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Full Length Research

Determination of the efficiency of combination therapies on the bioremediation of petroleum spill aquatic ecosystem

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ABSTRACT: Environmental degradation resulting from petroleum spills had become a major menace in most petroleum rich regions of the world. The remediation potential of individual green materials and their combinations were quantified in this study. Stimulated petroleum spill water was remediated with water lettuce, activated charcoal, rice husks and their combinations, within an experimental period of 40 days. The total petroleum hydrocarbons (TPH) value of the contaminated water and remediated contaminated water was determined in accordance with American Public Health Association's (APHA) approved procedures. Findings of this study depicted that the amendments used were able to reduce the TPH concentration of the contaminated water; with activated charcoal tending to have higher remediation efficiency than rice husk. The results revealed that the TPH concentration of the contaminated water treated with only water lettuce, declined from 3897 to 1296 mg/L; while the TPH value of contaminated water treated with water lettuce and activated charcoal (C_2 and C_3), dropped from 3897 to 535 mg/L and 382.33 mg/L respectively, depending on the charcoal quantity employed. It was also observed that the TPH of the contaminated water treated with water lettuce and rice husk (C_4 and C_5), dropped from 3897 to 864 mg/L and 680 mg/L respectively, depending on the quantity of rice husks used for the bioremediation program. Additionally, the study's findings revealed that the TPH of the contaminated water remediated with the combination of water lettuce, charcoal, rice husk and cassava starch (C_6 and C_7) declined from 3897 to 392 mg/L and 223 mg/L respectively. The study's findings had depicted that agricultural waste materials can be harnessed to remediate petroleum spill sites, and the remediation efficiency can be optimized through combined remediation methods/materials.

Keywords: Aquatic environmental, charcoal, petroleum production, water lettuce, waste materials.

INTRODUCTION

Petroleum is an organic oil which consists mainly of carbon and hydrogen, and minute quantities of iron, nickel, sodium, nitrogen, oxygen, chloride and sulfur (Varjani *et al*., 2015). The proportions of these components in the petroleum or its derivatives are dependent on the extraction location, the extracting process and the production procedures used. Despite the advocacy for green energy, the demand for petroleum-based energy is on the rise, due to exponential population growth. The Organization of the Petroleum Exporting Countries (OPEC), an international cartel in charge of petroleum production and exportation in member countries, reported 2.7% increment in the global petroleum production between 2020 and 2021; while the OPEC Reference Basket witnessed an increment of 68.5% (OPEC, 2020; Uguru and Udubra, 2021). Several petroleum derivatives widely utilized in the domestic, health, automobile and industrial sectors, include; petrol, diesel, paraffin wax, lubricating oils and jet fuel. Petroleum production activities are associated with spills, which are mainly caused by saboteurs and system failures

Petroleum spill causes significant negative alteration in water and soils' physiochemical properties and microbial population (Sun *et al*., 2016). Jia *et al.* (2017) stated that petroleum has the ability to block the soils' pores (voids), hindering the aeration and water infiltration and percolation processes in the soil. This can results in the death of most soil microorganisms and plants since they both require adequate air and water for their survival. Heavy metal toxicity resulting from oil spills can retard the performance of microorganisms, lead to death of plants and cause cancerogenous diseases in human beings (Singh and Kalamdhad, 2011; Akpokodje and Uguru, 2019a). Akpomrere and Uguru (2020a, b) in their investigation into the effect of oil theft on the environment reported that petroleum spills had negative impacts on the landmass. According to Akpomrere and Uguru (2020a, b) reports, the heavy metals concentrations in the sediments, water and vegetation of all the regions prone to petroleum spills, were above the maximum allowable limits approved by the World Health Organization.

Since petroleum and its products have several negative impacts on the environment, remediation of petroleum contaminated environments has therefore become inevitable. Some of the basic remediation methods for petroleum contaminated environment include; physical, thermal, biological and chemical methods: each of these methods has its own consequences on the environment (Yu *et al*., 2020). Although the chemical remediation method is one of the fastest procedures for rejuvenating petroleum spill sites, it comes with some serious consequences, such as; causing further alterations in the soil's physiochemical properties, high operating costs and the requirement for skilled workers. The bioremediation method which is considered to be a rather slow remediation process is rapidly becoming the most acceptable process due to its minimal consequences on the environment and its low working costs (Chao *et al*., 2017; Mujaheed *et al*., 2021). Phytoremediation, which uses green plants, with high growth rates, as a means of remediating polluted environments, is one of the economical and ecofriendly branches of bioremediation (Ashraf *et al*., 2019; Akpokodje *et al*., 2019).

It had been observed that combined therapies yield better remediation results, when compared to single treatments (Suresh and Ravishankar, 2004; Sidik *et al.,* 2012; Ramírez *et al.,* 2015; Yoo *et al*., 2017). Vaziri *et al*. (2013) reported that the introduction of natural absorbent (physical method) into contaminated water/soil will help to reduce the concentration of oil in the environment, which will pave way for the optimization of microbial degrading activities in the oil contaminated environment. Although, within the last two decades, numerous pieces of research (Arslan *et al*., 2017; Ashraf *et al.,* 2019; Rehman *et al*., 2019; Kafle *et al.,* 2022) had been done on the optimization of the remediation capacities of various plants. However, there is no recorded information on the combined therapy of water lettuce, activated charcoal, rice husk and cassava starch (in solution), for the remediation of petroleum contaminated water. Therefore, the main purpose of this research is to enhance the phytoremediation efficiency of water lettuce plants, by using cassava starch, activated charcoal and rice husks.

MATERIALS AND METHODS

Petroleum products

The kerosene, petrol and diesel were purchased from a filling station in Delta State, Nigeria.

Organic amendments and water lettuce

The rice husk was obtained from a local rice mill in Benue State, while the cassava starch was procured from a local market in Delta State, Nigeria. The water lettuce was obtained from the fresh water creeks of Delta State, Nigeria; while the activated charcoal was procured from a local chemical shop in Delta State, Nigeria.

Simulation of the petroleum spill water

Petroleum spill water was simulated by adding petrol, kerosene and diesel (at the rate of 5% the volume of the water) to the natural groundwater. The contaminated water with its content was stirred vigorously for two minutes, and poured immediately into already prepared awaiting containers at the rate of I L per container. The stirring was done in order to avoid the fractional separation of the water from the petroleum products.

Remediation setup

The containers containing the contaminated water were coded and arranged in seven rows as shown in Table 1. Each row contains three containers.

Remediation procedure

The activated charcoal was immersed in the beaker containing the contaminated water for six hour, before it was removed. This was closely followed by the introduction of the organic amendments (rice husk and cassava starch) in accordance with the experimental design (Table 1).

After one week, the water lettuce plants were transplanted from a freshwater reservoir, into the beaker containing the organic amendment(s), and placed under a partially shaded environment for 40 days. The one week window was allowed to enable the rice husk and cassava starch to start the decomposing process, thus leaching

Table 1. Remediation setup.

Sample code	Amendment material(s)
C1	Water lettuce
C2	Water lettuce + 100 g C
C3	Water lettuce + 200 g C
C4	Water lettuce + 100 g RH
C5	Water lettuce + 200 g RH
C6	Water lettuce + 100 g C + 100 g RH + 10 g CS
C7	Water lettuce + 200 g C + 200 g RH + 20 g CS

C = activated charcoal, RH = rice husk, CS = cassava starch.

some essential nutrients into the contaminated water.

Two hundred (200 ml) of fresh water was added to each experiment pot twice weekly, to augment for water lost through evapotranspiration. The remediation procedure for each set-up was carried out in triplicate.

Determination of the water Total Petroleum Hydrocarbons (TPH)

At the end of the experimental period, the water lettuce was removed from the containers, and the water was sieved with a 45 µm filter paper. Then the TPH concentration of the filtered water was determined in accordance with American Public Health Association (APHA) 23rd edition approved procedures (APHA, 2017).

Statistical analysis

The SPSS software (version 20.0) was used to analyze the influence of the various treatment therapies on the TPH concentration of the petroleum contaminated water.

RESULTS AND DISCUSSION

The effect of the petroleum products on the water's TPH concentration was plotted as seen in Figure 1. The TPH of the water was observed to increase dramatically from 0.25 mg/L to 3897 mg/L after the petroleum products contamination. This portrayed that the petroleum products had negative consequences on the simulated aquatic ecosystem. A similar trend was reported by Ogeleka *et al*. (2017), as a result of oil spills in Niger Delta waters. The TPH concentration of the contaminated water was higher than the maximum approved limit (10 mg/L) by the Nigeria Department of Petroleum Resources (DPR) for water bodies. Petroleum toxicity in water is dependent on; the TPH concentration and quantity in the water, the natural remediation agents present in the water and the sensitivity of the aquatic organisms to petroleum (Landis and Yu, 2004).

Impact of remediating therapy on the water TPH

The ANOVA result of the impact of the treatment options on the TPH concentration of the petroleum contaminated water was presented in Table 2. The findings revealed that treatment options had a significant ($p \leq 0.05$) effect on the water's TPH concentration, at the end of the 40 day experimental period.

The mean results of the water's TPH at the end of the experimental period are plotted in Figure 2. As presented in Figure 2, regardless of the treatment applied the water's TPH declined significantly during the course of the experimental period. Results obtained revealed that activated charcoal was a better water TPH attenuation agent, compared to rice husk. The TPH concentration of the contaminated water treated with water lettuce and 100 g of activated charcoal declined from 3897 to 535 mg/L, while the TPH concentration of the contaminated water treated with water lettuce and 200 g of activated charcoal declined from 3897 to 382.33 mg/L. Whereas, The TPH concentration of the contaminated water treated with water lettuce and 100 g rice husk declined from 3897 to 864 mg/L, while the TPH concentration of the contaminated water treated with water lettuce and 200 g rice husk declined from 3897 to 680 mg/L.

The results depicted that the remediation efficiency was higher in the combined treatment when compared to the single treatment, with water lettuce as a constant in all the cases. Observations made from the study revealed that the TPH concentration of the contaminated water treated with water lettuce, activated charcoal, rice husk and cassava starch (C_6 and C_7), declined from 3897 to 392 mg/L and 223 mg/L respectively, depending on their quantities in the combined therapy used. Green adsorbents played essential parts during bioremediation; apart from being ecofriendly, they helped to reduce the toxicity/concentration of the contaminants, thus providing an enabling environment for the degrading microorganisms to thrive (Ahmad *et al*., 2015; Obah *et al*., 2020).

The activated charcoal presence gave better remediation results, but it is however not suitable to use a large quantity of it or prolong its contact time. Charcoal

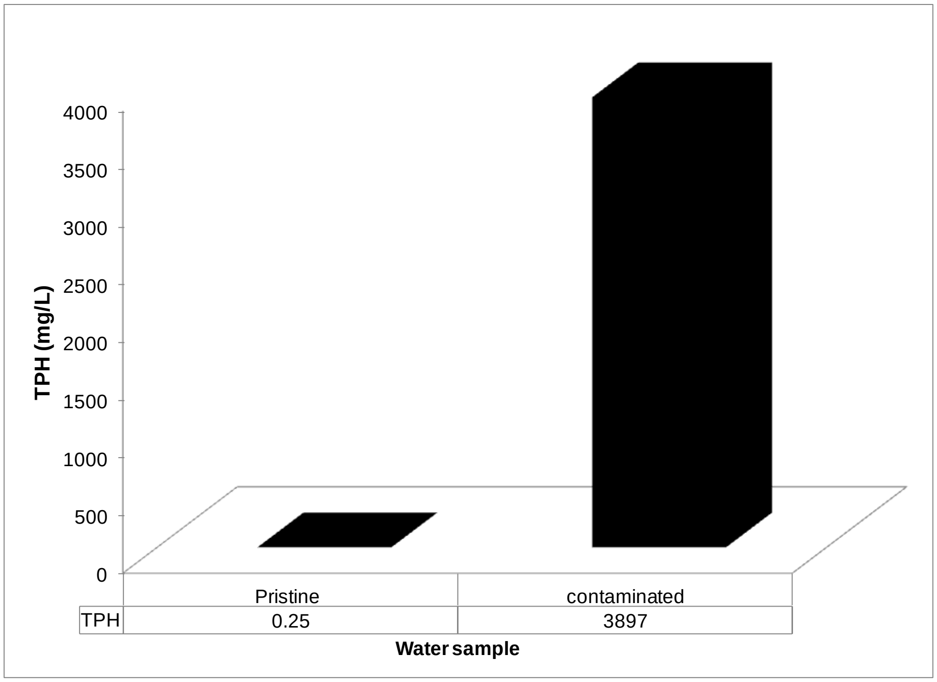

Figure 1. Effect of petroleum products on water TPH.

Table 2. One-way ANOVA of the effect of treatment options on water TPH.

Parameters	Sum of squares	df	Mean square	F	p-value
Between Groups	2379798.29	6	396633.048	3063.37	5.32E-21*
Within Groups	1812.67	14	129.476		
Total	2381610.95	20			

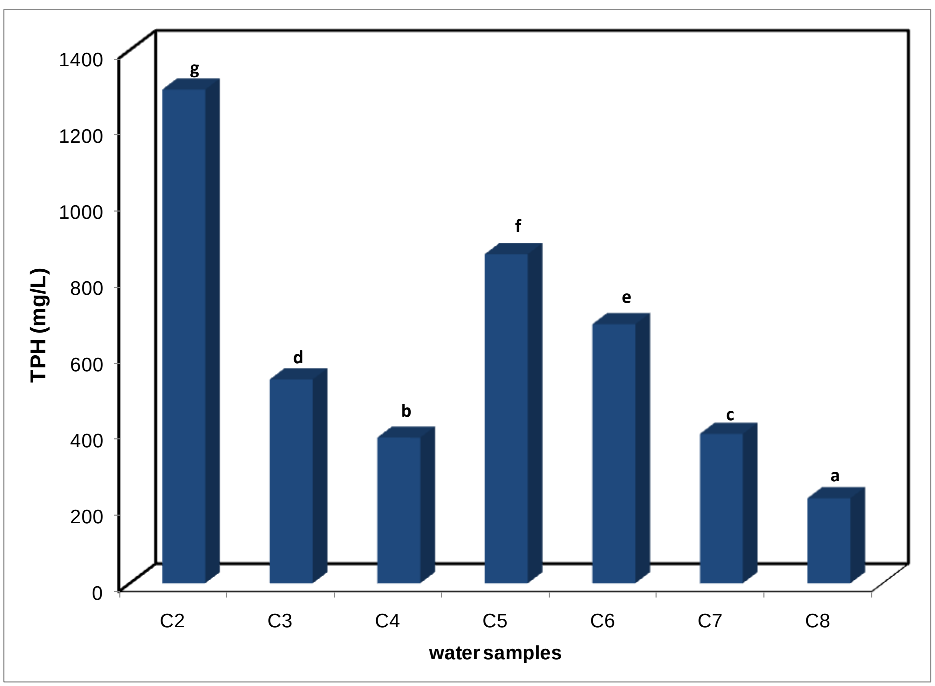

Figure 2. Mean results of the water TPH.

being an adsorbent has the tendency of absorbing both useful metals and other minerals – required by aquatic organisms for their proper growth and development - from the water (Kyzas and Kostoglou, 2014). The adsorption potency of adsorbents is highly dependent on the contact duration between the adsorbents and the contaminated medium (Sumalatha *et al*., 2014; Uguru *et al*., 2020).

Figure 2 shows that rice husk has the ability to degrade TPH in petroleum contaminated water, as the water's TPH content was reduced from 3897 to 680 mg/L, with the introduction of 200 g rice husk (by % weight of the water). This portrayed that rice husk is a good bioremediation agent, in the remediation of petroleum contaminated water. Similar observations were recorded by Akpokodje and Uguru (2019b) and Obah *et al.* (2020), which is attributed to the high nutrient content of the decomposing organic matter. The nitrogen concentration of rice husk tends to increase intensely during decomposition (Thiyageshwari *et al.,* 2018; Oluchukwu *et al.,* 2018). Apart from facilitating the growth of microscopic plants in the water, which will further enhance the phytoremediation of the contaminated water, the high nitrogen and phosphorous content of the rice husk aids the growth and performance of oil degrading microorganisms in the water.

The study results also portrayed that the addition of cassava starch in minute quantities to the remediation program further enhanced the phytoremediation potential of the water lettuce. It was observed that the remediation setup with cassava starch introduced had the lowest water TPH concentration (223 mg/L) at the end of the experimental period. Sorgatto *et al*. (2021) reported that cassava starch in solution helps in the growth of aquatic microscopic plants. These microscopic plants carry out phytoremediation, which lowers the water's TPH content, and reduces its biological oxygen demand and its chemical oxygen demand. These actions by the microscopic plants will optimize the phytoremediation potential of the water lettuce and the petroleum degrading activities of the microorganisms in the contaminated water. According to Shahid *et al*. (2020), aquatic plants weaken the concentration of pollutants in the water through adsorption and translocation of the pollutants, which are aided by the presence of microbes in the contaminated water.

Though the TPH concentration in the water declined drastically at the end of the short remediation period, it was still above the limit approved by DPR for water bodies. The decline in the water's TPH observed in this study signifies that agricultural materials can be used to remediate petroleum contaminated water, which has been a major problem in the oil-rich Niger Delta region of Nigeria.

Conclusion

This research was carried to appraise the remediation potency of different green materials, in order to optimize the phytoremediation potential of water lettuce. Stimulated petroleum spill water was remediated with water lettuce, and different proportions of activated charcoal, rice husks and cassava starch. At the end of the 40 day experimental period, the results revealed that all the green materials used had different degree of remediation efficiency. It was observed that the TPH content of the contaminated water remediated with only water lettuce declined by 66.7%, while the contaminated water remediated with 100 and 200 g of activated charcoal declined by 86.3 and 90.2%, respectively. Likewise, the TPH content of the contaminated water treated with water lettuce and rice husk therapy (C_4 and C_5) declined by 77.8 and 82.6% respectively. Furthermore, it was observed that the TPH content of the contaminated water treated with combined therapies of water lettuce, activated charcoal and cassava starch (C_6 and C_7) declined by 89.9 and 94.3%, respectively. These results revealed that green materials can be used to remediate petroleum spill sites, and combined therapies tend to yield better bioremediation results.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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