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REMEDIATION OF CONTAMINATED SOIL WITH CRUDE OIL BY COMPOSTING

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Abstract — In recent years, one of the primary issues noted worldwide in the environment is the contamination of crude oil in soil. In comparison to traditional methods, bioremediation offers a potential alternative for removing hydrocarbon pollution from the environment. This review paper gives an overview of the benefits, mechanism, and operation of aerobic composting remediation of soil contaminated with crude oil. Within this study, it was demonstrated that with composting technology, one could successfully treat crude oil contaminated soil with a > 90% removal efficiency. Aerobic composting utilizes aerobic bacteria and fungi that require oxygen to grow and biodegrade crude oil's biological component into carbon dioxide and water, whereas anaerobic composting utilizes anaerobic microbes that grow in the absence of oxygen and convert the crude oil's organic component primarily into methane. In terms of efficiency, biodegradation capacity, and rate, aerobic conditions outperform anaerobic conditions. Numerous parameters have been discussed and demonstrated to have an effect on the composting condition and also on the bacteria and fungi used to biodegrade crude oil contaminants at various stages of the composting process, including initial concentration, soil type, soil/compost ratio, aeration rate, moisture content, C/N ratio, pH, and temperature. Microbes use crude oil organic matter as carbon and energy sources during the composting process, whereas fungi produce enzymes that catalyze crude oil oxidation reactions. It is believed that the mutualistic and competitive interactions between bacteria and fungi maintain a robust biodegradation system. The thermophilic phase exhibited the highest rate of biodegradation. However, the presence of a diverse and dynamic microbial community throughout the composting process ensures that crude oil degradation occurs. The efficient composting processes using specific microbes need to be investigated.

Keywords: Composting, crude oil, remediation, contaminated soil

1.0 INTRODUCTION

Environmental quality has become a major concern in recent years for both developing and developed countries. Endocrine-disrupting chemicals (EDCs), persistent organic pollutants (POPs), synthetic dyes, microplastics, and heavy metals have been produced in large quantities as a result of the rapid development of industrial and agricultural activities [1-8]. Petroleum hydrocarbon contaminants had the most devastating environmental impact and posed a lethal health risk to humans and other living forms among all organic contaminants found in soil [1]. Oil spills can endanger sea life, disrupt a day at the beach, and contaminate seafood. Biomarker studies have revealed an irreversible injury to persons exposed to oil and gas spills. These effects can be classified as lung damage, liver damage, lower immunity, increased risk of cancer, reproductive damage, and increased levels of certain toxic substances. Crude oil can be considered as a mixture of different hydrocarbons that can be naturally degraded by specific microorganisms. The aquaphobic properties of crude oil with most of its components have low water permeability, causing crude oil particles to bind with soil particles. Hence, the result is not bioavailable to microorganisms. The presence of crude oil in the environment could be diversified into numerous sources as the number of individual hydrocarbon components is vast. The unintended or premediated and uncontrolled human-caused release of crude oil pollutants could be contributed to or caused by exploration, petroleum production, transport of crude oil, ship spills, and human-induced accidental environmental releases such as storage tank leakage, petrochemical industry effluent discharge, and bursts in aged underground pipelines [2]. One of the biggest crude oil contamination incidents was the explosion and sinking of the Deepwater Horizon rig in the Gulf of Mexico in April 2010 that released 24 to 38 million gallons of crude oil into the environment [3]. Hence, this incident has led to 1162 ha of land area in Mexico being contaminated with total petroleum hydrocarbons (TPH) during the 2015-time period [4]. There are a variety of physical and chemical methods for removing crude oil from the environment [13-18]. However, due to the inefficient, expensive, and not environmentally friendly majority of the technologies discussed above are not recommended treatment for crude oil contamination in soil. The use of biological pesticide remediation over other physical and chemical approaches have a number of advantages, particularly for toxins that are extensively dispersed and pesticides that are diluted [19,20]. Composting is an environmentally friendly way of waste management. Composting is a process that converts various biodegradable wastes into compounds that may be utilized safely and beneficially as biofertilizers and soil additions. In comparison to the landfilling technique of waste disposal, which may present a pollution danger to underground water, the composting process helps keep underground water from getting polluted. This is because composting eliminates microorganisms and chemical contaminants. These are pathogenic bacteria found in garbage that are potentially dangerous to people [21].

2.0 EFFECT OF CRUDE OIL POLLUTANTS ON ENVIRONMENT

Crude oil pollutants have the potential to affect soil properties such as texture, compaction, structural integrity, infiltration properties, saturated hydraulic conductivity, and soil chemical properties such as mineral and other metal ion concentrations and content levels [22]. Additionally, contamination of soil properties with crude oil pollutants inhibits microbial activity and reduces plant growth due to the soil's reduced availability of essential nutrients. As a result, this would impede or temporarily reduce the natural degradation rate of petroleum compounds [23]. Furthermore, crude oil also causes a reduction in water and air permeability in the soil pores, consequently affecting plant rooting [10]. According to a research report from Tang et al. [24], it was stated in the experimental report that germination of wheat and maize seeds could be inhibited with only 0.1% of TPH due to the water and nutrient transportation from the soil being prevented or hindered [24]. The microbial community is also being influenced by the interference of crude oil in the soil as the metabolic cycles and degradation of other organic compounds are being disrupted [19]. This is due to the fact that exposure to TPHs (crude oil) could damage the cell membrane, alter protein conformation, and hinder the enzymatic activities of certain microbes [25]. Therefore, treatment methods for crude oil contaminants have enticed various scientific interests for the past decades. Numerous physical or chemical types of treatment methods have been developed, such as thermal treatment, soil washing, and advanced oxidation processes, which have been proposed to treat crude oil contaminant soil [26]. These treatment methods are highly efficient and have a better recovery rate. However, these methods are expensive, not environmentally friendly, and require complicated technical experience. On the other hand, bioremediation methods are said to be environmentally friendly and cost-effective, as well as easy to use. They can be used to remove or store organic pollutants [27].

3.0 ENVIRONMENTAL FATE AND TRANSPORT OF CRUDE OIL IN SOIL

When crude oil pollutants infiltrate the environment, the petroleum components undergo multiple stages such as physical abiotic processes, biological or chemical interference with microorganism and weathering. Subsequently, in terrestrial conditions, crude oil would infiltrate downwards and gradually expand sideways as it reaches the groundwater [25]. Crude oil generally exists as non-aqueous phase liquid (NAPL) in soil. As illustrated in Figure 1, the distribution of crude oil can be classified into 3 different phase, portion of the crude oil are dissolved in water, some are sorption by solid organic particles, some are volatized into land gas and the remaining exists as non-aqueous phase. The process of diffusion and dispersion in groundwater, biodegradation in soil particles, volatilization that is released into the atmosphere [28]. Hence, the untreated crude oil in soil would not only contaminate soil conditions and disrupt groundwater environment conditions. Nonetheless, crude oil can be easily absorbed by organic materials and immobilize to soil and sediment due to hydrophobicity properties [29].

4.0 COMPOSTING

Composting is a preferred or endorsed bioremediation technique due to its high biodegradation efficiency and ease of operation [21]. Composting is defined as a biochemical process that employs microbes to absorb or stabilize crude oil pollution where organic substrates are mineralized to create more rigid substrates, impregnated forms, and organic products [30]. Thus, the predominant microbes responsible for pollutant degradation in composting are bacteria and fungi, which are also known to be the most critical factor governing composting remediation. It is classified into two types: aerobic and anaerobic [31]. Aerobic composting utilizes aerobic bacteria and fungi that require oxygen to grow and biodegrade crude oil's biological component into carbon dioxide and water, whereas anaerobic composting utilizes anaerobic microbes that grow in the absence of oxygen and convert the crude oil's organic component primarily into methane [32]. Thus, composting is divided into two phases: thermophilic phase and a maturation phase. Normally, during the thermophilic phase, microbes undergo a stabilization phase during which they engage in intense microbiological activity associated with the rise in temperature. This process indicates that the microbes are degrading the pollutants source. After passing through the thermophilic phase, the temperature gradually decreases and stabilizes, indicating that the feed or medium has been depleted or absorbed into simple molecules. This process is governed by the humification process, also known as the maturation phase [33]. In terms of efficiency, biodegradation capacity, and rate, aerobic conditions outperform anaerobic conditions.

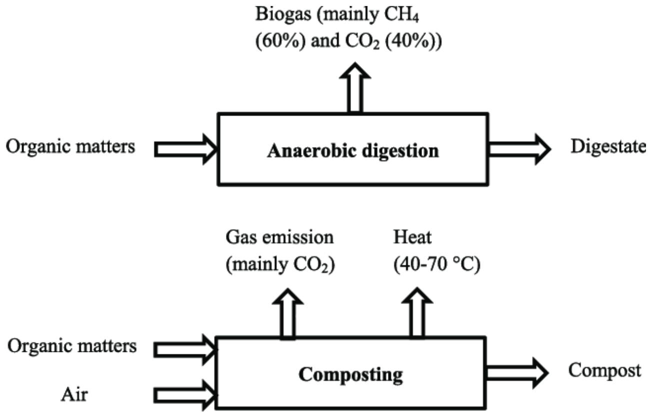

Figure 1 Illustration of Aerobic and Anaerobic composting

4.1. Aerobic Composting Phase

Aerobic composting can be classified into four stages, depending on the microbial metabolic rate and resulting heat production. During the mesophilic phase (45°C), the microbial community (mesophiles) adapts to the initial conditions and rapidly expands in population due to the abundance of degradable organic substrates [34]. Additionally, it would result in a rapid increase in pH, temperature, and carbon dioxide while decreasing the oxygen content significantly as a result of active microbial activity [16]. As microbial activity reaches a peak, the thermophilic phase begins, which favors thermophilic microbes such as *Pseudoxanthomonas* sp. and *Bacillus* sp. to thrive in that environment, thereby halting the growth and thriving of mesophiles [31]. Additionally, a longer thermophilic phase would aid in pathogen elimination and accelerate maturation. As it enters the cooling stage, microbes begin to slow their metabolism and activities due to a lack of organic substrates such as crude oil pollutants, causing the temperature to fall to an ambient level [34]. Thus, when the compost reaches the maturing stage, the composting process is complete [31].

4.2. Initial Composting Materials

Composting materials are primarily composed of raw organic matter, which serves as the fundamental nutrient and microbe component, as well as bulking agents that regulate the operational condition. As a result, co-compost materials are regarded as critical parameters affecting microbial metabolism during the composting process. Additionally, these materials have an effect on the physiochemical properties of the compost mixture as well as the structure of the microbial community. Generally, various types of organic wastes such as food waste, manure, or yard waste are used as compost materials. As illustrated in Table 1, some examples of compost materials and their characteristics that have been used to biodegrade TPH-contaminated soil are included. As illustrated in the Table 1, different organic wastes have varying C/N ratios, nutrient contents, and moisture contents. Aerobic microbes do not thrive in environments with a high moisture content, such as food waste with a C/N ratio of 20-40. As a result, additional feedstock is required to modify the carbon/nitrogen ratio, moisture content, and porosity of the co-composting materials [35]. Thus, bulking agents are required to alter the composting environment to one that is favorable for aerobic microbes, and inoculants can be added to stimulate the microbial population and community during the composting stage.

Table 1 Compost materials used in studies on TPH degradation

Organic Wastes	C/N Ratios	N/P/K	Moisture	Initial pH	Bulking Agent	References
Food Waste	21.5-39.2	-	71.4-79.8	N/A	Rice Husk, Saw Dusk, Rice Bran	[36]
Food Waste	38	1.1/0.6/1.2	72	4.0	Mature compost, Sawdust	[37]
Mature Compost	30	-	48	7.7	Wheat Straw	[35]
Synthetic food waste	13.3	-	80.5	5-6	Rice Husk, Sawdust, Rice Bran	[36]

To maximize the biodegradation of TPHs or crude oil pollutants, it is critical to consider the initial concentration of contaminants, the soil texture, the incubation time, and the C/N compost ratio. For example, Park et al. [38] reported that diesel with an initial concentration of 10,839 mg kg1 achieved a 96 percent removal efficiency after 30 days, whereas lubricant oil with a lower initial concentration of 2400 mg kg1 achieved only a 70 percent removal efficiency after 150 days, owing to the higher molecular weight and more complex molecular structure [38]. Additionally, other soil properties such as texture, porosity, and nutrient sources have an effect on the metabolism of TPH microbes. For example, clay with a low penetrability would result in decreased oxygen transfer, resulting in slower aerobic microbe growth. In comparison, sandy soils have a high permeability, which allows for increased oxygen content transfer and thus improves TPH mineralization [39].

4.3. Microbial Community

Within the microbial community of compost mixture consist of a diversify and robust system to degrade petroleum-related contaminants in soil. In numerous stages of composting would exists different types of bacteria and fungi that thrive on each phase and each serve different purposes. As seen on Table 2, on the types of bacteria and fungi species that aid in degradation process during composting. Fungi are proven to be excellent decomposition agent for crude oil pollutants as it produces extracellular enzymes, including lignin peroxidases (LiP), manganese peroxidases (MnP), and laccases (Lac).

Table 2 Microbes identified during organic pollutants degradation by composting

Phases	pH	Temperature	Name of Species		References
			Bacteria	Fungi	
Mesophilic	7.1-7.2	40–50 °C	*Pseudomonas* sp	-	[37]
		50 °C	*Bacillus pumilus*		[40]
Thermophilic	8.0–8.5	55–70 °C	*Pseudomonas* sp.	*Aspergillus sp*	[37]
		55–70 °C		*Penicillium* sp.	[37]
		60°C	*Geobacillus sp.*		[40]
		60°C	*Ureibacillus thermosphaericus*		[40]
Maturation	7.0-7.2	30°C	*Proteus mirabilis*		[37]
		27°C	*Bacillus amyloliquefaciens*		[40]
			Bacillus licheniformis		[40]

According to several research papers published in the last decades on the biodegradation of TPHs or crude oil that made use of composting [41,42]. Table 1 summarized six significant studies that conducted or utilized composition remediation to treat crude oil contaminants in soil. These research experiments were taken in laboratory, pilot, or field-scales. Composition remediation can be used to treat crude oil contaminants in sandy or sandy loam, based on the data in Table 1. In terms of efficacy, several research studies have demonstrated removal proficiency of greater than 70%, with a maximum of 99 percent. Additionally, this method is capable of treating crude oil with a concentration of 131,720 mg kg1. Additionally, all compost materials are readily available, environmentally friendly, and recyclable, such as wood chips or yard waste.

Table 3 Composting remediation efficiencies for petroleum hydrocarbon-contaminated soil

Initial Conc. (mg kg^{-1})	Soil texture	Compost material	Soil/Compost (dry wt.)	Duration (day)	Removal Efficiency (%)
18,000	Sandy	Maize straw and pine wood chips	1/1–3/1	150	26.8–38.6
1000	Sandy	Poultry manure and wood chips	1/2	300	77–99
25800-77200	Sandy loam	Chicken Manure and rice husk	1/1	53	38–57
10,000	-	Bagasse wastes and mushroom waste	1/3	15	40
131,720	-	Immature compost	1/2	84	93–95
2540	-	Yard waste and rumen waste	1/1	80	45.2

5.0 FACTORS AFFECTING COMPOSTING

The rate of biodegradation of crude oil pollutants is determined by two major factors: abiotic and biotic factors. Abiotic factors include pH, moisture content, temperature, TPH characteristics, nutrient, and C/N ratio; biotic factors include microbial diversity and hydrocarbon bioavailability.

5.1. Moisture

Moisture is a critical factor in the composting process because it has an effect on the microbial activity and physicochemical properties of TPH-contaminated soil. Moisture content increases the solubility of nutrients and minerals required by microorganisms during the cultivation phase. However, a high moisture content (>70%) reduces soil porosity, resulting in a decrease in microbial density. Again, a high moisture content prolongs the incubation period and promotes the growth of pathogenic bacteria. On the other hand, a low moisture content (40%) would allow for early dryness, affecting nutrient uptake during the composting process. As a result, microbial activity would be inhibited, resulting in inconsistency throughout the composting process. Thus, the optimal moisture content for aerobic composting is between 50-60%, as indicated by previous studies [43,44]. Guo et al. also reported that when the moisture content is prepared or maintained within a certain range, 90-94 percent of TPH biodegradation efficiencies are achieved after 84 days of composting [45].

5.2. Oxygen Content and Aeration

Oxygen acts as an electron receptor for aerobic microorganisms in aerobic composting. The soil texture (particle size), composting phase, and airflow rate all influence the oxygen content of the compost mixture [45]. For example, Tran et al. [31] reported that after 45 days of food waste composting, the removal efficiency of fuel oil was 93 percent in sandy soil and 82 percent in silt soil. Thus, increased porosity promotes oxygen penetration and enhances microbial metabolism and TPH oxidation [31]. Again, depending on the microbial activities, the oxygen concentration and requirement for the composition process varies. During the initial mesophilic stage, the microbes require a high concentration of oxygen to initiate metabolic activity. While more oxygen is required during the thermophilic stage, when microbial activity is at its peak, only a small amount of oxygen is required during the maturation stage. As a result, each stage's oxygen concentration and supply must be monitored. Aeration rates are critical because they affect the activity of microbes, the incubation temperature, and also the gaseous emissions produced during the composition process. Due to a lack of available oxygen, low aeration rates discourage the growth of aerobic microbes in the compost mixture. Additionally, low aeration rates increase total nitrogen through the nitrification process, lowering the carbon/nitrogen ratio. Nonetheless, an excessive aeration rate would result in a decrease of biodegradation rates [46].

5.3. C/N Ratio

Carbon to nitrogen ratios vary depending on the contamination level of the soil and compost materials. Typically, soils contaminated with crude oil have a C/N ratio of 30 to 150 and a low nutrient content, which inhibits microbial activity and diversity. Thus, organic wastes such as food scraps, manure, and crop residues are added to the initial compost mixture to increase its nutritional value. Koolivand et al. reported on the addition of immature compost to the oil sludge compost mixture during the compositing process in order to adjust the C/N ratio. After 82 days of incubation, the C/N ratio of 20 was found to be more efficient at degrading TPH than the C/N ratio of 10 [46]. Additionally, other research indicates that a C/N ratio of ten may inhibit microbe activity during composting due to the high concentration of salt, which alters the pH value, whereas a high C/N ratio results in insufficient nutrients for microorganism survival. Thus, a C/N ratio of 10–40 demonstrated high TPH degradation efficiencies [43].

5.4. Temperature

Temperature is a significant environmental factor in the composting process because it affects the physicochemical properties of pollutants, as well as the activity and structure of microorganisms [47]. Nonetheless, high temperature increase volatility and decrease the viscosity of TPHs. Hence, prompt bioavailability of crude oil pollutants. As composting is a self-heat generating biochemical process, whereby the heat is generated as a by-product of the biodegradation of organic materials. Hence if the temperature of the compost mixture is at (>50 °C), the survival of mesophiles and pathogenic bacteria is threatened. Additionally, ambient temperature has an effect on microbial activity and degradation rate, as low ambient temperatures decrease the bioavailability of TPHs [43].

5.5. pH

The pH value of a compost mixture is significantly influenced by co-composting materials and soil. As a result, the pH value of crude-oil contaminated soil can range between 1.3 and 9.2. Additionally, during the compost phase, the pH value varies according to the stage of composting. The pH value tends to be lower during the mesophilic phase, as organic acids are produced as the microbes decompose and utilize the crude oil contaminant. Following that, the pH value gradually increases as it enters the thermophilic phase due to the formation of NH4 and organic acids are degraded to form VOCs, CO2, and H_2O [43]. For soil contaminated with crude oil, a previous study reported on the effect of different pH values on the biodegradation efficiency of the crude oil contaminant. The results indicated that at pH 3, the efficiency achieved is 4%, while at pH 7, the efficiency achieved is 72 percent, while at pH 9, the efficiency achieved is only 30%. As a result, the optimal pH range is 5-7 [48].

Table 4 Composting optimal conditions for crude oil degradation.

Parameters	Optimal Conditions
Temperature	55–70 °C (in thermophilic stage)
Moisture	50–60%
pH	7.0
C/N Ratio	10–40
Oxygen Content	Minimum 10%, with aeration rate 1–2 L kg dry wt.$^{-1}$ min^{-1} for pilot and field-scale
Porosity (particle size)	<1 cm
Soil Texture	Sandy or low clay content

6.0 GASES EMISSION FROM COMPOSTING

Composting generates greenhouse gases (GHGs) generally as a result of the decomposition of organic wastes, which mostly consist of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). Carbon dioxide is often produced as a result of organic matter disintegration and microbial respiration, whereas nitrous oxide is produced as a result of the decomposition of nitrogen-rich organic materials [32]. Organic materials containing volatile sulphur compounds (VSCs), volatile fatty acids (VFAs), and a number of inorganic molecules such as NH_3 and H_2S release odorous gas emissions, resulting in an unpleasant odor [49]. Despite the fact that composting processes generate these odorous particles at a very low concentration, chronic exposure to these gases can result in headaches, anxiety, and depression. These compounds are emitted mostly during the mesophilic and thermophilic stages of composting, which are characterized by an insufficient supply of oxygen, a high moisture content, and a poor permeability of the compost mixture. As a result, the odorous gases produced during the composting of petroleum-related pollutants such as crude oil are considered possible air pollutants that must be monitored [50].

7.0 FUTURE RESEARCH AND PROSPECT

Additionally, the current composting approach for soil remediation has some limitations, necessitating further development and study to improve the approaches. The bioavailability of contaminants is a significant constraint; the extent to which organic pollutants can be absorbed and utilized in soil was determined by their accessibility for absorption, toxicity, and utilization; pollutants are typically inextricably linked to the soil. Thus, in the long term, bioavailability of contaminants will continue to be a more important limiting factor for bioremediation efficacy than the density of biodegrading bacteria. Thus, study is necessary to determine the best effective approach for determining the bioavailability of a variety of pollutants in soil prior to selecting a remediation strategy for soil pollution control. Second, there is a dearth of data and study on the enzymatic characteristics of contaminated soils that have been remedied through anaerobic composting or aerobic composting. As a result, it is vital to monitor the enzyme activity of these soils. Additionally, the effect of contaminants derived from the same soil/compost mixture on their degradation requires additional research [51]. This is because the growth and activity of composting microorganisms, which have a significant impact on the efficiency of TPH biodegradation, can be enhanced by optimizing operational parameters. The time-consuming nature of microbial activity has also been identified as a significant challenge for future study and development [31]. Additionally, existing research regarding the microbiological development of soils polluted with organic contaminants during composting is lacking [51]. Although extensive research and development have been conducted on a variety of pollutant-degrading microorganisms and have successfully oriented the microorganism degradation pathways, a more in-depth understanding of the microbial lifestyles and dynamics of communities found in soil/compost mixtures is required to further enhance the effect of composting or compost on soil remediation [51].

8.0 CONCLUSION

Composting is a potential bioremediation technology that has been shown to efficiently biodegrade crude oil contamination in soil due to the methodology's resilient, green, cost-effective, widely available resources, ease of operation, and high removal effectiveness. Nonetheless, the composting conditions and parameters must be closely monitored and optimized in terms of pH, moisture content, temperature, TPH properties, nutrient content, and C/N ratio. Additionally, present understanding is limited to laboratory, pilot, and field scales, necessitating additional research and development. Control mechanisms for greenhouse gases released during the composting process must also be incorporated into the design.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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