Comparison of physicochemical properties of contaminated and natural attenuation of Elele soil

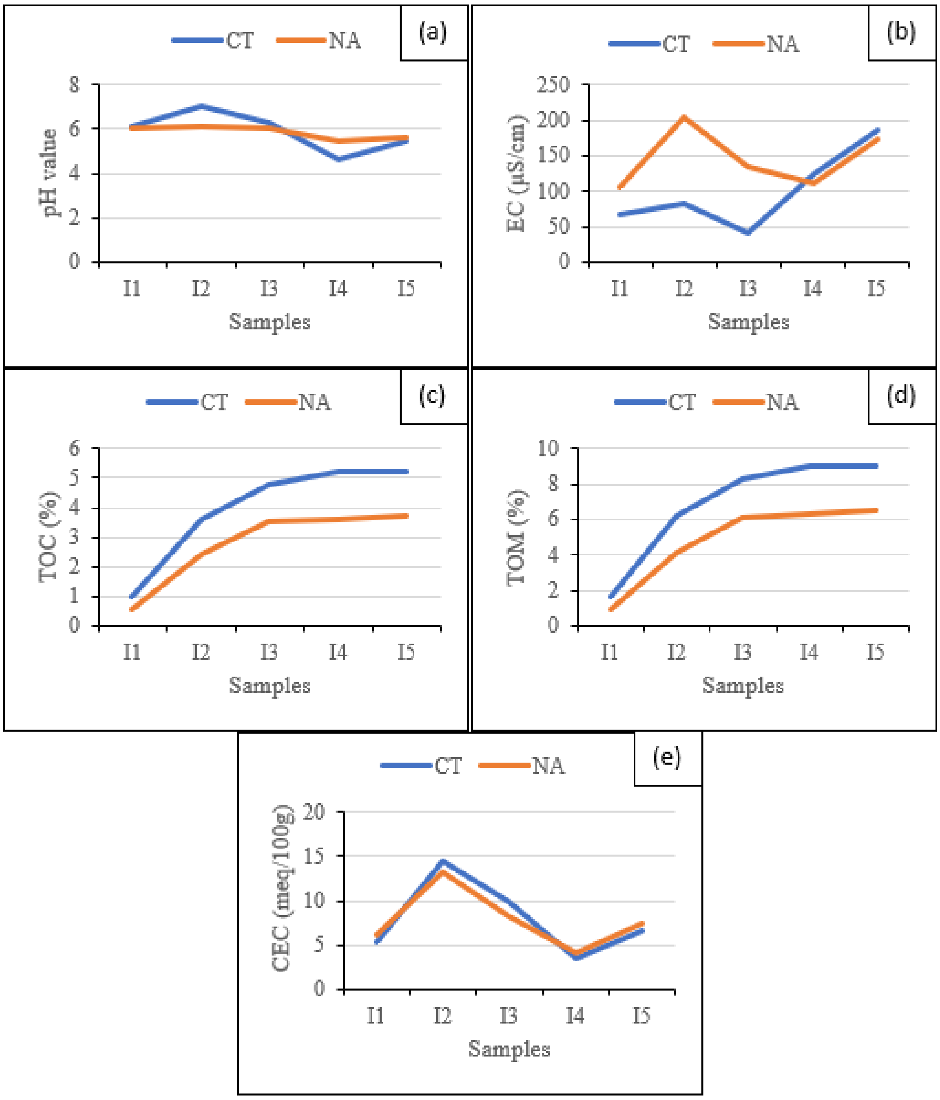


Fig. 5. Comparison of physicochemical properties of contaminated and natural attenuation of Isiokpo soil

3.2 Natural Attenuation of the Soil Physicochemical Properties and the Hydrocarbons

Result shown in Figs. 2 and 3 indicated a reduction in the concentration of THC and TPH in all the soil samples in response to natural attenuation. This indicated an effective natural attenuation process. In the Elele soil, the mean THC recorded 14.32% decrease, from 860.16 mg/kg to 736.97 mg/kg while the TPH decreased from 631.92 mg/kg to 552.72 mg/kg, indicating 12.53% decrease. In the Isiokpo soil, the mean THC value was decreased by 12.37% (from 757.62 mg/kg to 663.87 mg/kg) while the mean TPH was decreased by 9.75% (from 551.69 mg/kg to 497.88 mg/kg). Crude oil contamination of soil through spillage during exploration, production, refining, transportation and storage is a persistent environmental occurrence. Natural attenuation has been regarded as latent method of cleanup of petroleum hydrocarbon contamination owing to its nearly zero environmental impact and economic benefits (Wang et al., 2012; Hang Lv et al., 2018). Generally, the current study showed reduction in concentration of the contaminated soil subjected to natural attenuation. The attenuation rate reduced with increasing amount of crude oil spill indicating a time and volume dependent attenuation process. Though detailed study of the attenuation process was not conducted, however, it is well researched that biodegradation through microbial actions is the most prominent of natural reduction of the total mass of petroleum contaminants (Amadi et al., 2017; Hang Lv et al., 2018). According to Wang et al. (2010), natural attenuation, with any addition of organic material, of oil impacted soil depends on the ambient soil condition and the oil type. The natural attenuation occurs by complex mechanisms of physical, chemical, and biological processes that transform the hydrocarbon components by selectively reducing the easily degradable contaminants, while the more recalcitrant compounds which are typically non-volatile or semi-volatile linger in the soil.

Tables 1 and 2 also showed the NA results of the physicochemical properties and hydrocarbon contents of investigated soil. Figs. 4 and 5 showed the comparison of the physiochemical properties of the NA soil and that of the CT soil. As indicated, soil physicochemical properties, except EC, showed relative decrease in their values after NA remediation. It can be seen from the results that the values of NA soil pH decreased in almost all the soil samples except sample I4 and I5 from Isiokpo. In Elele soil, the mean pH value decreased from 6.11 to 5.60 with 8.25% decrease whereas in the Isiokpo soil, the mean pH decreased from 5.89 to 5.85, which was slightly minimal with 0.68%. NA remediation increased the EC of all soil from Elele from mean value of 56.40 μS/cm to 109.20 μS/cm with 48.35%. Soil I4 and I5 of Isiokpo show a decrease in EC after NA remediation, however, the mean EC of the Isiokpo soil increased by 46.29% from 99.80 μS/cm to 146.00 μS/cm. Natural attenuation remediation indicated an overall decreased in TOC, TOM and CEC values of investigated soils. From Figs. 4 and 5, TOC and TOM in all Elele and Isiokpo soils decreased in comparison with that of CT. The mean values show a decreased from 4.49% to 3.19% and 7.76% to 5.52% for TOC and TOM respectively in soil from Elele. That of Isiokpo soil decreased from 3.96% to 2.77% and 6.84% to 4.79% respectively. The CEC values of the soils after NA was almost the as that of CT.

Similar to the works of Bello and Anobeme (2015), Amadi et al. (2017), Idam et al. (2023), the current work revealed drastic restoration of soil properties after NA remediation. The higher pH values in the CT soil may be caused by the induction of potential drought in the surface and subsurface layers of the contaminated owing to the hydrophobic nature of crude oil which could intensify salinization, and thus raise the pH values as compared with that of the control soil (Njoku et al., 2009). The resultant decreased in pH caused by NA could by ascribed to the production of organic acids resulting from microbial degradation of the hydrocarbons (Osuji & Nwoye, 2007). The increase in EC of the NA remediated soil could be the caused by the initiation of cations by natural chemical processes after microbial degradation of hydrocarbon, hence the near stabilization of the CEC (Wang et al., 2010; Hang Lv et al., 2018). Crude oil pollution in soil has been shown to be related with the built-up of exchangeable base and effective cation exchange capacity (Agbogidi et al., 2007; Idam et al., 2023). The NA remediation of current study did not indicate significant improvement in the CEC of the remediated soils. It is noted that crude oil contamination contributed to total organic carbon of soil (Wang et al., 2010). The concentration of TOC and TOM in the study soil may have resulted from the elevated THC content recorded in the soil, hence the drastic reduction of both parameters after remediation period. Wang et al. (2010) had reported that crude oil contamination significantly increased the total organic carbon contents most probably because of the much higher THC concentration in spilled sites. The current work revealed both slight spatial increases and decreases in pH of soil after remediation.

4. CONCLUSION

Crude oil production has been associated with oil spill, contributing to the degradation of soil in most areas of their operation. This study shows that crude oil contamination elevated total hydrocarbon content and total petroleum hydrocarbon of soil and adversely impacted on the soil physicochemical properties. Natural attenuation, without any input, was effective remediation strategy in reclaiming soil physicochemical properties and decreasing hydrocarbon content. The method though may take a longer time but can possibly destroy completely the contaminants in the soil and potentially lead to a reduction in the cost of carrying out remediation in contaminated sites significantly. It is therefore recommended that natural attenuation integrated with enhanced bioremediation strategies with the introduction of organic nutrients and chemicals to stimulate indigenous microorganisms (biostimulation) and inoculation with exogenic microorganisms (bioaugmentation) be studied and compared.

DISCLAIMER (ARTIFICIAL INTELLIGENCE)

Author(s) hereby declare that NO generative AI technologies such as Large Language Models

(ChatGPT, COPILOT, etc.) and text-to-image generators have been used during the writing or editing of this manuscript.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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