



A systematic review on the effectiveness of remediation methods for oil contaminated soils

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ABSTRACT

There is no doubt that several soil remediation methods have been applied in removing oil contaminants in the soil. Selecting an appropriate method is critical for effective cleanup of a contaminated site. The aim of this study is to review field studies of soil remediation methods to determine their effectiveness in cleaning oil contaminated soils. A systematic search of literature was conducted in different databases to extract mainly field studies, few pilot studies, and greenhouse experiments. One grey literature (conference paper) was also selected. Studies selected were published between 2000 to 2021, and 51 literatures were chosen for this review.

The review of field application studies on soil remediation revealed that combined method has high oil removal efficiency, short cleanup duration, moderate remediation cost, and low environmental impact. Followed by biological method which is considered a green and cheap remediation method. While chemical method was reported as the least effective soil remediation method, concerns exist over their negative environmental impact, and high remediation cost. Similarly, physical method showed low remediation effectiveness resulting from high energy consumption of most technologies under physical method and the need for further treatment of gases produced during remediation. Therefore, combined method was considered the most effective remediation method for oil contaminated soil.

1. Introduction

Oil pollution has become a major source of environmental concern globally due to its severe human and ecological consequences. Oil is usually released accidentally or deliberately into the environment during extraction, production, transportation, storage, and processing (Berthe-Corti and Höpner, 2005). The movement of oil involves about 10-15 transfers with pipelines, tankers, tank trucks, and railcars, and these may cause accidental release of oil into the environment. A typical example is the Lac-Mégantic oil spill that happened in 2013 in Quebec, Canada when a train carrying crude oil derailed, and spilled 5416 tons of crude oil into the environment. In addition, human errors, and other man-made activities such as war, vandalism of oil facilities, and equipment failure have all been identified as major causes of oil spills. The deliberate release of crude oil into the environment during the Gulf war contaminating the soil and creating about 320 oil lakes across the Gulf desert (Radwan et al., 1995), and the incessant vandalism of oil pipelines and equipment in Niger Delta, Nigeria are typical examples, pictures of the devastation of oil pollution in Ogoni land Niger Delta,

Nigeria is shown in Fig. 1.

Petroleum hydrocarbons are a mixture of simple and complex hydrocarbons that consist primarily of carbon and hydrogen and some amounts of nitrogen, sulphur and oxygen (Chandra et al., 2013; Varjani and Upasani, 2017). They are categorized into four areas: the saturates, aromatics, asphaltenes (phenols, fatty acids, ketones, esters, and porphyrins) and the resins (pyridines, quinolines, carbazoles, sulfides and amides) (Huesemann and Moore, 1993; Adeniji et al., 2017), as shown in Fig. 2. They also contain many volatile compounds like benzene, toluene, ethylbenzene, and xylene (BTEX), and some toxic compounds including polycyclic aromatic hydrocarbons (PAHs) which bind to soil components and are difficult to degrade (Samanta et al., 2002; Pino-Gonzalez et al., 2018). Their toxicity, mutagenic and carcinogenic nature as well as their persistence in the environment may affect human health and the ecosystem, thus they are regarded as priority and environmental contaminants by the United States Environmental Protection Agency (USEPA).

The devastating effects of oil pollution to the soil is enormous, oil alters the soil microbial population, composition and structure of soil

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organic matter and enzymatic activities within the soil, thus impeding the growth and development of plants. They bind to soil components and are usually difficult to remove or degrade, causing contamination of surface and ground water systems (Huesemann and Moore, 1993; Samanta et al., 2002; Abioye et al., 2011; Adeniji et al., 2017; Pinedo-Gonzalez et al., 2018). With the growing interest in soil remediation, different remediation methods have been proposed for oil contaminated sites. The traditional soil remediation methods of excavation, landfilling and incineration were considered not efficient. Excavation and landfilling could not remove pollutants from soil, while incineration is expensive and can lead to formation of secondary pollution, such as the formation of volatile organic compounds (Chu and Kwan, 2003; Greene, 2005; Lemming et al., 2010). In recent years, many laboratory and field studies have been conducted to develop remediation methods that can effectively remove oil contamination from soil. Bioremediation which includes the use of plants and microorganisms to remove contaminants from the environment has received considerable attention as the most promising method for oil spill cleanup. Proponents of phytoremediation have argued that it is aesthetically pleasant, economical, and simple (Pilon-Smits, 2005; Prasad and Singh, 2011; Ali et al., 2013). The mechanism of phytoremediation involves uptake of contaminants by plant roots for stabilization, volatilization, or degradation, however, most plant roots are sensitive to high oil concentration and cannot penetrate deep soils (Alkorta et al., 2004; Yoon et al., 2006; Doty, 2008; Wu et al., 2010; Singh and Singh, 2017).

Furthermore, several authors have studied the degradation of oil by various microorganisms in the soil. Chen et al. (2014) investigated the ability of three bacterial strains (*Pseudomonas* spp. *Ochrobacterium tritici* and *Bordetella petrii*) isolated from oil-contaminated soil to degrade petroleum lubricating oil, the result showed excellent ability of the three bacterial strains in oil degradation. While Nna Orji et al. (2018) studied the ability of fifteen microorganisms to detoxify crude oil contaminated soil, following bioaugmentation with each isolate five microorganisms were identified as best oil degraders (Samanta et al., 2002; Maila and Cloete, 2004; Basumatary et al., 2012; Cubitto and Gentili, 2015; Adeniji et al., 2017). Many critics of microbial remediation have opined that they require longer time and may not efficiently remove high concentration of oil in the soil (Margesin et al., 2003).

However, some scholars believe a combination of bioaugmentation, and biostimulation will reduce cleanup duration and enhance remediation efficiency (Lin et al., 2009; Nikolopoulou et al., 2013). Nikolopoulou et al. (2013) studied the effect of alternative nutrients on the detoxification of crude oil contaminated beach sand, the result revealed that adding nutrients to the oil contaminated beach sand significantly increased the activity of indigenous microorganisms and subsequent decontamination of the beach sand within 30days. In addition, fungi have shown more tolerance to high concentration of pollutants than bacteria due to their aggressive growth, wide biomass output, and long hyphae reach (Potin et al., 2004; Wu et al., 2008). According to several

literatures, the strategy involved in fungal degradation of oil is enzymatic transformation by intracellular cytochrome P450 and extracellular ligninolytic enzymes. Rao et al. (2014) identified three enzymes including lignin peroxidase (Lip), Manganese peroxidase (Mnp), and Laccase as important enzymes in oil degradation, and several authors have reported the success of these enzymes in removing oil pollutants from soil (Wu et al., 2008), however this approach is limited due to difficulties in purification and cost of enzymes.

Also, some scholars have argued that chemical remediation methods such as chemical oxidation and surfactant washing are fast in removing high concentrations of pollutants from soil and can be used to overcome limitations of microbial degradation, nonetheless the toxic effect of chemicals on the environment is a major constraint to their application in oil spill removal (Lee and Jones-Lee, 1997; Cho et al., 2002; Ward et al., 2004; Mecozzi et al., 2006). While several others have considered thermal methods such as electrokinesis, microwave heating, and thermal desorption very effective due to their high remediation efficiency within short duration (Lee and Jones-Lee, 1997). Thermal methods involve heating the soil to transform pollutants by changing their chemical compositions and have shown to remove high concentrations of oil in soil, however the application of heat to soil during thermal remediation can be expensive and can cause damage to the soil structure (Xu et al., 2019).

Recently, many scholars have suggested that applying air to soil in biosparging and bioventing techniques can stimulate the degradation activities of microorganisms and enhance the removal of deep soil oil pollutants. Kao et al. (2008) reported that biosparging which involves transferring contaminants from aqueous phase to gaseous phase via injected airstream can only remove volatile organic compounds (VOCs) such as BTEX, leaving nonvolatile compounds and heavy oil in the soil (Johnson et al., 2001; Kao et al., 2008). While bioventing has been shown to remove soil pollutants ranging from light to middle distillate oil (Lee and Swindoll, 1993; Mosco and Zytner, 2017). However, Mosco and Zytner (2017) conducted a bioventing experiment using aged soil and noted that sufficient oil pollutants were still bound to the soil, indicating that bioventing could not remove heavy and aged oil in soil. The production of off-gas a secondary pollution is a major limitation of bioventing (Lee and Swindoll, 1993).

There is no doubt that various soil remediation methods have been used in removing oil pollutants in the soil (Musterait, 2001; Kao et al., 2008; Wu et al., 2008; Cubitto and Gentili, 2015; Song et al., 2019), as summarized in Table 1. Selecting an appropriate remediation method is critical for effective clean-up of oil polluted sites. Most remediation experts believe the most effective remediation method will depend on the type of oil spilled, quantity spilled, and the environmental medium impacted (Samanta et al., 2002; Pugazhendhi et al., 2018). Some others believe the cost of clean-up, duration, and the impact of a remediation method on the environment are major factors that determine selection of a remediation method. While Martin (1998) believe the combination of



Fig. 1. Oil Pollution in Ogoni land Niger Delta, Nigeria.

two or more methods may result in a synergistic effect and subsequent clean-up efficiency. Therefore, this systematic review aims at synthesizing field application studies of different remediation methods and evaluate their effectiveness in removing oil contaminants in the soil. The effectiveness of a remediation method will depend on cost of application, impact on the environment, clean-up duration and clean-up efficiency. This review is imperative because it will help environmental managers and users to make right decisions on the appropriate methods to apply in oil spill cleanup. Fig. 3 is a representation of studies in the literature review.

1.1. Major land oil spills in history

Each year, thousands of oil spills occur globally on land. Although some of these spills are minor oil spills, many are major environmental disasters caused by accidents, equipment failure, pipeline leakage, vandalism etc. Table 2 shows some major land oil spills in history, and remediation methods applied to remove the oil spill and clean the soil.

1.2. Remediation methods

The process of removing harmful substances from environmental medium or changing them to less harmful substances is referred to as remediation. They are techniques or skills employed to reduce or eliminate contaminants from soil and other environmental medium, ranging from simple to complex techniques involving both *in situ* and *ex situ* methods (Kuppusamy et al., 2017). Remediation methods include mechanical recovery and containment, incineration, solidification/stabilization, capping, vitrification, encapsulation, bioremediation, electrokinesis, thermal desorption, surfactant washing, and oxidation, among others. According to the United States Environmental Protection Agency (USEPA) remediation techniques can be categorized into biological, chemical, physical, and thermal methods. In this study remediation techniques are categorized into biological, chemical, physical, and combined methods, as presented in Fig. 4.

Physical remediation method uses physical and mechanical barriers to isolate, recover and separate pollutants from the soil, they include capping, soil replacement, soil washing, and thermal desorption (Ossai et al., 2020). Soil replacement involves removing and replacing contaminated soil with new one, while soil washing is an *ex-situ* pretreatment method used to mechanically separate contaminants from the soil. In thermal desorption the contaminated soil is treated with heat using steam, microwave, and infrared radiation to make the

contaminants volatile. The advantage of physical remediation is that treated soil can be reused after remediation process.

Chemical method can offer rapid and aggressive alternatives that are not sensitive to concentration of the contaminant and may offer alternative treatment to overcome some limitations of bioremediation (Kim et al., 2017). Chemical method involves the application of chemicals in soil remediation process and can be done *in-situ* or *ex-situ*. Chemical remediation method includes chemical augmentation, chemical leaching, and chemical oxidation. Chemical augmentation is the addition of chemicals to the soil to stimulate activities of degrading microorganisms in the soil. Some common chemicals used in soil remediation include phosphoric acid, potassium phosphate, sulfuric acid, nitric acid, and hydrogen chloride (Hong et al., 2002; Lee and Lee, 2012). In chemical leaching, the contaminated soil is washed with water, reagents, and other fluids that can leach the pollutants from the soil through ion exchange, precipitation, adsorption, and chelation. While chemical oxidation involves the use of oxidants including permanganate, sodium persulfate, and ozone in soil remediation. The advanced oxidation process uses oxidizing agents such as hydrogen peroxide, and ultraviolet lights to treat contaminated sites. Toxicity and high cost of chemicals are the major disadvantages of chemical method (Yao et al., 2012).

Biological method refers to remediation techniques that use living organisms such as plants and microorganisms to remove, degrade or reduce contaminants in the soil. It is described as the detoxification of polluted sites with biological processes (Enyiukwu et al., 2021). The focus is to optimize soil parameters such as soil temperature, pH, porosity, nutrients, moisture, C:N ratio, microbial population, and diversity in soil remediation (O'Brien et al., 2017). Biological techniques include bioaugmentation, biostimulation, vermiremediation, and phytoremediation (Samanta et al., 2002). Bioaugmentation is applied when indigenous microbes such as bacteria may not be efficient to degrade contaminants, the introduction of microbes to supplement indigenous population increases the rate of degradation. Apart from bacteria, fungi and earthworms have proved to possess abilities to degrade contaminants (Adedokun and Ataga, 2014). While biostimulation modifies the environmental medium to stimulate indigenous microorganisms to carry out degradation of contaminants. Nutrients such as oxygen, nitrogen, carbon, and phosphorus are usually introduced to stimulate activities of bioremediation microbes to degrade contaminants. The major advantage of biostimulation is that indigenous microbes will carry out the degradation of contaminants. In phytoremediation, plants are employed to detoxify contaminated soils, several plants possess the

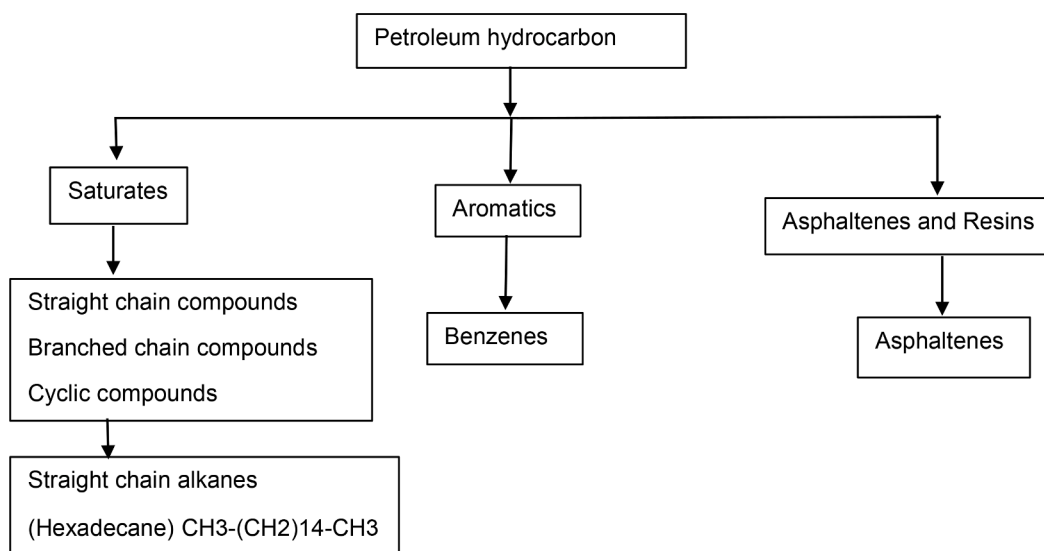


Fig. 2. Components of petroleum hydrocarbon composition.

Table 1
Literature review.

S/ no	Study type	Title	Refs.	Remediation method
1	Microbial degradation	The enhanced biodegradation of petroleum lubricant by bacterial strains harvested from oil contaminated soil.	Chen et al., 2014	Bioremediation
2	Microbial degradation	Pollution Status of Heavy Metals in Spent Oil-Contaminated Soil in Gwagwalada.	Nna Orji et al., 2018	Bioremediation
3	Microbial degradation	Bioremediation of crude oil-contaminated soil by immobilised bacteria on an agro-industrial waste-sunflower seed husks.	Cubitto and Gentili, 2015	Bioremediation
4	Vermiremediation	Bioremediation of petroleum hydrocarbons from crude oil-contaminated soil with earthworms: <i>Hyperidrosis africanus</i> .	Ekperusi and Aigbodion, 2015	Bioremediation
5	Microbial degradation	Bioremediation of crude oil contaminated soils using surfactants and hydrocarbon clastic bacteria.	Etok et al., 2015	Bioremediation
6	Microbial degradation	Dissolution and removal of PAHs from a contaminated soil using sunflower oil.	Gong et al., 2005	Bioremediation
7	Microbial degradation	Removal of polycyclic aromatic hydrocarbons from manufactured gas plant-contaminated soils using sunflower oil: laboratory column experiments.	Gong et al., 2006	Bioremediation
8	Microbial degradation	A study on hydrocarbon degradation by biosurfactant producing <i>Bacillus cereus</i> in oil contaminated soil samples.	Janaki et al., 2016	Bioremediation
9	Biotechnology	<i>Ex-situ</i> bioremediation of contaminated soils: from biopiles to slurry-phase bioreactors.	Kalogerakis, 2012	Bioremediation
10	Biotechnology	Application of <i>in situ</i> bioparging to remediate a petroleum-hydrocarbon spill site: field and microbial evaluation.	Kao et al., 2008	Bioremediation
11	Microbial degradation	Evaluation of bioremediation effectiveness on crude oil-contaminated sand.	Kim et al., 2005	Bioremediation
12	Bioslurping/ biotechnology	Remediation of petroleum hydrocarbon-contaminated sites by DNA diagnosis-based bio slurping technology.	Kim et al., 2014	Bioremediation
13	Microbial degradation	Bioremediation of petrol engine polluted soil using microbial consortium and wheat crops.	Kumar et al., 2017	Bioremediation
14	Fungi/enzyme degradation	Potential role of polycyclic PAHs oxidation by fungal laccase in the remediation of an aged, contaminated soil.	Wu et al., 2008	Bioremediation
15	Microbial degradation	<i>Ex situ</i> bioremediation of oil-contaminated soil.	Lin et al., 2010	Bioremediation
16	Microbial degradation	Isolation and characterisation of indigenous soil bacteria for the bioaugmentation of PAH contaminated soil in semiarid Patagonia, Argentina.	Madueño et al., 2011	Bioremediation
17	Phytoremediation	Germination of <i>Lepidium sativum</i> as a method to evaluate (PAHs) removal from contaminated soil.	Maila and Cloete, 2002	Bioremediation
18	Microbial degradation	<i>Lysin bacillus sphaericus</i> and <i>geobacillus sp</i> biodegradation of petroleum hydrocarbons and biosurfactant production.	Manchola and Dussán, 2014	Bioremediation
19	Cold-adapted microbes	Field scale <i>ex-situ</i> bioremediation of petroleum contaminated soil under cold climate conditions	Gomez and Sartaj, 2013	Bioremediation
20	Microbial degradation	Crude oil degradation efficiency of a recombinant <i>Acinetobacter baumannii</i> strain and its survival in crude oil-contaminated soil microcosm.	Mishra et al., 2004	Bioremediation
21	Bioventing/biotechnology	Large-scale bioventing degradation rates of petroleum hydrocarbons and determination of scale-up factors	Mosco and Zytner, 2017	Bioremediation
22	Microbial degradation	Enhanced <i>ex situ</i> bioremediation of crude oil contaminated beach sand by supplementation with nutrients and rhamnolipids.	Nikolopoulou et al., 2013	Bioremediation
23	Biotechnology	An integrated environmental biotechnology for enhanced bioremediation of crude oil contaminated agricultural land.	Onwurah, 2003	Bioremediation
24	Microbial degradation	Biodegradation of petroleum hydrocarbons by Actinobacteria and Acinetobacteria strains producing biosurfactant.	Pidgorskyi and Nogina, 2016	Bioremediation
25	Microbial degradation	Isolation and characterisation of naphthalene-degrading bacteria from sediments of the Cadiz area (SW Spain).	Nair et al., 2008	Bioremediation
26	Microbial degradation	Exploring the potentials of <i>Nypa palm</i> (<i>Nypa fruticans</i>) ash and rabbit droppings for enhanced <i>ex situ</i> bioremediation of crude oil contaminated soil.	Solomon et al., 2018	Bioremediation
27	Composting/phytoremediation	Inherent Bacterial Diversity and Enhanced Bioremediation of an Aged Crude Oil-contaminated Soil in Yorla, Ogoni Land Using Composted Plant Biomass	Solomon et al., 2018	Bioremediation
28	Biostimulation and bioaugmentation	Bioremediation of 6% [w/w] Diesel-Contaminated Mainland Soil in Singapore: Comparison of Different Biostimulation and Bioaugmentation Treatments.	Mathew et al., 2006	Bioremediation
29	Microbial degradation	Proteolytic activity of some cold-tolerant bacteria from arctic sediments	McDonald et al., 1963	Bioremediation
30	Cold-adapted microbes	Cold-adapted bacteria for the bioremediation of crude oil-contaminated soil.	Wang et al., 2016	Bioremediation
31	Cold adapted microbes	Cold Region Bioremediation of Hydrocarbon Contaminated Soils: Do We Know Enough?	McDonald and Knox, 2014	Bioremediation
32	Biotechnology	Two-phase partitioning bioreactors in environmental biotechnology.	Quijano et al., 2009	Bioremediation
33	Biotechnology	Petroleum-contaminated soil remediation in a new solid phase bioreactor.	Rizzo et al., 2010	Bioremediation
34	Vermiremediation	Earthworm Ecology: From Darwin to Vermiculture.	Pearce and Satchell, 1984	Bioremediation
35	Vermiremediation	Vermiremediation strategy for remediation of Kuwaiti oil contaminated soil	Almutairi, 2019	Bioremediation
36	Phytoremediation	Effectiveness of phytoremediation as a secondary treatment of (PAHs) in composted soil.	Parrish et al., 2010	Bioremediation
37	Enzymes	Enzymes as useful tools for environmental purposes	Rao et al., 2014	Bioremediation
38	Enzymes	Enhancing Bioremediation with Enzymatic Processes: A Review.	Ruggaber and Talley, 2006	Bioremediation
39	Chemical oxidation	Remediation of fuel oil contaminated soils by activated Persulphate in the presence of MnO_2 .	Mazloomi et al., 2016	Chemical remediation
40	Chemical augmentation	Chemical augmentation for the remediation of a hydrocarbon-contaminated soil.	Osuji and Raji, 2007	Chemical remediation
41	Chemical oxidation		Tsai and Kao, 2009	Chemical remediation

(continued on next page)

Table 1 (continued)

S/ no	Study type	Title	Refs.	Remediation method
42	Chemical oxidation	Treatment of petroleum-hydrocarbon contaminated soils using hydrogen peroxide oxidation catalysed by waste basic oxygen furnace slag.	Wang et al., 2015	Chemical remediation
43	Chemical augmentation	Hydrogen peroxide (H ₂ O ₂) requirements for the oxidation of crude oil in contaminated soils by a modified Fenton's reagent.	Kim et al., 2017	Chemical remediation
44	Chemical oxidation	Effect of chemical amendments on remediation of potentially toxic trace elements (PTEs) and soil quality improvement	Yen et al., 2011	Chemical remediation
45	Chemical oxidation	Application of persulfate to remediate petroleum hydrocarbon-contaminated soil: feasibility and comparison with common oxidants.	Ferrarese et al., 2008	Chemical remediation
46	Chemical oxidation	Remediation of PAHs-contaminated sediments by chemical oxidation.	Alderman et al., 2007	Chemical remediation
47	Direct current treatments	Effective treatment of PAHs contaminated superfund site soil with the peroxy-acid process.	Streche et al., 2013	Physical remediation
48	Thermal desorption	The treatment of diesel contaminated soils by dccts (direct current treatments) methods.	Araruna et al., 2004	Physical remediation
49	Thermal desorption	Oil spills debris clean-up by thermal desorption.	Musteraït, 2001	Physical remediation
50	Thermal desorption	Removal, handling, and thermal desorption treatment techniques for manufactured gas plant wastes.	Smith et al., 2001	Physical remediation
51	Electrokinesis	Thermal desorption treatment of contaminated soils in a novel batch thermal reactor.	Chung and Kamon, 2005	Physical remediation
52	Microwave heating	Ultrasonically enhanced electrokinetic remediation for the removal of Pb and phenanthrene in contaminated soils.	Chien, 2012	Physical remediation
53	Enhanced vapor extraction	Field study of <i>in situ</i> remediation of petroleum hydrocarbon contaminated soil on site using microwave energy.	Harmon et al., 2001	Physical remediation
54	Biostimulation, bioaugmentation, fungi remediation	Thermally enhanced vapour extraction for removing PAHs from lampblack contaminated soil.	Gao et al., 2014	Combined
55	Bioremediation, chemical, physical remediation	Effect of different remediation treatment on crude oil-contaminated saline soil.	Agarry, 2017	Combined
56	Bioremediation, physical remediation	Enhanced <i>ex-situ</i> bioremediation of soil contaminated with petroleum refinery waste effluent by stimulation through electro kinetics and organic fertilizer.	Abreu et al., 2009	Combined
57	Bioremediation, chemical, physical remediation	Simulation of soil vapour intrusion attenuation factors including biodegradation for petroleum hydrocarbons.	Almutairi, 2018	Combined
58	Bioremediation, chemical, physical remediation	An assessment of remediation strategies for Kuwaiti oil lakes.	Chien et al., 2011	Combined
59	Bioremediation, chemical, physical remediation	Development of a four-phase remedial scheme to clean up petroleum hydrocarbon-contaminated soils.	Coria et al., 2009	Combined
60	Bioremediation, biotechnology, chemical remediation	Hydrocarbon contaminated soil: geophysical-chemical methods for designing remediation.	Effendi and Aminati, 2019	Combined
61	Bioremediation, chemical, physical remediation	Enhancing bioremediation of crude oil-contaminated soil by combining with a photocatalytic process using TiO ₂ as a catalyst.	Robinson and Angyal, 2008	Combined
62	Bioremediation, chemical remediation	Use of mixed technologies to remediate chlorinated DNAPL at a brownfield site.	Tsai and Kao, 2009	Combined
63	Bioremediation, physical remediation	Treatment of fuel-oil contaminated soils by biodegradable surfactant washing followed by Fenton-like oxidation.	Kim et al., 2010	Combined
64	Bioremediation, physical remediation	Effect of electro kinetic remediation on indigenous microbial activity and community within diesel- contaminated soil.	Gomes et al., 2012	Combined
65	Biotechnology, physical remediation	Electrokinetic remediation of organochlorines in soil: enhancement techniques and integration with other remediation technologies.	Iturbe et al., 2004	Combined
66	Nanotechnologies	Remediation of contaminated soils using soil washing and bio pile methodologies at a field level.	Medina-Pérez et al., 2019	Combined
67	Bioremediation, Physical remediation	Remediating polluted soils using nanotechnologies: environmental benefits and risks.	Haapea and Tuhkanen, 2006	Combined
68	Bioremediation, chemical, physical remediation	Integrated treatment of PAHs contaminated soil by soil washing, ozonation and biological treatment	Huang et al., 2000	Combined
69	Bioremediation, Chemical remediation	Combining remediation techniques increases kinetics for the removal of persistent organic contaminants from soil.	Valderrama et al., 2009	Combined
70	Bioremediation, Physical remediation	Oxidation by Fenton's reagent combined with biological treatment applied to creosote- contaminated soil	Reddy et al., 2006	Combined
71	Vermiremediation	Enhanced electrokinetic remediation of a contaminated manufactured gas plant (MGP) soil.	Wegwu and Onyeike, 2006	Bioremediation
		Growth performance and proximate profile of <i>Telfairia occidentalis</i> Hook F. (cucurbitaceae) grown in crude oil-contaminated soil.		

ability to grow in contaminated soils, and extract contaminants from polluted soils. While some plants can accumulate toxic substances in their tissues, others can transfer them to less harmful substances. Phytoremediation also adds aesthetic beauty to the environment.

Combined method involves various combination of biological, chemical, and physical methods used in conjunction with one another to remove contaminants from environmental medium. Most single remediation methods are limited and cannot be used for all contaminants type. Combined remediation method integrates the strength of two or more methods, thereby limiting their individual weaknesses. Examples

include physical-chemical, physical-biological, chemical-biological, and physical-chemical-biological methods (Gomes et al., 2012). In the physical-chemical combined method, contaminants are removed from the environment by separating, fixing, or exchanging existing state of contaminant. Remediation is fast and simple to operate, however the technique has negative impact on the environment (Zhou et al., 2012). While the biological method in chemical-biological, and physical-biological combined methods may serve as polishing agents after the application of physical or chemical method. In addition, combining physical-chemical-biological methods have the advantage of

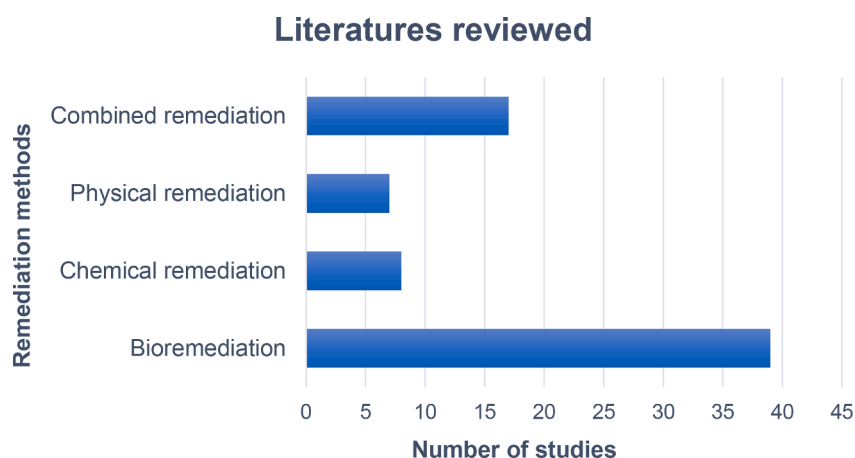


Fig. 3. Literature representation.

Table 2
Major inland oil spills.

S/ No	Oil Spills	Cause of Spill	Quantity of oil Spilled	Date of Spill	Clean-up Method Applied	Refs.
1.	Lake view gusher in California, USA	Eruption of pressurized oil well	2million	1910-1911	Sand embankment around the well	Read, 2011
2.	Kuwait desert pollution	Arabian Gulf war	4million	1991	Bioremediation	Radwan et al., 1995
3.	Mingbulak oil spill, Uzbekistan	Well blow-out	285,000	1992	Natural Attenuation	Tula and Tokhtakhunova, 2020
4.	Ogoni oil spill, Niger Delta, Nigeria	Pipeline leakage, equipment failure, and sabotage	10.5million	1976-1991	Bioremediation Ongoing	Ya'u et al., 2019 (International conference on humanities)
5.	Lac Megantic spill Quebec, Canada	Freight train accident	5,416	2013	Bioremediation	De Santiago-Martin et al., 2015
6.	South Dakorta oil spill Amherst, South Dakorta USA	Pipeline leakage	281	2017	-	Belvederesi et al., 2018
7.	North Dakorta oil spill, USA	Pipeline leakage	1,157	2013	-	Belvederesi et al., 2018
8.	Little Buffalo oil spill, Alberta Canada	Pipeline leakage	1,572	2011	-	Datta and Hurlbert, 2019
9.	Prudhoe Bay oil spill Alaska, USA	Pipeline leakage	359	2006	Combined	Gunsch et al., 2017
10.	Cheshire oil spill, CT USA	Rollover truck accident	2,012	2022	-	Okeke et al., 2022
11.	Colonial pipeline spill, North Carolina USA	Pipeline leakage	378,506	2020	Bioremediation	Belvederesi et al., 2018
12.	Isreali oil spill Be'er, Ora Isreal	Pipeline leakage	5,511.56	2014	Combined	Benson, 2014

fast remediation, and reduced impact on the environment.

2. Method

2.1. Literature search

A Systematic literature search is a vital component of systematic review. It involves a thorough search of literature that aims for a transparent study identification report, leaving readers informed about what was done to identify the studies and how the findings of the review are situated in the relevant evidence. A systematic search of literature was conducted in different database including Scopus, Wiley online library, Multidisciplinary Digital Publishing Institute (MDPI), to extract mainly peer reviewed literatures and a few grey literatures (conference papers and PhD thesis) published from 2000 to 2021. The search was restricted to studies in English language, this had an insignificant impact on the selection process as very few literatures were published in other languages. Key words were developed to identify research studies, initial key words were centered on the different soil remediation methods, clean-up of oil contaminated soils, and oil polluted soils, words were modified to suit each database. The search strategy was based on Boolean search keywords using an “AND” strategy, essentially this was to gain wider range of results. Meanwhile, the “OR” strategy was used to combine terms within each group for flexibility to bring the number of unrelated papers down (examples are soil or site, contaminants or

pollutants, remediation, or clean-up). Titles, abstracts, bibliographies, and subject headings were also searched, essentially to ensure relevant and vital literatures were collated. In addition, research studies of known soil remediation experts were searched, this provided valuable knowledge on the different methods employed in cleaning oil contaminated soils. Furthermore, references of selected papers helped in identifying other research studies that offered valuable data and information on soil remediation methods. However, some database did not yield vital result due to lack of access to publications and lack of adequate information.

Consequently, a total of 950 papers were initially selected. Duplicate copies were removed, and 935 papers were remaining. After removing papers on surface and ground water remediations a total of 505 papers were selected. Titles, abstract, aim and objectives of selected papers were used for screening and 135 papers were selected for this study. Selected papers were further screened to remove papers on soil remediation of heavy metals, industrial wastes, and dyes, and 85 papers were selected. In line with the aim of this review, selected papers were also assessed for eligibility based on their methodological quality. Papers that were considered for in-depth analysis consisted of those that studied field application of remediation method for oil contaminated soil with well-structured and consistent methodology that allowed replication of the research results. Finally, 51 papers were chosen for this study, Fig. 5 is a flow diagram of the literature search.

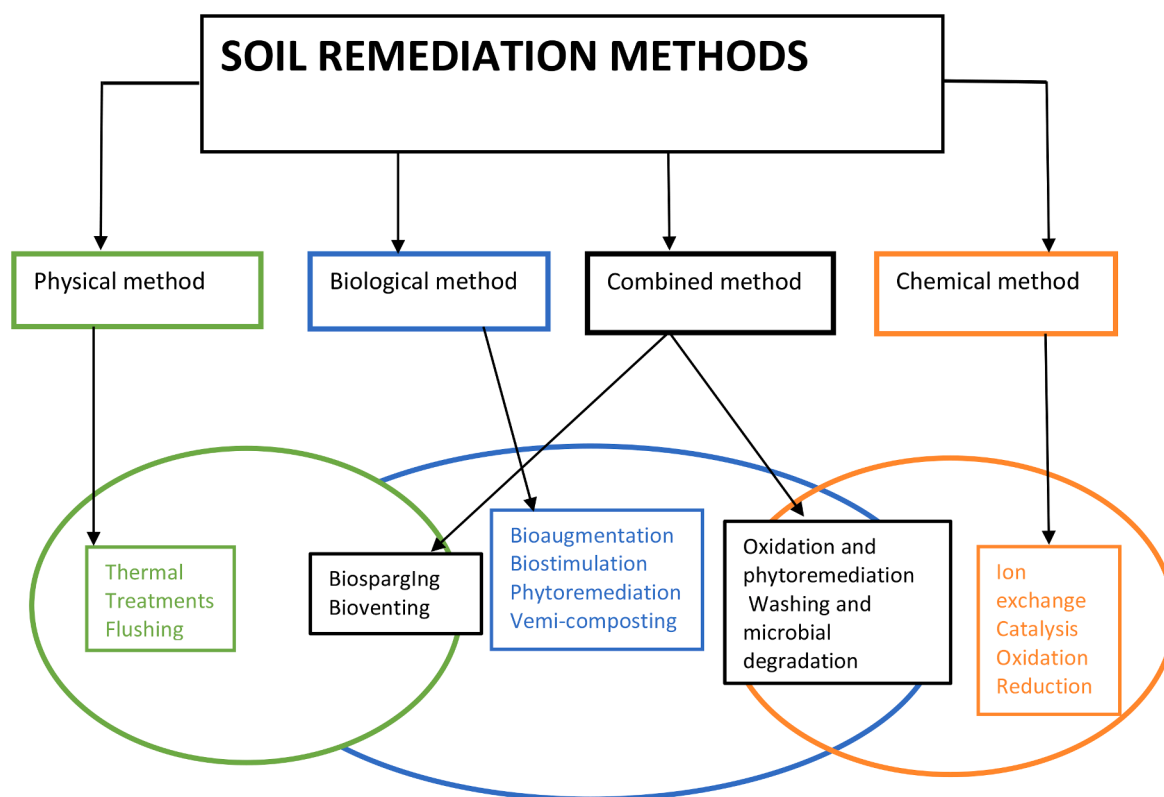


Fig. 4. Remediation methods.

2.2. Data analysis

This review adopted comparative analysis approach for data analysis (Ragin, 1981; Wagemann and Schneider, 2010). Soil remediation methods were compared to determine the most effective method for remediation of crude oil contaminated soils. The cost of a remediation method, clean-up duration, simplicity of remediation technology, and environmental compatibility of method were used to compare the effectiveness of different remediation methods. Also, advantages and disadvantages of different remediation methods were used for comparison.

3. Result

3.1. Results of inclusion and exclusion

The database search yielded 950 publications, most of the studies were retrieved from Wiley online library. After removing duplicate copies and papers on surface and ground water publications, 505 papers were selected for this study. Titles and abstracts of selected papers were used for screening and 135 papers were selected. In addition, full text of selected papers was read to assess their eligibility. 85 papers passed the eligibility assessment, while 34 papers on soil remediation of heavy metals, industrial wastes, and dyes were excluded. Finally, study methods and designs of selected papers were further used for assessment and 51 literatures were selected for this review. Studies selected include field studies, pilot studies, green house studies and grey publication (conference paper).

All the papers selected were published in English language from 2000 to 2021. 29 papers are studies on biological methods, 10 papers investigated the effectiveness of combining different soil remediation methods in oil spill removal, while 7 papers are on chemical remediation methods, and the remaining 5 papers studied the effectiveness of physical methods in oil spill remediation. Majority (37) of selected

literatures are field studies, 11 are pilot studies, while 2 papers are greenhouse experiments, the remaining 1 paper is a conference paper. Table 3 shows field application studies selected for this review, while Fig. 6 is a representation of literatures for each remediation method.

4. Selected literatures

This systematic review identified 51 field publications on soil remediation methods used in removing oil contaminants in the soil, and categorized them into biological, chemical, physical, combined methods for better understanding.

4.1. Biological methods

Biological method also known as bioremediation is the use of biological systems such as plants and microorganisms to destroy or reduce contaminants and concentration of toxic substances in polluted environment (Abioye et al., 2011). The aim of bioremediation is to reduce organic and inorganic contaminants to undetectable concentrations below the limits set by regulatory agencies as safe or appropriate using plants and microorganisms. Presently, a range of bioremediation methods exist, new and innovative solutions have also been developed for oil spill remediation. Many are *in situ* while others are *ex situ* remediation methods (Samanta et al., 2002). Numerous microorganisms, prokaryotes and eukaryotes are capable of degrading hydrocarbons in the soil. The degradation ability of microorganisms has been extensively studied by several authors, and the most studied microorganisms are bacteria, their numerous strains, and their capacity to detoxify contaminated soils. The most frequently identified active members of bioremediation consortia include these genera: Acinetobacter, Actinobacter, Alcaligenes, Arthrobacter, Bacillus, Berjerinckia, Flavobacterium, Methylosinus, Mycobacterium, Mycococcus, Nitrosomonas, Nocardia, Penicillium, Phanerochaete, Pseudomonas, Rhizoctonia, Serratia, Trametes and Xanthobacter (Samanta et al., 2002;

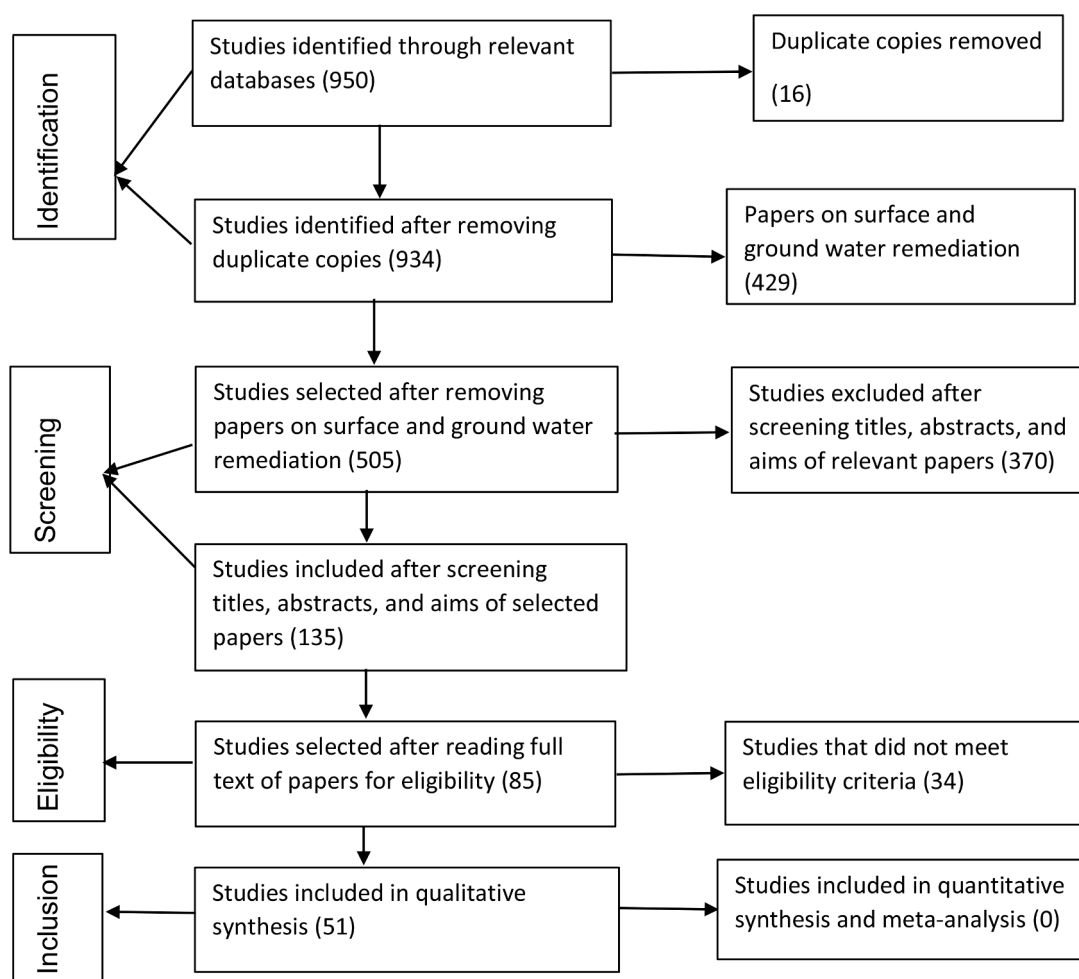


Fig. 5. Flow diagram of the literature search.

Kao et al., 2008; Chen et al., 2014; Madueno et al., 2014; Cubitto and Gentili, 2015; Janaki et al., 2016).

Most studies on bioremediation are laboratory studies, field applications of bioremediation are limited. However, this study identified some field applications of bioremediation in oil spill cleanup, ranging from simple to complex technologies, including the combined use of two or more bioremediation techniques. Gargouri et al. (2013) and Bello-Akintoshio et al. (2017) conducted field experiments to investigate the ability of various bacteria consortium in degrading hydrocarbon contamination in the soil and reported that bacteria showed significant hydrocarbon removal efficiency. Most microbial degradation of oil in the soil are largely controlled by the rate of nutrients and oxygen supplied. Several researchers have stimulated the degrading ability of indigenous microorganism through nutrients additions. They observed that the total petroleum hydrocarbon (TPH) concentration in the soil reduced when nutrients such as urea and fertilizers were added to the soil (Solomon et al., 2018; Remelli et al., 2020). Onifade and Abubakar (2007) simulated the activities of indigenous soil microorganisms and their results revealed that the total hydrocarbon concentrations in the soil reduced. While Okparanma et al. (2017) applied enhanced natural attenuation for oil spill degradation. In their study, fertilizers and windrows were used to supplement the nutrients and oxygen requirements of hydrocarbon degrading bacteria (HDB) to breakdown contaminants. Their results revealed that concentration of TPH in the soil was reduced to 94.3%, signifying the effectiveness of remediation by enhanced natural attenuation (RENA).

Furthermore, a full-scale biosparging and microbial study was conducted by Kao et al. (2008) at a petroleum contaminated site. They

evaluated the effectiveness of *in situ* biosparging in controlling BTEX at the spill site and determined dominant native microorganisms at the spill site through microbial identification. Results revealed that natural attenuation caused the initial decrease of BTEX concentration, however the application of biosparging caused further degradation of BTEX in the soil, and more than 70% of BTEX was removed from the soil after 10 months remediation time. Bioremediation has also shown to be a viable strategy for remediation of cold regions. Studies have identified some cold-tolerant bacteria, their adaptations and ability to degrade hydrocarbons in very cold regions (Snape, 2008; McDonald and Knox, 2014). In cold regions such as Antarctica, introducing non-native microbial amendments and nutrients violate Antarctica Treaty restrictions (Gomez and Sartaj, 2013). McWatters et al. (2016) successfully conducted bioremediation with biopile using indigenous bacteria without additional heating of the soil, the result showed that the biopile removed more than 60% of the contaminants, and the remediated soil was reused in a building foundation. Also, many scholars have reported hydrocarbon degrading ability of different fungi including mushrooms and yeasts. The strong potentials of fungi in bioremediation hinges on their aggressive growth, wide biomass output and long hyphae reach in the environment (Andersson et al., 2003; Potin et al., 2004; Wu et al., 2008, 2010). The strategy involved in fungal degradation of hydrocarbons is enzymatic transformation by intracellular cytochrome P450 enzymes and extracellular lignolytic enzymes comprising of Lignin peroxidase (LiP), Manganese Peroxidase (MnP) and Laccase. These three enzymes have been identified as very important for the remediation of hydrocarbon-contaminated soils (Gianfreda and Rao, 2004; Ruggaber and Talley, 2006). Wu et al. (2008), and Hestbjerg et al. (2003)

Table 3
Selected studies for systematic review.

Title	Refs.	Methods	Clean-up time	Study design	Clean-up efficiency	Operability	Cost	Impact on ecosystem	Remarks
1 Application of <i>in situ</i> bioparging to remediate a petroleum-hydrocarbon spill-site: Field and microbial evaluation	Kao et al., 2008	Biological	10 months	Field	70%	Simple	Low	None	Very effective
2 Potential role of polycyclic aromatic hydrocarbons (PAHs) by fungal laccase in the remediation of an aged-contaminated soil.	Wu et al., 2008	Biological	2-14 days	Field	80%	Complex	Low	None	Effective- high cost of enzyme
3 <i>Ex situ</i> bioremediation of oil-contaminated soil	Lin et al., 2010	Biological	28 days	Field	70%	Simple	Low	None	Effective
4 Removal handling and thermal desorption treatment technique for manufactured gas plant waste	Musteraït, 2001	Physical	-	Field	-	Simple	High	Negative	High energy cost, and impact on the environment
5 Rhamnolipids and nutrients boost remediation of crude oil-contaminated soil by enhancing bacterial colonisation and metabolic activities	Tahseen et al., 2016	Biological	30-60 days	Field	95%-97%	Simple	Low	None	Very effective
6 Chemical augmentation for the remediation of hydrocarbon contaminated site	Osuji and Raji, 2007	Biological	-	Field	100%	Simple	Low	None	Very effective
7 Use of mixed technology to remediate chlorinated DNAPL at a Brownfield site	Robinson and Angyal, 2008	Combined	-	Field	-	Complex	Moderate	Slight impact	Not very effective
8 Effect of remediation on growth parameters, grain, and dry matter yield of soybean (Glycine, max) in crude oil contaminated soils in Ogoni land, south-eastern Nigeria	Ayolagha and Peter, 2012	Biological	60days	Field	60%-79% (Growth rate)	Simple	Low	None	Effective
9 Zero-valent iron activated persulfate remediation of PAHs contaminated soil: An <i>in-situ</i> pilot study	Song et al., 2019	Chemical	104 days	Pilot-scale	62%-83%	Complex	High	Negative	Not very effective-complex technology
10 Field study of <i>in situ</i> remediation of petroleum-hydrocarbon contaminated soil on site using microwave energy	Chien, 2012	Physical	3.5h	Field	90%	Simple	High	Negative	High energy cost
11 Integrated treatment of PAHs contaminated soil by soil washing, ozonation and biological treatment	Haapea and Tuhkanen, 2006	Combined		Field	90%	Complex	Moderate	Slight impact	Not very effective-complex technology and high remediation cost
12 Remediation of petroleum hydrocarbon-contaminated sites by DNA diagnosis based bioslurping technology	Kim et al., 2014	Biological	2 years	Field	93%	Simple	Low	None	Very effective
13 Assessment of the effectiveness of on-site <i>ex-situ</i> remediation by enhanced natural	Okparanma et al., 2017	Biological	11 months	Field	94%	Simple	Low	None	Very effective

(continued on next page)

Table 3 (continued)

Title	Refs.	Methods	Clean-up time	Study design	Clean-up efficiency	Operability	Cost	Impact on ecosystem	Remarks	
14	attenuation in the Niger Delta region, Nigeria Effectiveness of phytoremediation as a secondary treatment of (PAHs) in composted soil	Parrish et al., 2010	Biological	12 months	Field	-	Simple	Low	None	Effective
15	Combining remediation techniques increases kinetics for removal of persistent organic contaminants from soil.	Huang et al., 2000	Combined		Field	95%	Simple	Moderate	Slight	Effective
16	Oxidation of Fenton's reagent combined with biological treatment applied to creosote-contaminated soil	Valderrama et al., 2009	Combined	-	Field	75%-80%	Complex	Moderate	Slight	Not very effective
17	Inherent bacterial diversity and enhanced bioremediation of an aged crude oil-contaminated soil in Yorla, Ogoni land using composted plant biomass	Solomon et al., 2018	Biological	70 days	Field	99%	Simple	Low	None	Very effective
18	Effective treatment of PAHs contaminated superfund site soil with the peroxy-acid process	Alderman et al., 2007	Chemical		Field	-	Complex	High	Negative	Not very effective
19	Biostimulation proved to be the most efficient method in the comparison of <i>in situ</i> soil remediation treatments after a simulated oil spill accident	Simpanen et al., 2016	Combined	16 months	Pilot-field study	89%-99%	Simple	Moderate	None	Effective
20	Enhanced electrokinetic remediation of contaminated manufactured gas plant (MGP) soil	Reddy et al., 2006	Combined	-	Field	-	Complex	Moderate	Negative	Not very effective
21	Oil spill in Lac-Megantic, Canada: Environmental monitoring and remediation.	De Santiago-Martin et al., 2015	Physical	4years	Field (conference paper)	90%	Simple	High	Slight impact	Effective
22	Crop production on heavily disturbed soils following crude oil remediation	Croat et al., 2020	Biological	3 years	Field	-	Simple	Low	None	Effective
23	A study evaluating the effectiveness of bioremediation of oil-contaminated Lena District, the Republic of Sakha (Yakutia), after an oil spill.	Lifshits et al., 2017	Biological	-	Field	87.1%	Moderate	Low	None	Effective
24	Bioremediation of Petroleum hydrocarbons-contaminated soil by bacterial consortium isolated from an industrial wastewater treatment plant.	Gargouri et al., 2013	Biological	30days	Field	63.4%	Simple	Low	None	Effective
25	Remediation of vehicle wash sediments contaminated with hydrocarbons: A field demonstration	Karthikeyan et al., 2011	Biological	3 yrs	Field	75%	Simple	Low	None	Effective
26	Theory and Application of Landfarming to Remediate Polycyclic	Harmsen et al., 2007	Biological (Landfarming)	10-15 yrs.	Field	100%	Simple	Low	None	Effective

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Table 3 (continued)

Title	Refs.	Methods	Clean-up time	Study design	Clean-up efficiency	Operability	Cost	Impact on ecosystem	Remarks
Aromatic Hydrocarbons and Mineral oil-contaminated sediments; Beneficial Reuse									
27 Determination of a suitable TPH remediation approach via manova and inferential statistics assessment	Okonofua et al., 2020	Combined (chemico-biological)	-	Field	90%	Moderate	Moderate	-	Effective
28 Microwave in soil remediation from VOCs to build up of a dedicated device	Acierno et al., 2003	Physical		Field	-	Complex	High	Negative	High cost of remediation
29 Seeding Growth Agronomic Crops in Crude Oil Contaminated Soil	Issoufi et al., 2006	Biological	28days	Green house	-	Simple	Low	None	Effective
30 Bioaugmentation of tar-contaminated soils under field conditions using <i>Pleurotus ostreatus</i> refuse from commercial mushroom production	Hestbjerg et al., 2003	Biological	3 weeks	Field	78%	Simple	Low	None	Very effective
31 Estimating remediation and contaminant respiration emissions for alternatives comparisons at petroleum spill sites	McAlexander and Tuggle, 2015	Combined	10yrs	Field	-	Complex	Moderate	-	Not very effective
32 Impact of ectomycorrhizal colonization of hybrid poplar on the remediation of diesel contaminated soil	Gunderson et al., 2007	Biological	12 weeks	Pilot study	354.1mg kg ⁻¹	Complex	Low	None	Not very effective
33 Enhanced land treatment of petroleum-contaminated soils using solid peroxygen materials	Davis et al., 2006	Chemical	6 weeks	Pilot study	50%-70%	Complex	10% cost reduction	None	Not very effective
34 Bioremediation of Benzene, toluene, ethylbenzene, xylene contaminated soil: a biopile pilot experiment	Genovese et al., 2008	Biological	15 days	Pilot study	90%	Moderate	Low	None	Very effective
35 Feasibility analysis of the remediation of fuel oil-contaminated soil and ground water around the railroad station of Ycity, Korea with surfactant aided soil flushing	Lee and Lee, 2012	Chemical	5 h	Field	53%	Complex	High	Negative	Not very effective
36 <i>Pseudomonas</i> sp. (strain 10-1B): A potential inoculum candidate for green sustainable remediation	Bello-Akinosho et al., 2017	Biological	10 weeks	Pilot study	-	Simple	Low	None	Effective
37 Growth performance and proximate profile of <i>Telfainia occidentalis</i> Hook F. (Cucurbitaceae) grown in crude oil-contaminated soil	Wegwu and Onyeike, 2006	Biological	6 weeks	Field	-	Simple	Low	None	Effective
38 Community composition of Arbuscular mycorrhizal	Garcés-Ruiz et al., 2018	Biological	-	Field	-	Simple	Low	None	Effective

(continued on next page)

Table 3 (continued)

Title	Refs.	Methods	Clean-up time	Study design	Clean-up efficiency	Operability	Cost	Impact on ecosystem	Remarks
fungi associated with native plants growing in a petroleum-polluted soil of the Amazon region of Ecuador									
39 Characterization and application of surfactant foams produced from ethanol-sodium lauryl sulfate-silica nanoparticle mixture for soil remediation	Chattopadhyay and Karthick, 2017	Chemical	-	Pilot study	94%	Complex	High	Negative	Not very effective
40 Petroleum-contaminated soil remediation in a new solid phase bioreactor	Rizzo et al., 2010	Biological	42 days	Pilot study	20-35%	Complex	Low	None	Not very effective
41 <i>In situ</i> radio-frequency heating for soil-remediation at a former service station	Huon et al., 2012	Physical (Thermal)	80%-time reduction	Field	1100kg	Complex	High	Negative	High cost of application
42 Advanced oxidation of polycyclic aromatic hydrocarbons in soils contaminated with diesel oil at pilot scale	Cavalcanti et al., 2021	Chemical	-	Pilot study	80%	Complex	High	Negative	Not effective
43 Plant growth, soil properties, and microbial community, four years after thermal desorption	Bartsch et al., 2021	Biological	4 years	Field	-	Moderate	Low	None	Effective
44 Wheat growth in soil treated by <i>ex situ</i> thermal desorption	O'Brien et al., 2017	Biological		Green house	-	Simple	Low	None	Effective
45 Bioremediation of 6% [w/w] diesel-contaminated mainland soil in Singapore: comparison of different bio stimulation, bioaugmentation treatment	Mathew et al., 2006	Biological	60 days	Field	48%-53%	Simple	Low	None	Effective
46 Bioremediation of oil-contaminated soils by stimulating indigenous microbes	Williams et al., 1998	Biological	56 weeks	Field	83%-84%	Simple	Low	None	Very effective
47 Remediation of soil contaminated by PAHs and TPH using alkaline activated persulfate enhanced by surfactant addition at flow conditions	Lominchar et al., 2017	Chemical	-	Pilot study	-	moderate	High	Negative	Not effective
48 Pilot plant investigation of thermal remediation of tar-contaminated gravel	Anthony and Wang, 2006	Combined	-	Pilot study	-	-	Moderate	Sight impact	Not effective
49 On site remediation of a fuel spill and soil reuse in Antarctica	McWatters et al., 2016	Combined	5 years	Field	907 ±22mg/kg	Complex	Moderate	None	Effective
50 Long-term dynamics of plant communities after biological remediation of oil-contaminated soils in far north	Novakovskiy et al., 2021	Biological	12 years	Field	55%-90%	Simple	Low	None	Very effective
51 Bioremediation of soil contaminated by hydrocarbons with the combination of three technologies: bioaugmentation, phytoremediation and vermiremediation.	Rodriguez-Campos et al., 2018	Biological	-	Field	86.4%	Simple	Low	None	Very effective

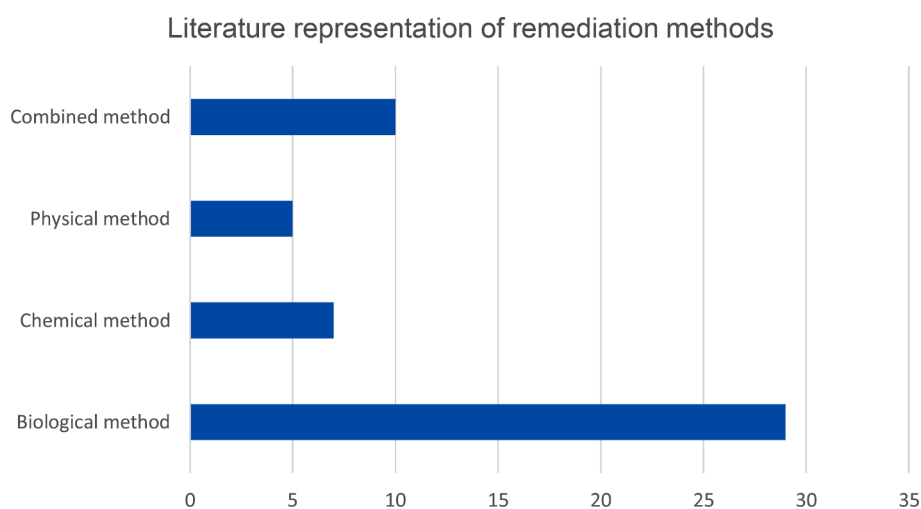


Fig. 6. Literature representation of remediation methods.

conducted field studies to investigate the activities of Laccase enzymes in degrading petroleum hydrocarbons. Their results showed that laccase enzymes can degrade hydrocarbons and have no effect on native bacteria in the soil. While [Garcés-Ruiz et al. \(2018\)](#) demonstrated the degrading potentials of Arbuscular mycorrhizal fungi (AMF) in roots of diverse plants species colonizing a weathered crude oil site in the Amazon of Ecuador. In their study, large numbers of new AMF species were identified and their potentials in phytoremediation was confirmed.

Most of the selected bioremediation papers for this study involves phytoremediation or the combined application of phytoremediation and other soil remediation methods. Phytoremediation is the use of plants in cleaning polluted environments, it is a cost-effective remediation method that employs different strategies such as phytoextraction, phytodegradation, phytostabilization, phytovolatilization, and rhizofiltration ([Singh and Singh, 2017](#)). [Issoufi et al. \(2006\)](#) conducted a cost-effective soil remediation study of hydrocarbon contaminated soil using 6 different seedlings. Their results suggests that phytoremediation is a cost-effective soil remediation method that can effectively clean up contaminated soils. While [Harmsen et al. \(2007\)](#) and [Lifshits et al. \(2017\)](#) combined the use of landfarming and microorganisms in oil spill clean-up. Results obtained showed successful remediation of the contaminated soils using this combined method. Also, studies have shown that phytoremediation can be used as an effective tool in determining the impact of a remediation techniques on the soil. [Ayolagha and Peter \(2012\)](#) studied the growth performance of different plants including *Telfainia occidentalis* and Soybean in crude oil polluted soils treated with organic nutrients (poultry manure) and inorganic fertilizers. Results obtained showed that addition of nutrients enhanced germination rate of the plants, promoted greenish coloration of leaves and increased levels of macronutrients (carbohydrates, proteins, and lipids). While [O'Brien et al. \(2018\)](#) used plant growth index to determine the impact of thermal desorption on soils. Their result revealed that thermal desorption did not decrease the soil microbial population, suggesting that the method does not have negative impact on the soil. However, [Croat et al. \(2020\)](#) compared plant growths of two soils; a thermal desorption treated soil and a non-thermal desorption soil. Their results revealed that crop production was lower in soil treated with thermal desorption compared to the non-treated soil, suggesting that soil organic carbon was likely responsible for yield decline.

Furthermore, Vermiremediation or Vermi-composting is one of the most promising bioremediation technologies. It uses earthworms to remove contaminations from soil. The idea of using earthworms in vermicomposting has been known for centuries, but their application in bioremediation was incidentally discovered following the 1976 explosion of the Seveso chemical plant in Italy, when a vast area was

contaminated with extremely toxic chemicals such as 2, 3, 7, 8-tetrachlorodibenzo-p-dioxin (TCDD). Several faunas perished except for certain surviving species of earthworms which ingested TCDD-contaminated soils and were shown to bioaccumulate dioxin concentrates in their tissues ([Satchell et al., 1984](#)). Studies have shown that vermiremediation is effective in decontaminating hydrocarbon contaminated soils. [Rodriguez-Campos et al. \(2018\)](#) conducted bioremediation study of hydrocarbon contaminated soil using vermiremediation, phytoremediation, and bioaugmentation individually and in combinations. Results obtained after 112 days showed that the 3 techniques were able to degrade hydrocarbon in the soil. However, the highest TPH removal was with earthworms and bacteria (86.4%), followed by earthworms plus plants plus bacteria (82.7%), and bacteria (82.6%). Suggesting that the soil microorganisms are necessary for earthworms' survival and colonization. Although there are many field application studies on bioremediation as shown in this review, most biological techniques are effective when combined or with other methods.

4.2. Chemical methods

Chemical remediation involves the use of chemicals to remove pollutants from the environment, it offers rapid and aggressive alternative treatment to overcome some limitations of bioremediation. Some chemical remediation techniques reviewed in this study include, surfactant washing, chemical oxidation, and nanoparticle foams. Soil washing/flushing is a mechanical process used to extract chemical contaminants from soils using liquids (water, water + additives), typically aqueous solutions ([Mulligan et al., 2001](#)). Pollutants bind to soil particles and have low solubility in water, making them difficult to remove with water, therefore additives like acids, surfactants and chelating agents are often added to degrade contaminated soils. Various scholarly reports reveal that surfactants can be used to enhance the removal of petroleum hydrocarbons from polluted soils by increasing the solubility of petroleum components or lowering the interfacial tension to enhance mobility of the hydrocarbons ([Abdul et al., 1992](#); [Joshi and Lee, 1996](#); [Fountain et al., 1996](#); [Finzgar and Lestan, 2008](#)). [Joshi and Lee \(1996\)](#) believe the pre-treatment of hydrocarbon contaminated soils with surfactant washing (Igepal CA-720) will solubilise PAHs and enhance biodegradation of the contaminants. In [Lee and Lee \(2012\)](#) and [Lominchar et al. \(2017\)](#) surfactant washing, and chemical oxidation were combined in remediation process of hydrocarbon contaminated soils. [Lee and Lee \(2012\)](#) carried out the clean-up of a diesel contaminated railway station in Korea using combined techniques of surfactant aided soil washing and hydrogen peroxide. Results showed TPH removal

of 30% and 18% respectively using hydrogen peroxide and tween 80 after 1h, and 53% and 41% after 5h. Indicating that time is an important factor in soil flushing. While Lominchar et al. (2017) studied the use of alkaline activated persulfate and surfactant washing in cleaning oil polluted soil. Results revealed that applying surfactant washing and persulfate to the soil completely removed the contaminants in the soil, suggesting that surfactants and alkaline persulfate produces better remediation results.

Chemical oxidation technique involves the application of chemical oxidants to transform contaminants to a more soluble substance for further degradation. The common oxidants applied in soil remediation are ozone, hydrogen peroxide, permanganate, and persulfate. Hydrogen peroxide (H_2O_2) oxidation has been identified as an efficient technology for the treatment of hydrocarbon-contaminated soils. The two types of iron-catalysed Hydrogen peroxide oxidation processes include the Fenton oxidation which uses soluble iron such as ferrous ion (Fe^{2+}), and the Fenton-like process which employs the use of iron oxyhydroxide such as goethite. Cavalcanti et al. (2021) investigated the remediation of diesel contaminated soil using advanced Fenton's oxidation method. Results revealed 80% PAHs removal efficiency, the toxicity test conducted using earthworms as bioindicators further proved the mineralization of PAHs from the soil. According to Chuang et al. (2009), a significant portion of soil contaminants are oxidised by hydrogen peroxide without the addition of soluble iron. In Alderman et al. (2007), Peroxy acid process was used to degrade PAHs contaminants at a superfund site. Hydrogen peroxide, acetic acid, and deionised (DI) water solution were applied to 5g of superfund soil at a ratio of 3:5:7 or 3:3:9. The experiment with 3:5:7 ratio resulted in almost complete degradation of 14 EPA regulated PAHs, while 3:5:9 ratio did not show any degradation. After a scaleup experiment, results showed significant degradation of PAHs with the 3:5:9 ratio. Also, Davis et al. (2006) evaluated the potentials of peroxygen materials to enhance TPH removal from soil and reduce treatment time. Their results revealed that peroxygen compounds effectively reduced TPH concentrations in the soil to 70% and reduced clean-up time. Recently, a body of scholars suggest that the injection of hydrogen peroxide may facilitate a cost-effective remediation design for sensitive and impermeable soils including treatment of petroleum contaminated soils (Baciocchi et al., 2003; Ferguson et al., 2004; Mecozzi et al., 2006).

Over the past two decade, the use of nanoparticle in soil remediation have received substantial attention by many scholars due to their physical and chemical properties (Zhang, 2003; Liu, 2006). Several types of nanoparticles have been studied including Iron nanoparticles ($FeNP$) and Silica Oxide nanoparticles ($SiONP$). Karthick et al. (2019) applied silica nanoparticle stabilized anionic surfactant foams for remediation of diesel contaminated soil, the nano-foam showed maximum oil removal efficiency of 94.5%. Also, Zero-Valent iron persulfate (ZVI/PS) is widely applied in decontamination of pollutants. The development of the Zero-Valent Iron Persulfate (ZVI/PS) has provided new ways of resolving the problem of quenching SO_4 by Fe^{2+} . Song et al. (2019) conducted field experiment to evaluate the effectiveness of different zero-valent iron (ZVI) to activate persulfate in cleaning PAHs-contaminated soils and their impact on the soil microbial community. Three reaction pits were excavated and used to test the activation ability of three zero-valent irons. After 104days, the PAHs removal efficiency of persulfate was recorded for different zero-valent iron activated persulfate. The results showed an 82.21% removal for n ZVI activated persulfate, 62.78% for c-n ZVI activated persulfate and 69.14% for mZVI activated persulfate. The soil's microbial community decreased after remediation and the soil PH also decreased due to release of hydrogen ion (H^+) during persulfate degradation. Although this technique has shown ability to remove high concentrations of hydrocarbons from soil, it is limited by its resultant salinization of soil caused by residual sulfate after remediation which may result in decrease of soil microorganisms. In general, chemical methods are limited due to their negative impact on the environment and high cost of

application.

4.3. Physical methods

Physical remediation involves the physical isolation of contaminants from environmental media such as soil. Physical treatment technologies carry out remediation by taking advantage of both the physicochemical properties of the pollutants (density, solubility, viscosity, volatility) and the environmental media (bulk density, moisture content, permeability, porosity, particle size, heat, and electric current conductivity). Examples include product recovery, encapsulation, soil capping, pump and treat, soil vapor extraction, airsparging, thermal desorption and microwave heating. This study reviewed five field studies of physical methods including thermal desorption and microwave heating. Studies have shown that physical remediation can successfully be applied in cleaning crude oil contaminated soils (Acierno et al., 2003). The Lac-Megantic oil spill that occurred in Canada was cleaned by physical remediation method. According to De Santiago-Martin et al. (2015), recovery wells, and absorbent socks were used to contain crude oil from leaching into the groundwater, and 90% remediation efficiency was recorded. While Huon et al. (2012) reported the successful remediation of a former petrol station in Kent, United Kingdom using *in-situ* radio frequency heating (ISRFH) and soil vapor extraction. Results revealed that about 1100kg of hydrocarbons was removed from the soil, and there was 80% reduction in remediation time, while keeping total energy consumption almost constant.

Microwave heating is effective, simple, and robust to use, and can be used to treat different types of soil pollution. The use of microwave energy in cleaning petroleum-contaminated soil was demonstrated in a field-scale study by Chien (2012). A constant microwave power of 2KW was applied to the hydrocarbon-contaminated soil for 3.5 h without water input. The results showed very efficient remediation of PAHs using the microwave heating technique. While Acierno et al. (2003) designed a microwave opened applicator useful to perform microwave steam distillation soil remediation process. Experimental research conducted *in situ* on soils contaminated with VOCs using this technique confirmed the heating-degradation process already observed in the closed cavity experiments, and the feasibility of microwave technique in field remediation. Furthermore, Anthony and Wang (2006) conducted a pilot scale study to investigate the potentials of using incineration in a fluidized bed combustor in remediation of oil contaminated soil. However, the major limitations of physical method include high cost of application and the need for further treatment of the gases produced during remediation.

4.4. Combined methods

Combined remediation describes the concurrent use of multiple remediation technologies to enhance the overall performance of the environment. Through exploiting the capabilities of each technology, remediation practitioners may incorporate biological, chemical, and physical treatments in cleaning a wider variety of soil conditions and contain concentrations. Most studies have investigated the effectiveness of combining two or more remediation methods in overcoming the limitations of different remediation methods. Robinson and Angyal (2008) studied the application of different remediation methods in cleaning a chlorinated organic compounds and petroleum-hydrocarbon impacted site at a former chlorofluorocarbon manufacturing facility under the New Jersey department of environmental protection's brownfield redevelopment initiative. The remediation process involved an initial excavation and on-site treatment of more than 95% of contaminated soil mass by adding calcium oxide and lime kiln dust to the contaminated soil. Treated soil was reused onsite, while residual soil and groundwater contamination was treated *in situ* by adding emulsified oil to induce anaerobic biodegradation and emulsified oil/zero-valent iron to reduce residual contaminants chemically, and finally

engineering caps and administrative controls (deed restrictions) were also applied. The results showed a cost reduction of 50%, suggesting that integrated technology is an effective soil remediation method. While Huang et al. (2000), and Haapea and Tuhkanen (2006) reported that combining different remediation methods can increase the kinetics for removal of organic pollutants from the soil. They combined soil washing, ozonation and biological treatments in cleaning an aged oil-contaminated PAHs soil. Three different doses of ozonation and soil washing at different PH were studied to assess their effects on PAHs degradation. The results of their experiment revealed a 90% reduction in PAHs concentration using combined soil washing, ozonation and biological treatment techniques.

In addition, Okonofua et al. (2020) conducted an experiment using landfarming, chemo-biological, and physical methods to remove hydrocarbon contaminants in the soil. Results obtained showed over 90% TPH and PAHs removal, and landfarming recorded the highest TPH and PAHs removal. While Simpanen et al. (2016) combined chemical oxidation and bioremediation in an *in-situ* remediation. Biostimulation proved to be the most efficient method with 68.2% TPH removal. Similarly, Valderrama et al. (2009) conducted three series of experiments to determine the optimal dose of reagents in the Fenton's reaction. The study reported a maximum 80% PAHs degradation from the Fenton treatment with a reagent ratio of 90:1 for H₂O₂: Fe. Additional iron was included to enhance the removal efficiency of PAHs, but there was no change in the rate of degradation, showing that the efficiency of PAHs chemical oxidation will depend on the physico-chemical properties of different PAHs and the characteristics of the soil. Smaller PAHs with three rings were more susceptible to chemical oxidation than those with a high molecular weight (4-5 rings PAHs). Also, biological treatment of the pretreated soil resulted in 45% degradation of PAHs in the soil, indicating that maximum efficiency of degradation was achieved by the Fenton's reaction combined with chemical and biological treatments.

4.5. Comparative analysis of remediation methods

The first evaluation criterion used in this comparison is cost of remediation methods, cost data of environmental remediation is limited and varies from project to project depending on the environmental media (air, water, soil), the physicochemical properties of the medium, the type and concentration of contaminants. Remediation costs include capital and operational costs, however operational costs are the most significant costs in each soil remediation method. In this review, the costs of clean-up published by Dadrasnia and Agamuthu (2014) presented in Table 4 was used as standard to compare and rank remediation costs. Result of the cost comparison revealed that biological method has very low remediation cost, ranging from \$5 to \$266 per tons soil.

Other criteria used for comparison include cleanup efficiency, and cleanup time. Cleanup efficiency compares the quantity of contaminant a method can degrade from the soil and provides the level of degradation of hydrocarbons by each method, the higher the clean-up efficiency, the higher the effectiveness of a method. Also, methods were compared using the time required by a method to decontaminate polluted soils, cleanup time depends on the method selected and the concentration of contaminants released to the environment. The result of comparative analysis of remediation methods using cost, cleanup efficiency, and cleanup time, as shown in Fig. 7 reveals that combined remediation is a

Table 4
Summary of remediation costs (Dadrasnia and Agamuthu, 2014).

Remediation method	Cost (US\$/tons soil)	Ranking
Biological	5 - 266	Low - 5
Chemical	19 - 940	High - 2
Physical	50 - 330	Moderate - 3
Integrated	30 - 500	Moderate - 3

very effective soil remediation method followed by bioremediation.

In addition, the simplicity of a method can be used to determine effectiveness of a remediation method, complex technologies are difficult to operate compared to simpler technologies. Most bioremediation technologies are easy to operate compared to chemical and physical methods that may require skills and training. Also, environmental impact of each remediation method was considered and used for this comparison. Chemical and physical methods have negative environmental impact, most chemicals used for soil remediation are toxic to the soil microbes. Furthermore, the advantages and disadvantages of different remediation methods were considered and presented in Table 5.

5. Discussion

There are limited field application studies on soil remediation, most of the existing field studies are bioremediation studies followed by combined remediation studies, very few studies on the field application of chemical and physical methods were identified and selected for this review, as shown in Fig. 6. Generally, the review reveals that combined method is a more effective soil remediation method followed by bioremediation. Result of the comparative analysis in Fig. 7 shows that combined method has very high cleanup efficiency, short cleanup duration, and moderate remediation cost that may be as low as \$30 depending on the technology (Dadrasnia and Agamuthu, 2014). While low remediation cost and less environmental impact are the major strengths of bioremediation, however the inability of bioremediation to detoxify high concentrations of toxic substances within a short time is a major limitation of the method. Chemical remediation was reported as the least effective soil remediation method followed by physical method. The high cost of chemical application and negative effects of chemicals on soil are the major limitations of chemical method. While the major limitation of physical method hinges on high energy consumption of some technologies under physical method.

Most soil remediation methods are applied as single technologies only, despite their various successes, they have several limitations which include high cost of remediation, long clean up duration, and negative environmental impact. In addition, some methods have shown high clean-up efficiency in toxic and low concentration of contaminants but would show low clean-up efficiency in large and field scale implementations. However, these limitations can be overcome by combining two or more remediation methods (Huang et al., 2000). Correlating to this, many researchers have reported that integrating two or more remediation methods is a more effective soil remediation method (Kim et al., 2010). Combined remediation method is a tool used to improve soil efficiency by combining any or all the soil remediation methods to harness their advantages while reducing their limitations. According to literatures, Physical remediation methods are suitable in removing high concentrations and toxic contaminants. Chemical method is used to remove high concentrations of toxic contaminants and to treat the contaminant source, while biological method is suitable for degrading low concentration of contaminants. Typically, physical, and chemical methods remove pollutants faster from the soil than biological method, especially high contaminant concentrations.

There are several options for combining remediation methods, the options chosen will depend on the contaminant type, the concentration of pollutants and the pollutant matrix. Physical, and chemical methods can be combined to decontaminate high concentrations and toxic contaminants (Reddy et al., 2006). While biological method is usually employed as a polishing stage in combined remediation and can be used after physical or chemical remediation, especially when the concentration of contaminants has been reduced. Five of the papers on combined remediation discussed in this study integrated biological methods in their treatment train, as their enhancement is often a suitable approach in soil remediation to restore the soil flora and fauna. Haapea and Tuhkanen (2006) studied the combined treatment of PAHs

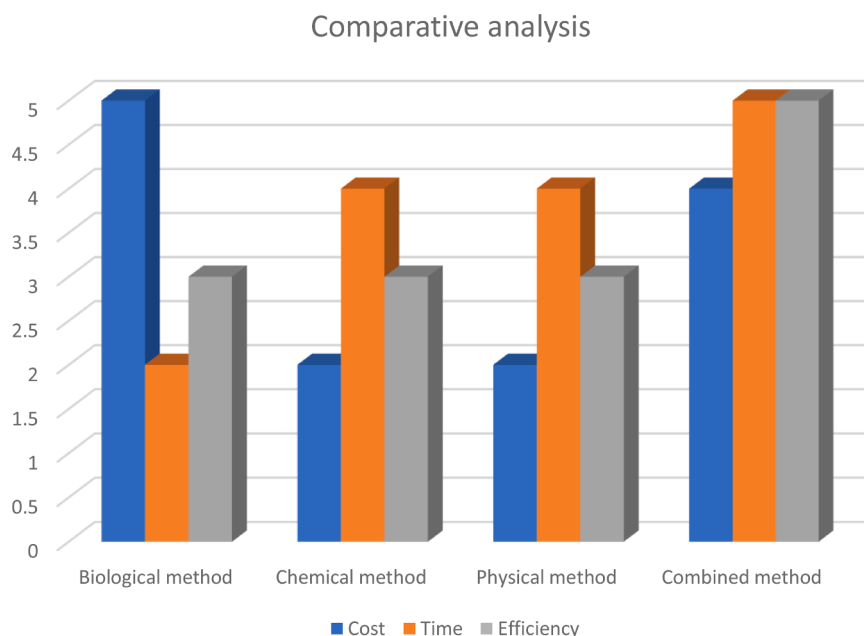


Fig. 7. Comparative analysis of remediation methods.

contaminated soil by soil washing, ozonation, and biological treatments. None of the single treatments studied could reach the clean-up target of 85% alone, however 90% degradation was achieved by combining the three treatment methods, the study further observed that ozone consumption was 5-10 times lower with soil washing. Robinson and Angyal (2008) also reported that combining physical, chemical, and biological remediation methods reduced the contaminants level at a brownfields site to acceptable regulatory limit, including 50% reduction of remediation cost. While Huang et al. (2000) reported 95% clean-up efficiency of PAHs contaminated soil by combining land fanning, microbial degradation, and phytoremediation.

In addition, biological method can be used in natural attenuation after chemical or physical remediation to restore soil flora and fauna. Valderrama et al. (2009) reported 80% degradation of PAHs in soil by chemical oxidation, however the maximum cleanup efficiency was achieved by combining chemical and biological methods. Similarly, Tsai and Kao (2009) reported 90% clean-up efficiency by combined methods of chemical and biological remediation. The studies show that combined remediation is an effective soil remediation method, integrating biological method in the treatment train enhances remediation process and restores soil flora and fauna.

6. Conclusion

There are several soil remediation methods available, the selection of one or more will depend on several factors including cost, environmental impact of method, cleanup duration, and efficiency. This study systematically reviewed remediation methods to evaluate their effectiveness in cleaning hydrocarbon-contaminated soils. Fifty-one field application studies were selected through a systematic search and categorized into four methods: biological, chemical, physical and combined remediation methods for easy assessment, as presented in tables 3. Brief description of selected studies was also presented in the study. Some set criteria including cost of remediation method, cleanup duration, cleanup efficiency, simplicity of method, and environmental impact of remediation method, were used in comparing the different soil remediation methods. In addition, advantages and disadvantages of the different methods were also considered.

The reviewed studies show that combined remediation is a more effective soil remediation method, followed by biological method. All

the studies on combined remediation method recorded high clean-up efficiency of 70% and above by optimizing the strength of two or more methods to overcome limitations of each method. However, the options chosen will depend on the contaminant type, concentration, and pollutant matrix. Physical, and chemical methods can be combined to decontaminate high concentrations and toxic contaminants, while biological method can serve as polishing agent after physical or chemical remediation. In addition, integrating biological, chemical, and physical methods can increase the clean-up efficiency of soil to 90% and above, including cost reduction of 50% and above. Although biological method is a green inexpensive soil remediation method, the inability of biological method to cleanup high concentration of toxic contaminants is a major limitation of the method. While the poor performance of chemical remediation method stem from high cost of chemicals, and concern over the negative and toxic impacts of chemicals on the environment. Similarly, high costs of remediation due to high energy consumption, and the need for further treatment of gases produced during remediation are the major limitations of physical remediation method, hence they are not considered effective soil remediation methods.

7. Recommendation

A good number of existing studies on soil remediation are laboratory based, involving the use of spiked soil or real contaminated soils for laboratory experiment. Very few fields application studies on remediation of contaminated soil exist, most technologies that have shown great successes in laboratory studies may not perform well when applied in the field. This is because real contaminated soils are more complex to treat than spiked soil, and soils studied in the laboratory. In addition, laboratory conditions are different from real field conditions. Therefore, more field application studies of soil remediation should be conducted.

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CRedit authorship contribution statement

Uloaku Michael-Igolima: Conceptualization, Visualization,

Table 5
Advantages and disadvantages of remediation methods.

Remediation methods	Advantages	Disadvantages
Biological Remediation	<ul style="list-style-type: none"> • Cost effective • Environmentally friendly (mineralizes oil to CO₂ and H₂O) • Minimal site disruption • Simple to operate • Less manpower needed • Useful for low concentrations of pollutants 	<ul style="list-style-type: none"> • Requires longer time for clean-up • Low predictability • Reliant on environmental factors • Cannot clean high concentrations and recalcitrant contaminants
Chemical Remediation	<ul style="list-style-type: none"> • Reactions are fast and can result in complete degradation of contaminants • Ideal for cleaning high concentration of pollutants • Does not generate high volumes of waste materials • It can be applied to a wide range of contaminants 	<ul style="list-style-type: none"> • Complex technology • Requires large volumes of reagents, therefore it is expensive to use • Low predictability • Reliant on environmental factors • Not environmentally friendly • It may form toxic intermediate breakdown products
Physical Remediation	<ul style="list-style-type: none"> • Fast • Clean-up efficient • Permanent removal of pollutants • Ideal for cleaning high concentrations of pollutants. 	<ul style="list-style-type: none"> • Expensive to use • Destructive to the ecosystem • Can cause secondary pollution
Combined Remediation	<ul style="list-style-type: none"> • Application to a wide range of contaminants and contaminated materials. • Clean-up efficiency is enhanced. • Able to remove difficult and recalcitrant contaminants. • Can remove high concentration of contaminants. • Treatment material can be reused or reclassified depending on regulatory approval. • Physical properties of soil are sometimes improved. • Fast. 	<ul style="list-style-type: none"> • Complex technology. • Expensive to use • Complex technology.

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Declaration of Competing Interest

The authors declare no conflict of interest.

Data availability

No data was used for the research described in the article.

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