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Treatment of Oil Sludge Contamination by Composting

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Abstract

Sludge is a complex emulsion of various petroleum hydrocarbons (PHCs), water, heavy metals, and solid particles. Polycyclic aromatic hydrocarbons (PAHs), which are components of crude oil sludge, constitute serious environmental concerns, as many of them are cytotoxic, mutagenic and potentially carcinogenic. It also affects the soil fertility. Treatment of oil sludge includes physical, chemical and biological process but still cost effective method is needed. Composting can serve as remedy to treat the sludge provided factors such as nutrients, pH, moisture, aeration and temperature within the compost pile. For maintaining the humidity different bulking agent can be used. High microbial diversity and activity during composting, due to the abundance of substrates in feedstocks, promotes degradation of xenobiotic organic compounds, such as pesticides, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs). Microorganisms can also bio transform pollutants into less toxic substances. The exhaustive investigation of oily sludge treatment methods will provide researchers to have a thorough understanding of recent developments and future research directions.

Keywords: Oil sludge; Contamination; Composting; Bioremediation

Introduction

Oil sludge is a thick, viscous mixture of sediments, water, oil and high hydrocarbon concentration, encountered during crude oil refining, cleaning of oil storage vessels and refinery-wastewater treatment. The chemical composition of oil sludge is complex and depends on the source. Oil sludge is mainly composed of alkanes, aromatics, asphaltenes and resin [1]. It has high content of aromatic hydrocarbons in the range of 1-40 carbon atoms [2]. The two major sources of oil sludge are oil storage tanks and refinery- wastewater treatment plants [3]. World growing environmental concern had led to put a check on oil sludge being discarded out of petroleum industries and manage it to sustainable level by improving plant design or by installing end of the line remediation processes. Dumping oil wastes or burning them with no previous treatment has serious environmental consequences and presents a risk to both ecosystems and human health [4]. Sludge produced in the oil industry can contain up to 80% oil and 40%solids. There has been a wide range of bioremediation processes applied to petroleum sludge clean-ups. Studies have shown microorganism is versatile in degradation of organic compounds. Composting is an aerobic biological process that converts organic matter into a more stable material with a lower content of degradable organic matter, thus giving rise to less phytotoxicity towards plants [5]. Composting has been practiced to reduce volume and water content of feedstock, to destroy pathogens, and to destroy odor-producing nitrogenous and sulfurous compounds [6,7]. Composting may accelerate the destruction of contaminants [8,9]. Despite its application in the treatment of soils contaminated with organic compounds, the use of composting in bioremediation has received little attention [10]. Much of the work on treatment of contaminated soils by composting has been done on soils with lower concentrations of the contaminating substances [11-13] (Table 1).

Compounds present in oil sludge

Oil sludge is mainly composed of alkanes, aeromatics, asphaltenes and resin [1]. Oil sludge contains volatile organic carbons (VOCs) and semivolatile organic carbons (SVOCs) (for example, PAHs) which over the years have been reported as being genotoxic [14,15]. The oil from the pyrolysis was composed basically of PAHs such as naphthalene, acenapthylene, phenanthrene, fluoranthene, benzo[*a*] anthracene, benzofluoranthenes, benzopyrenes, indenepyrene, benzo[*ghi*]perylene, and anthanthrene [16]. Quantification of the main compounds showed that sewage sludge pyrolysis oils contain significant quantities of potentially high-value hydrocarbons such as mono-aromatic hydrocarbons and phenolic compounds [17]. Based on the EU guidelines and the mean concentration values for metals found in the oily sludge, e.g., Pb (135.4 ± 125.8), Cu (105.2 ± 79.1), Hg (42.8 ± 31.3), Ni (320 ± 267.4), and Zn (1321.7 ± 529.9 mg kg^{-1}), disposal of oily sludge even in landfills for hazardous waste is not allowed [18]. One-time composting in static-aerated biopiles with organic amendments as the sole strategy to treat oily sludge is very effective in reducing the content of 2–4 rings PAH, but it is not effective in reducing the content of 5–6 ring PAHs, even after a relatively long time span (370 d) [19] (Figure 1).

Naphthalene: It is an aromatic hydrocarbon, with molecular formula $C_{10}H_8$ and the structure of two fused benzene rings. The thermophilic aerobic bacterium *Bacillus thermoleovorans* Hamburg 2 grows at 60°C on naphthalene as the sole source of carbon and energy. Naphthalene degradation by the thermophilic*B. thermoleovorans* differs from the known pathways found for mesophilic bacteria [20]. Naphthalene-degrading strain with high activity was isolated from soil polluted by cooking oil,which was identified and named Hydrogenophaga Palleronii LHJ38. naphthalene-degrading activity of this strain was developed and the optimum growth conditions of this strain were studied. Under the optimum conditions of 28°C,initial pH(6.6) and mol ratio of carbon to nitrogen 4 and naphthalene mass concentration 2 000mg/L,the degradation rate of naphthalene is more than 98% in 96 h [21].

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Treatment	Effect on soil chemistry	Effect on physical structure	Effect on microorganism	Approximate remediation cost (£/tonne)
Removal to landfill	UN	UN	UN	Up to 100
Soil washing	Y	N	N	25-150
Physico-chemical	Y	N	N	50-175
Vapour Extraction	Y	Y	Y	75
Solvent Extraction	Y	N	UN	50-600
Chemical n	Y	N	UN	175-450
In situ Flushing	Y	Y	UN	25-80
Surface amendments	Y	Y	Y	10-25
Thermal Desorption	Y	N	N	25-225
Incineration	N	N	N	50-1200
Windrow Turning	Y	N	Y	Oct-50
Land Farming	Y	N	Y	Oct-90
Bioventing	Y	Y	Y	15-75
Bioslurry	Y	N	Y	50-85
Biopiles	Y	N	Y	15-35
In Situ Bioremediation	Y	Y	Y	175

Table 1: Effects of remediation methods on soil characteristics and the estimated costs of treatment [13] (adapted from Houghton, 1996 [13]).

Anthracene: This is a polycyclic aromatic hydrocarbon consisting of three fused benzene rings. It is also component of coal tar [22]. The initial reactions in the bacterial degradation of anthracene involve the formation of *trans*-1, 2-dihydroxyanthracene prior to ring fission [23].

Phenanthrene: Phenanthrene is a polycyclic aromatic hydrocarbon composed of three fused benzene rings. Many species of bacteria found in soil are capable of utilising phenanthrene as a growth substrate [24].

Fluoranthene: Fluoranthene, a nonalternant PAH containing a five-membered ring, has been shown to be metabolized by a variety of bacteria, and pathways describing its biodegradation have been proposed. Fluoranthene has been used as a model compound in studies which have investigated the effects of surface-active compounds on PAH biodegradation [25].

Fluorene: Fluorene is a polycyclic aromatic hydrocarbon and has been found to be susceptible to microbial degradation to varying extent [23,26].

Pyrene: Pyrene is a PAH consisting of four fused benzene rings. It is the smallest peri-fused PAH (the rings are fused through more than one face). Many microorganisms have shown the capability of utilising four ringed aromatic hydrocarbons such as pyrene [27,28]. Bacteria such as Rhodococcus sp. strain UW1 are capable of growing on pyrene as sole carbon source. This organism was found to mineralise up to 72% of pyrene to CO_2 within two weeks [29].

Composting of oil sludge

Composting offers an economical and effective way to treat oil sludge. Composting process which involves the careful control and addition of nutrients, watering, tilling, addition of suitable microflora and bulking agents (wood-chips or hay) were considered an alternative option to improve the bioremediation of oil sludge [30]. Composting could be divided with respect to modes of operations such as batch operation and continuous or semi-continuous operation. When temperature is the basis, composting can be divided into mesophillic composting (25-40^{0}C) and thermopihillic composting (50-65^{0}C). The main advantage of is composting is waste stabilization [31]. Composting matrices and composts are rich sources of xenobiotic-degrading microorganisms including bacteria, actinomycetes and lignolytic fungi, which can degrade pollutants into compounds such as carbon dioxide and water. These microorganisms can also biotransform pollutants into less toxic substances and/or lock up pollutants within the organic matrix, thereby reducing pollutant bioavailability [32]. Composting of contaminated soil in bio piles is an

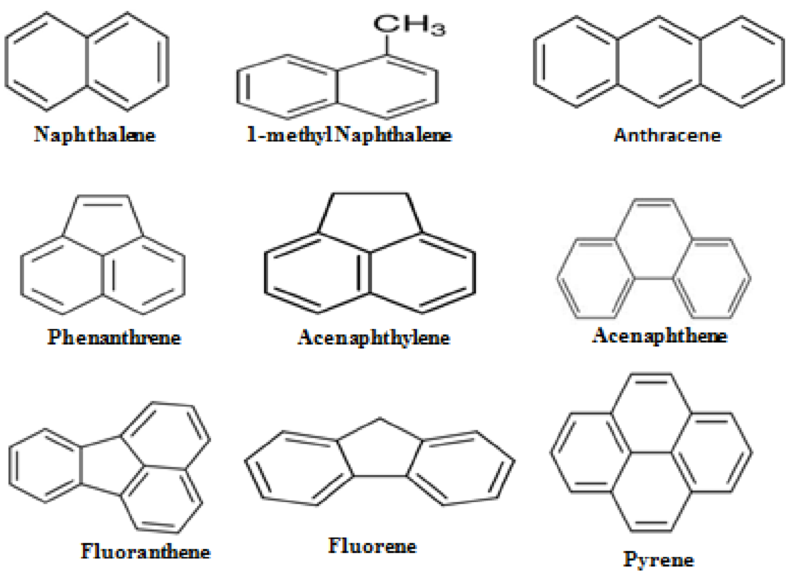

Figure 1: Some polycyclic aromatic hydrocarbons (PAHs) of environmental concern present in oil-sludge.

ex situ technology where organic matter are added to contaminated soil as a bulking agent. Heavy mineral oil degradation was much faster and more complete in compost-amended soil than in hay-, sawdust-, and mineral nutrient-amended soils. The enhanced degradation of heavy mineral oil in compost-amended soil may be a result of the significantly higher microbial activity in this soil [33].The composting process used to stabilize organic materials can be considered as a bioremediation process. Organic residues constitutes a medium in which the microbial population present can remedy the said sludge as long as the conditions support the microbial activity [34]. In composting factors such as aeration, use of inorganic nutrients or fertilizers and the type of microbial species play a major role in the remediation of oil contaminated sites. Experiments for bioremediation of oil sludge-contaminated soil in the presence of a bacterial consortium, inorganic nutrients , compost and bulking agent (wheat bran) showed 76% hydrocarbon removal compared to 66% in case of inorganic nutrients amended soil [35]. A study reported that oil wastes sludge from petrol stations and petroleum residues from a refinery decomposed to 78-93% during 4.5 months of composting with horse manure. At the end of the experiment, most of the polyaromatic hydrocarbons had been degraded except pyrene, chrysene and dibenz(ah)anthracene [36]. Ecotoxicity tests using luminescent bacteria and tests on plants in Petri dishes demonstrated that the composting process undoubtedly led to the biodegradation of toxic compounds. One of the most effective treatment is adding the bulking agent, where the initial hydrocarbon content was reduced by 60% in 3 months, compared with the 32% reduction achieved without the bulking agent [34]. Soil contaminated with diesel oil sludge (10,000 mg/kg sample on a dry weight basis) can also be supplemented with sewage sludge or compost for composting of contaminated soil in the ratio of 1:0.1, 1:0.3, 1:0.5, and 1:1 as wet weight basis. The degradation of diesel oil was significantly enhanced by the addition of these organic supplements relative to straight soil with degradation rates of total petroleum hydrocarbons (TPH) and n-alkanes the greatest at the ratio of 1:0.5 of contaminated soil to organic amendments on wet weight basis. The first order degradation constant of n-alkanes was about twice TPH degradation constant regardless of the kind and the amount of organic supplements [37]. In an experiments for the 90-day period bulked soil showed more rapid degradation of oil compared to all other amendments. Wheat bran-amended soil showed 76% hydrocarbon removal compared to 66% in the case of inorganic nutrients-amended soil due to a corresponding increase in the number of bacterial populations. Addition of the bacterial consortium in different amendments significantly enhanced the removal of oil from the petroleum sludge from different treatment units [38]. In a field-scale study in China bioremediation by augmentation of biopreparation was compared with a conventional composting using oily sludge and oil-polluted soil received from an oil production plant. The total hydrocarbon content (THC) varied from 327.7 to 371.2 g kg−1 of dry sludge and the THC in contaminated soil was 151.0 g kg−1. The. The sludge was mixed and watered every 3 days, biopreparation was applied every 2 weeks and experiment lasted 56 days under the ambient temperature. The THC decreased by 46–53% in the oily sludge and soil, while in the positive controls (activation of indigenous microorganisms) the THC decreased by 13–23%. After composting, the THC decreased by 31% in the oily sludge. The planting of Tall Fescue (Festuca arundinace) revealed a decrease of sludge toxicity after application of both bioremediation technologies and additionally decreased the THC by 5–7% [39].

Application of composting to treat oil sludge

Bioremediation has been accepted as an important method for the treatment of oil sludge by employing indigenous or extraneous microbial flora. The bioremediation of a dehydrated oil sludge of 960 m3 in volume was carried out in prepared bed in Binyi oil-containing sewage disposal station, of the oil contaminants, 52.75% was degraded within 160 days when treated in a greenhouse, while the oil contamination decreased by only 15.46% in the untreated sludge [30] composting of horse manure was used as a means of degradation of two oil wastes, oil sludge from petrol stations and petroleum residues from a refinery. Oil wastes decomposed to 78-93% during 4.5 months of composting. The degradation of the waste oils was higher than that of the reference paraffin oil [36]. In a study [40] contaminated soil (FAO: Lithosol) containing >380 000 mg kg-1 total petroleum hydrocarbons (TPH) was bio remediated by composting. The soil was inoculated with sewage sludge and incubated for 19 months. The contaminated soil-wood chips mixture was mixed in a ratio of 4:1 with sewage sludge. Control experiments containing the contaminated soil and wood chips but without sewage sludge were set up in triplicate. Total petroleum hydrocarbons (TPH) was reduced by 17% in the control experiments and 99% in the sewage sludge compost at the end of the incubation period. The concentrations of most of the selected hydrocarbon components were reduced by up to 100% within the same period.

Vermicomposting represents a cost effective and speedy biotechnological process to convert organic substances into stabilized humus like byproducts using worms [41]. Soil microorganism, such as earthworms have considerable potential to boost the decomposition process, consequently it reduces the waste volume and resulted into the value added product [42]. In a study, the oil sludge from the oil storage tank of a typical petroleum refinery plant located in northern Taiwan is used as the raw material of thermal treatment using oxygen-containing gas. The treatment of oil sludge is conducted by the use of carrier gas with different concentrations of oxygen (4.83, 8.62, 12.35, and 20.95 vol % O_2) in the temperature range of 380−1123 K and at various constant heating rates of 5.2, 12.8, and 21.8 K/min. The significant reactions occur in the range 415−931 K. Below a temperature of 613 K, pyrolysis reactions are predominant [43]. Addition of palm oil mill effluent (POME) anaerobic sludge into the pressed-shredded empty fruit bunches composting showed biodegradation of composting materials by reduction of cellulose (34%) and hemicellulose (27%) content towards the end of treatment [44].

Fate of organic and inorganic compound

Biodegradation is the natural breaking down organic or inorganic matter into nutrients using living organisms under aerobic or anaerobic conditions. Various onsite techniques such as land farming, composting and soil piles as well as advanced ex situ methods such as bioreactors with better control of temperature and pressure that enhance the degradation of PAHs in soil has been reported by reference [45]. The biodegradation (transformation or mineralization) of a wide range of hydrocarbons, including aromatic, halogenated, aliphatic, and nitrated compounds, has been shown to occur in extreme environmental conditions [46]. When oil sludge containing large quantities of hydrocarbons is degraded, microorganisms use those hydrocarbons as substrates [47]. The initial step in degradation is the catabolism of oil sludge by bacteria and fungi, which involves the oxidation of the substrate by oxygenases, in which molecular oxygen is required. Aerobic bacteria use oxygen as an electron acceptor to break down both the organic and inorganic matters into smaller compounds, producing carbon dioxide and water as final products [48]. Oil sludge exhibits some biodegradable properties in the environments such as transformation, conversion or mineralisation, specific adhesion mechanisms and production of extracellular emulsifying agent by micro- organisms [49,50]. With the help of enzymes these contaminants are degraded to simpler, lower molecular chains and less toxic compounds (CO_2 and H_2O) are synthesized [51]. Hence aerobic conditions and specific micro-organisms are required for the bioremediation of PAHs contaminated soils. Low oxygen content was found to limit the bioremediation of PAHs contaminated soils [52]. Also the oxygen content in soils depends on microbial activity, soil texture, water content and depth [53]. The most rapid and complete degradation of the majority of organic pollutants is brought about under aerobic conditions [54].

Low molecular weight petroleum hydrocarbons (4-ring or less), are first removed through evaporation. As the molecular sizes increases, biodegradation rates become slower. Reference [55], reported the successful degradation of 16 priority PAHs using the injection of bacteria (the phenanthrene- and anthracene-degrading strain Sphingomonas *paucimobilis)*, fungi and bacteria – fungi microbial groups. Two white rot fungi were reported to achieve 58 -73% degradation of 3- and 4 – ring PAHs. Oil sludge, albeit very slow, is susceptible to degradation by naturally occurring microflora, but this process reduces nutrient and oxygen level in soil which in turn impedes other environmental processes such as transformation or mineralisation. In order to enhance the oil sludge biodegradation processes the bioavailability of hydrocarbons present in the oil sludge matrix can be increased by biostimulation, which is simply the addition of nutrients to stimulate the growth and degradative capabilities of the indigenous microorganisms present [56]. A study [57] showed enhanced remediation of oil sludge contaminated soil with bacterial consortium, bulking agents, inorganic supplements and compost. An inorganic nutrient was found to produce little influence on oil removal compared to soil amendment without inorganic nutrients. Soil microbial population was found to enhance the removal of hydrocarbons from soil. The treatment of 16 polycyclic aromatic hydrocarbons (PAHs), targeted by the US Environmental Protection Agency (USEPA), by composting led to a decrease of all PAHs mainly in the stabilization phase. The PAHs with three or fewer aromatic rings ($N \leqslant 3$) (except phenanthrene) exhibited a continuous decrease, while those with four or more ($N \geqslant 4$) showed increases in the intermediate stages (30–60 days) [58]. Toxicity tests on the aqueous soil extracts as well as plant growth and worm tests on the landfarm soil showed no striking negative effects of residual hydrocarbons. Oil, nitrate and phosphate to the groundwater showed minimal migration in case of large-scale landfarming [59]. The analysis of individual PAH using static-aerated biopiles in the mixture suggested that mechanisms of volatilization is responsible for the reduction of 2 ring PAH and biodegradation for 3–4 ring PAH [20].

Factors affecting composting

Temperature: An increase in temperature in the compost pile increases solubility of contaminants and induces higher metabolic activity of the compost [45]. Oil sludge contains highly degradable materials, these microorganisms accept the hydrocarbons as substrates, which enhance their activities leading to the higher increase in temperature [34]. Low temperature also affects microbial growth, propagation and subsequently results in decrease in the rate of degradation [45].

pH: Oil sludge degrading bacteria and fungi performance are affected by pH level; while on other hand nutrients like nitrogen and phosphorous have great effect on microbial degradation of oil sludge constituents . The mineralization of hydrocarbon components in the environment is generally optimal at pH 7 to 7,8, thus overall biodegradation process is enhanced [60]. Fungal decomposition of PAHs may produce mutagenic intermediates [61]. In such instance, liming may be used to increase the pH from acidic to alkaline state so that bacterial growth may be favored than fungal growth.

Moisture level: Moisture required for enzymatic activity. Elemental uptake by microorganisms is by absorption and transportation of solubilized molecules across the cell membrane. The availability of target molecules to the microorganisms depends on the amount of water present in the treatment matrix. Optimal water content for aerobc bioremediation treatment matrix is usually between 10% and 20% by mass [62]. Optimal activity occurs when the soil moisture and water content for aerobic bioremediation treatment matrix is usually between 50 and 80% of saturation (moisture holding capacity) [63].

Oxygen level: the breakdown of oil sludge components may possibly involve the utilization of oxygenase, in which molecular oxygen is required. Great efficiency of natural microbial hydrocarbon degradation occurs mostly when oxygen is available [64]. Aerobic biodegradation is the most effective pathway for bioremediation. This means that the presence and concentration of oxygen is the rate-limiting parameter in the biodegradation and catabolism of cyclic and aromatic hydrocarbons by bacteria and fungi [27]. Oxygen uptake rate (also referred to as dynamic respirometric index) provides the most reliable values of microbial activity in a compost environment [65].

Carbon/Nitrogen nutrient ratio: Different lignocellulosic residues are mixed with the sludge in order to obtain a suitable moisture and C/N ratio. Experiments were conducted to investigate the effects of C/N (carbon/nitrogen) ratio, C/P (carbon/phosphate) ratio and iron concentration on fermentative hydrogen production from POME. C/N ratio and Fe concenteration had larger effect on hydrogen production ($P<0.05$) while C/N and C/P ratio had effect on COD removal ($P<0.05$) [52]. Composting of oil palm empty-fruit-bunches and of oil palm empty-fruit-bunches in supplementation with various bulking agents differed in the resulting C: N ratios. Initially C:N ratios were 52:1, 35:1, 48:1, 47:1 respectively for the four compost heaps which were significantly reduced to 24:1, 14:1, 18:1 and 12:1 after composting for 60 days, resulting in the production of a stable humus that can be used for crop production. The increase in the nitrogen content of the compost gave a positive correlation with rate of utilization of cellulosic material [66]. In a pilot plant, four olive mill wastewater (OMW) composts, prepared with two different bulking agents and three N-rich organic wastes, were studied using the Rutgers system. Only the composts prepared with OMW, cotton waste (CW) and poultry manure or sewage sludge reached water-soluble organic C (C_W) and NH_4^+-N concentrations and C_W/N_{org} and NH_4^+/NO_3^-ratios within the established limits, indicating the effect of C/N ratio for a good degree of compost maturity [67] .

Conclusion

Compostable substrates contain metabolizable carbon, which will enhance microbial diversity and activity during composting and will promote degradation of xenobiotic organic compounds. PAH constituent of the oil sludge poses a serious threat to environment can render harm to humans, livestock, wildlife, crops, or native plants. A lot of physical, chemical and biological methods have been developed by none is as economical and environment friendly as composting. Still it has not been practiced widely although its potential is immense in terms of bioremediation. The general conclusion is, however, that composting degrades or binds pollutants to innocuous levels or into innocuous compounds and has substantial potential for remediation of polluted materials.

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