



Remediation of soil polluted with petroleum hydrocarbons and its reuse for agriculture: Recent progress, challenges, and perspectives

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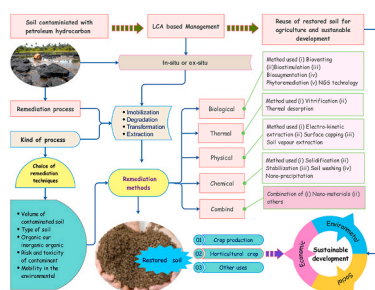
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HIGHLIGHTS

- PHs pollution can deteriorate soil properties and adversely affect the environment, humans, and other organisms' health.
- Various emerging remediation technologies, internal and external factors that affect the soil restoring process are reviewed.
- Nanotechnology looks very effective for remediation, but its success depends on its cost-effectiveness.
- Bio-electrochemical system is viewed as an eco-friendly and sustainable approach to restoring polluted soil with PHs.
- Next-generation sequencing (NGS) technologies mediated biodegradation processes by targeting rRNA are also discussed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling Editor: Y Yeomin Yoon

Keywords:

Polluted soil
Biodegradation
Petroleum hydrocarbons

ABSTRACT

Petroleum hydrocarbons (PHs) are used as raw materials in many industries and primary energy sources. However, excessive PHs act as soil pollutants, posing serious threats to living organisms. Various *ex-situ* or *in-situ* chemical and biological methods are applied to restore polluted soil. However, most of the chemical treatment methods are expensive, environmentally unfriendly, and sometimes inefficient. That attracts scientists and researchers to develop and select new strategies to remediate polluted soil through risk-based analysis and eco-friendly manner. This review discusses the sources of PHs, properties, distribution, transport, and fate in the environment, internal and external factors affecting the soil remediation and restoration process, and its effective

Abbreviations: BTEX, Benzene, toluene, ethyl benzene and xylene; BES, Bio-electrochemical systems; DNA, Deoxyribonucleic acid; EC, Electrical conductivity; EB, Electro-bioremediation; GC, Gas chromatography; HPLC, High-performance liquid chromatography; LTTD, low-temperature thermal desorption; NGS, Next-generation sequencing; PHs, Petroleum hydrocarbons; PAHs, Polycyclic aromatic hydrocarbons; PCR, Polymerase chain reaction; rDNA, Ribosomal DNA; SOD, superoxide dismutase; UV, Ultraviolet.

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<https://doi.org/10.1016/j.chemosphere.2022.133572>

Received 13 December 2021; Received in revised form 6 January 2022; Accepted 6 January 2022

Available online 8 January 2022

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re-utilization for agriculture. Bioremediation is an eco-friendly method for degrading PHs, specifically by using microorganisms. Next-generation sequencing (NGS) technologies are being used to monitor contaminated sites. Currently, these new technologies have caused a paradigm shift by giving new insights into the microbially mediated biodegradation processes by targeting rRNA are discussed concisely. The recent development of risk-based management for soil contamination and its challenges and future perspectives are also discussed. Furthermore, nanotechnology seems very promising for effective soil remediation, but its success depends on its cost-effectiveness. This review paper suggests using bio-electrochemical systems that utilize electro-chemically active microorganisms to remediate and restore polluted soil with PHs that would be eco-friendlier and help tailor-made effective and sustainable remediation technologies.

1. Introduction

Many industries use PHs as a raw material for manufacturing various goods and consumers products. PHs consumption is forecasted to increase from 85 million barrels in 2016 to 106.6 million barrels at the end of 2030 (Igunnu and Chen, 2014). However, its use varies from 32% for Europe and Asia, and 53% for the Middle East. Petroleum hydrocarbons include alkanes, cycloalkanes, polycyclic aromatic hydrocarbons, and many other organic pollutants. They are classified as major environmental pollutants due to their stability, durability in the environment (Song et al., 2021; Uddin et al., 2021). For instance, Dadrasnia and Agamuthu. (2013) reported that globally, about 8.8 million metric tonnes of oil are annually discharged in the aquatic environment. About 90% is directly linked with human activities due to the oil spill in the terrestrial and marine environment.

Excessive use of petroleum hydrocarbons leads to high pollution to the water and soil environment (Song et al., 2021; Uddin et al., 2021). Spillage of crude oil contamination is now common during offshore oil drilling, transport, and transfer to onshore in many parts of the world. The inherent problem is its complex composition with alkanes, aromatic hydrocarbons, resins, and asphaltene. They are very toxic to organisms due to their environmental transport, chemical properties, and pollutant nature (Mirjani et al., 2021). Their fate in the environment is not fully understood and is still very challenging. Consequently, characterization of petroleum hydrocarbon is also a crucial factor for evaluating and estimating the pollutant's behavior and chemical properties with the long- and short-term impact on the contaminated environment to decrease the animal's exposure's humans to the contaminant (Keramea et al., 2021).

Petroleum hydrocarbon and oil spills dissolution, dispersion, photo-oxidation, and biodegradation naturally occur very slowly, and these toxic pollutants keep going with adverse outcomes in contaminated exposed areas. Hence, they cause high potential risks and hazardous impacts on humans and other living organisms surrounding the polluted aquatic and terrestrial ecosystems (Haider et al., 2021). In addition, soil contamination with petroleum hydrocarbon has been reported to alter top-soil and sub-soil physico-chemical properties and phytotoxic impact on seed germination, crop growth, and yields (Grifoni et al., 2020; Haider et al., 2021). Therefore, to lower the danger of exposure, restore soil function, and provide ecosystem services, remediation of the contamination, it is required to implement risk-based remediation strategies for restoration and reclamation of affected soil for various uses, including agriculture (Zhang et al., 2021). For this reason, different treatment ex-situ or in situ methods include separation and destruction through ultrasonic, acoustic, electromagnetic and electric, heat and thermal, physio-chemical, chemical, and biological treatment methods are involved as strategies to restore soil polluted through petroleum hydrocarbon and to use in agriculture purpose. However, most of the treatment methods are expensive, environmentally unfriendly, and ineffective. Still, there is a high health risk to society due to the pollution of petroleum hydrocarbon. This leads scientists and researchers to develop and select new strategies to remediate the contaminated environment through risk-based analysis (Dubey et al., 2021; Koshlaf, 2020; Nwankwegu et al., 2022; Popoola et al., 2022; Ruley et al., 2022).

This review focused on emerging novel technologies for the remediation of petroleum contaminated soils and their effective utilization for growing crops and other plants. The internal and external factors that affect the restoring process, treatment methods, source, properties, distribution, and transport in the environment are discussed precisely. Furthermore, the mechanism of various petroleum hydrocarbon degradation, plants, microorganisms, their relationships, and interactions, including the methods for monitoring the remediation process such as chemical, biological, and NGS methods, are also presented. Finally, challenges and future perspectives are discussed in the recent development in petroleum hydrocarbons eco-friendly management and sustainable remediation of soil contaminants.

2. Sources of PHs, their fate into the environment

Naturally, PHs are the most commercially exploited fossil fuels (Adelaja, 2015). However, according to Environmental Protection Agency (2005), accidental leakage from oil and gas tankers, leakage during loading, transport, storage, and distribution, or in the form of oil spills can pollute the environment. Annually, 1.7–8.8 million metric tons (MMT) of PHs are discharged into the environment, almost 90% related to human activities (Shahzadi, 2021). In addition, they can negatively impact aquatic and terrestrial life (Adetunji et al., 2021; Chokor, 2021; Dadrasnia et al., 2013; Gao et al., 2022; Haider et al., 2021; Konur, 2021; Marvin et al., 2021; Rahimi Moazampour et al., 2021; Raja et al., 2022; Saeed et al., 2022). Generally, fossil based PHs are classified into aliphatic hydrocarbon heterocyclic and aromatic (Balint, 2021; Bojan et al., 2021; Speight, 2014). These PHs are usually transported to the environment, often starting from production, transportation, storage operations, refining, and consumption, then move to the air, water, soil, and entire terrestrial and aquatic ecosystems. The foremost route by which PHs permeate and contaminate soil is shown in Fig. 1.

In addition, the hydrophobicity property of various PHs enhances their affinity towards the water, air, and soil, as shown in Fig. 2.

3. Effect of petroleum hydrocarbon (PHs) on the environment and human health

PHs concentration in the environment has increased with various types of petrochemicals use and related industrial waste products and effluents. They are reported to pollute soil and emerging threat to the environment, neurotoxic and carcinogenic towards a human being (Abbasian et al., 2015; Acharya et al., 2022; Asejeje et al., 2021). Most PHs enter the environment through biological transformation (Abdel-Shafy and Mansour, 2021; Hassanshahian and Cappello, 2013). Moreover, they may also affect various environmental and many living organisms' biological processes. Hence, it needs more research and investigation into the environment's fate and transport to reduce its negative impacts (Abdel-Shafy and Mansour, 2016; Biache et al., 2014; Wartell et al., 2021; Yadav et al., 2021).

3.1. Effect of petroleum hydrocarbon (PHs) on agriculture soil

The effect of petroleum hydrocarbon on the environment and soil

depends on the chemical and physical characteristics and the various hydrocarbon components, primarily affecting the soil properties through adsorption, biodegradation, and leaching (Clay, 2014; Logeshwaran et al., 2018). They may enter the food chain and create toxic effects on humans. Agriculture soil plays a vital role in providing food for all and maintains balance in the ecosystem environment; however, contamination of soil adversely affects soil fertility and physical and chemical properties (Steliga and Kluk, 2020). The release of petroleum hydrocarbon into topsoil and subsoil has been reported to destroy the soil texture, structure and decreased pore spaces, saturated hydraulic conductivity (Hou et al., 2021; Steliga and Kluk, 2020). It can also affect the soil's biological properties, especially soil microbial population, and enzymatic activities, indirectly affecting plants' nutrient availability. Al-Joumaa (2009) conducted a study to evaluate the impact of petroleum hydrocarbon on soil physical and chemical properties. Results indicated that the polluted soil had relatively higher bulk density and absolute density than unpolluted soils. Percent clay fraction and calcium carbonate content was found low in the contaminated soil. EC values were higher in the polluted than unpolluted soil. Soluble Ca and SO_4^- ions were dominant in unpolluted soil.

3.2. Effect of PHs on plants/vegetation

A higher petroleum hydrocarbon concentration has been reported toxic to crop plants and much other vegetation. Soil aeration, water infiltration clogged by oil may severely affect plant growths (Hou et al., 2021; Steliga and Kluk, 2020), primarily reducing the root length and leaf area of sunflower crops (Hou et al., 2021; Steliga and Kluk, 2020). Al-Joumaa (2009) also reported no barley seed emergence after 14 days of exposure in the polluted soil, even after leaching and alleviating salts from contaminated soil. Petroleum pollution cause deformations of leaf pigments, H_2O_2 accumulation, oxidative stress, even plant cell death (Al-Joumaa, 2009). Inhibition in root-shoot growth, leaf lengths, and delay in cell expansion of maize has been reported by (Al-Joumaa, 2009; Hou et al., 2021; Steliga and Kluk, 2020). However, the phytotoxicity of petroleum hydrocarbons differs among various plant species. For instance, some crop species like wheat, rye, and oat could considerably tolerate and accumulate these pollutants and are used for

phytoremediation, discussed separately elsewhere (Al-Joumaa, 2009).

3.3. Consequences of PHs on human's health and other organisms

PHs are characterized by having high boiling points with a wide range of high relative molecular weight, which causes a different degree of toxicological effect to the environment and human and animal health. Depending on their susceptibility and exposure time, petroleum hydrocarbon molecules have diverse toxicity levels to the environment, people, and wildlife. They can damage various human body systems, i. e., nervous, circulatory, immune, and endocrine systems, and cause many metabolic, hormonal disorders and diseases. However, its lethal bioavailability to a substrate and toxicological effect depends upon the physical state, chemical nature, exposure concentration, exposure time, exposure mode, chemical properties, compositional fractions of petroleum hydrocarbon (Priyadarshane et al., 2022).

Consequently, it causes a different range of toxicological effects to human and animal health, such as mutagenicity, haemotoxicity, genotoxicity, carcinogenicity, ocular toxicity, cardiotoxicity, hepatotoxicity, nephrotoxicity, immunotoxicity, neurotoxicity, cytotoxicity, and teratogenicity, as shown in Fig. 3 (Azeez et al., 2015; Ossai et al., 2020; Premnath et al., 2021; Zheng et al., 2014). Besides, it also affects plants' growth due to inhibiting the uptake of mineral salt and water, which causes halting plant metabolic processes, further enhancing the shortage of nutrients and chlorophyll, leading to decreased resistance to disease pests. As a result, the plant's growth becomes stunted. In addition, the root becomes deformed well, as the leaves and flowers become necroses and chlorosis (Rusin et al., 2015; Saeed et al., 2022; Shan et al., 2014).

Petroleum hydrocarbons contain a diverse range of chemical constituents. However, the proportion, features, method of degradation and toxicological effect of small fraction chemical compounds on human and animal health have not been well understood and investigated. Hence, it needs more research to study the toxicological effects, mechanisms, and degradation of all the compounds present in the petroleum hydrocarbon to develop sustainable remediation technology. Li et al. (2020a,b) studied the ecotoxicological effects of petroleum-polluted soil on the earthworm. They reported that high petroleum concentrations in contaminated soil might significantly damage the cocoon and negatively

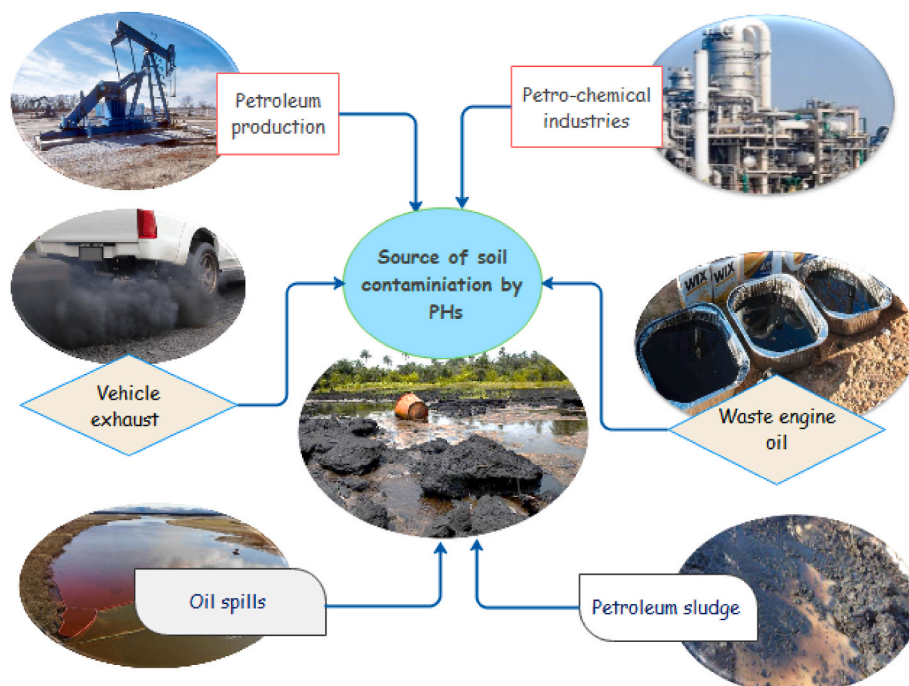


Fig. 1. Source of soil contamination by PHs.

impact DNA in the earthworm's seminal vesicles and reduce their body weight. The biological and some of the enzymatic activities like peroxidase, superoxide dismutase (SOD), and catalase activities were inhibited that indicating petroleum pollution could induce oxidative stress.

4. Remediation technologies for soil polluted with PHs

As discussed above, petroleum hydrocarbons have a massive impact on the environment and human health. It is necessary to clean and remove the contaminated environment's pollutants through suitable and sustainable remediation approaches. Hence the best remediation approach that can remove petroleum hydrocarbons from the polluted environment must be selected. Variables such as contaminant composition and type, polluted environment, biological and chemical conditions, and microbial species required for augmentation, as well as time, cost, regulatory requirements, and procedures, must all be considered. Moreover, particular attention must be given to adapt novel sustainable approaches towards reducing major risks of soils contaminated with toxic petroleum hydrocarbons (Ossai et al., 2020). In this context, various physical, chemical, biological, and other equivalent technologies are also discussed below.

4.1. Physical methods for PHs removal from soil

Worldwide several physical treatment methods to degrade and remove petroleum hydrocarbon from polluted soil have been reported. These methods include methods of separation and recycling i.e., soil flushing, soil washing, solidification, vitrification, stabilization, thermal desorption, incineration, and vapor extraction (Dadrasnia and Agamuthu, 2013; Raja et al., 2022). Petroleum-hydrocarbon polluted soil can be generally reclaimed and decontaminated by widely used (i) *in-situ* and (ii) *ex-situ* methods. Here, *in-situ* means in the original place, while *ex-situ* means outside the original place. Generally, the *in-situ* method is practiced on-site such as unmodified in their natural setting. In contrast, the *ex-situ* method is practiced off-site, such as using biodiversity outside their natural habitats in a laboratory or aquarium. To remove lighter molecules, especially (BTEX), polluted soil air extraction, air sparging, ignition, or combinations of these methods are used. *Ex-situ* soil cleaning treatment methods are carried out off-site by excavation, detoxification, destruction technology and can be applied to remove the petroleum hydrocarbons from soil and groundwater (Abdelhafeez et al.,

2022).

4.1.1. Excavation of contaminated soil for PHs removal

Excavation is a conventional and mechanical removal of petroleum hydrocarbon from polluted soil subsurface. It consists of (i) excavation and (ii) removal of the contaminants from polluted soil, primarily industrial and military bases. After excavation, polluted soil is transported to treat or destroyed at remediation sites. This method has been reported to create problems during the excavation due to the polluted soil's risks through handling and transportation. However, it can be avoided by adequately delineation the site before excavation. Sometimes, finding new landfill sites for the material's final disposal is more expensive and challenging (Ferronato and Torretta, 2019). Therefore, this method is considered an interim solution to decontaminate polluted areas because it constantly monitors potential risk, costs, and other related liability (Sohoo et al., 2021).

4.1.2. Surface capping of the contaminated soil

Surface capping is used to conserve the contaminated soil with petroleum hydrocarbon by the physical barrier between the polluted media and the surface. It helps to isolate petroleum hydrocarbon polluted soil from potential receptors. It also helps to prevent water pollution from rainfall water infiltration due to scientific surface soil capping. Contaminated media is covered with sufficiently thick and impenetrable barriers that minimize the transport of toxic petroleum hydrocarbons. Thus, surface soil and water pollution can be controlled, and direct or indirect contact of humans and animals with the contaminated media also be minimized to ensure their health safety (Li et al., 2022).

4.1.3. Prevention of soil contamination by physical barriers

Physical barriers are applied to minimize and control the transport of PHs in the soil profile. They also limit pollutants' flow away from the contaminated site and restrict their flow to groundwater and other surface water bodies (Wagner and Yarmak, 2017). Physical barriers are installed in front (down-gradient) of the polluted region. In a novel strategy, vertical barriers are used. It includes layering slurry, making walls and curtains, plastic cut-off walls, steel shoring, cement, and bentonite on top of background soil to create a barrier. In the case of softer soil, primarily slurry walls are applied. This system mainly consists of a trench having a down-gradient around a polluted zone. They

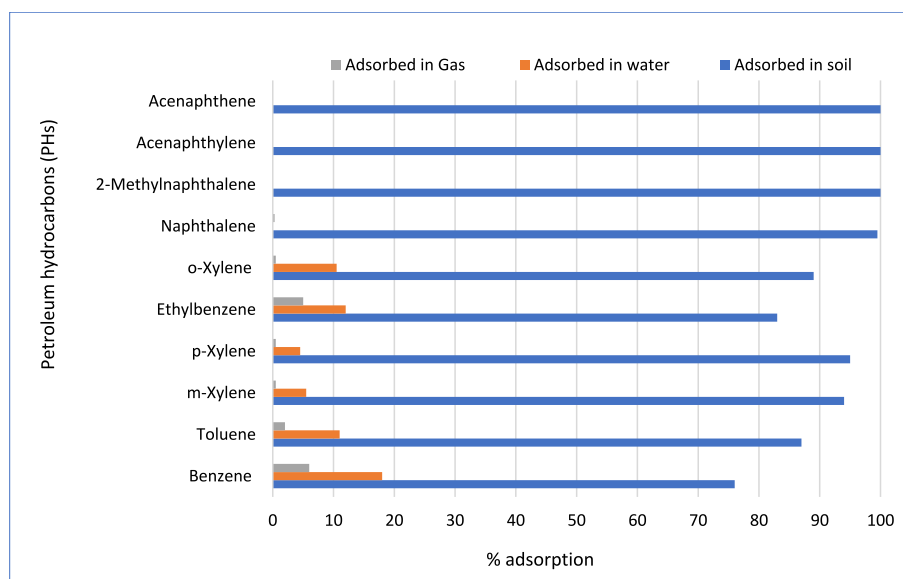


Fig. 2. Distribution of petroleum hydrocarbons on water, air, and soil. Source: Adapted from Chartered Institute of Environmental Health (<https://slidetodoc.com/contaminated-land-dealing-with-hydrocarbon-contamination-remediation-options/>).

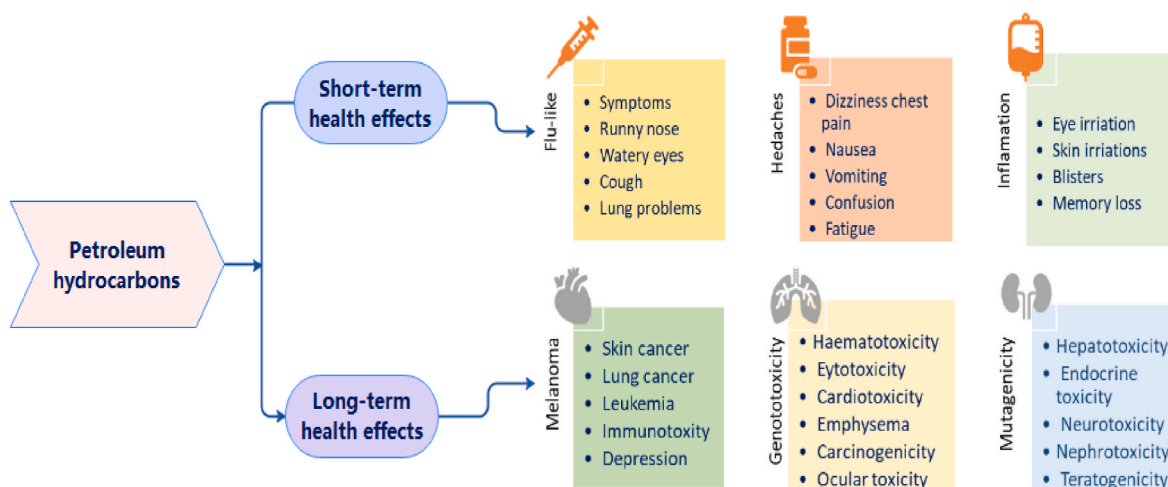


Fig. 3. Consequences of petroleum hydrocarbon exposure on human health adapted from (Ossai et al., 2020).

are usually filled with bentonite, cement, or hard clay soil. Sheet piling cut-off walls are water retaining structures installed into the impacted polluted soil to a depth of around 12–30 m. However, physical barriers do not eliminate pollutants but restrict leakage and lateral movement of petroleum hydrocarbons (Wagner and Yarmak, 2017).

4.1.4. Contaminated soil-washing

It is a crucial ex-situ method applied based on the physico-chemical separation and leaching process. In this method, dispersion of toxic PHs occurs between soil clay particles and the washing solution (Tran et al., 2022). PHs molecules bind to soil silts and clay particles, disperse water, and raise groundwater pollution levels. Some chemical additives like surfactants are also applied (Fabbricino et al., 2018; Kang, 2014). A study conducted by Murena and Gioia (2009) showed 97% petroleum hydrocarbon removal. To remediate contaminated soil, extraction solvent chlorinated organic compounds, hexane, and acetone ethyl acetate-acetone are used as washing solutions.

4.1.5. Thermal/heat desorption treatment methods

Under ex-situ methods, low-temperature thermal desorption (LTTD) is crucial to remediate toxic petroleum hydrocarbons. In this method, heat is used to separate and disperse pollutants from excavated contaminated soils physically. This method has a specific design for heating excavated polluted soils to volatilize organic constituents, entirely or partially, at 1200 °F. The vaporized hydrocarbons are treated in a catalytic oxidation chamber before discharge into the air. Then, soils are kept to cool and stabilized, then prepared to dispose or reuse in other possible work. However, soil types, such as coarse to fine soils, need proper, proven treatment and are accepted universally. LTTD is a practical remedial alternative to volatilize, stripping pollutants. However, its efficiency is reported site-specific and depends on various characteristics of petroleum pollutants, locations, contaminated soil volume, and layout. Its success is determined by the regulatory framework, logistical infrastructure, and economic considerations (Kuppusamy et al., 2017; Riser-Roberts, 2020).

4.1.6. PHs removal by microwave heating

Microwave heating is reported as a cost-effective and time-saving method. It involves homogeneous heating of PHs polluted soil by electro-magnetic irradiation, especially infrared and radio frequencies, ranging from 300 MHz to 3000 GHz. Its mechanisms solely depend on the interaction of dielectric materials and electromagnetic irradiations (Aguilar-Reynosa et al., 2017). Heat proportion during polluted soil heating is produced by bipolar rotation and ionic conduction. However, its efficiency depends on soil permeability and particle size, which play a

significant role in the vaporization of hydrocarbons. In the ionic conduction process, free ions are the key factor in the remedial process, which is placed by ionic motion produced via electric fields that lead to fast heating (Falciglia et al., 2018). Most of the chemical methods, including microwave heating, are energy-consuming and technologically complex and lack public acceptance.

4.2. Chemical methods for PHs removal from soil

Chemical methods used for the removal of PHs from the soil are described as follows.

4.2.1. Mechanisms of PHs degradation through a chemical process

Mechanisms for PHs degradation from contaminated soils through the chemical process take place by various abiotic and biotic transformations (Aeppli et al., 2014). In this process, these molecules are broken down into smaller parts via abiotic chemical changes without microorganisms' help through oxidation-reduction and hydrolysis (Almutairi, 2022; Cabral et al., 2022a,b; Kim et al., 2022). Whereas the biotic chemical transformations include both biotransformation and biodegradation processes. They might occur under aerobic conditions in which the degradation is activated by the oxygenase enzymes and oxidized into water and carbon dioxide. It can also take place in anaerobic conditions (Hsia et al., 2021).

4.2.2. Chemical oxidation-reduction

This is an advantageous technique based on in-situ chemical oxidation reactions. Various chemical oxidants like MnO_4^- , $\bullet\text{OH}$, $\bullet\text{SO}_4$, O_3 , Fe_2^+ , $\text{S}_2\text{O}_8^{2-}$, H_2O_2 , and O_2 are used in this method. They react on contaminated soil subsurface after application, where PHs are converted into non-hazardous, less toxic, less mobile, and inert compounds (Besha et al., 2018). However, oxidants are used up by dissolved heavy metals and soil organic substances instead of PHs during chemical oxidation reactions, resulting in ineffective treatment. Chen et al. (2016) conducted a chemical oxidation study to remediate diesel from contaminated soil and reported 48–93% diesel removal efficiency using MnO_4^- , $\text{K}_2\text{S}_2\text{O}_8$, H_2O_2 , under 1, 3, 5, and 10% concentrations. The removal efficiency for the PHs was in the order of $< \text{MnO}_4^- < \text{K}_2\text{S}_2\text{O}_8 < \text{H}_2\text{O}_2$.

4.2.3. Ultraviolet and photocatalytic oxidation of PHs

UV and photocatalytic oxidation methods involve UV irradiation and chemical oxidants like O_3 and H_2O_2 to degrade and mineralize petroleum hydrocarbon compounds (Pisharody et al., 2022). High-intensity UV irradiation is used by employing Hydrogen peroxide (H_2O_2) to oxidize contaminants into water, CO_2 , and salts. In UV-photocatalytic

oxidation methods, UV irradiation reacts with H_2O_2 by activating atomic bonds. It makes the molecules very oxidizable to produce reactive $OH\bullet$, which is highly reactive to the petroleum hydrocarbon for subsequent destruction. Hu et al. (2008) conducted photo-catalytic remediation of methyl tert butyl ether (MTBE). UV- irradiation on titanium dioxide (TiO_2) was found to catalyze and generate reactive oxidizing $OH\bullet$ in aerobic conditions and achieved 80% MTBE removal efficiency from contaminated media within 48 h. Gholami et al. (2020a) studied the photocatalytic performance of biochar-incorporated Zn-Co-LDH nanostructures, and they reported 92.7% of gemifloxacin degradation. A novel biochar-incorporated **zinc-cobalt layered double hydroxide** (Fe-Cu-LDH) nanocomposite was used, best sonocatalytic efficiency of 97.6% cefazolin sodium degradation was achieved (Gholami et al., 2020b). A newly synthesized 3-dimensional photocatalyst, WO_3 nanorods, and **N-Doped Magnetic WO_3 - x@Mesoporous Carbon** (NM- WO_3 -x@MC) nanocomposite degraded 90% of antibiotics and 76% of total organic carbon, respectively (Gholami et al., 2021). In another study, Moradi et al. (2021) used photocatalytic Fe TiO_3 /**graphene oxide** (GO) nanocomposite and showed significant phenol degradation under visible light irradiation.

4.2.4. Activated carbon adsorption of PHs

Activated carbon adsorption was also found helpful in the

decontamination of petroleum hydrocarbon compounds from surface soil. Many researchers have reported its efficiency in the removal of organic pollutants and reducing their bioavailability. It was reported as the most versatile adsorbent due to its micro-porous structure, large internal surface area, and hydrophobicity, making it a very strong, highly effective, and suitable *adsorbent* for petroleum hydrocarbon removal (Wang et al., 2022a,b,c,d). The adsorption process is controlled by the van-De-Waal force, and pollutants desorption occurs at an equilibrium state. For instance, Kalmykova et al. (2014) had conducted a study on the PAHs sorption using activated carbon. They showed around 50–63% sorption and removal of PAHs from leachate collected landfill sites.

4.2.5. Electrochemical systems for PHs removal

Electrochemical systems have been reported to remediate cyclic and aromatic hydrocarbon from polluted media, including soil. In this system, electric current serves either as electron donor or acceptor for oil spill remediation. Electro-osmosis, electromigration, and electrophoresis are the primary mechanism of transport and mobilization of pollutants. Electrical potential or electromagnetic waves are green technologies for the effective remediation and low molecular weight hydrocarbons volatilization. Wang et al. (2022) conducted an experiment using single-chamber air-cathode microbial fuel cells for the

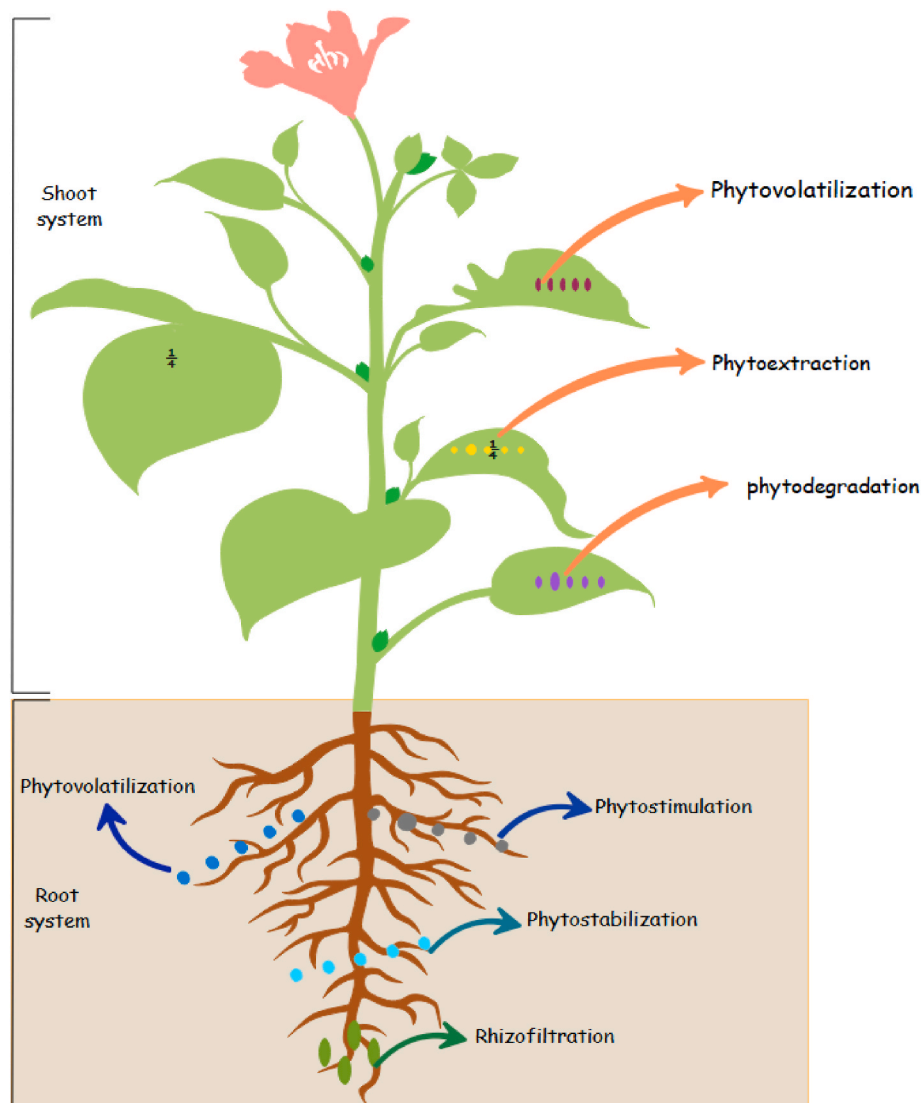


Fig. 4. Main mechanisms involved in phytoremediation of petroleum hydrocarbon.

degradation of pyrene and found it very effective. Also, the addition of pyrene influenced the electrical properties of micro fuel cells and this leads to an increase in the density of the anodic biofilm.

4.3. Biological methods for PHs removal

Biological methods for petroleum hydrocarbon removal are grouped into (i) Bioremediation. Microorganisms, especially bacteria and fungi, are applied to remove petroleum hydrocarbons, (ii) Phytoremediation, where plants and symbionts species are used to remove PHs from the contaminated environment. It also facilitates their removal via amendments of soil and agronomic practices (Ruley et al., 2022). A principal mechanism involved in phytoremediation i.e., phytovolatilization, phytodegradation, phytoextraction, phytostabilization, Phyto stimulation, and rhizofiltration, are shown in Fig. 4.

Due to the complex nature of petroleum hydrocarbons, the degradation process depends on the concentration, composition, and nature of the petroleum hydrocarbon present in the contaminated environment. Therefore, its degradation process results from biodegradation and chemical transformations (Ruley et al., 2022). As shown in Fig. 5, various physical and biological factors affect the biodegradation rate of petroleum hydrocarbons. Therefore, the physicochemical qualities and nature of the pollutant are some of the essential variables to consider when developing appropriate remediation methods for PH-polluted soil. For example, aliphatic hydrocarbons have a higher degradation rate than aromatic hydrocarbons such as PAH and BTEX compounds due to having less structural complexity and low molecular weight because the molecular weight and structure of the petroleum hydrocarbon increase leads to less biodegradable compounds. However, the addition of nutrients or co-substrates such as biosurfactants to complex hydrocarbons can enhance their degradation rates and other environmental factors, e. g., temperature and pH, salinity, pressure (Ambaye et al., 2021a; Cui et al., 2022).

In addition, PAHs degradation rates at anaerobic conditions are affected by the availability of terminal electron acceptors like SO_4^{2-} , NO_3^- , and O_2 (Sun et al., 2022). Especially in the area where the availability of oxygen is very low leads to a decrease in a continuous supply of nutrients like SO_4^{2-} , NO_3^- as well as it decreases the availability of terminal electron acceptors, which can be used for microbial metabolism during the biodegradation process (Sun et al., 2022). Due to this, several efforts have been made to supply alternative terminal electron acceptors to the soil, showing increased mineralization of petroleum hydrocarbons.

Temperature is another environmental component that could affect the PHs-biodegradation rates of petroleum hydrocarbons. It ranges from (-10 to 55 °C), affecting the diversity of microbial communities, physiology, and chemistry of the pollutant, and the pace of

contaminating biodegradation (Li et al., 2022; Okonkwo et al., 2021). Moreover, the degradation rate and solubility of polycyclic aromatic hydrocarbons also decrease with decreasing in temperature.

4.3.1. Phyto-degradation of PHs

Recent studies on phytoremediation of soil contaminated with PHs are reported in Table 1. Phytoremediation conventional studies have been associated with biostimulation via the addition of fertilizers (Tang, 2019; Tang and Law, 2019) but lately linked to bioaugmentation with bacteria or fungi (Asemoloye et al., 2017; Hou et al., 2015). There is also interest in examining the effect of Se in increasing the phytoremediation plant's efficiency (Huang et al., 2019). *Sorghum bicolor*, *Zea mays*, *Helianthus Annuus*, and *Lolium multiflorum* are typical species utilized in phytoremediation investigations. New phytoremediation-capable plants are being added to the list as a result of screening investigations. *Epipremnum aureum*, *Azolla filiculodes*, *Mucuna bracteata*, and *Epipremnum aureum* are the most recent additions.

Table 1 shows some potential methods and plants that have been used for the removal of soil polluted with petroleum hydrocarbons using phytoremediation in the duration of 20 to 150 days. They have a more significant influence on the reduction of hydrocarbons. For instance, Baoune et al. (2019) reported 70% of crude PHs and 61% of phenanthrene removal from the soil in 14 d. Kösesakal et al. (2016) reported 94% of total aliphatic hydrocarbons removal from liquid medium contaminated.

In general, phytoremediation is a term that refers to a group of technologies that use plants' natural capacity to clean up soil that has been contaminated by petroleum hydrocarbons. Phytoremediation is the most successful remedial strategy for reducing persistent organic compounds, and it has recently attracted a lot of interest due to its ability to degrade contaminants across a large area, broad applicability, eco-friendliness, and cost-effectiveness. However, only a few plants can absorb these organic chemicals from the soil, and plants cannot break down or detoxify persistent organic compounds due to a lack of fixed enzymes. Only a few plant species, such as Cucurbita species, have demonstrated a unique ability to absorb a significant amount of persistent organic chemicals from the soil and are considered persistent organic compound hyperaccumulators. Furthermore, genes encoding and expressing persistent organic compound-degrading enzymes have been discovered and extracted from persistent organic compound-degrading microorganisms. Transgenic hyperaccumulator plants encoding persistent organic compound-degrading enzymes could help with the phytoremediation of persistent organic pollutants (Malik et al., 2022).

4.3.2. Microbial degradation of PHs

Various researchers have widely reported microbial contribution in

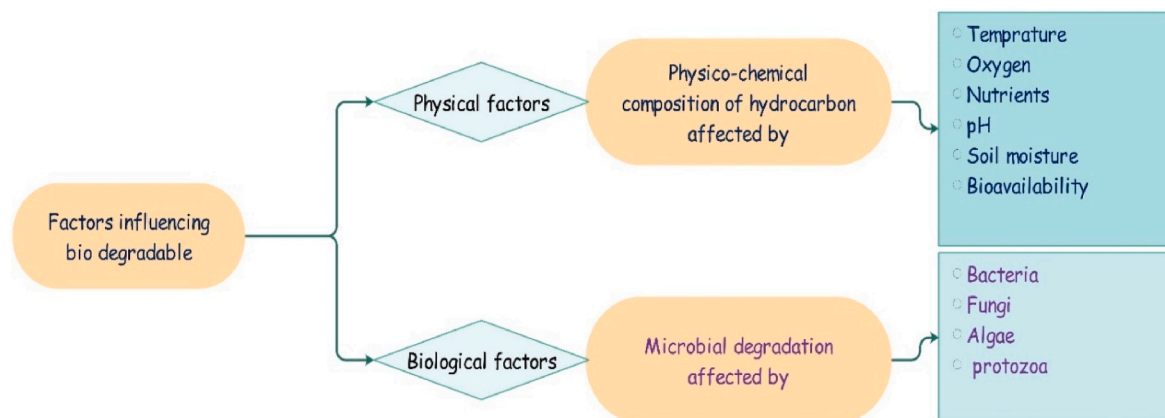


Fig. 5. Physical and biological factors affecting the biodegradation rate of petroleum hydrocarbon from the contaminated environment.

Table 1
Phytoremediation (using plant species) of soil contaminated with PHs.

Soil contaminant	Plant species	Methods	Removal efficiency	References
Crude petroleum oil	<i>Phragmites australis</i> and <i>Juncus maritimus</i>	Attenuation, bio stimulation in marshy sediment	PHs removal efficiency achieved best with <i>P. australis</i> (16%) by bio stimulation within 150 d	Ribeiro et al. (2014)
Petroleum hydrocarbons in crude oil contaminated soil	wheat (<i>Triticum aestivum</i>), maize (<i>Zea mays</i>), white clover (<i>Trifolium repens</i>), alfalfa (<i>Medicago sativa</i>), and ryegrass (<i>Lolium multiflorum</i>)	soil amendments (biochar and compost) on plants belonging to Poaceae and Fabaceae families.	The highest TPH removal (68.5%) by ryegrass with compost, white clover with biochar (68%). Without any soil amendment, ryegrass and alfalfa showed 59.55 and 35.21% degradation of TPHs, respectively. Biochar and compost alone removed 27.24% and 6.01% TPHs, respectively, within 62 d	Yousaf et al. (2022)
Crude oil	<i>Imperata cylindrica</i> ; <i>Mucuna bracteata</i> ; <i>Pteris vittata</i> ; and <i>Epipremnum aureum</i> ;	Screening of plants in Malaysia at 5% crude oil contamination by weight	<i>Mucuna bracteata</i> (31%), <i>Pteris vittata</i> (36%); <i>Imperata cylindrica</i> (40%); <i>Epipremnum aureum</i> (50%); to remove total PHs within 42 d	Tang and Angela. (2019)
Petroleum hydrocarbons	<i>Medicago sativa</i> L.	microcosm experiments involving <i>Medicago sativa</i> L. and isolated bacterial consortium <i>Acidocella aminolytica</i> and <i>Acidobacterium capsulatum</i>	<i>M. sativa</i> + Consortium resulted in 91% removal of diesel hydrocarbons in just 60 d	Michael et al. (2022)

degrading PHs from soil and water in the last few decades (Cabral et al., 2022a,b; Mohapatra et al., 2022; Rafeeq et al., 2022). Hydrocarbon-degrading microorganisms play an essential role in effective bioremediation methods, cleaning polluted soil, water, sediments, and other contaminated media (Mohapatra et al., 2022). Oleophilic microorganisms for degrading and cleaning petroleum pollutants from

terrestrial and aquatic systems are considered eco-friendly and economical. Therefore, microbial degradation is the ultimate natural mechanism for maintaining ecosystem and environmental sustainability (Cabral et al., 2022a,b). One of the widest mechanisms that can be used for the degradation of petroleum hydrocarbon is biodegradation. In this process, the compounds are degraded via specific anaerobic and aerobic

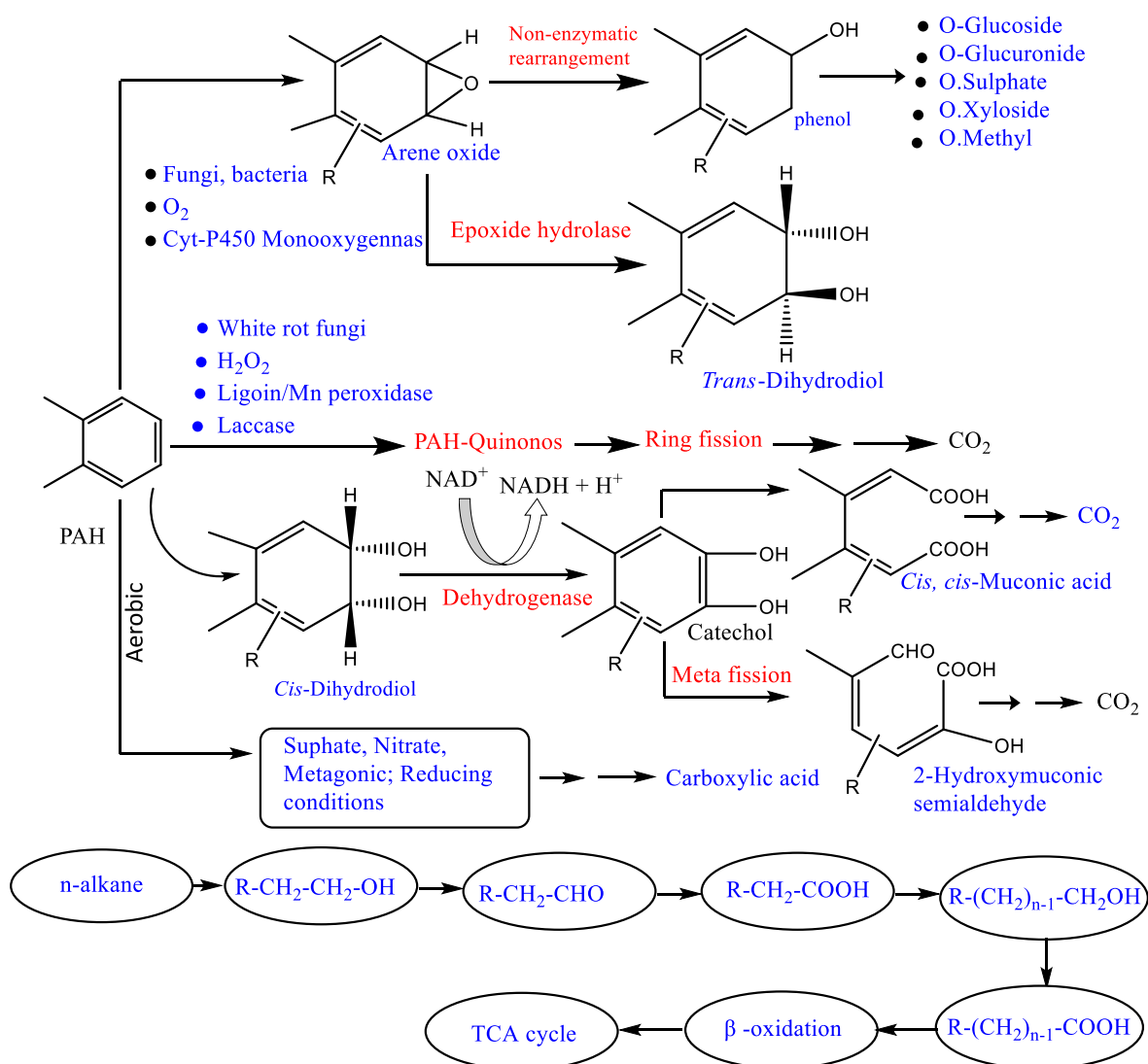


Fig. 6. Graphical flowchart for PHs and alkanes degradation using Microbial degradation adapted from (Koshlaf and Ball, 2017).

enzymes and use emulsifiers and biosurfactants, at a certain condition, to attach microbial cells to the substrates. The general process for the aerobic degradation of petroleum hydrocarbon using microorganisms is shown in Fig. 6 (Koshlaf and Ball, 2017).

In aerobic transformation, first, the petroleum hydrocarbon is attacked by using an enzyme that mediated the oxidative process (oxygenase), then it will be followed by the cleavage of the ring. The polycyclic aromatic hydrocarbons can also be oxidized into phenolic and quinone derivatives using fungi by the cation of peroxides (Koshlaf and Ball, 2017). The enzymes use the molecular oxygen produced from the aerobic transformation process to oxidize the substrate of polycyclic aromatic hydrocarbons to hydroxylation rings and other forms of intermediate oxidized products (Imam et al., 2022). The list of some potential microorganisms which can degrade petroleum hydrocarbon is shown in Table 2.

Degradation of PHs is reported faster by bacteria. However, fungi take a long time to acclimate to the environment and have low, competitive capabilities. Thus, degradation by fungi depends on the species capacities and is mostly limited to the soil environment, which is oxidic (Magan et al., 2022). The other mechanism used for the degradation of BTEX and polycyclic aromatic hydrocarbons in anaerobic degradation. There is carboxylation of the substrate in this process, followed by the cleavage aromatic ring of the phenanthrene aromatic compound. This pathway shows how the degradation route of PHs using an anaerobic process differs from the aerobic process route (Mohapatra et al., 2022).

Tsai et al. (2009) conducted research to degrade phenanthrene and fluorene through an anaerobic bio-transformation route coupled with sulfate-reducing bacteria. Results showed the formation of molybdate, which indicates that phenanthrene and fluorene are bio-transformed via hydration and hydrolysis processes followed by decarboxylation with p-cresol and phenol formation. Hence, these metabolites' formation can be considered a new biotransformation route of phenanthrene and fluorene. However, most petroleum hydrocarbons' degradation mechanism modes are not fully understood; more research and investigation are needed in the future, especially by considering the complexity of the field scale, as shown in Fig. 7.

Table 2
Summary of some potential petroleum hydrocarbon-degrading bacteria and fungi.

Microorganisms	Compound	Removal rate	References
<i>Aspergillus</i> spp.	Crude oil	59–87.7% of crude oil recovery	Zhang et al. (2016)
<i>Aspergillus ochraceus</i> CBMAI 849	Benzo [a] pyrene	76.6–99.7% bioremediation from saline environments	Passarini et al. (2011)
<i>Penicillium</i> sp. RMA1 and RMA2	Crude oil	Degradation achieved by RMA1-57% and RMA2 55%	Al-Hawash et al. (2018)
<i>Aspergillus</i> sp. RFC-1	Different PHs	60.3–97.4% removal of PHs from aqueous environments	Al-Hawash et al. (2019)
<i>Ochrobactrum</i> sp.	crude oil	83.49% degradation of crude oil equivalent to 3% v/v	Varjani et al. (2015)
<i>Candida tropicalis</i>	Diesel	Degradation of 83% diesel	Adams et al. (2015)
<i>Pseudomonas</i> sp.	Petrol, diesel, and waste engine oil	Degradation achieved petrol-76%, diesel-83%, and waste engine oil –69%	Ruley et al. (2020)
<i>Bacillus sorenness</i> D11, <i>Pseudomonas stutzeri</i> D13, and <i>Bacillus cereus</i> D12,	Petroleum-diesel TPH-DRO.	Strains consortium removed up to 80% petroleum- diesel, extract at 31.1 mg g ⁻¹ TPH-DRO.	Alsayegh et al. (2021)

4.4. Other bioremediation technologies

4.4.1. Land farming

This is a process in which the soil polluted with petroleum hydrocarbon is aerated and regularly spread out over thin layers, facilitates the degradation of the contaminant species, and adds nutrients. In addition to this, this treatment is a solid-phase approach in which the contaminated soil is immobilized and biodegraded using microbiological processes and oxidation, which can be done either ex-situ or in situ (Brown et al., 2017; Guarino et al., 2017).

4.4.2. Composting

Compositing can mainly be used to degrade targeted soil pollutants by combing the cured organic material (mature) with contaminated soil. This technology uses composted organic matters such as fishbone compost, spent mushroom, and bark compost supplemented with urea. Hence, adding urea-enriched composted organic matter to the contaminated soil gives a source of carbon supplements of nutrients to the microorganisms, increasing the metabolic capability and, therefore, the biodegradation of the organic compounds found in the soil matrix (Chen et al., 2015; Leech et al., 2020).

4.4.3. Bio-slurry system

It is one of the best ex-situ bioremediations technology used to remediation soil contaminated with PHs under controlled conditions. To aid in pollutant biodegradation, the soil is mixed with water to make a slurry, which is typically 10–60% by weight by volume. Indeed, increasing the mass transfer rates and the contact between contaminants, nutrients, and microorganisms present in a slurry phase is achievable. Bioreactors are typically run in batch mode to make slurries easier to handle and increase process yields (Avona et al., 2022; Geng et al., 2022; Kuppusamy et al., 2016b; Thenmozhi et al., 2022). However, the hydrocarbon availability for microorganisms to consume as a carbon and energy source is one of the most significant factors influencing their biodegradation rates because PHs are more stable in the soil matrix, and their bioavailability is hindered by physical contact. Moreover, due to partial oxidation of PHs, they form more toxic intermediates, which leads to poor performance during the treatments processes by microorganisms (Abena et al., 2019). To increase the bioavailability and solubility of hydrocarbons, it is necessary to have a liquid phase in the slurry bioreactors and add additives, e.g., biosurfactants, that can improve the bioremediation of soil polluted with PH. The optimizations of the operational parameters deserve in-depth investigation in the future, including pH, oxygen, nutrients, the type of additive used, and autochthonous bacterial diversity towards commercial scales and appropriate management of its effluents (Wu et al., 2017). Some recent studies on remediation of PHs contaminated soils using bio-slurry are summarized in Table 3.

4.4.4. Bioventing and bioparging

This is a process in which the polluted soil is degraded using in-situ remediation by supplying air or oxygen to existing soil microorganisms. This promotes microbial activity and generally leads to increased pollution from soil or wastewater (Hussain et al., 2022). In addition to this, nutrients can also be applied when required to promote the indigenous microbial community's growth. Constructed bioventing wells may be mounted horizontally or vertically, depending on the location's depth and geology (Hussain et al., 2022). While bioparging has a similar working principle and design to bioventing, they differ in the active region. That is, bioparging is mainly used below the groundwater level in the saturated zone, and compared to the bioventing systems, it has a high air injection flow rate. Moreover, in actual use, bioparging and bioventing systems are combined with various treatment approaches such as permeable reactive barrier technologies and soil vapor extraction (Chatterjee et al., 2022).

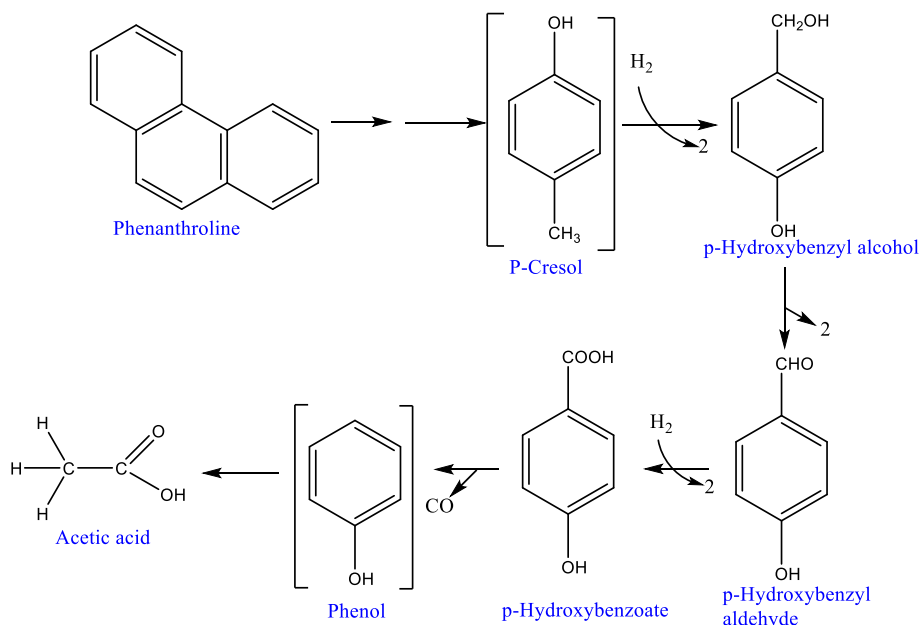


Fig. 7. Biotransformation route of phenanthrene using anaerobic and sulfate-reducing bacteria adapted from (Tsai et al., 2009).

4.4.5. Permeable reactive barriers

It's a subterranean wall designed to clean up polluted groundwater. The wall is "permeable," meaning it allows groundwater to move through it. To be treated, water must flow through the permeable reactive barriers. The wall's "reactive" materials either trap or decrease the toxicity of harmful pollutants. The groundwater is treated and then discharged on the other side of the wall (Obiri-Nyarko et al., 2014). Compared to the other traditional groundwater remediation technologies, permeable reactive barriers have the following benefits: having low maintenance cost and treating the pollutants in-situ approach and rejecting groundwater, and absence of above-ground infrastructure. On the other hand, permeable reactive barriers are the only suitable approach for shallow aquifers and need more capital-intensive costs (Andrade and dos Santos, 2020).

4.4.6. Natural attenuation

In natural attenuation, indigenous microbes degrade PHs as they live solely on carbon (C), readily available even in polluted media. When a site is contaminated, indigenous PH-degrading microbes multiply rapidly and adapt to the newly introduced petroleum hydrocarbons, resulting in the entire degradation of PHs (Zhang et al., 2022). This remediation process arises naturally in polluted sites, particularly those with lower light PH, usually used in zones where other restoration mechanisms are unsuccessful (Okparanma et al., 2017). Natural attenuation is suitable for treating petroleum hydrocarbon-contaminated places because it acts without human intervention and requires limited monitoring. Natural attenuation is effective in managing petroleum hydrocarbons-polluted sites in a biodegradation study. For instance, Lv et al. (2018) stated an about 60% decrease in PHs pollution plume due to natural attenuation of polluted soil. Natural attenuation was used to treat 25% of PH-polluted soils worldwide (Marić et al., 2018). Natural attenuation is more effective than other bioremediation strategies because it reduces the bulk, toxicity, mobility, volume, and concentration of pollutants in soil without requiring human involvement (Šrédlová and Cajthaml, 2022).

4.4.7. Reductive dechlorination and dehalogenation

In this process, PHs are removed by the reductive chemical processes. The dechlorination and dehalogenation process is used to lower toxic compounds' levels through progressive chlorine replacement, in

which halogen atoms are replaced by hydrogen atoms either by decomposition or partial volatilization (Zhu et al., 2022). Various studies have reported that bacteria and a few mesophilic and thermophilic methanogens can act during reductive dehalogenation at anaerobic conditions. For example, according to Zanolli et al. (2015), their microbial dehalogenation of organohalides showed that 98% of debromination was effective in the degradation of the marine in mangrove sediments after 90 days of incubation. In general, halogen compounds can exacerbate the harmful biological effects of living compounds by increasing their toxicity, mutagenicity, and other harmful forces, as well as affecting their hidden as bioremediation lacks certain halo-organic compounds that are generally biodegradable, moderately, as a completely broken apart body. Despite facing metabolic hurdles posed by halo-organic substances, bacteria have developed a strong desire to biodegrade them.

5. Emerging remediation technologies for soil polluted with PHs

5.1. Nanotechnology applications for petroleum hydrocarbon removal

Nanomaterials have unique properties such as high anion and cation adsorption capacity, high porosity and specific surface area, as well as high thermal and chemical stability. These properties make nanomaterials good candidates for environmental remediation technologies (Ambaye et al., 2021b; Amabye et al., 2018). These exceptional properties can also solve drawbacks of corresponding bulk material of surface/interface, quantum size, and high surface reactivity. This unique property makes them best suited for in situ remediation applications (Rani et al., 2017, 2021). Datsyuk et al. (2008) claimed acidic pre-treatment could increase hydrophilic functional groups, leading to nano-material dispersion and immobilization, which enhanced the performance efficiency of carbon nanotubes as adsorbents material. Oxidation and adsorption are the two main processes in which nanoparticles remove PHs from the contaminated soil matrix (Usman et al., 2022).

In general, since their reaction is much more efficient than that of other materials, nanosized particles provide a solution for quicker and more cost-effective site remediation. For instance, Jiemvarangkul et al. (2011) reported that about 88% of PHs are removed from contaminated water using nanosized oxide material prepared from iron and extract of

Table 3
Studies on remediation of PH-contaminated soils using bio-slurry.

Type of Pollutant	Working conditions and type of bacteria	Removal efficiency	Reference
PAHs	Soil contaminated with PAH: 10,973 mg kg ⁻¹ with soil RO water ratio of 30%; microbial consortium	93.4% removal of PAH in 12 d	Lewis (1993)
PAHs	Microbial consortium, PAHs: 4494 mg kg ⁻¹ . Soil/water: 20%, Temperature: 21 ± 2 °C; pH: 6.5–8	90% removal of PAH in 35 d	Lee et al. (2001)
PAHs	Indigenous soil bacteria, PAH: 13 g kg ⁻¹ . Soil/water: 25%, Temperature: 20 ± 2 °C; pH: 7.5	58.2% removal of PAH in 150 d	Fava et al. (2004)
PAHs	Natural soil bacteria, HCs: 500 µg g ⁻¹ , PAHs: 6.1 µg g ⁻¹ , Soil/water: 20%; Temperature: 30 °C	>70% removal of HCs and 40% for PAHs removal in 35 d	Beolchini et al. (2010)
Phenanthrene	Microbial consortium, Phenanthrene: 75 mg kg ⁻¹ . Soil/water: 18%, Temperature: 25 °C	100% removal of phenanthrene in 6 d	Woo et al. (2004)
PAHs	Microbial consortium, Pyrene: 100 mg L ⁻¹ . Soil/water: 50%, Temperature: 25 °C	77% removal of PAHs in 60 d	Thavamani et al. (2012)
PAHs	Microbial consortium, PAH: 3967 mg L ⁻¹ . Soil/water: 50%, Temperature: 25 °C	90% removal of PAHs in 60 d	Kuppasamy et al. (2016c)
Total petroleum hydrocarbons (TPHs)	Microbial consortium, Soil 1: 9.45 g kg ⁻¹ . Temperature: 27 °C	72.7% removal of TPHs in 54 d	Bento et al. (2005)
TPHs	Microbial consortium, TPH: 10 g kg ⁻¹	79% removal of TPHs in 70 d	Suja et al. (2014)
TPHs	Microbial consortium, TPH: 38.08 g kg ⁻¹ . Temperature: 25 °C	79.8% removal of TPHs in 84 d	Hesnawi and Mogadami, 2013
TPHs	Microbial consortium, TPH: 10 g kg ⁻¹ . Temperature: 30 °C	61.9% removal of TPHs in 60 d	Zhao et al., 2011
TPHs	Microbial consortium, TPH: 42 g kg ⁻¹ . Temperature: 25–30 °C, pH: neutral	76% removal of TPHs in 120 d	Farahat, and El-Gendy. (2007)
TPHs	Microbial consortium, TPH: 39–41 g kg ⁻¹ . Temperature: 25 ± 2 °C	74% removal of TPHs in 210 d	Ramadass et al. (2018)

vaccinium floribundum. However, 32 h of treatment with the same nanoparticles can remove about 82% of petroleum hydrocarbon from soil (Murgueitio et al., 2018a,b). Pavia-Sanders et al. (2013) used hybrid nanoparticles to remove polycyclic hydrocarbon from contaminated soil made from combining hydrophobic polyacrylic acid block copolymers with oleic acid-stabilized Fe₂O₃ nanoparticles with tetrahydrofuran as PAH sequestering agents from crude oil and hydrophilic polystyrene material, showing that the hybrid material can remove the pollutants more than ten times compared its weight. Hu et al. (2014) showed that compressible carbon nanotube-graphene hybrid aerogels could be used for oil sorption and removal from polluted media.

Cobalt and manganese nanoparticles facilitated the degradation of PHs in soils within 3 h (Nador et al., 2010; Zhou et al., 2019). In UV radiation, Saïen and Shahrezaei. (2012) used TiO₂ metal nano-oxide to remediate pollutants from petroleum refinery wastewater. In UV, 100 mg L⁻¹ catalysts degraded 78% PHs within 60–90 min. Alizadeh Fard

et al., 2013 successfully degraded PHs using TiO₂ nanopowder film, while Ziolli and Jardim. (2002) used colloidal TiO₂ metal nano-oxide and successfully photo-catalyze and degraded crude oil from sea seawater. In general, nanotechnology gives more benefits in the bioremediation of soil contaminated with petroleum hydrocarbons.

The use of nanotechnology in bioremediation is playing a vital role (Table 4). However, it also has some concerns regarding living biota, including human health. It is essential to evaluate their ecological risk and consequences. Currently, many studies emphasize the need to understand toxicity, mobility bioavailability, and potential risks of nanoparticles. The applicability of nanoparticles to large-scale remediation of PHs from polluted soil is another issue due to the recalcitrance nature and stability of manufactured nanoparticles (Vázquez-Núñez et al., 2020). Before these nanoparticles are used on a broad scale, it is important to understand their environmental fate better (Cecchin et al., 2017). Furthermore, nanotechnology's successful and long-term implementation would be dependent upon the use of low-cost, environmentally friendly nanotechnology in fields as diverse as biomedicine, agriculture, and bioremediation.

5.2. Electro-bioremediation (EB)

EB is a technology where electromigration or electroosmosis is used. It improves in situ pollutants biodegradation using a low voltage applied by electrodes positioned inside polluted soil. This technology is incredibly successful in polluted soil with a high surface area and low hydraulic conductivity (Camacho, 2021). For this reason, various biological growth parameters such as microbial population, nutrients, and surfactants can all be moved in various orientations within the groundwater matrices and soil (Li et al., 2020). Despite its effectiveness, this technology has many disadvantages, including energy consumption, only applicable to specific contaminated soils, low hydraulic conductivity, contamination availability, and soil matrix heterogeneity (Li et al., 2020). However, some studies are currently boosting these techniques by using additives, e.g., biosurfactants, to increase the availability of contaminants during the treatments processes. Even though other biological methods have some advantages, they also have disadvantages like lack of terminal electron acceptors, lower biokinetics, pollutants toxicity and partial degradation of materials, and high demands of capital and maintenance costs. Hence, they required more effective and long-term approaches to removing hydrocarbon pollutants from the environment (Baniasadi and Mousavi, 2018).

5.3. Bio-electrochemical systems (BESs) applications

BES is evolutionary new bioengineering technology that converts organic compounds present in wastewater into biogenic electricity for wastewater bioremediation. One of the exceptional properties of this technology is the microorganism that participated have the ability transferring of electrons extracellularly to solid anode electrode materials, compared to the microbial metabolism of using intracellular mechanisms as well as compared to low-cost conventional wastewater technologies, they produce less sludge, and it can also produce various high-value fuels and chemicals such as polyhydroxyalkanoates, methane, and ethanol (Adelaja, 2015; Baniasadi and Mousavi, 2018; Espinoza-Tofalos et al., 2020; Daghighi and Franzetti, 2020).

As a result, in the last ten years, bioelectrochemical systems such as microbial fuel cells have been employed to treat wastewater discharge from several industrial and municipal sources, including the chocolate industry, sewage, brewery, and swine. They have a high elimination of nitrate and sulfates and a 95 percent reduction in chemical oxygen requirement (Mohyudin et al., 2022; Prathiba et al., 2022). This has opened a new chapter to use these systems to remediate soil polluted with PHs by the electro-chemically active microorganism: the anode of the microbial fuel cells that can catalyze organic electron donor oxidation, such as hydrocarbons, and transfer electrons to the anode

Table 4
Application of Nanomaterials for removal of petroleum hydrocarbons from soil.

Type of PHs	Nanomaterials	Conditions	Remediation Efficiency (%)	References
Total PHs	Biogenic iron oxide	Total PHs are degraded in 14 h and 5 d at pH 6.0 and 0.74 M oxidant.	95	Bolade et al. (2021)
Total PHs	Biogenic iron oxide	Total PHs are degraded in 14 h and 5 d at pH 6.0 and 0.74 M oxidant.	95	Bolade et al. (2021)
Total PHs and PAHs n-alkanes	Ferrous sulfate enriched with biochar and activated persulfate	It removes the difference at pH 7 with shaking incubator at 25 °C in 7 d	62 78 92	Xia et al. (2021)
Total PHs	Iron nanoparticles coated with mortiño berry (<i>Vaccinium floribundum</i>)	It removes the total PHs in 32 h at pH between 9 and 109 with initial concentration of 94.20 mg L ⁻¹	88	Murgueitio et al. (2018)
Total PHs	Green mango peel-nanozero valent iron	Total PHs removal over one-week treatment at the temperature of 20 °C on a rotating shaker (200 rpm)	90	Desalegn et al. (2018)
Diesel-oil	nanoscale zero-valent iron activated with peroxymonosulfate	It removes the aged petroleum hydrocarbon in 2 h	61	Bajagain and Jeong, (2021)
Petroleum Hydrocarbons	Goethite-chitosan Nanocomposite	The nanocomposite increases the biodegradation process in 3 d of incubation	93	El-Sheshtawy et al. (2021)

extracellularly. Electrons are moved to a cathode via an external circuit, where O₂ is usually reduced to H₂O. The microbial fuel cell's anode can act as the non-exhaustible electron acceptor to transfer electrons from the polluted site to the source having high terminal electron acceptors.

Furthermore, the BESs electrodes could enhance petroleum hydrocarbon degradation without any energy input or chemical addition. That could reduce the operating costs than other methods of remediation as well as it is used as generates an electrical current, which can not only act as a bio-remediation indicator in real-time but also drive wireless sensors for remote online tracking (Gambino et al., 2021; Li et al., 2021; Wang et al., 2021). Several research studies have been carried out on bioelectrochemical systems to remove petroleum hydrocarbon from the contaminated soil in the past ten years, which are summarized in Table 5. Accordingly, much attention should be dedicated in the future to set those technologies at larger and green scales with more standardized engineering protocols.

Besides the above studies on PHs remediation from contaminated soil, for instance, Yu et al. (2021) have investigated the degradation of petroleum hydrocarbon polluted soils as substrates using double microbial fuel cells and showed that the pollutant degradation has

increased by about 59% compared to the open circuit control within 115 d. In another study, Morris and Jin. (2012) reported that using a modified single microbial fuel cell can increase by about 12% the removal rate of total petroleum hydrocarbon from contaminated soil to about 12% compared to the initial biodegradation rate. Finally, some reports showed that the use of microbial fuel cells and BESs could improve the degradation of petroleum hydrocarbons, particularly asphaltene fractions, aliphatic and aromatic compounds (Jin and Fallgren, 2022; Wang et al., 2022).

In both ex-situ and situ methods, microbial fuel cells and BESs can be used to remediate soil polluted with PHs (Adelaja, 2015). These technologies are reported biocompatible, robust, versatile, and eco-friendly to bioremediate various pollutants. However, due to diversion and competition among sulfates and nitrates electron acceptors, low microbial growth rate, low bioavailability of the substrate, high cost of the catalyst material, and overpotential losses at electrodes are considered as the main obstacle that restricts its efficiency for bioremediation processes at higher scales (Singh and Kumar, 2022). For example, Zepilli et al. (2021) reported that phosphate buffer in the anode could maintain the neutrality of the solution's pH for short periods. However,

Table 5
Application of BESs for removal of petroleum hydrocarbons from soil.

Type of pollutant	Configuration of the BESs and operating condition of the reactor	Microorganisms	Remediation efficiency rate (%)	Reference
Petroleum hydrocarbons	Petroleum hydrocarbons act as electron donor and glucose, with Tween 80 in single-chambered microbial fuel cells	Activated Sludge	45	Zhao et al. (2019)
Nitrobenzene C ₆ H ₅ NO ₂	Acetate acts as electron donor and nitrobenzene as acceptor in continuous dual-chambered microbial fuel cells	Mixed cultures	1.3	Mu et al. (2009)
Petroleum hydrocarbons	Petroleum hydrocarbons act as electron donors and oxalic water as an electron acceptor in Bottle-type dual-chamber reactors microbial fuel cells		29	Wang et al. (2020)
Furfural C ₅ H ₄ O ₂	Furfural act as electron donor and oxygen as an electron acceptor in batch Single-chambered microbial fuel cells	Mixed cultures	95	Luo et al. (2010)
Diesel oil	Diesel act as electron donor and oxygen as acceptor in Tubular microbial fuel cells with air-cathode	Microbial Anaerobic Consortium	59	Wang et al. (2019)
1,2-Dichloroethane C ₂ H ₄ Cl ₂	Dichloroethane act as electron donor and oxygen as acceptor in continuous dual-chambered microbial fuel cells	Microbial Anaerobic Consortium	95	Pham et al. (2009)
Diesel oil	Diesel act as electron donor and oxygen as an electron acceptor in batch dual-chambered microbial fuel cells	Mixed cultures	83	Cheng et al. (2017)
4-nitrophenol	Glucose acts as electron donor and 4-nitrophenol with H ₂ O ₂ in batch dual-chambered microbial fuel cells	Sludge from anaerobic	100	Zhu and Ni. (2009)
Petroleum hydrocarbon	Petroleum hydrocarbon act as an electron donor, and oxygen act as an acceptor in continuous dual-chambered microbial fuel cells	-	17	Li et al. (2019)
Petroleum hydrocarbons	PHs act as electron donors and oxygen as an electron acceptor in batch U-tube Single-chambered microbial fuel cells.	Microbial flora is mixed and extracted from the soil contaminated with saline and petroleum	15.2	Wang et al. (2012)
Phenanthrene	Phenanthrene acts as an electron donor, and oxalic water acts as an electron acceptor in batch sediment microbial fuel cells	Mixed microbial extracted from the polluted site	99	Yan et al. (2012)

this develops overpotential at the anode, resulting in acidification, leading to microbial activity inhibition. The other problems in which BESs hindered applying on a large scale are the ohmic losses, low substrate bioavailability, and a clear understanding of the microorganism with the transfer of electrons (Gupta et al., 2022). The use of bio-cathodes, biosurfactants, electro-chemically active microbes, and redox mediators can solve the electrical problems encountered to remediate soil polluted with PHs. However, the application of use materials integrated is still in the initial stage and needs more research and investigation.

6. Characterization of remediation processes

The effectiveness, predictability, and reliability of remediation technology are key factors in its performance. These criteria must be monitored regularly to be adopted on a broader scale. In addition, Treatment-specific monitoring protocols that are statistically sound are needed. Also, replication, proper sampling, and data processing are critical components of monitoring. There is a range of main biological and analytical approaches available today to track soil remediation processes (Kour et al., 2021).

6.1. Biological methods

Environmental remediation technologies have grown in prominence as a long-term strategy for restoring contaminated areas. To enable the proper implementation of such protocols, efficient and accurate techniques for monitoring environmental clean-up must be created. Several analytical approaches have been established over time to monitor the environmental clean-up process. These approaches, however, have several flaws, including poor efficacy and the inability to discern between biological and non-biological deterioration. As a result, biological or molecular methods are considered an excellent tool for microbial identification. It also provides information on the concentration of substrates and their fate. They are primarily dependent on the metabolic and abundance activities of microorganisms involved in bioremediation. Hence, there are two types of methods that can be used for this: culture-based and culture-independent (Kour et al., 2021). Culture-dependent methods like plate count, carbon source utilization tests, most probable number, transcriptomics, and proteomics are also considered methods for microbial identification and bioremediation (Daveray et al., 2019). The enzyme-related immunosorbent assay is the most widely used immunochemical tool for identifying and monitoring bioremediation techniques. For instance, according to Ko et al. (2019), their research to analyze and identify the microcystin formed by the cyanobacteria during the bioremediation process of eutrophic water using immunochemical tools shows high efficiency for controlling the bioremediation process. Culture-dependent methods cannot analyze the entire population at contaminated sites because only about 1% of all microorganisms are culturable. As a result, a modern technique known as the culture-independent approach replaces conventional recognition methods. These fingerprinting approaches do not need to cultivate and include a complete profile of microbial communities and have a high throughput, effective and repeatable such as metagenomic approaches. In addition to this, polymorphic DNA random amplification, restriction fragment length polymorphism, sequence typing, single-strand conformation polymorphism, fluorescence in situ hybridization, repetitive sequence-based PCR, reverse sample genome probing, PCR, and phospholipid fatty acid analysis can also use for monitoring and characterization of bioremediation process that includes microorganisms (Srivastava et al., 2019). However, a lack of standardization and reliability is also noted among molecular techniques when applied to complex and long-term contaminated soils and wastewaters. Another effective alternative is widely used to characterize the bioremediation process is the enrichment methods. The basic idea behind bromodeoxyuridine methods enrichment is that metabolically active organisms can

integrate a branded nucleotide bromodeoxyuridine method into their DNA, which can then be extracted based on the inserted mark and mainly used to enrich microbes that have been grown on xenobiotic compounds (Bhadury and Ghosh, 2022). Besides, another enrichment process is stable-isotope probing, which includes soil bacterial species providing a ^{13}C -labeled substrate. This proves that the microorganism using substrate integrates ^{13}C in their DNA, making it denser than standard DNA containing ^{12}C , allowing the metabolically active cells to be identified. The technique of stable isotope probing is also applied to monitor biological effects (Wang et al., 2022). For instance, Kasanke et al. (2019) used this technique to identify and characterize *Rhodoferrax* sp, degrading sulfolane successfully. Recently, microbial biosensors and microbial fuel cells are also used to assess bioremediation processes (Jadhav et al., 2021). Tang et al. (2008) developed a laccase enzyme-based biosensor to monitor catechol in bioremediation. Hence more research and investigation need in synthetic biology and genetic engineering to facilitate the design and development of microbial biosensors for efficient and accurate monitoring and assessment of bioremediation as well as to employ a sustainable tool for the rehabilitation of environmental remediation technologies.

6.2. NGS technology

NGS technology provides unique opportunities in PHs bioremediation and biodegradation research (Mangse et al., 2020). It offers new insights into the ecology of microbially mediated processes that influence the degradation of pollutants in the soil. NGS technology enables parallel analysis of DNA sequence information about nucleic acids, used for environmental quality assessment. It is applied to identify novel genes, pathways, and even microbial shifts involved in the degradation of hydrocarbons. In addition, NGS technology provides reliable information about critical enzymes involved in various detoxifying types of pollutants. Thus, they became a valuable tool for quality monitoring and data management based on contaminated soil sites (Chandran et al., 2020). NGS technology consists of library preparation, sequencing, base calling, established genome alignment, and assorted annotation. Library preparation starts with multiple DNA fragments, enzyme digestion, followed by sequences (López, 2020). However, they are typically more costly and need more complex devices and reagents than other advanced statical and bioinformatics tools.

6.3. Analytical methods

Analytical methods are at the core of remediation science, and it is difficult to assess without them. In situ monitoring of bioremediation with electrochemical electrodes is the preferred process, particularly in anoxic environments (Kour et al., 2021). They may detect several cations and as well anions (Yan and Reible, 2015). However, this technique was less successful in the case of hydrophobic organic compounds, such as PAHs (Jasmine and Mukherji, 2019). Pollutants, various intermediates contaminants can be quantified using spectrometry spectroscopic and chromatographic techniques. This is another way to track the bioremediation method. These entail thorough chemical and physical characterizations of the target molecules and their degraded intermediates (or final products), with high specificity, sensitivity, and reproducibility (Kour et al., 2021).

Gas chromatography (GC), high-performance liquid chromatography (HPLC) is among the chromatographic methods that are widely used to track the remediation of pesticides, petroleum products, oily sludge, hydrocarbons, and heavy metals (Ambaye et al., 2021a). The combination of GC and mass spectrometry is an excellent way to identify pollutants and their intermediates with both standardized and appropriate methods of purifications of soil samples (Ambaye et al., 2021a; Patil et al., 2022). In addition to this, various spectroscopy techniques like nuclear magnetic resonance, atomic absorption Fourier transform infrared, and 3D-fluorescence spectroscopy, X-ray diffraction,

spectroscopy, and Raman spectroscopy are used to monitor and characterize various pollutants, including PAHs, PHs, and petroleum sludge (Ambaye et al., 2021; Patil et al., 2022; Shrivastava et al., 2019). From the above spectroscopy techniques, atomic absorption spectroscopy and Fourier transform infrared spectroscopy are two of the most cost-effective, responsive, and reliable but less accurate methods available. Another way to classify the pollutants is by using the pollutants' physical and chemical properties due to thermal treatment (Ziglio et al., 2019). In general, petroleum pollution has a significant negative impact on humans and the environment. Researchers from all over the world are looking into bioremediation to minimize this type of pollution safely. Analytical scientists can provide a variety of answers in this area with the use of modern analytical techniques. Various analytical techniques have evolved because of the extensive research conducted in this area, and it has become clear that proper sample preparation is critical in determining the validity of quantitative hydrocarbon assay results. The use of GC-based analysis for monitoring analytes and their metabolites is obvious. Based on metabolomic profiling through Gas chromatography-mass spectrometry and Liquid chromatography-mass spectrometry, degradation pathways are determined. Other analytical techniques such as Fourier-transform infrared spectroscopy, Nuclear magnetic resonance, Liquid chromatography-mass spectrometry, and high-performance liquid chromatography are being used more frequently in bioremediation research to determine petroleum pollutants.

7. The challenges in adopting sustainable remediation technology

Selecting and adopting an appropriate sustainable remediation technology is dependent upon various factors which can affect the remediation process, efficiency, and suitability, including abiotic and biotic, scientific, and non-scientific factors, as well as having a shortage of clear scientific evidence regarding these factors, how they can affect the efficiency and remediation process during real application (Ossai et al., 2020). Hence it is necessary to have a complete understanding and knowledge of Physico-chemical properties, contaminate and environmental properties, biological properties, source, composition, age, type,

heterogeneity, concentration, site conditions, process, geohydrologic location, monitoring difficulties, space requirement, extended treatment time, required level of clean-up standard, remedial approach, and strategy, risk management strategy, life-cycle analysis, cost-benefit ratio, outcome, ability to achieve the limitations and favorable regulatory perception is necessary to select and predict sustainable remediation technology for soil polluted with petroleum hydrocarbon (Kuppusamy et al., 2016a; Ossai et al., 2020). In addition to the above factors, to achieve complete application of the concepts of sustainable remediation, it should be addressed as a new way of thinking about contaminated area rehabilitation, where the integration of economic, environmental, and social variables is all considered as a basic element in decision-making. Academics, governments, and industry all play a critical role in ensuring that sustainable remediation progresses and overcomes existing problems so that both developing and developed countries can effectively include sustainability into remediation initiatives. Proposed strategies for sustainable remediation and management of soil contaminated with petroleum hydrocarbon are shown in Fig. 8 (Kuppusamy et al., 2020; Thomé et al., 2018).

8. Conclusion and future perspective

This review paper addresses the leading available soil-remediation technologies to restore soil used at both lab and field scales. Various remediation technologies are being practiced, but no single approach can be applied to all environmental conditions to reduce PHs from polluted soil and other contaminated sites. Hence, this review paper provides, first, a better understanding of PHs compositions, properties, main transportations routes, and their fate into soil and environment. It is essential to mention that all remediation process parameters considerably impact the decision-making process and enhance remediation effectiveness. Therefore, nowadays, eco-friendly approaches and inventing novel PHs-degrading mechanisms towards restoring contaminated soils with cost-effectiveness are highly requested. Essential strategies for managing PHs and the design-making process are also needed for removing pollutants from the contaminated sites to achieve long-term sustainability goals. Parallely, the development of risk-based management tools based on databases, machine learning platforms,

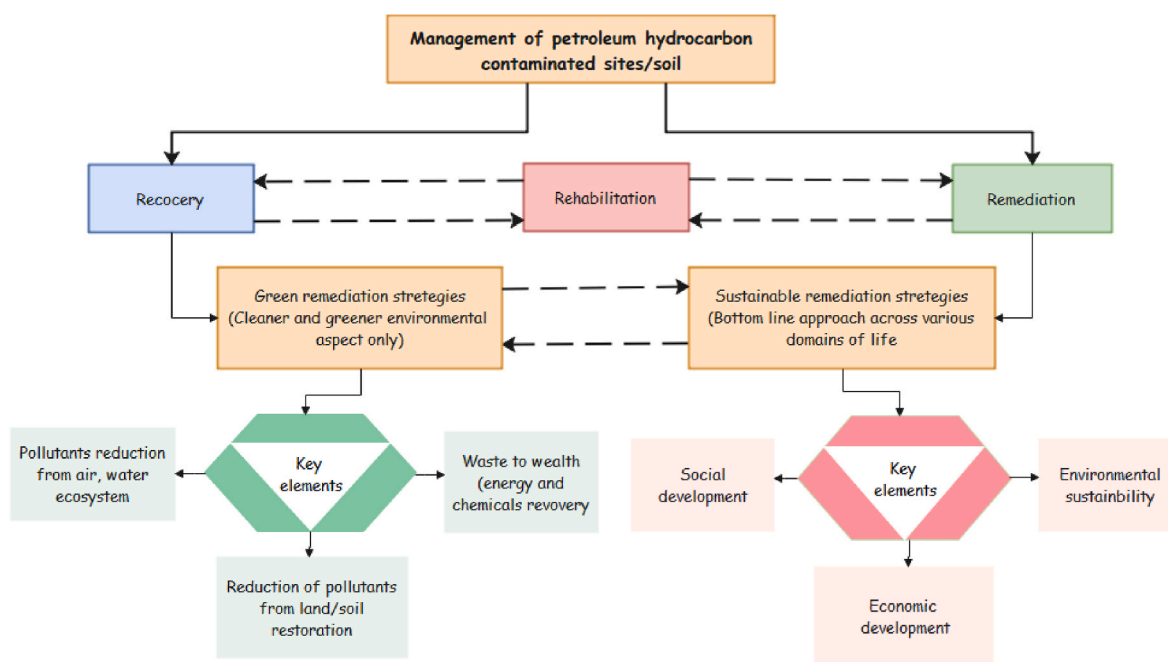


Fig. 8. Proposed approaches for sustainable remediation and management of petroleum hydrocarbon contaminated soil (Kuppusamy et al., 2020; Thomé et al., 2020).

LCA and LCCA studies, use of next-generation sequencing (NGS), the cost-effectiveness of nanotechnology and electrochemically active microorganisms-based, bio-electrochemical systems must be given more emphasis to design the appropriate solutions for soil contamination and with better future perspectives. More focus should be given on assessment approaches to determine the efficacy of remediation in the development of new *in-situ* remediation technologies and as well as in the development of new open-source software (for low-income countries). That can be applied to reduce PHs from polluted soil in all environmental conditions as sustainable and eco-friendly soil remediation technologies that can be used at several sites in the world with different contamination levels.

Author contributions statement

Teklit Gebregiorgis Ambaye: Conceptualization, Writing – original draft & editing last version, Alif Chebbi: reviewing & editing the last version, Francesca Formicola: reviewing Shiv Prasad: reviewing & editing the last version, Franco Hernan Gomez: reviewing, Andrea Franzetti: Conceptualization, Supervision, Writing – review & editing last version. Mentore Vaccari: Conceptualization, Supervision, Writing – review & editing last version.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the Italian Agency for Development Cooperation through the partnerships for knowledge (PFK) program and Fondazione Cariplo, which funded the project CREIAMO (D76C19000230007).

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