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Review

# Plant-assisted remediation of hydrocarbons in water and soil: Application, mechanisms, challenges and opportunities

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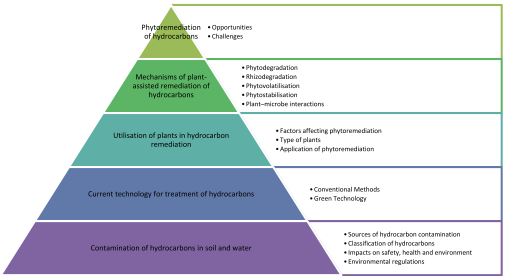
## HIGHLIGHTS

- A comprehensive knowledge about the hydrocarbons removal from ecosystem is important.
- Processes to remove hydrocarbons in the contaminated water and soil are presented in this review.
- Plant-assisted remediation can be potentially effective to treat water and soil contaminated with hydrocarbons.
- Challenges and opportunities in utilising phytoremediation technology for hydrocarbon removal.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Due to the increasing importance of diesel and petroleum for industrial development during the last century, petrochemical effluents have significantly contributed to the pollution of aquatic and soil environments. The contamination generated by petroleum hydrocarbons can endanger not only humans but also the environment. Phytoremediation or plant-assisted remediation can be considered one of the best technologies to manage petroleum product-contaminated water and soil. The main advantages of this method are that it is environmentally-friendly, potentially cost-effective and does not require specialised equipment. The scope of this review includes a description of hydrocarbon pollutants from petrochemical industries, their toxicity impacts and methods of treatment and degradation. The major emphasis is on phytodegradation (phytotransformation) and rhizodegradation since these mechanisms are the most favourable alternatives for soil and water reclamation of hydrocarbons using tropical plants. In addressing these issues, this review also covers challenges to retrieve the environment (soil and water) from petroleum contaminations through phytoremediation, and its opportunities to remove or reduce the negative environmental impacts of petroleum contaminations and restore damaged ecosystems with sustainable ways to keep healthy life for the future.

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**Contents**

## 1. Introduction

The crude oil (petroleum) industry produces waste, such as wastewater, sludge and, to a lesser extent, gas. If improperly handled, these wastes will contaminate the environment and damage industrial activity. According to Nadim et al. (2000), crude oil is a complex mixture of hydrocarbons comprising an elemental composition that includes carbon, hydrogen, oxygen and sulphur, together with trace amounts of nitrogen, metals and other elements. Modern refineries utilise a cracking process that heats hydrocarbon compounds to high temperatures above 230 °C at different pressures with or without a catalyst, to remove unused components. This process transforms the complex hydrocarbons into simpler molecules with lower boiling points through several dehydrogenation processes. During the refinery process, various petroleum products, ranging from methane to asphaltene, are produced (Nadim et al., 2000; Kronenberg et al., 2017; Zhang et al., 2017).

Newman and Reynolds (2004) argue that most problems with petroleum compounds are associated with contaminated soil and groundwater. Due to the industrial use of petroleum products and a long history of oil production, particularly in production and pipeline areas (White et al., 2006), petroleum hydrocarbons are the most commonly detected organic pollutant in the environment (Collins, 2007). Disposal of waste from the petroleum industry and oil spills from well explosion, power plants and pipeline breaks are the predominant causes of petroleum contamination, with effects on plants, animals, humans and the environment. Crude oil extraction may even have an impact on climatic change (Offor and Akonye, 2006). As Yakubu (2007) mentioned, released oil is a well-recognised problem in today's world, especially for oil-producing countries that extract and process oil (Ogbo and Okhuoya, 2008). The most common groups of organic contaminants are total petroleum hydrocarbons (TPH), which are known to be toxic to many organisms. Petroleum extraction and refining constitute the greatest source of TPH contamination in soils (Huang et al., 2005). This contaminant can affect human health following indoor and outdoor exposure. Indoor exposure occurs when contaminated tap water is used, while outdoor exposure can occur through inhalation and dermal absorption. A wide range of volatile compounds present in water supplies can impose risks on humans exposed to these contaminants during indoor activities (López et al., 2008), including polycyclic aromatic hydrocarbons (PAHs) that can have serious effects if released into the environment (Augulyte et al., 2008) and contaminate sediments (Bert et al., 2009).

Several methods to treat contaminated soil with organic compounds are thermal desorption (Vidonish et al., 2016), soil washing (Kumpiene et al., 2017), incineration, landfilling and microbiological treatment. Petroleum hydrocarbons and PAHs are susceptible to microbiological treatment. However, applying this method under field conditions cannot always treat the pollutant successfully since changes in weather conditions may not allow the microorganisms to completely remove pollutants from the environment (Singh and Jain, 2003). Moreover, nutrient and oxygen limitations (Cohen et al., 2002; Adam and Duncan, 2003; Hawrot and Nowak, 2006), bioavailability (Hawrot and Nowak, 2006), moisture and, occasionally, temperature (Singh and Jain, 2003) are critical factors that affect microbiological treatment.

There are many traditional methods to treat water or soil contaminated with organic matter, with low and high removal efficiency. Choi et al. (2016) observed the treatment of PAHs-contaminated sediment using retrievable activated carbons, with 50–60% PAHs removal efficiency. An appropriate technique for treating marine sediments is electrokinetics, although, Yan and Reible (2015) noted its limitation in PAH removal of less than 60%, together with high costs. Ferrarese et al. (2008) and Wang et al. (2016) achieved comparatively higher removals of PAHs through chemical oxidation and extraction processes, but these methods are costly due to chemical usage. Falciglia et al. (2018) proved the concept of a combined membrane, microwave heating with ultra-violet irradiation (MW-UV-A) for the successful remediation of PAH-contaminated marine sediments.

Varjani (2017) described the physical factors of hydrocarbon degradation. Temperature can affect petroleum biodegradation by altering the physical nature and chemical composition of the oil, the metabolism rate of hydrocarbons by microbes and the composition of the microbial culture. Low temperatures can increase the viscosity of the oil, reduce the volatilisation of toxic short-chain alkanes and increase their water solubility, thereby retarding the biodegradation process. Rates of degradation generally decrease with reducing temperature. The maximum rate of hydrocarbon metabolism is between 30 and 40 °C, but above 40 °C, the poisoning of microbes by hydrocarbons is increased. Oxygen is generally required for biodegradation processes since major degradation pathways for microbial oxidation of both aromatic and saturated hydrocarbons rely on molecular oxygen and oxygenases. The oxygen availability in soils is determined by the rates of microbial oxygen consumption, the soil type and the presence of utilisable substrates that increase microbial oxygen consumption, promoting oxygen depletion.

An alternative method to treat contaminated water and soil with organic compounds is phytoremediation or phytotechnology, which uses plants to detoxify contaminants. This technology has been used widely and successfully, mainly in developed countries, like Europe, the USA and Japan, to treat organic and inorganic wastes in the form of liquid as wastewater and solids in sludge or contaminated soil. This review paper will focus on the water and soil contamination with hydrocarbons. In Malaysia, several studies have examined the remediation of both organic (Al-Baldawi et al. 2013b; Almaamary et al., 2017; Almansoory et al., 2017; Alanbary et al. 2018, 2019; Sanusi et al., 2016; Kadir et al., 2018; Yusoff et al., 2019) and inorganic contaminants (Ismail et al., 2019; Selamat et al., 2018; Titah et al., 2018; Tangahu et al., 2013) through phytoremediation. Microorganisms that surround plant roots are known as rhizobacteria. These bacteria have a significant role in phytotechnology, as they work symbiotically with plants in the phytoremediation process to degrade organic compounds. Petroleum hydrocarbons are rapidly degraded in the rhizospheric region (Singh and Jain, 2003; Al-Baldawi et al., 2017). Additionally, there are also fungi and other types of organisms that play a role in phytoremediation (Singh and Jain, 2003; Hamdi et al., 2007; Ogbo and Okhuoya, 2008; Badri et al., 2009). Some literature reviews about the remediation technologies for organic pollutants include treatment technologies for PAH-contaminated sites (Gitipour et al., 2018), thermal treatment of hydrocarbon-impacted soils (Vidonish et al., 2016) and biodegradation of PAHs (Haritash and Kaushik, 2009).

To explore the application of phytotechnology in removing hydrocarbons from contaminated soil and water, information searching was conducted from Google Scholars and ScienceDirect database. Search terms including [source of petroleum hydrocarbon contamination], [impacts of hydrocarbon contamination], [current technology to remove hydrocarbon from soil/water], [phytoremediation/phytotechnology of hydrocarbons], [mechanisms of phytoremediation/phytotechnology], [application of phytoremediation for hydrocarbon removal], [constructed wetlands] and [plant-microbe interaction in phytoremediation] were searched to gather basic information related to the petroleum hydrocarbon contamination and its impact, and current technology to treat hydrocarbon in soil and water, and also the application of phytoremediation for petroleum hydrocarbon removal. All the information obtained are summarised in tables compiling research findings on the application of phytoremediation to remove hydrocarbons, and also simplified in figures to illustrate the source of hydrocarbon pollution in the ecosystem, classification of hydrocarbon, current technology to remove hydrocarbons and factors affecting phytoremediation. Based on the gathered information, the manuscript was written and arranged to firstly discuss on the possible source of hydrocarbon contamination in water and soil, classification of hydrocarbons, its impact on safety and environment, current technology to remove hydrocarbon including phytoremediation. Later the manuscript focusses on the utilisation of plant-assisted remediation for hydrocarbons, its mechanisms, finally the challenges and opportunities of this technology.

This review addresses the challenges and opportunities for petroleum phytoremediation as an alternative biotechnology approach or as an enhanced treatment system for soil or wastewater. The topics covered include common treatment methods for hydrocarbon wastes, definitions and mechanisms of phytoremediation, interactions between plants and microbes to degrade hydrocarbons and, also, the application of various plant species for hydrocarbon remediation. This information is part of our team initiative to search for native plants in Malaysia suitable for hydrocarbon phytoremediation in the petroleum industry. The conclusions obtained in this review will be used to support plant utilisation for the degradation of petroleum hydrocarbons.

## 2. Sources of hydrocarbon contamination

As illustrated in Fig. 1, the discharge of oil-containing sludge and wastewater to the environment increases annually due to urbanisation and industrial development activities from petrochemical industries, oil transportation and accidental petroleum spills for soil contamination, and from oily wastewater discharge, petroleum drilling/refining processes and leakage to underground storage tanks for water contamination (Cai et al., 2010; LeFevre et al., 2012). Hydrocarbons in water can be in the form of free-floating, emulsified, dissolved or adsorbed to suspended solids. A hydrocarbon is fundamentally an organic chemical compound composed of hydrogen, carbon and trace elements of heavy metals. Mixtures of compounds with carbon numbers ranging from $C_5$ to $C_{36}$ originating from petroleum, otherwise known as TPH, are typically analysed using the USA Environmental Protection Agency (USEPA) method 3510C (USEPA, 2011). The petroleum oil can be classified into three types, based on its components: (i) saturated hydrocarbons, which include non-cyclic hydrocarbons (paraffins), cyclic hydrocarbons (cycloalkanes) and olefinic hydrocarbons (alkenes); (ii) aromatics, such as benzene; (ii) non-hydrocarbons, such as sulphur compounds, nitrogen–oxygen compounds and heavy metals (Cote, 1976). According to Barrutia et al. (2011), diesel contains short-chain ($C_{12}$–$C_{16}$), medium-chain ($C_{18}$–$C_{22}$) or long-chain ($C_{24}$–$C_{30}$) alkanes.

Petroleum hydrocarbons are organic compounds naturally found in the earth and as crude oil, asphalt and coal. Petroleum hydrocarbons can also be in the form of gas (as natural gas), liquid (as crude oil) and solids. Petroleum hydrocarbons consist mainly of hydrogen and carbon, but sometimes include nitrogen, sulphur and oxygen (Nadim et al., 2000; Kirk, 2005). Posthuma (1977) described the general composition of crude oil (in % mass) as 70–80% carbon, 10–15% hydrogen, 0–10% sulphur, 0–1% nitrogen and 0–5% oxygen.

Crude oil can be described as a combination of hundreds of hydrocarbon compounds, which differ in size from the smallest, such as methane with only one carbon atom, to large compounds having 300 or more carbon atoms (Jones and Pujado, 2006). Paraffins or paraffin isomers are the main hydrocarbon compounds. Almost all of the remaining hydrocarbons are either cyclic paraffins, known as naphthenes or heavily dehydrogenated cyclic compounds, such as the aromatic family of hydrocarbons. Thereby, Jones and Pujado (2006) classified hydrocarbons in crude oils into four groups: paraffins, cyclic paraffins, aromatics and the unsaturated or olefinic hydrocarbons. As expected, the unsaturated hydrocarbons do not exist during the handling of crude oil to refined products.

Dowty et al. (2001) categorised crude oil compounds according to solubility, volatility and susceptibility of the hydrocarbons to degradation by microorganisms. The extent of degradation decreases when the number of condensed rings in the aromatic hydrocarbon structure increases, while the aliphatic fraction of hydrocarbons, such as alkanes and alkenes, are the most easily

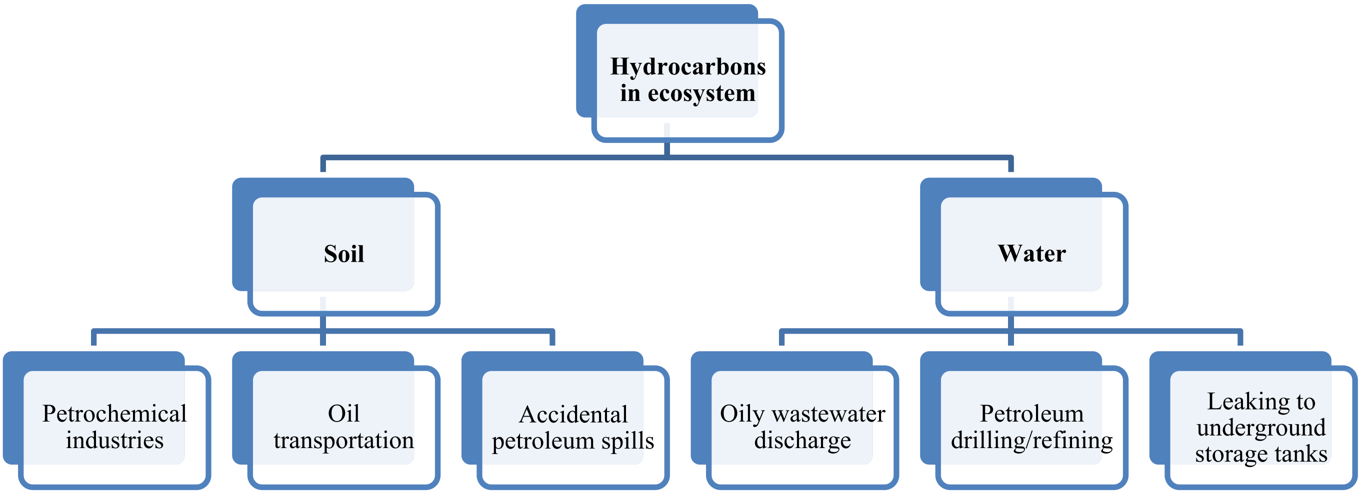

**Fig. 1.** Major sources of hydrocarbons in the ecosystem.

degraded (Dowty et al., 2001). Conversely, the asphaltic fraction is very complex and not easily degraded by microorganisms due to the increased presence of double covalent bonds and branching.

Several products are obtained by the refinement of crude oil and described depending on the intended use (Kirk, 2005). Diesel, gasoline, kerosene and heating fuels are refined petroleum products containing a complex mixture of hydrocarbons. Refined petroleum products are also categorised based on the carbon range of the components and the boiling point. For example, gasoline is usually composed of compounds with C5−C10 and has a boiling point of 30−200 °C, while diesel is usually composed of C12−C18 compounds, with a boiling point of 160−400 °C. The source of crude oil also has a significant influence on the composition of each finished product.

### 2.1. *Classification of hydrocarbons*

The two primary categories of hydrocarbons, aliphatic and aromatic, are sub-divided according to the general chemical structure of their constituent chemicals. Aliphatic hydrocarbons contain chains of linked carbon atoms, whereas aromatics contain one or more benzene rings bonded together (Epps, 2006; Hunt et al., 2019), as depicted in Fig. 2. Aliphatic hydrocarbons can be further divided into three main groups: alkanes, alkenes and cycloalkanes. Chemically, aliphatic and aromatic compounds can be differentiated by the patterns of bonding between adjacent compounds (The Interstate Technology and Regulatory Council 2003).

Alkanes (both linear carbon-chained and branched carbon-chained) are simple compounds that are characterised by single carbon−carbon bonds (Tara et al., 2014; Befkadu and Quanyuan, 2018). The more common alkanes include methane, butane and propane, and are components of gasoline, jet fuel, diesel and kerosene. Alkanes can be chlorinated with one or more chlorine atoms, forming a category of chemicals called volatile organic compounds, which includes common environmental pollutants, such as trichloroethylene, tetrachloroethylene and vinyl chloride. The general formula of alkanes is $C_xH_{(2x+2)}$. Alkenes have one or more double bonds between carbon atoms, while cycloalkanes are alkanes in which the carbon atoms form a ring. Alkenes and cycloalkanes are found almost exclusively in gasoline and jet fuel. The general formula of alkanes is $C_xH_{2x}$ (Hou et al., 2015; Zhang et al., 2013).

Aromatic compounds have one or more benzene rings as structural components. Benzene is a carbon ring that always consists of six carbons atoms and six hydrogen atoms ($C_6H_6$). The more common and simple aromatics encountered as environmental pollutants include benzene, toluene and xylene. A ubiquitous group of aromatic compounds is the PAH compounds, which occur as a result of chemical manufacturing or naturally in the environment as the result of organic degradation or incomplete combustion (The Interstate Technology and Regulatory Council 2003).

According to Balseiro-Romero et al. (2018), the qualitative hydrocarbon content of the petroleum mixture influences the degradability of individual hydrocarbon components. Biodegradation potentials decrease in the order of hexadecane > naphthalene ≫ pristane > benzanthracene. Alkanes and low-molecular-weight aromatics (benzene, toluene, naphthalene and methylnaphthalene) can be degraded to $CO_2$ by microorganisms in river water, but higher-molecular-weight aromatics are relatively resistant to microbial degradation (Jabbar et al., 2017). Polyaromatic hydrocarbon turnover times in sediments contaminated with hydrocarbons vary from 7.1 h for naphthalene to 400 h for anthracene, 10,000 h for benz[a]-anthracene and over 30,000 h for benz[a]-pyrene. Polynuclear aromatic compounds tend to be only partially, rather than completely, degraded to $CO_2$ (Daccòa et al., 2020). In agreement with this, saturated compounds are described as the most biodegradable among hydrocarbons, followed by high-molecular-weight, single-ring aromatics and then polar compounds as the least biodegradable. Hydrocarbons within the saturated fraction include *n*-alkanes, branched alkanes and cycloalkanes (naphthenes). The *n*-alkanes are considered the most easily degraded in a petroleum mixture. The bacterial degradation of aromatic compounds normally involves the formation of a compound with two hydroxyl groups, known as a diol, followed by cleavage and release of a diacid, such as *cis*,*cis*-muconic acid. Contradictorily, aromatic hydrocarbons can be oxidised in eukaryotic organisms to form a *trans*-diol. The metabolic pathways for the degradation of asphaltic components of petroleum are probably the least understood. These asphaltic components are complex structures, which are difficult to analyse by current chemical methods (Atlas, 1981; McIntosh et al., 2017). The terminology widely used to describe hydrocarbon groupings is summarised in Fig. 3 and it includes BTEX (benzene, toluene, ethylbenzene, xylene), organic and inorganic, PAHs, TROG (total recoverable oil and grease), TPH and TRPH (total recoverable petroleum hydrocarbons).

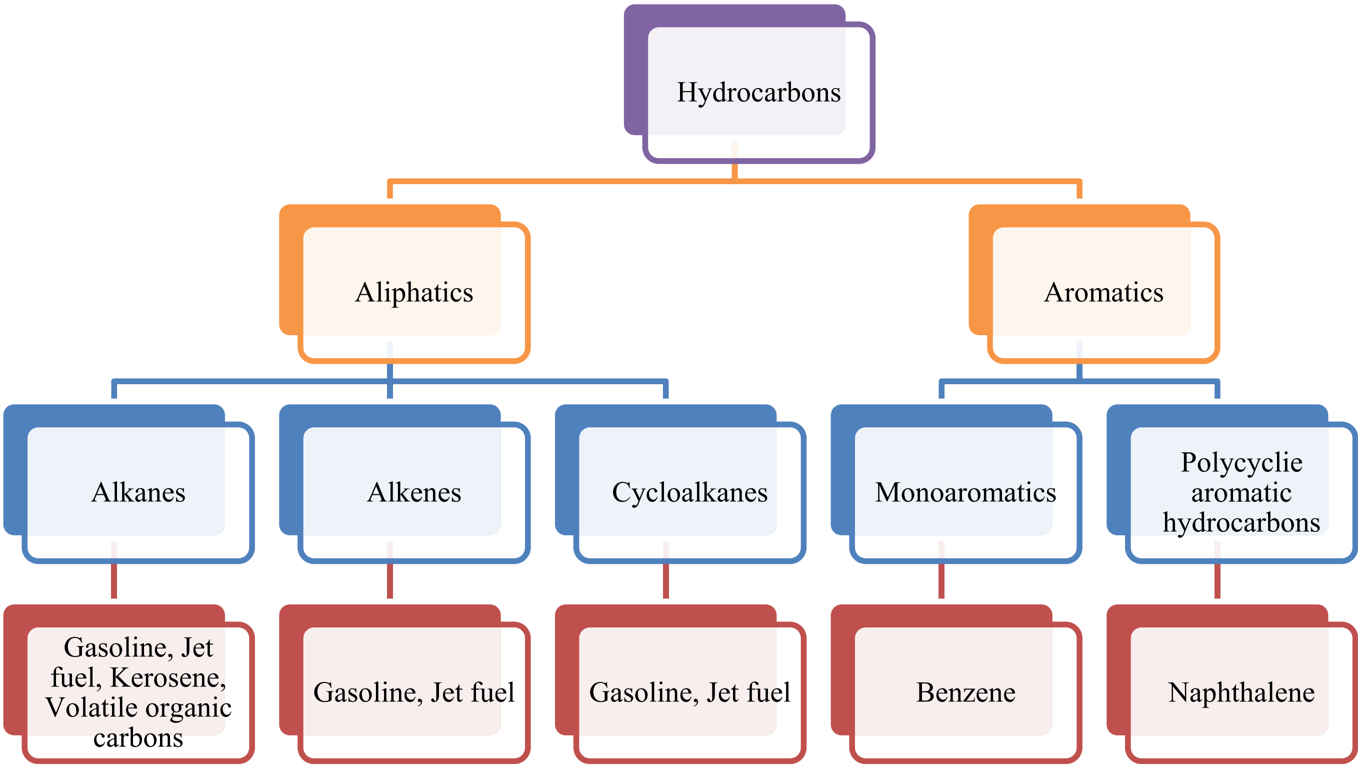

**Fig. 2.** Classification of hydrocarbon compounds.

### 2.2. Toxic effects of hydrocarbons on health and the environment

Hydrocarbons are the most common category of environmental pollutants traced in industrialised countries. During exploration, production, refining, transport and storage of petroleum and petroleum products, there are always possibilities for leakage, pipe rupture and accidental spills. Hydrocarbons are a pollution problem for marine ecosystems and impact almost directly on human health (Cazoir et al., 2012). Hydrocarbons can become hazardous, especially if they enter the food chain since several hydrocarbons are persistent, including polycyclic PAHs and polychlorinated biphenyls, which can contribute to toxic, mutagenic and carcinogenic effects (Perelo, 2010; Lors et al., 2012). Many reports around the world have shown the adverse effects of petroleum hydrocarbons available in air, water and soil on people health such as psychological problems, respiratory tract irritation, skin and kidney problems, and disturbance of blood profile (Jeevanantham et al., 2019; da Silva and Maranho, 2019). The toxicity of petroleum hydrocarbons has effects not only on human health but also on plants, soil microorganisms and the sustainability of ecosystems. Toxic effects are seen in plants grown on oil-contaminated soil, such as beans, with an oil concentration of 10,000 mg/kg (Baek et al., 2004), which showed slow growth and a reduced percentage of seed germination. These effects are also found in some plants due to diesel oil contamination in soil (Adam and Duncan, 2002). White clover (*Trifolium repens*) exhibited a significant reduction in photosynthetic pigments after 2 months in diesel oil-contaminated soil (Barrutia et al., 2011). Effects on invertebrates often occur at TPH concentrations lower than those associated with effects on plants (Efroymson et al., 2004). Vandermeulen et al. (1983) stated that the marine unicellular alga *Pavlova lutheri* Droop responded to changing patterns of petroleum hydrocarbons with variable motility. In an earthworm bioassay, oily soil was acutely toxic to

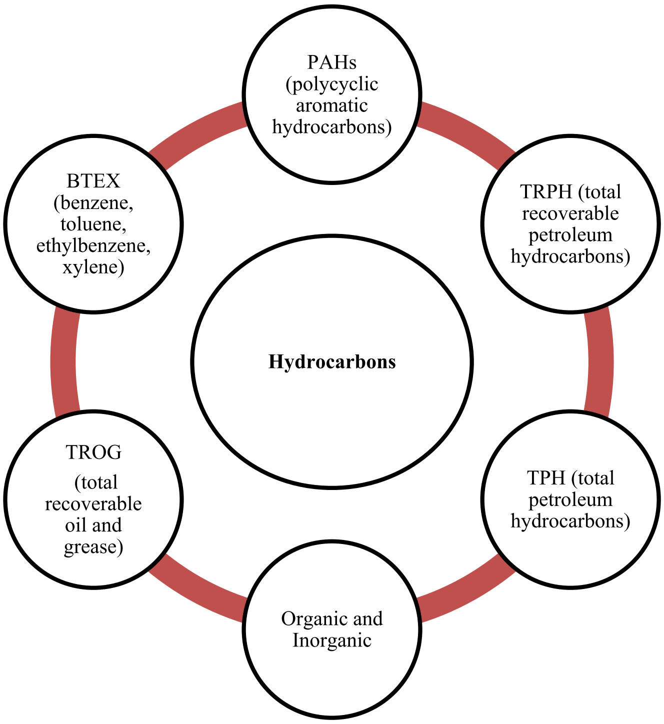

**Fig. 3.** Classification of hydrocarbon groupings.

*Eisenia* in the first 2–4 weeks of bioremediation experiments (Salanitro et al., 1997).

### 2.3. Environmental regulation of hydrocarbon contamination

Release of process wastewater from utility operations to surface water must not result in contaminant concentrations in excess of local ambient water quality criteria. Standards of permissible concentrations for each toxic substance are set ultimately for the protection of human health and preservation of the ecosystem. The level of hydrocarbons permissible in polluted soil (Table 1) and water (Table 2) differs by country. Generally, there are only concentrations for oil and grease. No hydrocarbon specifically exists in the environmental regulations of hydrocarbon contamination.

## 3. Current technology for treatment of hydrocarbon-contaminated water and soil

Some criteria for selecting technology or land treatment are the effectiveness of short-term and long-term treatments to meet the goals of rehabilitation, the effectiveness of pollutant reduction, reduction of pollutant toxicity and cost-effectiveness (Pavel and Gavrilescu, 2008). Several methods of decontamination of contaminated land are land farming, bioventing (Azubuike et al., 2016), natural attenuation (Pavel and Gavrilescu, 2008), biosparging, vitrification, incineration, physical–chemical soil washing, soil vapour extraction and electrokinetic methods (Gomes et al., 2013). Existing methods for treating contaminated soil can be categorised into *in situ* and *ex situ* techniques. Fig. 4 simplifies the treatment methods for contaminated water and soil treatment. *In situ* techniques include bioventing, biosparging, bioslurping and phytoremediation, which do not incur high costs, as the cost of excavation and transportation can be avoided. *Ex situ* techniques include landfarming, biopiling and bioreactor processes, which are more comprehensive remediation methods and incur high costs due to the requirement for excavation and ground transportation. For water treatment, there are various treatment systems applied for hydrocarbon contamination, such as chemical/physical methods through skimming, and biological approaches through booms, biosparging and bioremediation (Prince, 2014). These treatment systems can be grouped into *in situ* (on contaminated site itself) and *ex situ* (the wastes are shifted to another place for treatment) (Yu et al., 2011; Jabbar et al., 2018). Phycoremediation refers to the use of microorganisms or microalgae for the removal or biotransformation of contaminants from water, soil and air (Peng et al., 2009). Phycoremediation is a sustainable contamination remediation technique by natural resources (Yang et al., 2009).

Contamination of petroleum hydrocarbons exists not only in soil but also in groundwater (Erakhrumen, 2007; Zhang et al., 2017; Ossai et al., 2020). The existing treatment methods for petroleum hydrocarbon-contaminated groundwater include air sparging, vacuum-enhanced recovery, aqueous pumping, *in situ* chemical reduction or oxidation and *in situ* biodegradation with microorganisms (Befkadu and Quanyuan, 2018; Ossai et al., 2020). These methods act positively either to treat and destroy hydrocarbon compounds or both. Treatment methods for petroleum hydrocarbons are cost-expensive and energy-demanding due to the long-term and multimedia problem. Furthermore, many existing places have a comparatively low danger, as most seriously contaminated places have been remediated through existing technologies once the places are identified. However, issues still prolong for numerous, comparatively low-risk places with many contaminated media that still require treatment.

Phytotechnology is a promising and alternative technique to remediate and recover contaminated environment with petroleum. Phytoremediation process is capable to fulfil this role, contributing as a long-term, and is relatively cost-effective treatment option (Burken and Ma, 2006). It involves the use of plants and microorganisms associated in rhizosphere zone to reduce environmental influence. In rhizosphere, bacteria, fungi and yeasts are capable to degrade petroleum toxic compounds by consuming them as nutrients and energy source and converted them to less toxic compounds to the ecosystem (Fan et al., 2018). The related mechanism of hydrocarbon phytoremediation such as TPH with the involvement of rhizosphere microbes in contaminated water or soil is through rhizodegradation. In rhizodegradation, plants provide suitable habitation for the growth of microorganisms in the rhizosphere zone (Fahid et al., 2020; Zhang et al., 2020). Thus, the rest of this review paper will focus and elaborate on the utilisation of phytotechnology for petroleum hydrocarbon removal and degradation based on the up-to-date research findings, its mechanisms, and finally challenges and opportunities of applying this remediation technology.

## 4. Utilisation of plants in hydrocarbon remediation

Various factors affect the accumulation, distribution and transformation of organic compounds by a plant, as illustrated in Fig. 5. Ossai et al. (2020) described some of the environmental parameters that influence the mechanisms of phytoremediation as the type of soil and organic matter, which can hinder the availability of petroleum hydrocarbon pollutants, availability of water and oxygen in the soil, temperature, nutrients, sunlight and climate. Soil texture can also impact on phytoremediation performance by influencing the contaminant bioavailability. The bioavailability of contaminants may be lower in soils with high clay contents since clay can bind molecules more readily compared with silt or sand. The addition of compost and aeration to soil can enhance phytoremediation efficiency (Al-Valdawi et al. 2013a; Robichaud et al., 2019). Water plays an important role in transporting nutrients for plants and eliminating wastes. If the moisture content of the soil is low, there will be a loss of microbial activity and dehydration of plants. Adequate soil nutrients are required to support the growth of plants and their associated microorganisms, most notably during phytoremediation efforts when the plant–microbe community is already under stress from the contaminant. The ratio of the plant number to the total mass of contaminant should be considered if the phytotechnology is intended to treat waste, including hydrocarbons, especially on a large scale (Al-Baldawi et al. 2015b). Plant characteristics, such as the root system and enzymes, are key players in phytoremediation (Susarla et al., 2002; Truu et al., 2015; McIntosh et al., 2017; Jeevanantham et al., 2019).

**Table 1**
Level of hydrocarbon concentration in soil according to local regulation act.

| Contaminant | Concentration (mg/kg) | Country | Reference |
|---|---|---|---|
| Benzene | 1.1 | Malaysia | Malaysian Recommended Site Screening Levels for Contaminated Land (2009) |
| Benzene | 1.1 | New Zealand | Environmental Data Management Software (2014) |
| Hydrocarbons | 1.2 | Canada | |
| Benzene | 5 | USA | |

**Table 2**
Level of oil and grease concentration in water according to local regulation acts.

| Contaminant | Concentration (mg/L) | Country | Reference |
|---|---|---|---|
| Oil & grease | 10 | Malaysia | Malaysia Environmental Quality Act 1974 (2009) |
| Aliphatics | | | |
| $C_5-C_8$ | 0.3 | USA | Environmental Data Management Software (2014) |
| $C_9-C_{12}$ | 0.7 | USA | |
| $C_9-C_{18}$ | 0.7 | USA | |
| $C_{19}-C_{36}$ | 14 | USA | |
| *Aromatics* | | | |
| $C_9-C_{10}$ | 0.2 | USA | |
| $C_{11}-C_{22}$ | 0.2 | USA | |
| Benzene | 0.15 | Canada | |
| Benzene | 1 | | |
| Organic compound | 1 | Japan | Water Environment Quality Standards (EQS, 2012) |
| Oil & grease | 10 | India | The Central Public Health and Environmental Engineering Organisation (CPHEEO, 2012) |

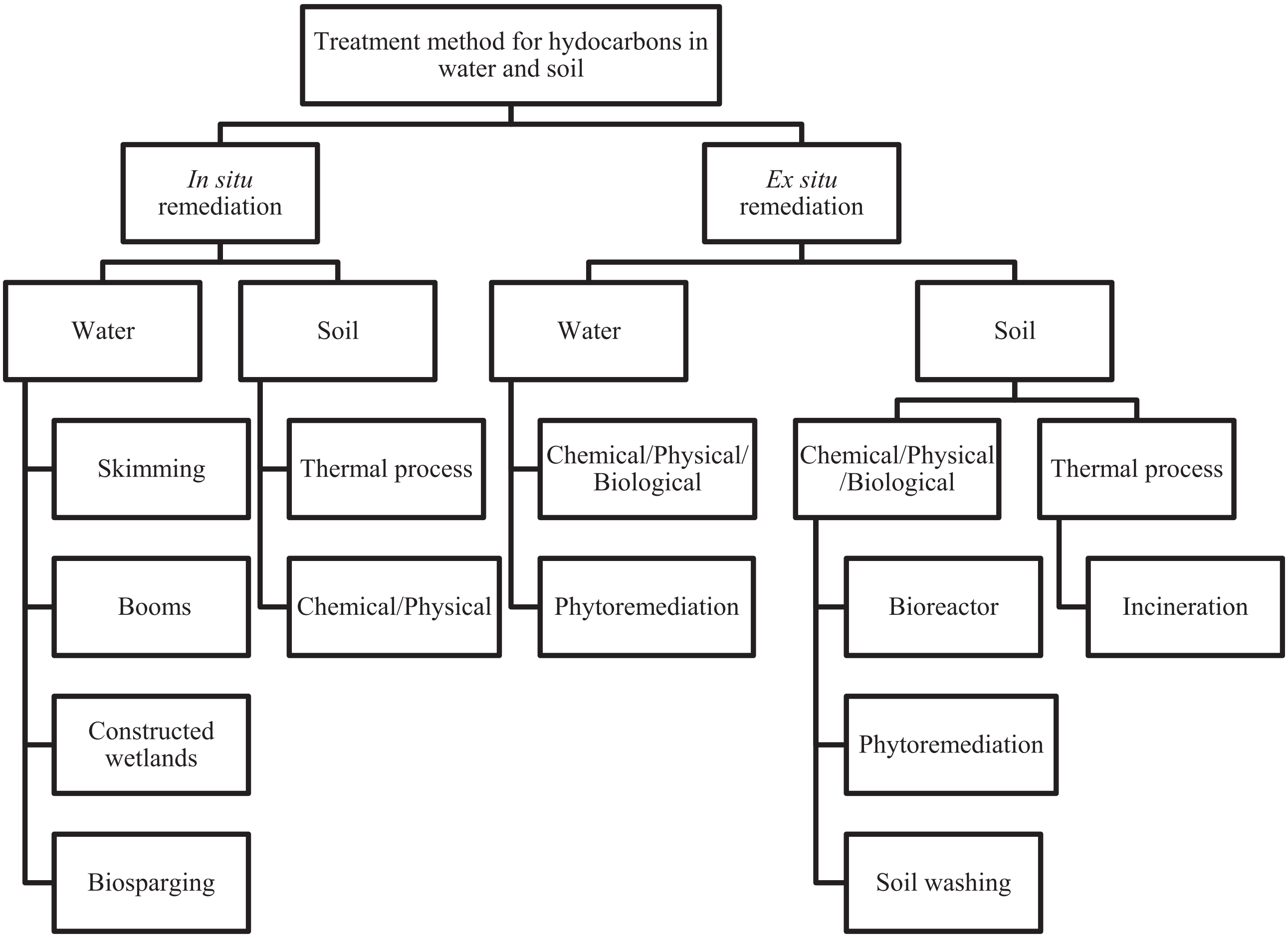

**Fig. 4.** Treatment methods for hydrocarbons in water and soil.

### 4.1. Type of plants used in phytoremediation

Many plants have been used in phytoremediation for treating municipal, livestock and industrial wastewaters. Some examples of plants used for the degradation of petroleum hydrocarbons are listed in Table 3. Most of the plants used for petroleum hydrocarbons degradation are terrestrial plants since the majority of the research has focused on the phytoremediation of hydrocarbon-contaminated soil. Hence, the use of aquatic plants is rare. Different type of plants, including *Scirpus* (bulrush) and *Eichhornia* (water hyacinth), and perennial plants, such as *Typha* (cattail) and *Phragmites* (common reed), are applied in CWs due to their wide distribution, high biomass, tolerance and resistance to numerous environmental and contaminant toxicity and resistance to contaminants (Tam et al., 2009; Jeevanantham et al., 2019).

Based on the data in Table 3, Fig. 6 shows the percentage of plant types used in phytoremediation. Most of these plants are terrestrial plants (62%), while 33% are aquatic plants, and ornamental plant types comprise 5%. It was found that more research was conducted to treat hydrocarbon contamination in soil rather in water; hence, more terrestrial plants were used, as listed in Table 3. In addition, plant-soil microbe interactions in the rhizosphere of terrestrial

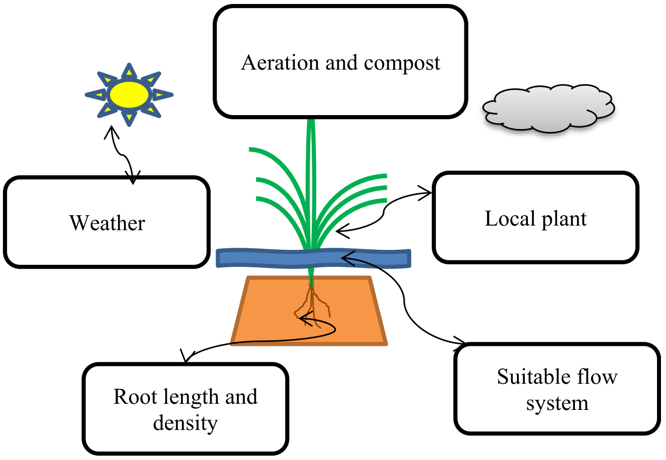

**Fig. 5.** Factors affecting phytoremediation.

plants with denser root system, are dominant in degrading hydrocarbons, and thus contributing to the enhancement of pollutant degradation and uptake.

Fig. 7 summarises the percentage of countries that have research in phytoremediation of hydrocarbons based on the compilation data in Table 3. The compiled data indicate that phytoremediation technology has not yet been widely adopted for hydrocarbon degradation in South-East Asian countries, since not many records are found. It also might be due to the lack of formal publications recording the research work on hydrocarbon degradation in this region. Majority of the research (54%) in Asia is concentrated in Japan, India, Iran, China, Indonesia, and Malaysia.

The ability of the plant to remove different types of petroleum hydrocarbons is illustrated in Fig. 8. The highest percentage of petroleum hydrocarbon removal has been achieved by the plants of *Juncus roemerianus* and *Phragmites australis* (more than 90%), both aquatic plants (Dowty et al., 2001; Lin and Mendelssohn, 2009).

### *4.2. Application of phytoremediation in treatment wetlands*

The performance of constructed wetlands (CWs), human-made design of phytotechnology simulating natural wetlands, to remove heavy metals and hydrocarbons from wastewater has been widely investigated (Tromp et al., 2012; Langergraber, 2013). The CWs used for the removal of hydrocarbons from wastewater can be classified as flow into free-water surface constructed wetlands (FSCWs), horizontal sub-surface flow constructed wetlands (HSCWs), vertical sub-surface flow constructed wetlands (VSCWs) and hybrid constructed wetlands (HCWs) which are a combination of HSCW and VSCW systems (Verlicchi and Zambello, 2014). The FSCW tank is designed for shallow free-flow water over the substrate planted with suitable plants or tanks with water only including floating plants in which free wastewater flows (Fig. 9a). For HSCW systems, wastewater is fed to the tank from the influent fed horizontally through the substrate below the surface of the wetland bed, which is planted with plants (Fig. 9b). For the VSCW systems, wastewater flows vertically from the surface of the wetland down to the substrate (Fig. 9c). The hybrid CW systems constitute two or more wetlands or the arrangement of wetlands with other pond systems, such as lagoons and facultative ponds, in parallel or series (Fig. 9d). To improve the treatment efficiencies, hybrid systems can be designed in two or three stages (Li et al., 2014).

Van Afferden et al. (2011) undertook field studies on the treatment of methyl *tert*-butyl ether (MTBE) and benzene from polluted groundwater in full-scale CWs as a biological cost-effect treatment technology. The 100% removal efficiency of MTBE and benzene from polluted groundwater was achieved using a roughing filter coupled with a polishing filter with plants (willows). The types of plants used in CWs can be divided into four groups: emergent plants, floating-leaved plants, submerged plants and freely-floating plants (Saeed and Sun, 2012). Emergent plants can grow in a water depth of 0.5 m or more above the substrate surface, while for floating-leaved plants, the root is grown in submersed sediments with a water level of 0.5–3.0 m and the leaves are free-floating. In addition, the submerged plants are immersed in water, and the floating plants are free-floated on the water surface with only the roots under water. Organic compounds can be degraded aerobically and anaerobically in CWs, depending on the design system. The oxygen available in wetlands depends on the atmospheric oxygen diffusion and the transfer of oxygen from plant roots into the substrate for aerobic degradation (Al-Baldawi et al. 2015a), while anaerobic degradation can proceed inside the substrate where a lack of oxygen exists.

## 5. Mechanisms of plant-assisted remediation of hydrocarbons

Phytoremediation is a sustainable and promising technology for the elimination of contaminants from a polluted ecosystem. In phytoremediation, plants assisted by other microbes play a key role to degrade, remove, convert, assimilate, metabolise or detoxify harmful contaminants from soil, water and air. Phytoremediation has many technical benefits and aesthetical values when compared with traditional *ex situ* remediation techniques, such as soil washing, excavation and incineration, and an off-site secured landfill (Zhang et al. 2010). Particularly, it is low-cost, simple to operate in the field and efficient (Gurska et al. 2009).

Phytoremediation is a biological technology process applying natural plants to augment the process of degradation, transformation, extraction, containment, accumulation or immobilisation of pollutants in soil and water (USEPA, 2000; Kamath et al., 2004). This technology has received considerable attention as an innovative method of treatment, as it is a cost-effective substitute for the more developed treatments used at contaminated sites (Alkorta and Garbisu, 2001). In general, phytoremediation can save the cost of a large area with high levels of organic pollution, low waste, pollution, nutrients or metals, for which the contamination does not pose an imminent danger (Kamath et al., 2004). In addition, phytoremediation is widely accepted by the public (Alkorta and Garbisu, 2001; Ossai et al., 2020). Phytoremediation encompasses a wide range of research areas, including constructed wetlands (CWs), oil spills and the accumulation of heavy metals by plants.

An overview of the general mechanisms occurring in the phytoremediation of contaminants is summarised in Fig. 10. Phytoremediation mechanism can be divided into (i) the degradation of pollutants, (ii) extraction and (iii) suppression, or a combination of these three methods (Santos and Maranho, 2018). Phytoremediation can also be classified based on its mechanisms to remove or decontaminate the pollutants. These mechanisms include contaminant extraction from soil or groundwater, contaminant concentration in plant tissue, contaminant degradation by various biotic or abiotic processes, evaporation or transpiration of volatile contaminants from the plant into the air, as well as immobilisation of contaminants in the root zone (USEPA, 2000).

As mentioned in the introduction, this review focuses on phytoremediation as a method for treating soil and water contaminated by oil (petroleum hydrocarbons). There are four identified processes involved in the phytoremediation of petroleum hydrocarbons, namely phytodegradation, phytoextraction/plant uptake,

**Table 3**
Plants used for the degradation of petroleum hydrocarbons.

| Plant Species | Plant type | Medium of pollutants | Country | Main findings | Reference |
|---|---|---|---|---|---|
| Fescue (*Lolium arundinaceum* Schreb.) KY31<br>Ryegrass (*Lolium multiflorum* L.) Marshall<br>Bermuda grass (*Cynodon dactylon* L. Pers.) Alicia | Terrestrial | Soil | USA | • High vegetation at the site with an initial TPH concentration of 9175 mg/kg.<br>• Alkylated two-ring naphthalenes were degraded in all treatments.<br>• Higher degradation of the larger three-ring alkylated phenanthrenes-anthracenes and dibenzothiophenes in the planted fertilised plots compared to the non-planted and non-fertilised plots. | White et al. (2006) |
| Black rush/needlerush (*Juncus roemerianus*) | Aquatic | Soil | USA | • Removal percentage of n-alkanes and PAHs using *J. roemerianus* was higher than that of the unplanted treatment.<br>• Phytoremediation by *J. roemerianus* was more active for degrading PHAs than *n*-alkanes. | Lin and Mendelssohn (2009) |
| Bermuda grass (*Cynodon dactylon* L. Tifway), alfalfa, crabgrass, fescue and ryegrass | Terrestrial | Soil | USA | • Hardwood sawdust + inorganic fertiliser-amended soil had a lower TPH value than the inorganic fertiliser-amended, paper mill sludge-amended and non-amended control soil due to the more available forms of nitrogen present in the fertiliser and its higher phosphorous values. | White et al. (2003) |
| Three grasses:<br>- *Pennisetum glaucum* (L.) R. Br.<br>- *Brachiaria ramosa* L. Stapf.<br>- *Sorghum sudanense* (Piper) Stapf.<br>Legume (*Aeschynomene americana* L.) | Terrestrial | Soil | USA | • In a greenhouse study of four warm-season plants, increasing nitrogen application rates in soil contaminated with 3% weathered crude oil increased plant growth.<br>• Vegetation was successfully established at a field site contaminated with 2.5% weathered crude oil.<br>• Total bacterial, fungal and PAH degradation levels were significantly higher in vegetated fertilised plots than in non-vegetated, non-fertilised plots.<br>• The greatest degradation of oil and enhancement of microbial activity included mineral soil augmented with time-release fertiliser, vegetated with *P. hemitomon* or *S. lancifolia* combined with substrate aeration. | Thoma et al. (2002) |
| Alligator weed (*Alternanthera phyloxeroides*) | Semi-aquatic | Soil | USA | • Phytoremediation was assessed by combining plants, fungi (*Trametes versicolor*) and compost to clean up a 40-year polluted site in the Canada. | Dowty et al. (2001) |
| Common reed (*Phragmites australis*)<br>Grass (*Panicum hemitomon*)<br>Bulltongue arrowhead (*Sagittaria lancifolia*) | Terrestrial | | | • Petroleum over 150 g/kg can be decreased by 65–75%, without fertilizers. | |
| Willow species (*Salix planifolia* and *Salix alaxensis*) | Terrestrial | Soil | Canada | • No significant difference in PAH degradation between the presence or absence of plants. | Robichaud et al. (2019) |
| Tall fescue (*Festuca arundinacea* Schreb.)<br>Brown mustard (*Brassica juncea* (L.) Czern.)<br>Basket willow (*Salix viminalis* L.) | Terrestrial | Soil | Canada | | Roy et al. (2005) |
| Ryegrass (*Lolium perenne* Affinity)<br>Alfalfa *Medicago sativa* L. | Terrestrial | Soil | Canada | • Significantly higher number of petroleum-degrading bacteria in the rhizosphere of perennial ryegrass.<br>• No statistically significant difference between plant treatments for both heterotrophic and petroleum-degrading fungi. | Kirk et al. (2005) |
| Perennial bunchgrass (*Vetiveria zizanioides* (L.) Nash) | Aquatic | Soil | Venezuela | • No difference in plant growth between the fertiliser treatments in petroleum hydrocarbon-contaminated soil.<br>• The presence of plants did not result in lower TROG concentrations. | Brandt et al. (2006) |
| Bread grass (*Brachiaria brizantha*) | Terrestrial | Soil | Venezuela | • Positive effect of fertiliser levels on microbial counts. | Merkl et al. (2006) |
| Three legumes:<br>- *Calopogonium mucunoides* Desv.<br>- *Centrosema brasilianum* (L.) Benth.<br>- *Stylosanthes capitata* Vogel<br>Three grasses:<br>- *Brachiaria brizantha* (Hochst. ex A. Rich.) Stapf.<br>- *Cyperus aggregatus* (Willd.) Endl.<br>-*Eleusine indica* (L.) Gaertn. | Terrestrial | Soil | Venezuela | • All plants died within 6–8 weeks.<br>• 50% removal of total oil and grease content (TROG) by *B. brizantha.*<br>• 30% removal of TROG by *C. aggregatus.*<br>• TROG content in soil planted with *E. indica* and its unplanted control was reduced below than with other species and their corresponding controls. | Merkl et al. (2005a), Merkl et al. (2005b) |
| Perennial grass-like plant (*Cyperus laxus* Lam.) | Terrestrial | Soil | Mexico | • Phytoremediation rate of inoculated plants was three times higher than that obtained with non-inoculated plants and was reached before flowering.<br>• *Cyperus* laxus Lam. can be selected for phytoremediation of tropical swamps polluted with hydrocarbons. | Escalante-Espinosa et al. (2005) |
| Tall fescue (*Festuca arundinacea*)<br>Yellow sweet clover (*Melilotus offcinalis*) | Terrestrial | Soil | Indiana | • Plant root density differed between the non-vegetated and vegetated soil.<br>• Plant root density contributed to the reduction in PAHs. | Parrish et al. (2005) |
| Scots pine (*Pinus sylvestris*)<br>Poplar (*Populus deltoides* × *Wettsteinii*)<br>Grass mixture (*Festuca rubra*; *Poa pratensis*, *Lolium perenne*, *Trifolium repens* and *Pisum sativum*) | Terrestrial | Soil | Finland | • All plant types tolerated a diesel fuel level of 0.5% (w/w).<br>• Phytoremediation using legume species was the most effective method for diesel fuel elimination from the soil.<br>• Treatment with pine and poplar also improved diesel fuel removal. | Palmroth et al. (2002) |

(*continued on next page*)

**Table 3** (*continued* )

| Plant Species | Plant type | Medium of pollutants | Country | Main findings | Reference |
|---|---|---|---|---|---|
| Italian ryegrass, bird's-foot trefoil and alfalfa | Terrestrial | Soil | Austria | • *Enterobacter* degraded hydrocarbons and efficiently colonised the rhizosphere microbes.<br>• Maximum degradation of diesel fuel was 68%, using a combination of Italian ryegrass and alfalfa. | Yousaf et al. (2011) |
| Rushes (*Typha latifolia*, *Typha angustifolia*, *Scirpus lacustris*, *Juncus acutus*), reed (*Phragmites communis*) | Semi-aquatic | Water | Bulgaria | • Oil content of the water after treatment was decreased to less than 0.2 mg/L.<br>• Oil removal was associated with its degradation by the indigenous microflora. | Groudeva et al. (2001) |
| Maize plants | Terrestrial | Soil | Poland | • PAH degradation (60%) was achieved by microbe-assisted phytoremediation. | García-Sánchez et al. (2018) |
| Sorghum (*Sorghum bicolor* (L.) Moench | Terrestrial | Soil | Russia | • 100 mg/kg of phenanthrene inhibited the accumulation of plant biomass, causing decreases in carboxylic acid, carbohydrates and amino acids in the rhizosphere of sorghum.<br>• 10 mg/kg of phenanthrene did not significantly affect plant growth and the total amount of root exudation. | Muratova et al. (2009) |
| Maize (*Zea mays*) and cowpea (*Vigna unguiculate*) | Terrestrial | Soil | Nigeria | • Sawdust and chromolaena leaves have potential in protecting and maintaining optimum growth for plants in a polluted environment. | Offor and Akonye (2006) |
| Mangrove | Aquatic | Soil | Nigeria | • Chromatium species, well adapted to the epiploic sediment of a mangrove ecosystem, are sensitive to hydrocarbon accumulation in sediments.<br>• Sulphur bacteria might indicate crude oil contamination in mangrove ecosystems. | Essien and Antai (2008) |
| Soybean (*Glycine max*) | Terrestrial | Soil | Nigeria | • No statistical difference between the number of pods produced by *G. max* grown in contaminated soils mixed with cow dung and those without cow dung. | Njoku et al. (2008) |
| Wildflowers (mainly *Senecio glaucus*) | Terrestrial | Soil | Kuwait | • Took up and detoxified alkanes and aromatic hydrocarbons. | Trapp et al. (2001) |
| *Eucalyptus camaldulensis* | Terrestrial | Soil | Iran | • The highest reduction, 82% in oil hydrocarbon was observed in treatment of 1% oil pollution and the lowest (44%) was recorded for the treatment of 4%. | Taheri et al. (2018) |
| Tall fescue (*Festuca arundinacea* Schreb.) and meadow fescue (*Festuca pratensis* Huds.) | Terrestrial | Soil | Iran | • Infected plants contained more root and shoot biomass than non-infected plants and created higher levels of water-soluble phenols and dehydrogenase activity in the soil.<br>• 80–84 and 64–72% removal for PAH and TPH in the rhizosphere of plants, respectively, compared with only 56 and 31%, respectively, in the controls. | Soleimani et al. (2010) |
| Tall fescue (*Festuca arundinacea*) | Terrestrial | Soil | Iran | • The growth and dry biomass of the plant decreased by increasing the crude oil concentration in the soil.<br>• The length of the leaves was reduced with increased crude oil concentration.<br>• All planted samples had higher crude oil reduction than non-planted samples. | Minai-Tehrani et al. (2007) |
| Alfalfa (*Medicago sativa*) | Terrestrial | Soil | Iran | • In 120 days, the maximal reduction was observed in the 1% planted sample, while the 10% planted sample showed the least reduction. | Shahriari et al. (2007) |
| Carpet grass (*Axonopus affinis*) | Terrestrial | Water | Pakistan | • Carpet grass (*Axonopus affinis)* was planted in soil spiked with diesel (1% w/w) for 90 days and inoculated with bacterial strains, *Pseudomonas* sp. ITRH25, *Pantoea* sp. BTRH79 and *Burkholderia* sp. PsJN<br>• Maximum hydrocarbon degradation (89%) was achieved with the three strains inoculum, higher than the non-inoculated plants (46%) | Tara et al. (2014) |
| *Phragmites australis* | Aquatic | Water | Pakistan | • *P. australis* was planted on a floating mat for the remediation of diesel (1%, w/v) contaminated water inoculated with three bacterial strains (*Acinetobacter* sp. *BRRH61, Bacillus megaterium RGR14 and Acinetobacter iwoffii AKR1).*<br>• Maximum hydrocarbon reduction (95.8%), chemical oxygen demand (98.6%), biochemical oxygen demand (97.7%), total organic carbon (95.2%), and phenol (98.9%) were accomplished. | Fahid et al. (2020) |
| Ryegrass (*Lolium multiflorum*) | Terrestrial | Soil | Pakistan | • Maximal hydrocarbon removal (85%) occurred in spiked soil amended with compost, biochar and consortia. | Hussain et al. (2018a) |
| *Scirpus triqueter* | Aquatic | Soil | China | • The effect of the plant-growth-promoting rhizobacteria (PGPR) on phytoremediation in the pyrene-Ni co-contaminated soil was investigated.<br>• PGPR-inoculated S. triqueter increased pyrene and Ni removals. | Zhang et al. (2020) |
| *Scirpus triqueter* | Aquatic grass | Water | China | • Maximal removal ratio of diesel in the contaminated soil was 55% after 60 days. | Zhang et al. (2014) |
| Alfalfa plants | Perennial flowering plant | Soil | China | • 2,4-DCP removal by transgenic alfalfa plants was 98.8% at 144 h of treatment. | Wang et al. (2015) |

**Table 3** (*continued* )

| Plant Species | Plant type | Medium of pollutants | Country | Main findings | Reference |
|---|---|---|---|---|---|
| Seepweed (*Suaeda glauca*), sea lavender (*Limoniumbi color Kuntze*), Central Asia saltbush (*Atriplex centralasiatica* Iljin)<br>and reed (*Phragmites communis Trin*) | Aquatic | Water | China | • TPH degradation of 40% by seepweed was achieved after 90 days.<br>• Seepweed roots significantly decreased the surface and volume of soil micro-pores.<br>• Increased bioavailability of TPH. | Wang et al. (2011) |
| Maize (*Zea mays* L.) | Terresterial | Soil | China | • Two biosurfactants (rhamnolipid and soybean lecithin) and a synthetic surfactant (Tween 80) were used to enhance phytoremediation of crude oil contaminated soil by maize (*Zea mays* L.)<br>• TPHs removal in the treatments with soybean lecithin, rhamnolipid and Tween80 were 62%, 58% and 47%, respectively, and for the control (without surfactant) was only 52%. | Liao et al. (2016) |
| Ryegrass (*Lolium perenne*) | Terrestrial | Soil | China | • The mixture of ryegrass with diverse microbial strains provided the best result, with a degradation rate of 58% after 162 days.<br>• Saturated hydrocarbon degradation happened most with the mixture of microorganisms and ryegrass. | Tang et al. (2010) |
| Marvel of Peru/four o'clock flower (*Mirabilis jalapa*) | Terrestrial | Soil | China | • Degradation rate of 60.25–73.11% was achieved.<br>• *Mirabilis jalapa* could be effectively applied in phytoremediation of ≤10,000 mg/kg petroleum-contaminated soil. | Peng et al. (2009) |
| Rice | Aquatic | Soil | China | • The concentration of PAHs in rice lateral roots was higher than in nodal roots.<br>• PAHs were more easily absorbed in the inside of rice roots than on the outside surface. | Jiao et al. (2007) |
| Bermuda grass (*Cynodon dactylon*) and lawn grass (*Zoysia japonica*),<br>Creeping bentgrass (*Agrostis palustris* Huds.),<br>White clover (*Trifolium repens* L.) | Terrestrial | Soil | Japan | • The highest microbial population was found with White clover rhizosphere soil.<br>• White clover was selected for degradation of chlorinated dioxins. | Wang and Oyaizu (2009) |
| Maize (*Zea mays*), red bean (*Phaseolus nipponesis* OWH1) | Terrestrial | Soil | Korea | • Removal of TPH after 120 days was 86.5%. | Baek et al. (2004) |
| Bulrush (*Scirpus grossus*) | Emergent wetland plant | Water | Malaysia | • Degraded TPH when the concentration of diesel in water was up to17,400 mg/L. | Al-Baldawi et al. 2013a |
| Mexican primrose-willow (*Ludwigia octovalvis*) | Terrestrial tropical plant | Soil | Malaysia | • Removed TPH during 72 days; 79.8% removal efficiency for 2 g/kg of gasoline. | Al-Mansoory et al. (2015) |
| Mexican primrose-willow (*Ludwigia octovalvis*) | Terrestrial tropical plant | Sand | Malaysia | • TPH degradation were 67.0, 42.4 and 46.2% in sand spiked with real crude oil sludge at 10, 50 and 100% (v/v) respectively. | Alanbary et al. (2019) |
| Grey sedge (*Lepironia articulate*) | Aquatic | Water | Malaysia | • Degraded PAHs from wastewater; 80% removal efficiency. | Al-Sbani et al. (2016) |
| Grass (*Paspalum scrobiculatum* L. Hack) | Terrestrial tropical plants | Sand | Malaysia | • Optimum conditions were found to be at a diesel concentration of 3%, 72 sampling days, and an aeration rate of 1.77 L/min with a 76.8% maximum TPH removal. | Sanusi et al. (2016) |
| *Scirpus grossus* and *Lepironia articulata* | Aquatic | Soil | Malaysia | • *S. grossus* and *L. articulata* were exposed to 3 kg of crude oil sludge under greenhouse conditions for 30 days.<br>• 100% *S. grossus* could survive in the sludge compared to only 55% L. articulate survived. | Sharuddin et al. (2019) |
| Mangrove (*Rhizophora* sp., *Avicennia* sp. and *Bruguiera* sp.) | Aquatic | Soil | Indonesia | • *Rhizophora* sp. had the highest tolerance and was able to reduce higher TPH in the media compared to those of *Avicennia* sp. and *Bruguiera* sp when exposed to 10, 20 and 30% of petroleum hydrocarbon in soil.<br>• Hydrocarbon was absorbed and translocated into the guard cell of the stomata. | Hidayati et al. (2018) |
| *Vertiver zizanioides* | Terrestrial | Water | Indonesia | • 6-plant pot with 1% crude oil concentration reduced the oil content to 91.39%, while 3-plant pot could decrease the oil content to 90.28%. | Effendi et al. (2017) |

Notes: 2,4-DCP = 2.4-dichlorophenol; TROG = total recoverable oil and grease; TPH = total petroleum hydrocarbons; PAH = polycyclic aromatic hydrocarbon.

phytovolatilisation and rhizodegradation. Phytodegradation is the breakdown of pollutants either internally, through metabolic processes, or externally, by enzymes released by plants into the ground. Sites contaminated with moderately hydrophobic organic chemicals are efficiently removed *via* direct uptake by plants. Other factors, including hydrophobicity, solubility, polarity and sorption properties of pollutants, especially petroleum hydrocarbons, can influence their accessibility by plant roots (Ossai et al., 2020). In order for the organic contaminant to be remediated using plants, the contaminants must come into contact with the plant roots and dissolve in the soil water (Interstate Technology and Regulatory Cooperation Work Group 2001), thus the existence of surfactants or biosurfactants expressed by rhizobacteria exposed to petroleum hydrocarbons can enhance further the degradation of hydrocarbons in plant rhizosphere (Almansoory et al. 2017, 2019; Liao et al., 2016). Furthermore, phytovolatilisation involves the movement of contaminants out of the soil or underground and, *via* plants, into the atmosphere. The following section will discuss in details on each related mechanisms of hydrocarbon phytoremediation.

### 5.1. *Phytodegradation*

Phytodegradation, or similarly acknowledged as phytotransformation primarily involves degradation of the contaminants

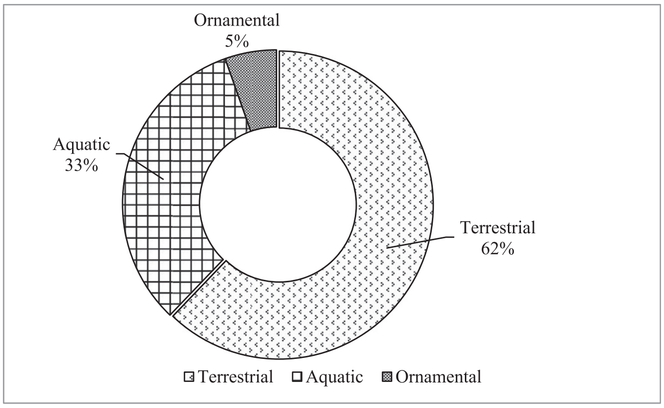

**Fig. 6.** Plants used in phytoremediation.

through metabolic processes inside the plant. To a lesser extent, degradation may external to the plant, mainly in the rhizosphere, by the release of chemicals, such as enzymes that cause transformation. Degradation caused by microorganisms at the plant root zone is known as rhizodegradation (USEPA, 2000). Kavamura and Esposito (2010) showed that organic compounds are degraded or mineralised by specific enzyme activity or rendered non-toxic *via* enzymatic modification. Gerhardt et al. (2009) indicated that organic pollutants might be affected by soil microbes that are stimulated by various root exudates (Germaine et al., 2009), but other organic compounds may be degraded because of enzymes, alcohols, sugars and acids released by plant roots. Organic compounds may also be stored before subsequent biochemical degradation into less harmful products (Wild et al., 2005; Brandt et al., 2006). In addition, research work by Al-Baldawi et al. (2017) and Alanbary et al. (2019) have proven that the longer carbon chains in TPH of petroleum hydrocarbon were degraded into lower carbon chains at the end of exposure.

As mentioned above, some enzymes secreted by plants into the soil and water can enhance the growth and activities of rhizosphere microorganisms. These enzymes also contribute to decompose organic pollutants. The rhizosphere plays a vital role in providing a habitat for microorganisms that can assist the plants in increasing the efficiency of phytoremediation (Chen et al., 2016). The availability of pollutants to the plants can be increased by nitrogen and phosphorus fertilisers, together with root exudates containing chelating agents, thereby enhancing the plant growth, as well as the capability of the plant to extract and accumulate contaminants. As highlighted by Gerhardt et al. (2009), microbe-assisted phytoremediation is a complex interaction involving roots, root exudates, rhizosphere soil and microbes and leads to degradation of organic compounds into non-poisonous or less poisonous compounds (García-Sánchez et al., 2018). Up to 40% of a plant's photosynthate can be precipitated and stored in the soil as sugars, organic acids and larger organic compounds that can be utilised as carbon and energy sources for soil microbes.

### 5.2. *Rhizodegradation (phytostimulation)*

Rhizodegradation, one type of microbe-assisted phytoremediation, is the breakdown of an organic pollutant in the soil or water through microbial action at the rhizosphere of plants. The microbes involved encompass bacteria (Al-Baldawi et al., 2017; Liao et al., 2016; Fahid et al., 2020), yeast and fungi (Robichaud et al., 2019). It improves the conditions in the rhizosphere zone and is a slower process than phytodegradation (USEPA, 1999). Microbial degradation in the rhizosphere might be the most significant mechanism for the elimination of diesel-derived organics in planted contaminated soils. Due to the extreme hydrophobicity of pollutants, such as PAHs, their sorption to soil reduces their uptake by plants and, consequently, phytotransformation (Kamath et al., 2004). The concentration of bacteria around the root zone (rhizosphere) is usually 10- to 1000-fold greater than the concentration of bacteria found in the soil mass (Glick, 2010). Roots penetrate soil, and through respiration, oxygen will be supplied to rhizobacteria, which simultaneously with enzyme exudates, can enhance the rhizodegradation of pollutants and later ease the process of uptake by plants through phytoextraction.

Rhizodegradation is one of the most effective mechanisms for plants to remediate organic pollutants, mainly high-molecular-weight recalcitrant compounds. The rhizosphere zone has an important role in the phytoremediation of organic contaminants *via* plant–microbe interactions. Many factors influence the rhizosphere zone efficiency, such as the size and rooting intensity of the rhizosphere (Joner et al., 2006; Hussain et al. 2018a, 2018b). Plants should be suitable to the oil-contaminated area and preferably be tolerant of the climate environments and soil characteristics. Due to the significant cost variable, less expensive plants are always favourable (Etim, 2012). The rhizosphere zone in the plant increases the number of microorganisms. Compared with the control

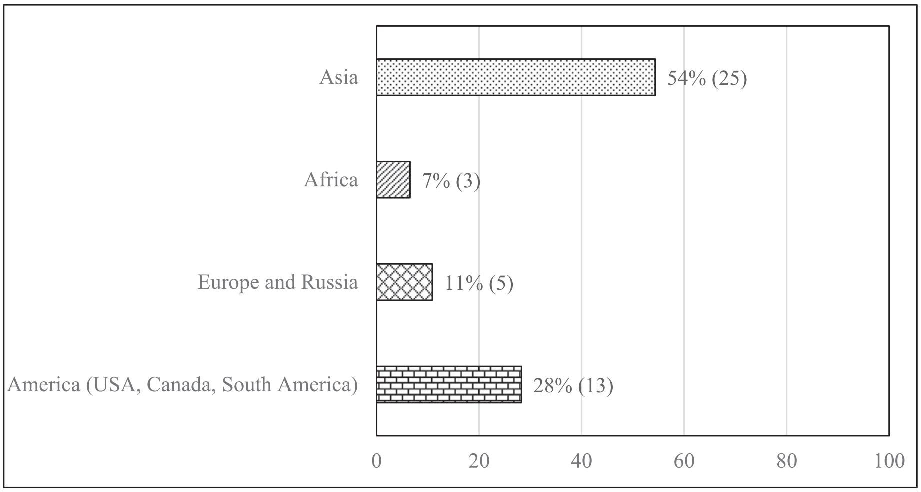

**Fig. 7.** Research records on hydrocarbon phytoremediation in different continents.

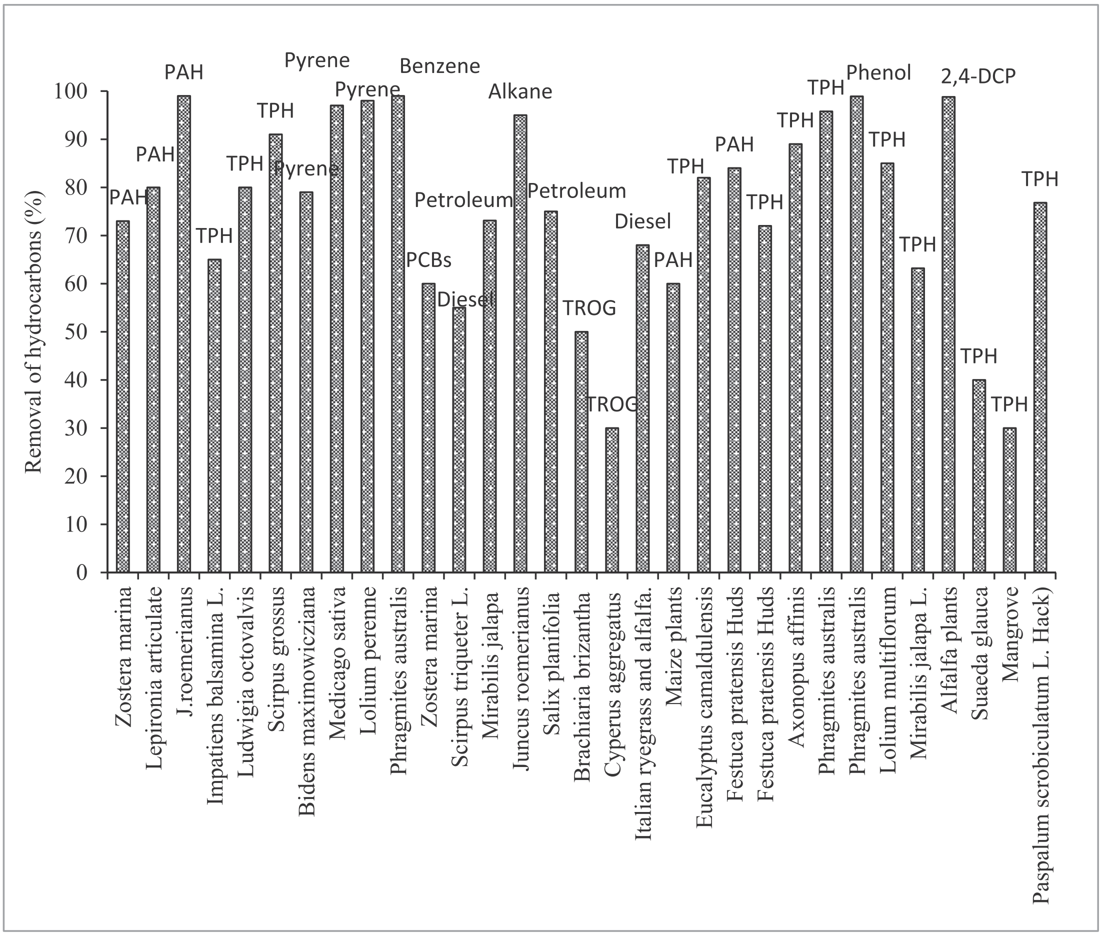

**Fig. 8.** Ability of plants to degrade petroleum hydrocarbons: polycyclic aromatic hydrocarbons (PAH); total petroleum hydrocarbons (TPH); polychlorinated biphenyls (PCBs).

(non-rhizosphere) soil, one study showed the microorganism action was 0.29–0.36 higher in the rhizosphere soil (Chen et al., 2003). The oily characteristics of petroleum hydrocarbon obviously increase the hydrophobicity of pollutants, and thus reducing their bioavailability to the plant roots and their associated rhizobacteria (Hussain et al., 2018a). As found by Almansoory et al. (2017, 2019), biosurfactant produced by hydrocarbon-degrading bacteria (HDB), *Serratia marcescens* exposed to the roots of *Luwidgia octavalvis*, could reduce surface tension of the gasoline and hence enhanced the TPH degradation through phytostimulation. Liao et al. (2016) also proposed biosurfactant-assisted phytoremediation as a useful biotechnological approach for the remediation of petroleum hydrocarbon in contaminated soil. LeFevre et al. (2012) examined the ability of microorganisms present in the soil samples to degrade naphthalene, categorised as one of petroleum hydrocarbons, in batch experiments, and observed that bacteria 16S rRNA genes inside the soil were able to mineralize naphthalene. Whilst, another research work conducted by Fahid et al. (2020) found that *Phragmites australis* could stimulate hydrocarbons degrading bacteria (*Acinetobacter* sp. BRRH61, *Bacillus megaterium* RGR14 and *Acinetobacter* iwoffii AKR1) to degrade hydrocarbons in water.

### 5.3. Phytoextraction/plant uptake

Some plant species together with rhizobacteria can express enzymes in the soil and groundwater to degrade hydrocarbons to simpler organic compounds or shorter carbon chain that will be adsorbed onto the root surface and accumulated in the roots, and extracted to the upper parts of plants (stem and leaves) through phytoextraction or through plant uptake (Hidayati et al., 2018; Al-Baldawi et al. 2015a; Hunt et al., 2019). This mechanism occurs due to root-zone microbiology activities and the chemical properties of the soil environment or contaminant. It can improve the solubility and bioavailability of organic compounds, and thus improving the degradation, allowing plants to uptake useful degraded components for their growth. Hidayati et al. (2018) found that TPH was translocated and absorbed into the stomata of mangrove, and Al-Baldawi et al. (2015a) also proved that TPH was detected in the upper part of *Scirpus grossus*. According to a review paper by Hunt et al. (2019) plants are capable to uptake petroleum hydrocarbon in their tissues.

### 5.4. Phytovolatilisation

The principle of phytovolatilisation is the uptake and transpiration of a pollutant through a plant, resulting in the release of the pollutant or the transformed form of the pollutant to the atmosphere (Erakhrumen, 2007). This mechanism is mostly applicable to those contaminants generated from traditional air-stripping with a Henry's constant ($K_H$) > 10 atm $m^3$ water $m^{-3}$ air that includes BTEX, trichloroethene, vinyl chloride and carbon tetrachloride (Kamath et al., 2004).

Plants can eliminate toxic compounds from the soil through phytovolatilisation. In this mechanism, the water-soluble pollutants are absorbed by the roots, translocated to the leaves and

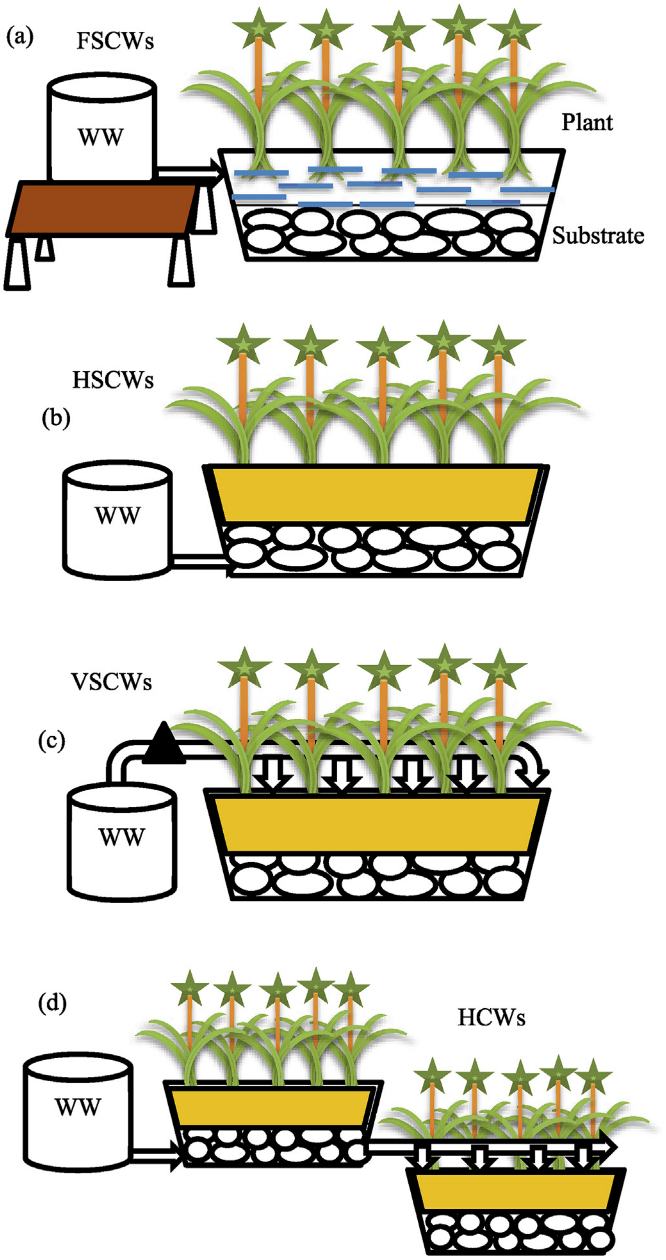

**Fig. 9.** Configuration of constructed wetlands: (a) free surface constructed wetlands (FSCWs), (b) horizontal sub-surface flow constructed wetlands (HSCWs), (c) vertical sub-surface flow constructed wetlands (VSCWs) and (d) hybrid constructed wetlands (HCWs) (WW = wastewater).

volatilised to the atmosphere through the pores in the epidermis of the leaf or stem of a plant (direct phytovolatilisation) or volatilised from the soil due to plant root activities (indirect phytovolatilisation) (Vanek et al., 2010). Phytovolatilisation occurs when growing trees and other plants uptake water, and the associated water-soluble pollutants are then transported to the leaves and evaporated to the air (USEPA, 2000). Many factors affect the evaporation of organic compounds and their direct degradation by plant enzymes through phytodegradation (Wild et al., 2005). Low-molecular-weight organic compounds can be transported from the soil through plant membranes and released across leaves through evapotranspiration processes (Gerhardt et al., 2009). In control studies of contaminants (without plants beds), the rate of evaporation is generally reliant on the weather temperature and relative air moisture and, thus, maximum during summer. Plant transpiration and evapotranspiration rates depend considerably on the plant type and propagation stage, which can significantly accelerate the water loss. The greater the plant size, the more intense the activity, and this can also contribute towards the increased evapotranspiration rate of microcosm wetlands during the summer. Phytovolatilisation in the sub-surface system, resulting from plant root activities, is limited due to the slow diffusion rates of pollutants through the unsaturated region that may result in comparatively low mass transfers. However, at the surface, in contact with the environment, water remains in direct contact with the atmosphere, and more contaminants will be released *via* volatilisation (Imfeld et al., 2009).

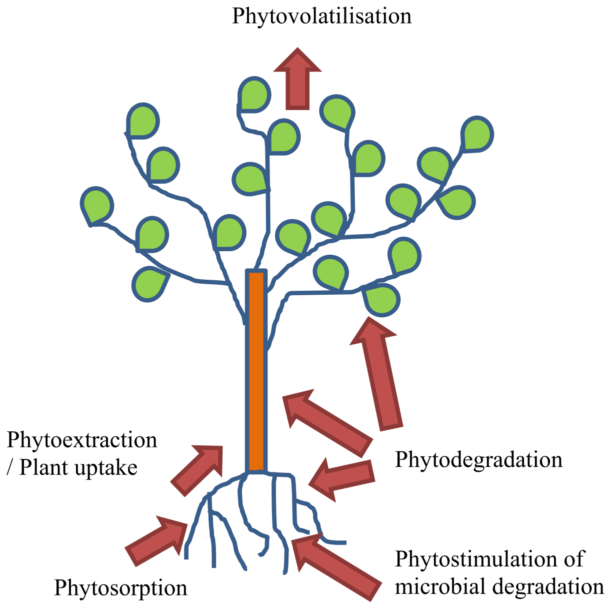

**Fig. 10.** Phytoremediation mechanisms of hydrocarbons.

### 5.5. Plant–microbe interactions in phytoremediation of hydrocarbons

Plant roots are hosts to thousands of microorganisms comprising consortia of rhizobacteria, yeast and fungi, which interact synergically and symbiotically with the roots, promoting plant growth and enhancing removal and detoxification of pollutants (Al-Baldawi et al. 2015a, 2017). The advantage of microbial consortia over a single strain of microorganism is the wide range of enzymatic capacity available to degrade crude oil. The rhizosphere microbial community can accelerate the degradation of organic pollutants particularly hydrocarbons while, simultaneously, promote the plant growth (Kirk et al., 2005). Co-oxidation usually occurs in petroleum mixtures in which compounds that cannot be degraded by microorganisms can be broken down enzymatically due to the capability of the microorganism to grow on other hydrocarbons. These compounds are transformed into smaller molecules without consumption of energy or carbon from the oxidation process (Al-Baldawi et al., 2017; Alanbary et al., 2019; Merkl et al., 2006). Organic pollutants, for example, petroleum hydrocarbons and PAHs, can be stabilised in the soil medium, extracted by plants and transformed or accumulated in a non-hazardous form.

The time required to degrade organic compounds generally depends on the relative complexity of the molecules and molecular weight (Varjani, 2017). High-molecular-weight compounds, such as aliphatic hydrocarbons, and compounds with a benzene ring, particularly grouped under aromatic hydrocarbons, require a longer time and even more time when more than one benzene ring is involved, such as PAHs (Al-Sbani et al., 2016). Fig. 11 classifies the organic compounds that can be transferred through plant membranes, as aromatic (low molecular weight), aliphatic (non-volatile) and others, according to the mechanism of phytoremediation. Low-molecular-weight compounds can be easily removed from the soil and released through leaves *via* phytovolatilisation processes. However, the non-volatile compounds can be degraded to non-toxic ones through phytodegradation and phytoextraction by microorganism enzymes. Through phytoextraction, the low molecular weight and less toxic hydrocarbons will be adsorbed to the root surface or translocated to the upper plant parts (stem and leaves) (Hunt et al., 2019).

The toxicity of hydrocarbons can negatively impact on plant growth. In addition, the ability of plants and microorganisms to absorb water and nutrients from the soil will be reduced due to hydrophobic properties of hydrocarbons (Lin and Mendelssohn, 2009). Some organic contaminants can be extracted from soil and water through plant roots. These compounds may have multiple terminations since they are metabolised in the root, translocated through xylem and embedded in cell wall materials, such as lignin.

## 6. Challenges and opportunities in phytoremediation application for hydrocarbon removal

The increase in industrial production due to global development over the last century has intensified the release of chemicals materials into the ecosystem. Phytoremediation has emerged as an encouraging approach for *in situ* elimination of many pollutants. Plant tolerance to hydrocarbons during phytoremediation of contaminated water and soils is crucial to the recovery of media and soil health. Treatment wetlands, especially for the petroleum industry, are few. Hence, more plant-assisted remediation systems should be applied on-site for water and soil to investigate the phytoremediation performance and compare the results with laboratory-scale and fill the data gaps on the use of treatment wetlands specifically for the petroleum industry (Zhuang et al., 2007). Phytoremediation is a sustainable option for water and soil organic elimination, but it also has some challenges, especially in large scale (on-site) application in which the performance might vary from the findings from the laboratory or greenhouse studies. The challenges in real or field application will be due to variations in weather and nutrients, moisture content, harmful insects and plant pathogens (Nedunuri et al., 2000). Any negative effect of these factors can reduce or prevent plant growth in the field and, in turn, impact on phytoremediation performance. Thus, a big challenge is awaiting for industry or any stakeholder in implementing phytotechnology to remove hydrocarbons from contaminated soil and water. Control over the moisture content of the plant growth medium, ether in soil or water, is challenging because drying agents can prevent growth (Khandare and Govindwar, 2015). The main obstacle faced in field studies is the distribution of contaminants in the soil treatment region, while in the laboratory and greenhouse, soils are usually well mixed, to achieve a homogeneous matrix. Similarly, to hydrocarbon-contaminated water, variations in hydrocarbon concentration from low to moderate strength from industrial effluent might affect the plant and rhizosphere microbe growth, and hence the overall remediation performance, with extreme case can cause plants and microbes to die. Phytoremediation is an appropriate approach for sites that have low-to-moderate levels of contamination by metals due to inhabited plant growth in highly contaminated water and soils (Zhang et al., 2017).

Some advantages and limitations of phytoremediation are summarised in Fig. 12. As Kirk (2005) highlighted, the main advantage of phytoremediation is that it is a green technology that aims for sustainable development. It uses natural resources of plants and microorganisms, reduces environmental degradation, improves health and lives, and protect ecosystems. Other advantages are that phytoremediation is effective for both organic and inorganic pollutants, and hence it is suitable to be used for mixed type of pollutants with several mechanisms (phytodegradation, phytoaccumulation, phytoextraction, phytotransformation, phytovolatilisation) to remove or detoxify the pollutants; it is effective for soil contaminated with large volumes and highly dispersed contaminants at low-to-moderate concentrations; it can be conducted *in situ* with the structure and texture of the soil is maintained; it is environmentally friendly and aesthetically accepted by the public with pleasing scenery view. At the end of remediation, contaminated soil can be reclaimed for agricultural (e.g. as nursery place for omental plants) or other development purposes, or treated effluent can be reused for cleaning or landscape purposes, minimising adverse impact on the ecosystem. Furthermore, the cost of phytotechnology is comparatively lower and cheaper than other chemical and physical treatment technologies, since it simply implemented and maintained.

Phytoremediation does also face some limitations. One of the major limitations of phytoremediation is the need for a lengthy time to reach a “clean” state. However, this obstacle can be reduced through soil amendments or modification of soil properties through fertiliser addition, and inculcation with microbes that have the ability to exudate enzymes, biosurfactant or bio-chelate which eventually can expedite the removal rate of hydrocarbons. Phytoremediation is not applicable if pollutants pose risk conditions to humans or water; and it is weather-dependent and thus is limited by climate, surrounding conditions, soil conditions and the availability of nutrients. For temperate countries, cold weather can totally stop or reduce the pollutant removal rate, but for tropical countries like Southeast Asia countries which receive sunlight throughout the year, it becomes a great opportunity to adopt phytoremediation to mitigate pollution. In addition, the hydrocarbon contaminants treated by phytoremediation must be non-toxic to plants and microorganisms, and most of the time not in high concentration. For the high concentrations of hydrocarbons which

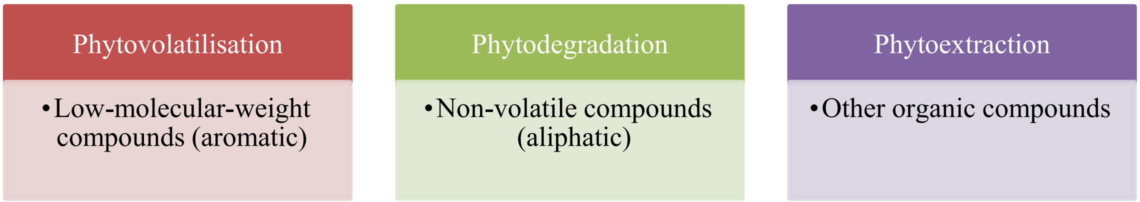

**Fig. 11.** Classification of phytoremediation mechanisms according to molecular weight of hydrocarbons.

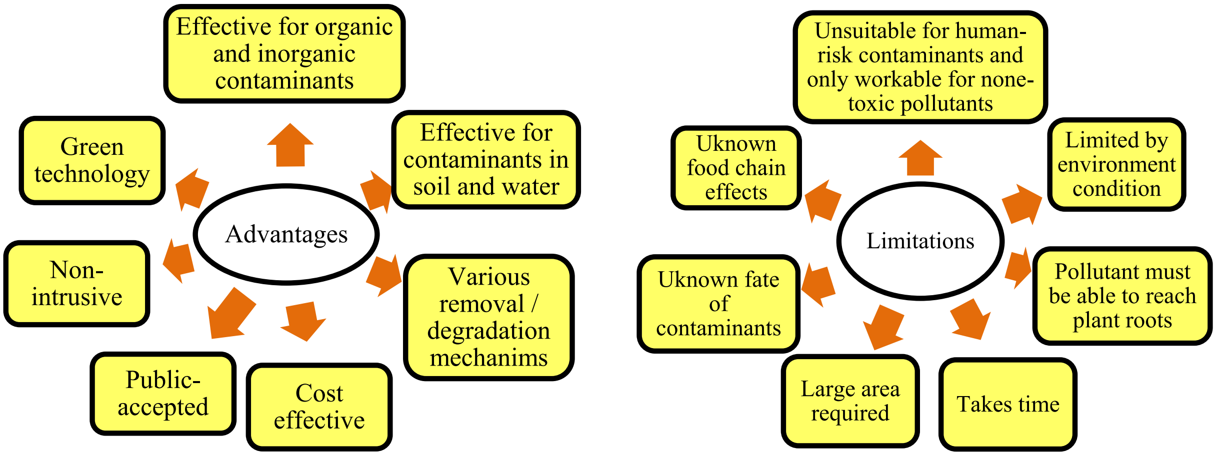

**Fig. 12.** Advantages and limitations of phytoremediation.

plants cannot resist, phytotechnology can be integrated with prior treatments of green physical and chemical technologies. In this situation, phytoremediation can function as a polishing treatment, offering a cheaper, environmentally friendly and simple method to reduce further the level of hydrocarbons. For phytoremediation to be effective, the hydrocarbon pollutants should be able to reach the plant roots and hence, it cannot work efficiently for deep contaminated soil. To resolve this problem, various types of plants with different type of root system can be applied including tap and fibrous root systems, to ensure the roots reaching all the involved pollutants. In case of wastewater treatment, large volume of contaminated water together with longer retention time require large area if phytoremediation technique were to be adopted. This limitation can be overcome by designing stages reed bed systems. By utilising plants to remediate contaminants, the final contaminant fate can be uncertain and thus the effects to food chain might be unknown because it also involves fauna and biota which are uncontrollable. Few mitigation steps need to be taken; non-edible plants should be prioritised to be used for phytoremediation in order to minimise the possibility entering the food web, and more research work should be conducted to fully understand and reveal the fate of degraded or extracted hydrocarbon contaminants by the plant-microbe interactions.

## 7. Conclusions

Up to now, phytoremediation of petroleum-polluted soils has been demonstrated at several sites and in greenhouse studies. However, less is reported for phytoremediation of hydrocarbon-contaminated water. Phytoremediation technology is appropriate for water and soil contaminated with hydrocarbons because it is simple in operation, easy to maintain in sites, environmentally-friendly and economical. Nevertheless, the performance of phytoremediation of organic compounds in water treatment is limiting due to the low aqueous solubility of petroleum hydrocarbon contaminants. CWs are an effective option for removing a range of organic compounds from wastewater, are low in cost and can be constructed near the contaminated field, especially for oil spills. The mechanisms involved in the remediation of toxic organic compounds include biosorption, biodegradation and decontamination. A number of opportunity strategies are offered by applying phytotechnology including elimination or reduction of pollutants, reclaim and reuse of the treated hydrocarbon-contaminated areas and convert them into recreational, agricultural, residential and commercial development after being declared totally free from pollutants. Similarly, treated effluent can be reused for cleaning or landscape purposes, thus minimising the adverse impact on the environment.

Phytoremediation is a promising method to degrade petroleum hydrocarbons in contaminated soil, with many advantages over other relevant treatments. Terrestrial plants are commonly used for petroleum hydrocarbon degradation, whereas the use of aquatic plants is rare. The effectiveness of the plants is associated with the existence of microbial growth on the plant roots. Several countries in Asia have applied phytoremediation to degrade hydrocarbons, but this process is not yet widely used in south-east Asian countries. Hence, phytoremediation should be promoted in this region.

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