

# A review- bioremediation of oil sludge contaminated soil

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**Abstract.** Petroleum oil as a vast source of energy widely used in the whole world in several sectors especially in industry and transportation. The leakage or contamination of oil from pipeline, tank, and industry as a form of oil sludge with soil can produce major environmental and health hazard. Bioremediation is one of the most economical and environmentally safe technology to prevent this contamination though it takes longer time. This paper reviews the basic processes involved in bioremediation, types and the factors affecting it. This study includes some previously adopted different bioremediation methods varies with different process material such as refinery treatment sludge, sewage sludge, microbial organism, bulking agents and different chemical additives. The comparison of these methods is presented in respect of the removal efficiency of an entire process as well as the TPH (Total Petroleum Hydrocarbon), aliphatic, aromatic, resins, asphaltene fraction of oil sludge within the different period of time.

## 1 Introduction

The importance of petroleum oil is increased day by day as it is used as major raw material to provide the energy and heating facility for running the huge number of industries around the world [1]. The consumption of petroleum oil was 93.60\*106 barrels per day in 2015 which may be increased about 38% at the end of 2040 [2, 3]. This petroleum oil is a composition of aliphatic hydrocarbon, aromatics, resin, asphaltene and other organic and organometallic compound [4]. Environment is seriously contaminated with petroleum products due to tanker accidents, storage tank ruptures, pipeline leaks, transport accidents, offshore and onshore petroleum industry activities etc. [5–7]. These petroleum products adversely affect the ecosystems and biological activity by changing the parameter such as pH, moisture content and aeration level of soil and introduce toxicity [8, 9]. Petroleum hydrocarbons exist in 60% of contaminated sites of whole Canada causing toxicity propagation in environment, continuation of soil degradation by affecting water and nutrient retention [10, 11]. There are some physical and chemical method such as landfilling, incineration to remove hydrocarbon contaminants. These methods often produce toxic chemical affecting the physicochemical properties of soil [4]. Bioremediation has the cost effective, eco-friendly and sustainability advantages comparable to these method [12]. The aim of this study is to give an overview of bioremediation technology highlighting the factors on which it depends and comparison of different strategies adopted by different researchers for this technology.

## 2 Overview of bioremediation technology

Bioremediation of contaminated soil is defined as the use of living organism (mainly bacteria, fungi, algae) to make environment free from toxicity of contaminant through transformation, degradation and mineralization of the contaminants to less harmful compound [13, 14]. There are three basic processes involved in this technology. These are bio-transformation which convert contaminant components into less or non-hazardous products, biodegradation which produces smaller organic or inorganic molecules from organic substances and finally mineralization which involves entire breakdown of organic materials into inorganic compounds such as CO<sub>2</sub> or H<sub>2</sub>O [15]. Each of these process can occur at contaminant site (in situ) or out of contaminant site (ex situ) as well as aerobic (with oxygen) and anaerobic (without oxygen) [16]. In situ bioremediation involves the treatment of organic pollutants under natural conditions at original contaminated sites without relocation [7, 14]. Whereas Ex situ bioremediation excavation and transportation of pollutants from original contaminated site to be treated elsewhere to maintain the proper conditions of the bioremediation process more easily [14]. Though ex situ bioremediation is faster than in situ, due to relocation of pollutants it has increased cost and the chance of direct exposure of toxicity to public generally worker [7]. Fig. 1 represents the types of bioremediation.

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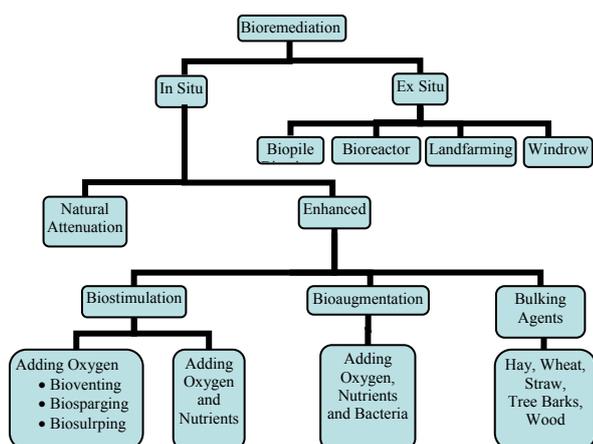


Fig. 1 Schematic diagram for types of bioremediation [10, 20].

### 3 Factors affecting bioremediation technology

#### 3.1 Nutrients

The acceleration and growth of microbial organism such as bacteria and fungi depends on carbon nutrient elements such as Nitrogen and Phosphorus, an electron acceptor and organic compound that supplies carbon and energy [17, 18]. The growth of the microorganism is prevented if there is shortage of organic and inorganic compounds such as nitrates, sulphates, carbon dioxide, ferric iron, amino acids, vitamins and the metabolism of microorganisms will occur [14]. Microorganisms facilitate degradation of oil sludge depending on some certain forms of nitrogen and phosphorus ( $\text{NH}_3$ ,  $\text{NO}^{3-}$ ,  $\text{NO}^{2-}$ , organic nitrogen and orthophosphate such as  $\text{KNO}_3$ ,  $\text{NaNO}_3$ ,  $\text{NH}_3\text{NO}_3$ ,  $\text{K}_2\text{HPO}_4$  and  $\text{MgNH}_4\text{PO}_4$ ) and nutrients in the form of fertilizers, slow release and oleophilic are added to meet this requirement [19].

#### 3.2 Effect of pH

Microorganisms and enzymes have pH dependency behaviour [21]. In most of cases microbial organism perform degradation activities at optimum nearly at neutral pH between 6 and 9 [22, 23]. In some cases, liming is introduced to transform from acidic to alkaline state to facilitate bacterial more than fungal growth to avoid mutagenic intermediates produced by fungal decomposition of PAHs [14].

#### 3.3 Temperature

There is direct effect of temperature on the properties of oil hydrocarbon in soil. Generally this hydrocarbon pollutant exists in soil with longer period of time at low temperature [21]. The viscosity is increased with decrease in temperature whereas the solubility in water and volatility is reduced which negatively affect the transformation of long chain n-alkanes from solid phase to water phase causing delaying the oil sludge biodegradation [24]. Though there is a variety range of

temperatures for effective biodegradation of hydrocarbon it is found to be optimum between 30 to 40°C [14].

#### 3.4 Water activity

Biodegradation rates are highly influenced by the microbial growth and their biochemical and enzymatic activities which are directly related to amount of water in soil. Sufficient moisture content is needed for transportation of both soluble pollutant molecules across the microbial cell membrane through absorption and final degraded products [7, 14 & 21]. Optimal bioactivity for effective aerobic bioremediation occurs when moisture is between 30% and 80% of the saturation (moisture holding capacity) available for plant usage [10]. Microbial activity becomes less effective when the moisture content is below 10% [10, 25]. However there is waterlogging problem if water holding capacity of soil rises above the optimal limit which develops anoxic condition and badly affects bioremediation rates [14].

#### 3.5 Oxygen

Oxygen is mostly important for aerobic biodegradation which is the more effective than anaerobic condition because the biodegradation rates in that condition is limited to halogenated aromatics compounds [26]. The molecular oxygen is needed for use of oxygenase to degrade the oil sludge component and higher efficiency is achieved with sufficient availability of oxygen. This means that, the presence and concentration of oxygen is important in such process [27]. Also, lack of aeration, in the system may be a rate-limiting parameter in the biodegradation and catabolism of hydrocarbons by bacteria and fungi [23].

#### 3.6 Texture, hydraulic conductivity and permeability

These three interrelated geotechnical properties of soil affect the supply of nutrient, oxygen & contaminants which in return affect the microbial activity and biodegradation of pollutant [10]. Low permeability fractured rocks are inefficient to supply the flushing solutions to the contaminants and flushed with surfactants and co-solvents [10]. Soil with hydraulic conductivity larger than  $10^{-4}$  cm/s is found to be sufficient for transport of nutrients and pollutants [28]. Finely texture soils (greater amount clay minerals) of low permeability will supply less nutrients and oxygen to microorganisms and due to their high surface area and chemical reactivity will cause biofouling as because soil pore spaces are plugged with microbial cells [29]. Sand and silt particles have less effect on the supply of nutrients and oxygen as they are less reactive chemically due to their smaller surface area [10].

### 3.7 Salinity

It was found from different studies that salinity maintains a positive dependency with rates of mineralization of polycyclic aromatic hydrocarbons [30]. It is also noted that hyper salinity with high temperature will decrease microbial metabolic rates, growth and their products [31].

### 4 Material Used

All of these types shown in Fig. 1 adopt the mechanism to stimulate the microbial community for degrading the contaminants of soil. Materials used to assist in bioremediation are generally based on these types for accumulating the growth of microbial community. Table I summarizes some materials used in different bioremediation methods by some researchers. Some use addition of nutrients such as slow release fertilizer in bio-stimulation to increase the efficiency of biodegradation by stimulating microbial growth [32]. Bulking agent such as wheat bran, wood chips, sawdust, leaves, hay, shredded rubber tires, cotton stalks, rice straw are used to increase the oxygen diffusion by increasing soil porosity and decreasing the soil bulk density and it facilitates microbial activity [33]. Inorganic salts with commercial fertilizers, manure and sewage sludge have an effect to increase the removal efficiency of hydrocarbons in soil up to two times [13].

### 5 Microbial communities and activities

Microbial community characterization with a combination of a single bacterial strain or in a syntrophic bacterial consortium, of selective degrading abilities can provide successful mechanism to biodegrade in a given environment [34]. Soil microbial activities are found to be negatively affected if hydrocarbon levels go beyond 10% [35]. Bio-surfactants an important agents produced by bacteria have the ability to emulsify petroleum hydrocarbon in oil sludge by increasing the surface area of the substrates and their solubility will be increased so that they can be easily biodegradable to bacteria and fungi [14]. These bio-surfactants extracted by bacteria can enhance hydrocarbon catabolism with bulking agents and has the advantage of being more cost effective, natural and non-toxic than chemical surfactants in biodegradation of petroleum hydrocarbons [36].

### 6 Comparison

Table 1 represents the different bioremediation strategies of soil contaminated with different pollutants such as crude oil, diesel oil, petroleum, sludge from different oil industry which performed by different researcher. It also shows that different materials such as different bacterial community, bulking agents, nutrients in the form of fertilizer and some chemical additives were used by different methods. All these methods were run for various duration from 38 days to 1 year and consequently achieve removal efficiencies from 58% to 99.9% with different contaminant concentration.

**Table 1.** Different Bioremediation Strategies of Oil Contaminated Soil.

Material or Additive Used	Oil Contaminant & Concentration	Maximum Duration	Maximum Removal Efficiency	Source
Two hydrocarbon clastic bacterial isolates	Crude oil tank bottom sludge (COTBS), 30.7 g/kg	90-days	96-97%	[3]
Nutrients as phosphate and nitrate salts	Maroon oil reservoir tank sludge, 5 % w/w	365-days	TPH 34%, Aromatic Fraction 43%, Aliphatic Fraction 64%, Asphaltene 4%, Resin 6%	[4]
Nutrients, inoculum and bulking agents	Diesel, 1 % v/v	90-days	96.6%	[10]
Landfarming with cotton stalks	Shengli oil field open storage sludge, 12.57 mg/g	1170-days	TPH 64.8%, Aromatic Fraction 85.5%, Aliphatic Fraction 90%	[37]
Bacterial consortium, inorganic nutrients, compost and a bulking agent of wheat bran	Petroleum refinery Sludge, 5% w/w	90-days	72%	[38]
Bacterial consortium, nutrients mixture	Refinery oil sludge, 18% w/w	365-days	TPH 89.7%, Aromatic Fraction 94.1%, Aliphatic Fraction 95.1%, Asphaltene + NSO 63.5 %	[39]
Sewage sludge, chemical additives	Diesel fuel, 30 g/kg & Black oil, 30 g/kg	120-days	90% & 63%	[40]
Manure with indigenous microbes and rice straw	Petroleum, 519 g/kg	360-days	TPH 58.2%, Aromatic Fraction 58.52%, Aliphatic Fraction 73.65%, Asphaltene 28.84%, Resin 16.85%	[12]
Oleophilic fertilizer	Petroleum, 2500 mg/kg	38-days	80%	[6]
Organic solid waste of soyacake	Diesel, 5% w/w	126-days	81%	[41]
Microbial consortium	Crude oil, 5000 mg/kg	40-days	64.4%	[42]

Fig. 2 shows the comparison between 3 strategies of diesel oil contaminated soil. The highest removal efficiency is 96.6% after final 90 days in the technology which uses Ottawa sand as bulking agent, poultry and cow manure as nutrient and microbial inoculum. Other two methods use organic solid waste of soy cake, sewage sludge and chemical additives. So, the interaction between bulking agent, microbial community and nutrient can be an important concern to achieve greater removal efficiency within less time period.

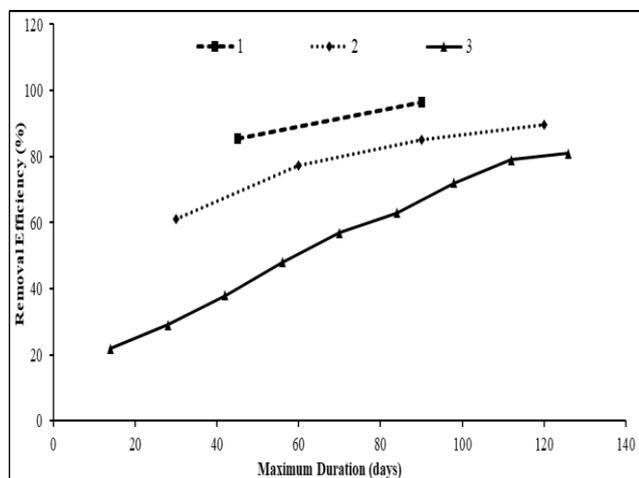


Fig. 2. Removal Efficiency vs Maximum Duration for Diesel Oil Contaminated Soil of three Strategies. Data for 1, 2, 3 from [10, 40, 41].

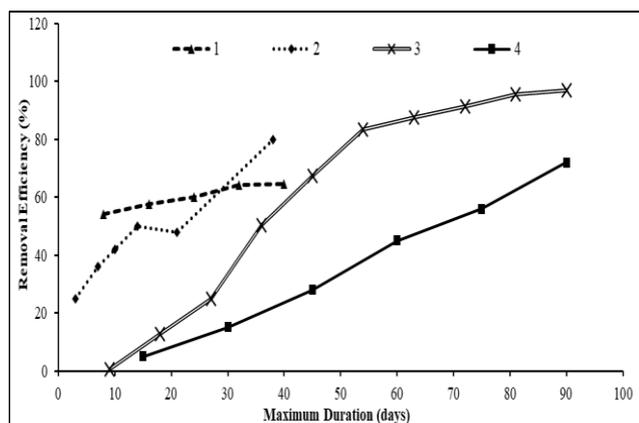


Fig. 3. Removal Efficiency of Total Petroleum Hydrocarbon vs Maximum Duration of four Strategies. Data for 1, 2, 3, 4 from [3, 6, 38, 42].

Fig. 3 represents the comparison between the removal efficiencies of total petroleum hydrocarbon in 4 strategies of bioremediation. Highest removal efficiency is 96.93 % which is for bioremediation method carried out with two hydrocarbon clastic bacterial isolates and 10 stages of 90 days duration in a crude oil bottom tank sludge contaminated soil. Removal efficiency with 4 stages of 40 days period in crude oil contaminated soil is 64.4% which is lowest among these four methods. So the amount of stages and duration of test involved in a specific bioremediation method can play a significant role to remove oil pollutants.

There is a comparison between maximum removal efficiency of TPH (Total petroleum hydrocarbon), aliphatic fraction, aromatic fraction, asphaltene, asphaltene combined with NSO (nitrogen, sulphur, and oxygen containing compound) and resin in 4 different methods shown in Figure 4. All of these methods were carried out for 1 year or more than 1 year.

Aliphatic fraction removal efficiency is highest in all four cases. Resin and asphaltene which are complex compound has the lowest removal efficiency as it takes larger time to break down than other compound. If asphaltene is combined with NSO, its removal efficiency as well as overall efficiency will be increased. In maximum cases, treated mechanism was found to be more effective than control mechanism.

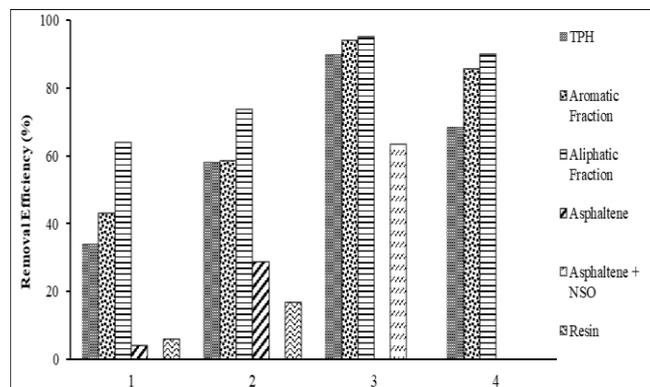


Fig. 4. Removal Efficiency of Total Petroleum Hydrocarbon with Different Fraction of four Strategies. Data for 1, 2, 3, 4 from [4, 12, 37, 39].

## 7 Conclusion

Bioremediation is considered as principal natural process to remove the petroleum hydrocarbon pollutants from the environment in the whole world. Factors which affect bioremediation technology should be kept favorable to carry out microbial activities for optimal result in this method. It is important to select appropriate combination of different materials such as nutrient, bulking agent, microbial community to remediate soil successfully contaminated with different contaminants. Time duration and stages in a method can affect the removal efficiency significantly and complex compounds in oil pollutant can delay the full reduction of contaminants.

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