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Mitigation of petroleum-hydrocarbon-contaminated hazardous soils using organic amendments: a review

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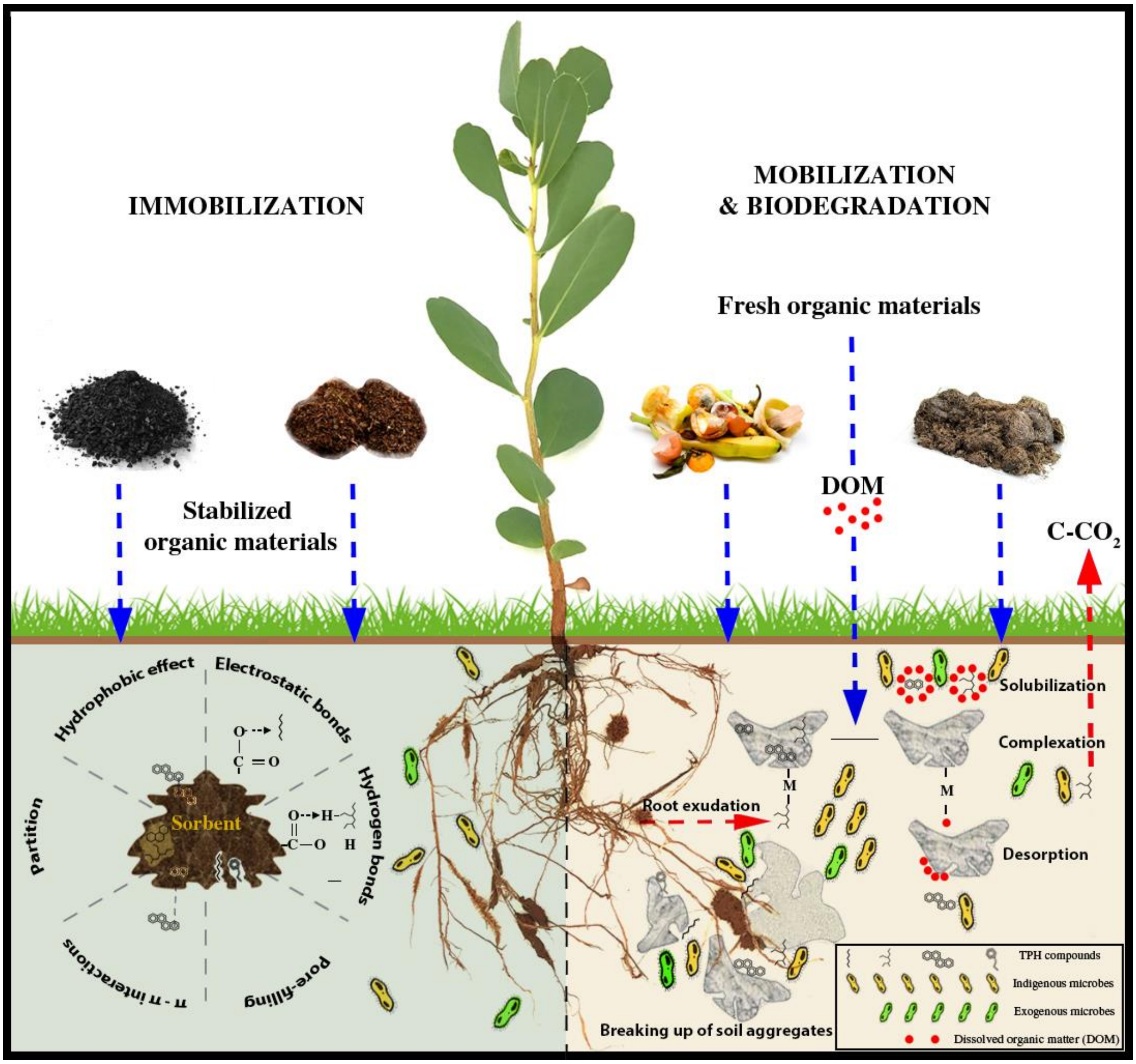
Contents

Abbreviations: BTEX - benzene, toluene, ethylbenzene, and xylene; DOC - dissolved organic carbon; DOM - dissolved organic matter; HMW - high molecular weight; LMW - low molecular weight; NAPL - non-aqueous phase liquid; PAH - polycyclic aromatic hydrocarbon; PCR - polymerase chain reaction; SOM - soil organic matter; TPH - total petroleum hydrocarbons.

Highlights

- The first-time review on role of organic amendments to remediate TPH-polluted soils.
- Organic amendments can cause either net immobilization or net mobilization of TPH.
- Plant-microbe based remediation systems benefit from organic amendment applications.
- Persistence of organic amendments on TPH remediation need further investigation.

Graphical abstract



ABSTRACT

The term “Total petroleum hydrocarbons” (TPH) is used to describe a complex mixture of petroleum-based hydrocarbons primarily derived from crude oil. Those compounds are considered as persistent organic pollutants in the terrestrial environment. A wide array of organic amendments is increasingly used for the remediation of TPH-contaminated soils. Organic amendments not only supply a source of carbon and nutrients but also add exogenous beneficial microorganisms to enhance the TPH degradation rate, thereby improving the soil health. Two fundamental approaches can be contemplated within the context of remediation of

TPH-contaminated soils using organic amendments: (i) enhanced TPH sorption to the exogenous organic matter (immobilization) as it reduces the bioavailability of the contaminants, and (ii) increasing the solubility of the contaminants by supplying desorbing agents (mobilization) for enhancing the subsequent biodegradation. Net immobilization and mobilization of TPH have both been observed following the application of organic amendments to contaminated soils. This review examines the mechanisms for the enhanced remediation of TPH-contaminated soils by organic amendments and discusses the influencing factors in relation to sequestration, bioavailability, and subsequent biodegradation of TPH in soils. The uncertainty of mechanisms for various organic amendments in TPH remediation processes remains a critical area of future research.

Keywords: Total petroleum hydrocarbons; Organic amendments; Immobilization; Mobilization; Bioremediation.

1. Introduction

It is estimated that the world's population will reach nine billion by the year 2050 (Gómez-Sagasti et al., 2018). The increasing population, together with continuously growing economies, expands the production, distribution, and utilization of petroleum products to meet the energy demand of the population, but it also leads undesirably to environmental pollution, especially in the soil environment. At least 342,000 sites in Western Europe are contaminated by petroleum hydrocarbons; in the US, 90% of contaminated sites are linked to petroleum-based contaminants (Panagos et al., 2013, Stroud et al., 2007). Globally, assessing the risk of soils impacted by petroleum products and determining their remediation are among the most complicated tasks in the environmental sector, especially for financial and technical implications (Hoang et al., 2020).

Total petroleum hydrocarbons (TPH) are the measurable amount of several hundred hydrocarbon compounds that have originated from crude oil, and they consist of predominantly hydrogen (H) and carbon (C). In many countries, the amount of TPH is a commonly used gross parameter for quantifying the degree of contamination to manage environmental remediation (Pinedo et al., 2013). TPH are made up of hydrocarbon compounds of two types: volatile petroleum hydrocarbons and extractable petroleum hydrocarbons. While the former group includes small chain hydrocarbons (C_6-C_{10}) such as benzene, toluene, ethylbenzene, and xylene (BTEX), the latter comprises long-chain hydrocarbons (C_{10}-C_{40}) and polycyclic aromatic hydrocarbons (PAHs) (Kamath et al., 2004). With respect to chemical structures, TPH include aliphatic hydrocarbons such as alkane, alkene, and alkyne hydrocarbons, and aromatic hydrocarbons (Supplementary Information, SI Figure 1), which are considered mainly recalcitrant in nature and classified as priority contaminants (Varjani, 2017). Once TPH contaminants enter the soil, they pose severe threats to humans, animals, and plants (Hoang et al., 2020). Hence, the selection of effective and low-cost technologies for remediating TPH-contaminated soils is an urgent need for the reclamation and restoration of the contaminated soil environment and for the reduction of the potential hazard to human and ecological health.

The fast demographic and economic growth also has produced a massive amount of organic wastes, causing environmental impacts and increasing disposal costs (Renaud et al., 2017). Organic wastes have been increasingly considered as potential materials for land application. Incorporation of different organic wastes to soils as conditioners or fertilizers has been proved to be an ecologically sound, socially acceptable, and economically attractive alternative to landfilling or incineration of the waste materials. Organic wastes also might enhance the remediation of contaminated soils (Wu et al., 2017, Wu et al., 2017). Incorporation of amendments that have originated from raw or properly treated organic wastes into TPH-contaminated soils include crop residues, green manures, animal manures, biosolids, composted organic materials, and biochar, was found to be effective in enhancing the remediation efficiency (Huang et al., 2019). The carbon substrates, nutrients, and active microorganisms added to soils via the amendments could favourably alter soil properties and TPH dynamics and stimulate the growth and activity of soil microbial populations that are beneficial for TPH remediation (Koshlaf et al., 2020, Nwankwegu et al., 2016).

Although the use of organic amendments for environmental remediation has been reported in a number of studies, most of the current research has focused on heavy metal(loid)s (Park et al., 2011), a particular group of TPH (i.e., PAHs) (Sayara and Sánchez, 2020), or one specific category of organic amendments (e.g., plant residues, composts, biochar) (Guo et al., 2020, Medina et al., 2015, Ren et al., 2018). No review article has discussed the remediation of TPH-contaminated soil with a range of soil organic amendments. The main aim of this work is to summarize the potential benefits of organic amendments for the remediation of TPH-contaminated soils. First, the abundance, suitability, and drawbacks of organic amendments for TPH remediation are critically discussed. Second, how organic amendments enhance the remediation of TPH-contaminated soil is considered. Finally, the factors affecting the ability of organic amendments to remediate TPH-contaminated soil are described, with the aim of determining the most sustainable outcomes including the the long-term effects of organic amendments on the mobilization and bioremediation of recalcitrant fractions of TPH.

2. Dynamics of TPH in contaminated soil environments

Once TPH contaminants enter the soil environment from natural and anthropogenic sources as depicted in Figure 1, the compounds undergo weathering that involves physicochemical and biological processes. The weathering of different TPH components depends largely on their physicochemical characteristics, initial concentration, and chemical composition (Balseiro-Romero et al., 2018). The process is also influenced by soil properties including physicochemical characteristics (e.g., particle size, porosity, organic matter content, permeability) and biological characteristics (e.g., indigenous microbial abundance and activity). In addition, environmental conditions, such as temperature, humidity, and precipitation, affect weathering (Truskewycz et al., 2019). Hence, fate of TPH in the soil environment are influenced by both abiotic and biotic processes.

2.1. Volatilization and dissolution

After oil spills on land surfaces, the gasoline compounds (i.e., a highly volatile fraction with low equivalent carbon number) can quickly volatilize due to their low boiling point (SI Figure 2) (Balseiro-Romero et al., 2018, Rivett et al., 2011). Simple aromatic compounds (e.g., BTEX) may evaporate completely under warm conditions if the compounds are not confined via environmental interactions (e.g., leaching to groundwater) (Simantiraki et al., 2013, Truskewycz et al., 2019). Conversely, only a small proportion of diesel compounds can escape to the atmosphere via volatilization (Balseiro-Romero et al., 2018). The volatilization of TPH contaminants changes the viscosity and density of the residual non-aqueous liquid, thereby affecting its transportation over time (Fine et al., 1997). Moreover, TPH compounds can move downward through the soil matrix and create a vapour plume in the saturated zone. Subsequently, the compounds can migrate to the atmosphere, or partition into the soil pore water or groundwater (Rivett et al., 2011). Petroleum hydrocarbon vapours are transported to the gaseous phase through diffusion or advection, and the process depends mainly on soil-pore characteristics (Balseiro-Romero et al., 2018). Gas-phase mass transfer in a TPH-contaminated soil consists of volatilization from the non-aqueous phase liquid (NAPL) and partitioning in the gaseous/aqueous interphase (Balseiro-Romero et al., 2018, Rivett et al., 2011).

Water-soluble TPH compounds with low molecular weight (LMW) or low equivalent carbon number can leach through the soil profile due to their high water solubility (SI Figure 2)

(Balseiro-Romero et al., 2018, Cipullo et al., 2018). The dissolution of TPH contaminants from NAPL and gaseous phases to soil water can be described by first-order mass transfer kinetics (Rahbeh and Mohtar, 2007). The dissolved pollutants can be transported through the available pore water in the soil matrix via advection, diffusion, and dispersion. Dispersion is the mechanical spreading of TPH contaminants resulting from non-uniform and indirect pore water movement within the heterogeneous soil system (Balseiro-Romero et al., 2018). The presence of polar molecules (e.g., alcohol, acetone) can increase the solubility of C_{34+} *n*-alkane hydrocarbons in soils (Truskewycz et al., 2019).

2.2. Degradation and sorption/desorption

Degradation is fundamental for the natural attenuation of TPH residues in soil, and it can occur via either abiotic (e.g., photodegradation by sunlight) or biotic (e.g., microbial degradation) pathways (Serrano et al., 2008). The process involves a variety of interactions among TPH characteristics, microorganisms, physicochemical properties of the soil, and environmental conditions (Hoang et al., 2020).

Sorption plays a crucial role in the advective-dispersive transport dynamics, persistence, transformation, and bioaccumulation of TPH contaminants (Rivett et al., 2011). Sorption of TPH contaminants to soil particles can occur through various mechanisms such as hydrogen bonding, covalent bonding, and hydrophobic interactions (Pierzynski et al., 2005). Typically, sorbed TPH contaminants are less accessible or available to microbes, thereby limiting degradation and transport of the compounds in soil (Khan et al., 2018). The sorption process increases with increasing organic matter, clay content, and hydrophobicity of hydrocarbon (Providenti et al., 1993). Soil organic matter (SOM) (both quantity and nature) has been recognized as one of the critical factors in determining sorption of TPH (Liu et al., 2013).

Generally, LMW TPH contaminants, when entering the soil, are relatively mobile and degradable due to low hydrophobicity (i.e., a low K_{ow} value), high water solubility, and high volatility (SI Figure 2). In contrast, high molecular weight (HMW) or high equivalent carbon number TPH contaminants are more persistent and resistant to degradation (Harmsen and Rietra, 2018). The degradation of TPH contaminants decreases as time passes due to the decline in bioavailable TPH fractions. The major means by which TPH contaminants are removed from soils is biotic through degradation or co-degradation processes mediated by soil microorganisms (Hussain et al., 2017). In this context, the removal of TPH is mainly limited by a low supply of contaminants to microorganisms due to TPH sorption and sequestration (i.e., aging) in soil. Yet, the aging process can also reduce the toxicity of the contaminants to receptors owing to their poor bioavailability (Ouvrard et al., 2013).

Nonetheless, the degradation of sorbed TPH contaminants is not necessarily negligible because, under certain conditions, microbes can access and degrade at least part of the compounds (Chen et al., 2010). On the one hand, microorganisms can secrete extracellular enzymes or modify their cell properties in order to take up or degrade the sorbed contaminants (Balaji et al., 2014, Xu et al., 2013). On the other hand, the TPH compounds sequestered by partitioning onto the soil, or by entrapment and diffusion into soil pores during the weathering process, could subsequently be released into the gaseous or aqueous phases (desorption), making them available for microbial degradation (Cornelissen et al., 2000). The presence of plant roots may further alter TPH bioavailability in soils due to root exudates (Martin et al., 2014). Indeed, LMW organic acids from root exudates can enhance not only TPH bioavailability by providing competing sorptive sites in soil particles, but also microbial growth and activity in the soil, favouring the biodegradation of TPH contamination (Hussain et al., 2017). Bioactive compounds secreted from plants and microorganisms are essential in desorbing bound TPH contaminants from soil matrices (Truskewycz et al., 2019).

3. Organic amendments for the remediation of TPH-contaminated soils

Organic amendments denote the materials derived from biomass or living beings (containing mainly C) that can enhance soil health and crop productivity. Hence, use of organic amendments in agriculture is a common practice (Hamid et al., 2020). Owing to their unique textural and surface properties, chemical composition, and the functional groups, their potential use has been extended for environmental remediation, and the research and development in this area has been gaining significant momentum since the last decade (Gómez-Sagasti et al., 2018, Park et al., 2011). Within the context of remediation of soil TPH contamination, a wide array of organic amendments, including crop residues, green wastes, manures, biosolids, and biochar (Figure 1 & SI Table 1) can be used due to their positive effects on soil health and plant growth. The amendments not only favourably alter soil physical (e.g., bulk density), chemical (e..g., cation exchange capacity) and biological (e.g., microbial activity) properties but also supply essential nutrients for enhancing plant growth.

Remediation of soil contaminated with TPH can be achieved by minimizing TPH bioavailability through chemical and biological immobilization in soils, consequently reducing TPH exposure to receptors (Cao et al., 2016). In contrast, localized TPH contamination in urban areas can be remediated by mobilizing the contaminants using phytoextraction (i.e., the use of plants to clean up pollutants via accumulation in harvestable tissues) and chemical washing (Huguenot et al., 2015). Moreover, TPH contaminants can undergo plant and microbial degradation (biodegradation), which could affect the concentration and bioavailability of the compounds (Dubrovskaya et al., 2017, Garcés Mejía et al., 2018). Immobilization, mobilization, and biodegradation are, therefore, the key remediation strategies for managing soils contaminated with TPH (Hussain et al., 2017, Lamichhane et al., 2016). Organic amendments increase surface area, surface charge density, and quantity of exchange sites in the impacted soil, leading to enhanced surface adsorption and complexation, thereby reducing

TPH mobility in the soil (Farzadkia et al., 2019) (Figure 2-A). For example, sorption coefficients for PAHs in soils were increased by ten-fold and up to hundred-fold after adding 10% (w/w) compost and 5% (w/w) biochar, respectively (Sigmund et al., 2018). Besides, organic amendments can redistribute TPH from solid to aqueous phases by influencing the sorptive sites in soil particles, consequently enhancing TPH availability for microbial degradation (Cai et al., 2017) (Figure 2-B). For instance, the addition of sewage sludge and cow dung in an used lubricant oil-contaminated soil (10%, w/w) resulted in 82 and 94% biodegradation, respectively, which were significantly higher than that in the control (without any amendment) setup (56%) (Agamuthu et al., 2013). Due to its dual effects, the selection of organic amendments should be carefully considered to suit the remediation purpose. A brief description associated with different organic amendments deployed for remediation of soil contaminated with TPH is provided below.

3.1. Bio-wastes

Bio-wastes consist of a wide range of readily available organic amendments, such as crop residues, green manures, animal manures, biosolids, and yard waste composts (Quilty and Cattle, 2011). The amendments have been used to remediate the soil contaminated with TPH (Sarkar et al., 2005, Shahsavari et al., 2013). Crop residues and green manures are generated at approx. 3.8 billion Mg annually, and they could supply a massive amount of C and essential nutrients to degraded soils if managed properly (Thangarajan et al., 2013). Indeed, returning crop residues to TPH-contaminated soils could be a sustainable method of nitrogen (N) fertilization to the soils (Che et al., 2018, Chen et al., 2014, Lal, 2005). The essential role of crop residues in TPH remediation has been reported in different studies (Shahsavari et al., 2013, Wang et al., 2016). The amendments immobilized TPH via sorption, which alleviated TPH toxicity, thereby enhancing microbial and plant growth (Medina et al., 2015, Nunes et al., 2020).

Similarly, a massive amount of animal manure can be recycled for environmental remediation. For example, a huge production of animal feces (~ 12 million tons) occurs on daily basis globally (Faostat, 2016). Apart from supplying essential nutrients such as N, P, K, S, and micronutrients, the presence of humic and fulvic substances, which are considered to be the most active compounds in animal manure, affects not only the soil characteristics but also TPH bioavailability (Ezenne et al., 2014). Animal manure incorporated in a TPH-contaminated soil enhanced dissolved organic matter (DOM), which was attributed to reduced sorption capacity of the clay fraction of the soil (Wei et al., 2014). DOM is considered as an effective desorbing agent for a variety of TPH compounds such as aliphatic hydrocarbons (e.g., 1,2-dimethylcyclohexane, pristane) (Chen et al., 2019, Kopinke et al., 2011) and PAHs (e.g., phenanthrene, pyrene) (Chen and Yuan, 2012, Zhang and He, 2013). In addition, meats produced from animal by-products have been shown to be a carbon and nutrient source that can be used for TPH remediation (Liu et al., 2019). The composition, nutrient value, and usefulness of animal manure depend on various production factors, such as animal types, manure collection, storage, and handling (Miller et al., 2003). Due to these factors, their effects on the impacted soils might be dissimilar (Olawale et al., 2020). For instance, Adesodun and Mbagwu (2008) found that the addition of poultry manure resulted in higher rate of TPH reduction in comparison to those in cow dung- and pig waste-treated TPH-contaminated soil. The differential performance of these animal manure was attributed to different C: N: P ratios in the amendments which affected differently on hydrocarbon degraders in the impacted soil (Adesodun and Mbagwu, 2008).

Other organic amendments, such as biosolids, paper mill wastes, and composts, also are applied for remediation of soils impacted by TPH contaminants (Dindar et al., 2017, Wang et al., 2012). The existence of humic substances in the amendments offers a feasible approach for remediation of TPH-contaminated soils (Dindar et al., 2017). Biosolids or slurries incorporated into a TPH-contaminated soil can alter TPH bioavailability or enhance microbial activity to remediate the soil (Rivera-Espinoza and Dendooven, 2004). For instance, the application of digestates from biogas production favoured TPH biodegradation (Gielnik et al., 2019). Likewise, sugarcane- and paper-industry waste stimulated biodegradation of TPH contaminants by improving soil properties and microbial activity (Babaei et al., 2020).

A variety of organic wastes from food scraps, leaves, cattle manure, or even sewage sludge can be composted and used in TPH-contaminated soils (Ren et al., 2018). The addition of composts is often linked to altered TPH availability due to formation of metal-humic complexes (Wu et al., 2013). The incorporation of composts can increase SOM content, which decreases TPH bioavailability in the contaminated soil (Puglisi et al., 2007). Furthermore, the cation exchange capacity of humic substances in composts is considered equivalent to that of clay minerals in soil, and it is beneficial in retaining essential nutrients in the soil (Kim et al., 2017).

3.2. Biochar

Another important organic amendment that can be used in TPH remediation is biochar. Biochar is basically a carbon-rich and porous material produced by the pyrolysis of various organic wastes under oxygen-limited conditions (Zhu et al., 2017). Its potential for remediation is well documented owing to its high surface area, nano- and micro-porosity, and functional groups (Li et al., 2019). Owing to the porous properties and the strong adsorption ability of biochar, it has been widely used as a TPH adsorbent in contaminated soils (Jia et al., 2020, Wei et al., 2020). Besides, biochar can alter soil physicochemical properties and supply a high amount of essential nutrients to favour plant growth and TPH microbial degradation (Gul et al., 2015). Therefore, amending contaminated soils with biochar is considered as an effective method for remediating soils contaminated with TPH (Lamichhane et al., 2016).

Khan et al. (2015) applied biochars produced from four feedstocks (sewage sludge, soybean straw, rice straw, and peanut shell) prepared at 500 ºC at 2 and 5% (w/w) to a PAH-contaminated urban soil. The application of biochars reduced significantly the bioaccessible PAHs in soil and their bioaccumulation in turnip (*Brassica rapa* L.,), with the highest efficiency at 5% biochar application. Amongst the organic amendments, peanut shell biochar was the most effective in reducing bioaccessible PAHs which was attributed to its least polar surface and highest aromatic functional groups (C – H) (Khan et al., 2015). In a long-term field experiment, soil amended with wheat straw biochar produced at 650 ºC at the rate of 30 and 45 ton ha^{-1} resulted in a reduction of dissolved PAHs by 25 and 22%, respectively (Oleszczuk et al., 2016). The authors reported that the strength of biochar-PAH interactions tended to reduce over time and there was no potential risk regarding the content of dissolved PAHs in the biochar-treated soil.

In addition to the "adsorption" effects, the application of biochar can enhance TPH biodegradation ("biostimulation" effects). Qin et al. (2013) studied the effects of addition of rice straw biochar (2%, w/w) prepared at 500 ºC on microbial composition and ultimate TPH biodegradation in a soil contaminated with TPH at 16,300 mg kg^{-1} during a 180-day period. The addition of biochar at 80th day promoted significant TPH removal (84.8%) due to higher sorption of metabolites which were toxic to soil microorganisms. The presence of 2% biochar reduced bioavailability and ecotoxicity of TPH, thereby stimulating the microbial community and subsequent TPH degradation. Similarly, Kong et al. (2018) applied sawdust and wheat straw biochars to a TPH-polluted soil and noticed that the addition of biochars provided a better habitat for PAH degraders resulting in accelerating PAHs biodegradation (Kong et al., 2018).

3.3. Drawbacks associated with the application of organic amendments

Despite the abovementioned advantages, use of organic amendments can exhibit some adverse effects. Application of crop residues containing a sizeable input of carbonaceous material could result in the immobilization of inorganic N in soils (Chen et al., 2014). Similarly, application of animal manures and biosolids in the soil can result in several potential side effects which include: (i) high concentration of aerobic and anaerobic decomposition products including N compounds (e.g., NH_4^+ -N, NO_3^- -N) and greenhouse gas emission (e.g., CO_2, CH_4); (ii) input of undesirable substances such as inorganic (e.g., heavy metals) or organic (e.g., PAHs, antibiotics, micro-plastics) contaminants, and infectious agents; and (iii) N and P runoff due to a surplus input of nutrients from the organic amendments (Park et al., 2011, Scotti et al., 2015, Thangarajan et al., 2013). In addition, heavy metals can be introduced into the soil following compost or biochar addition when feedstocks (e.g., biosolids, animal manure) contain a considerable amount of metals. For example, a high cadmium level (10 mg kg^{-1}) from animal manure was reported in China (Wang et al., 2014). Additionally, the use of composts and biochar as soil organic amendments has the potential to alter soil electrical conductivity and pH (Mohan et al., 2014, Scotti et al., 2015). As a result, studies exploring such effects of organic amendments may be required for obtaining the most desirable remediation outcomes.

In addition, in the case of biochar utilization, though bioenergy and syngas can be produced during the synthesis of biochar, whether the pyrolysis system is economically feasible remains unclear. In some instances, the production of biochar caused negative energy balances due to a higher energy consumption of the pyrolysis unit (Brassard et al., 2018). Therefore, to gain maximum benefits from biochar application for the remediation of contaminants including TPH, a life cycle analysis of biochar is necessary.

4. Enhanced remediation of TPH-contaminated soils by organic amendments

Broadly, two approaches, namely immobilization and mobilization, have been deployed to treat soils contaminated with TPH. Recent studies indicate that the addition of organic amendments reduces TPH contents in the contaminated soil (Kong et al., 2018, Ren et al., 2018). The enhanced remediation of soil contaminated with TPH is caused not only by elevated sorption of TPH to the newly added organic matter (immobilization), but also by desorption, solubilisation, or complexation (mobilization) derived from organic amendments. Once mobilized, TPH contaminants can become redistributed in the aqueous soil phase, which is available for biodegradation to occur.

4.1. Immobilization of TPH

The immobilization technique is applied to reduce TPH toxicity via reducing their solubility, mobility and bioavailability in soil. Various organic amendments have been deployed to enhance the remediation efficiency (Table 1), with the aim of reducing human and animal exposure, plant uptake, and leaching to groundwater.

4.1.1. Biochar

Due to its intrinsic nature, biochar is increasingly used in immobilization (Lamichhane et al., 2016, O'Connor et al., 2018, Palansooriya et al., 2020). Application of biochar, which was produced from harvested sugarcane residue and at a pyrolytic temperature of 550 ^{o}C, to a soil contaminated with crude oil increased the sorption of aromatic fractions through various surface interactions such as partition and π–π electron donor-acceptor interactions (Wei et al., 2020). Similarly, several studies have shown that biochar incorporated into TPH-contaminated soils can increase sorption of TPH contaminants to organic and mineral surfaces. The enhanced sorption could reduce the toxicity of TPH contaminants (Li et al., 2019, Qin et al., 2013, Zhu et al., 2017).

Different mechanisms are attributed for the increased sorption of hydrocarbon compounds onto biochar after incorporating it into TPH-contaminated soils. Firstly, the adsorption of TPH contaminants on carbonized fraction of biochar is achieved by electrostatic and van der Waals interactions (physical sorption) (Jiang et al., 2016), or via π–π bonds, hydrogen bonds, and coordination bonds (chemical sorption) (Yang et al., 2018). Secondly, the partition of TPH contaminants on biochar due to hydrophobic interactions is closely associated with the solubility and hydrophobicity of TPH (Kandanelli et al., 2018). Thirdly, the pollutants could bind strongly to biochar through micro-pore filling (Nguyen and Pignatello, 2013). Under various conditions, one of the abovementioned mechanisms predominates for the sorption process, and other mechanisms also are involved (Dai et al., 2019). It is well known that TPH bioavailability in the soil is readily affected after the addition of biochar.

4.1.2. Mature and stabilized bio-wastes

The organic matrix of compost enhances the remediation of soil contaminated with TPH because of enhanced sorption. Indeed, humic substances (e.g., fulvic acids, humic acids, and humin), which are formed from organic wastes during composting processes, could stimulate TPH immobilization in soil, thereby reducing both their leachability and uptake by plants (Wu et al., 2013). Therefore, amending the native soil with compost-derived organic matter might be employed to remediate the soil contaminated with TPH.

Humic acids, which are theoretically the soluble fraction of humic substances at an alkaline pH, show a strong ability to sorb hydrophobic organic pollutants (Chianese et al., 2020). Generally, mature compost, which is characterized by relatively high levels of humic acids, showed higher sorption capacity to TPH contaminants than immature compost (Barriuso et al., 2011, Farzadkia et al., 2019) (SI Figure 3). The sorbing efficiency of humic acids is associated with the hydrophobicity of the sorbate, as influenced by the functional groups. The interactions of humic acids with TPH contaminants are mainly attributed to hydrophobic effects, π–π bonds, and hydrogen bonding (Chianese et al., 2020). For instance, the application of mature olive mill waste compost reduced the mobility and toxicity of PAHs in a polluted mine soil and decreased PAH availability in a creosote-contaminated soil (García-Delgado et al., 2019). Similarly, the addition of 10% (w/w) compost as a soil amendment to a pyrene-contaminated soil resulted in a moderate increase in the sorption of the pollutant (Kah et al., 2018). Likewise, the addition of cotton gin crushed compost ameliorated the toxicity of gasoline to soil microorganisms (Tejada et al., 2008). The higher binding ability of humic acids than that of fulvic acids with the contaminants has been reported, and the disparity could be explained, at least partly, by the higher aromatic content and molecular weight of humic acids (De Paolis and Kukkonen, 1997, Plaza et al., 2009, Tejada et al., 2008). In addition to high levels of humic acids, the lignin-cellulosic residues in mature compost could serve as a major compartment for TPH sorption (Wu et al., 2013). Different compost amendments (composted biosolids, mushroom compost, and leaf compost) reduced both direct (soil-human) and indirect (soil-plant-human) exposure of TPH contaminants due to hindrance of their bioavailability (Attanayake et al., 2015). Similarly, several studies have illustrated that various organic wastes could be composted and used as sources for strong sorption of TPH compounds in the soil (Table 1). It is generally accepted that the more advanced the humification processes, the greater TPH sorption and the lower the availability.

Enhanced immobilization by incorporating the organic amendments protects plants and microorganisms from being overloaded with bioavailable TPH (Bushnaf et al., 2017). The approach is especially efficient when either there is high bioavailability of TPH or conditions become less supportive for microbial growth and activity in the contaminated soil (e.g., inorganic nutrient scarcity or low organic content is expected) (Bushnaf et al., 2011, Khan et al., 2015, Meynet et al., 2014). Despite that, the effect might be only for relatively short time periods (Bolan et al., 2014, Wu et al., 2013). Sorbed contaminants can be re-released into the soil through changes in soil properties or microbial action, causing the organic amendments themselves to be a source of TPH contamination in soils (Zama et al., 2018). These problems can be overcome by using the above amendments as pre-treatments to immobilize and concentrate TPH contaminants in soils (García-Delgado et al., 2015). After the pre-treatment, a slow release of sorbed TPH and an inoculation of hydrocarbon-degrading microbes are simultaneously deployed by introducing organic amendments, which can achieve the complete biodegradation of the contaminants in soil (Barati et al., 2017, Beesley et al., 2010).

4.2. Mobilization and bioremediation of TPH

Mobilization is a common approach to remove TPH contaminants in soil via washing (i.e., soil flushing) or by plant uptake (i.e., phytoextraction). When soils contaminated with TPH are being remediated using organic amendments, the process for mobilization is often bio-mineralization of the contaminants into innocuous products such as CO_2 and H_2O (Zama et al., 2018) (Table 2).

4.2.1. Mobilization

Contrary to the sorption effect discussed above, the addition of organic amendments can also enhance TPH availability in contaminated soil, and this increase might be due to the presence of DOM (Bolan et al., 2011, Chen and Yuan, 2012). A variety of organic amendments can produce DOM resulting in enhanced mobilization of TPH contaminants. For example, Kobayashi et al. (2009) found that covering top soil with compost in a PAH-impacted soil resulted in the leaching out of DOM, which contributed mainly to enhanced solubility and bioavailability of phenanthrene, pyrene, and benzo[a]pyrene. The dissolved organic fraction theoretically breaks the mineral-TPH association releasing the associated contaminants into soil solution (increased extractability and bioavailability) (Chen et al., 2019). Additionally, DOM can increase the solubility and mass transfer of TPH into the aqueous phase. In this phase, microbes have direct access to DOM-bound hydrocarbon compounds (Cai et al., 2017). Xiu-Hong et al. (2014) reported that pine needle litter-derived DOM reduced the strong sorption of phenanthrene and fluoranthene to soil particles. Similarly, desorption of PAHs from soil was facilitated by compost-derived DOM. The DOM could strongly interact with the sorbed PAHs (Yu et al., 2011). Returning crop residues to soils could enhance dissolved organic carbon (DOC) content (Zhao et al., 2018). The increased DOC could impede sorption and mobilize PAHs in the soil (Wang et al., 2019). The DOC is even effective in desorbing biochar-adsorbed PAHs (Bao et al., 2020, Hussain et al., 2018).

Soil microorganism-DOM associations can be formed via physical connections, and they facilitate the microbes' direct contact with the DOM-bound hydrocarbon compounds. Because there is competition for the interaction sites between DOM and microbial cells, microbes move to the hydrocarbon compounds (Haftka et al., 2008, Schaefer and Juliane, 2007). For example, after adding four types of DOM to the soil, the soil bacterium *Pseudomonas putida* G7 showed chemotaxis, and this resulted in bacterial DOM-enhanced mobility (Jimenez-Sanchez et al., 2015). Inoculating soil with microorganisms following the addition of organic amendments could enhance the solubility of highly hydrophobic TPH (e.g., HMW PAHs) by transforming the PAHs into comparatively polar metabolite(s), which can be further metabolised; naturally existing microbes may not readily metabolise them (Cébron et al., 2015).

Through rhizoremediation, incorporation of organic amendments can have indirect effects in controlling TPH bioavailability. By definition, rhizoremediation is the process in which plant-associated microorganisms degrade organic contaminants in the rhizosphere (Saravanan et al., 2020). Plant roots secrete LMW organic acids, which enhance TPH bioavailability in the rhizosphere because they desorb the contaminants (Jia et al., 2016, Liu et al., 2015). Ni et al. (2018) observed that corn-straw-derived biochar incorporated into a PAH-contaminated soil enhanced the desorption of PAHs through increased secretion of LMW organic acids in the rhizosphere of rice. It is assumed that organic acids can break the “bridges” of metal cation – TPH and soil mineral – TPH associations, leading to the release of TPH molecules into the aqueous soil phase (Gao et al., 2010, Ling et al., 2015). Moreover, the breaking up of soil aggregates is a physical effect of root tips that can release TPH entrapped in the aggregates into soil solution (Khan et al., 2013, Masciandaro et al., 2013). Addition of organic amendments can enhance plant growth in soils contaminated with TPH, which, in turn, increases root development and exudation and, consequently, TPH bioavailability in the soil (Barati et al., 2018, Bell et al., 2014, Eisenhauer et al., 2017).

4.2.2. Bioremediation

Degradation via microorganisms is considered as a green and sustainable method to remediate soils contaminated by TPH. However, natural biodegradation of TPH in contaminated soil is often limited due to low microbial quantity and activity, soil nutrients, and oxygen status, amongst other factors (Hoang et al., 2020). Apart from enhancing TPH bioavailability by supplying DOM and LMW organic acids, as mentioned above, organic amendments can stimulate TPH bioremediation when added to the soil. The bioremediation could be stimulated through various effects, as discussed below.

4.2.2.1. Bio-stimulating effects

Incorporation of organic amendments stimulates the capability of naturally existing microorganisms to biodegrade TPH in soils. Increased organic matter content by adding organic amendments in the contaminated soil could improve soil aggregate stability and other physical properties, such as soil porosity, available water, and water infiltration and percolation (Medina et al., 2015, Wang et al., 2016). The improvement of soil physical properties promotes microbial degradation of TPH in the impacted soils (Akbari and Ghoshal, 2015). SOM plays a fundamental role in making stable soil aggregates because it helps to form cationic bridges that bind mineral particles to organic polymers (Bronick and Lal, 2005, Qurashi and Sabri, 2012). Whereas easily decomposable, LMW substances are transient, more recalcitrant, HMW organic components (e.g., lignin, cellulose) have a smaller, but the longer-lasting effect, on aggregate stability (Six et al., 2004). Additionally, the enhanced biological activity derived from the decomposition of added organic matter, coupled with the release of microorganisms' agglutinants (e.g., exo-polysaccharides), likely contribute to increased soil aggregate stability (Qurashi and Sabri, 2012). The improved structural stability, in turn, results in a reduction in bulk density and an increase in porosity, hydraulic conductivity, and available water of the impacted soil (Wang et al., 2016).

The application of organic amendments (pea straw residues) to a PAH-contaminated soil increased porosity when compared to unamended soil (without pea straw addition) (Koshlaf et al., 2019). Soil aeration is of essential importance in the bioremediation process, because the initial degradation of petroleum hydrocarbons requires the activity of oxidative enzymes (dioxygenases and monooxygenases), thus making oxygen a limiting factor for biodegradation in many processes (Varjani, 2017). Indeed, incorporation of bulk materials (e.g., sawdust, coconut husk, straw, and woodchips) to TPH-contaminated soils improves soil properties including microbial activity and reduces bioremediation time (Ma et al., 2016, Nwankwegu et al., 2016, Wang et al., 2016). Similarly, the addition of organic amendments improves soil physical properties by providing a better balance between both macropores (among aggregates) and micropores (within aggregates), which contribute to permeability and available water, respectively. With optimal soil structure, water moves more easily into and through the soil (Gul et al., 2015).

Organic amendments also provide abundant labile carbon and nutrient sources to increase SOM and stimulate soil microbial biomass and activity in TPH-contaminated soils, thereby increasing natural attenuation potential of the soils (Krzebietke et al., 2020, Larkin, 2015, Liu et al., 2020, Lukić et al., 2016, Wang et al., 2016). Different sorts of organic amendments, including food waste, vegetable waste, animal manures, and sewage sludge have been shown to enhance the overall efficiency of PAH bioremediation by supplying carbon and nutrients to the PAH degraders (Lukić et al., 2016). Surprisingly, biochars (especially those produced from nutrient-rich feedstocks and at low temperature) can also provide a noticeable amount of labile components to enhance soil microbial growth and activity (Cross and Sohi, 2011). Furthermore, research has shown that increases in organic C stock following addition of organic material (e.g., cow dung, poultry waste, palm oil waste) resulted in enhanced nutrient retention in TPH-contaminated soils, thereby affecting the growth of microbes and plants (Alvarenga et al., 2017, Egobueze et al., 2019). The goal is to have a gradual and continuous release of mineral nutrients to support soil organisms' requirements (Scotti et al., 2015).

Organic amendments introduced into TPH-impacted soils favour the growth of indigenous microbial communities, which are indicators of soil health and quality in addition to the traditional physicochemical ones (Ge et al., 2013, Huang et al., 2019, Koshlaf et al., 2019, Zhang et al., 2020). Wheat straw added to crude oil-contaminated soils resulted in an increase in the cumulative CO_2-C respiration and microbial biomass after a 60-day incubation (Alotaibi et al., 2018). Similarly, using the real-time polymerase chain reaction (PCR) technique, Ros et al. (2014) reported that, after 6 months of incubation, there was an increase in bacterial abundance in an oil-sludge-contaminated semi-arid soil treated with biosolids compared to a control soil with no biosolids. In a recent study, Zhang et al. (2020) used two highly efficient oil-degrading bacterial strains and demonstrated, after 40 days of incubation, a higher oil degradation rate in the presence of wheat bran and swine wastewater compared to soil without the organic materials. The wastewater supplied a nitrogen source and process water for the biodegradation. The authors observed increases in enzymatic activities (dehydrogenase, peroxidase, and urease) and soil fertility and attributed them to the rise in microbial activity (Zhang et al., 2020).

Organic amendments also could induce changes in microbial composition due to competition for available substrates, antagonisms, or the degree of contaminant removal (Huang et al., 2019, Olawale et al., 2020, Ondoño et al., 2014). Consequently, the tendency of added organic amendments on the selective enrichment of soil microorganisms has been reported. For example, Gram-negative bacteria were enriched, and genus dominance was changed following the incorporation of plant residues (*Conocarpus* and *Tamarix*) to a soil contaminated with crude oil (Al-Saleh and Hassan, 2016). Similarly, the abundance of Gram-negative PAH-degrading bacteria (mainly *Pseudomonas* sp.) was increased in a pea-straw-amended PAH-contaminated soil (Koshlaf et al., 2019). A relative abundance of some petroleum degraders (e.g., bacteria *Sphingomonas*, *Idiomarina*, and *Phenylobacterium* and the fungi *Humicol*a, *Wallemia*, and *Graphium*) was also promoted by inputs of rice straw and sawdust in a TPH-contaminated soil (Huang et al., 2019) (Figure 3-A). Such a change in microbial composition was attributed as being the key contributor to enhanced TPH biodegradation in the soils (Gielnik et al., 2019, Koshlaf et al., 2019).

Apart from changes in microbial abundance, organic amendments could enhance microbes that are metabolically active in degrading TPH (Huang et al., 2019, Košnář et al., 2019). For instance, Bastida et al. (2016) reported that the abundance of catabolic enzymes participating in TPH biodegradation was increased in a compost-treated contaminated soil compared to an unamended treatment. Similarly, the number of *alkB* gene copies in a TPH-contaminated soil increased about 300-fold with the addition of pea straw compared to the number in an unamended soil (Shahsavari et al., 2013). Han et al. (2017) found increased abundances of the bacterial 16S rRNA and PAH-degrading genes (e.g., *pdo1* and *nah*) after adding three agricultural wastes (wheat stalk, cow manure, and spent mushroom) to soils contaminated by aged PAHs (Figure 3-B). Interestingly, the positive effect of organic amendments including compost in promoting the activity and abundance of microbial biomass was demonstrated to persist for more than five years (Hernández et al., 2015).

The presence of plants in soils contaminated with TPH could further enhance the positive effects induced by organic amendments. Enhanced root exudation (induced by the amendments) favours the activity of rhizosphere microbiota, and, in turn, some of the microorganisms can degrade TPH in the rhizosphere (Martin et al., 2014). The combined effect of root exudates and organic amendments could stimulate TPH biodegradation by regulating the community structure and function of soil microbiota (Guo et al., 2017, Hussain et al., 2018) and could induce co-metabolism in the rhizosphere (Correa-García et al., 2018). Spent mushroom compost incorporated in a black-oil hydrocarbon-polluted soil enhanced the survival of *Megathyrsus maximus* (Guinea grass) in the contaminated soil and promoted degradation of the hydrocarbon compounds (Guo et al., 2017). It is generally accepted that any soil amendment that is able to promote plant growth has promising potential for effective rhizoremediation (Agarry et al., 2014).

4.2.2.2. Bio-augmenting effects

Organic amendments incorporated in soils contaminated by TPH can help microorganisms in their bioremediation of the soil (Gielnik et al., 2019). For instance, aged refuse from a landfill containing an exogenous hydrocarbon-degrading microorganism was added to a TPH-contaminated soil, which resulted in a TPH removal rate of 89.83% (half-life: 13.86 days) (Liu et al., 2018). Similarly, the addition of biosolids containing introduced hydrocarbon degraders accelerated bioremediation of a diesel-contaminated soil (Rivera-Espinoza and Dendooven, 2004). Addition of hydrocarbon-degrading microbial communities in organic amendments could contribute to the enrichment of key microbial taxa in the contaminated soil, thereby accelerating TPH biodegradation (Gielnik et al., 2019, Huang et al., 2019). Once in the soil, the microbial reinforcement could make adaptive changes when exposed to the TPH. The changes might include morphological (e.g., formation of mycelia), physiological (e.g., adjustment of cell surface to become more hydrophobic; electron shuttles facilitating biotransformation), and behavioural (e.g., chemotaxis and swimming modes) adaptations (Ren et al., 2018).

Nevertheless, there are debates as to whether the increase in the competent microorganisms in the receiving soil is primarily from the exogenous organic matter. This concern is probably due to the relatively short survival time of the introduced microorganisms following land application (Shahi et al., 2016). To enhance the survival of the introduced microorganisms, bio-carriers such as plant residues and biochars could be used (Chen et al., 2012, Tao et al., 2019). These organic amendments can interact favourably with the exogenous microorganisms in different ways. These may include supplying a habitat, adsorption of nutrients through cation exchange capacity, and providing slow-release hydrocarbons for biodegradation (Kołtowski et al., 2017, Zhu et al., 2017).

Biodegradation of HMW fractions of TPH (e.g., > 3-ring PAHs) typically requires the concerted efforts of microbial consortia with desired catabolic activities (Ghosal et al., 2016). However, such conditions may not normally exist in most TPH-contaminated soils (Safdari et al., 2018). Therefore, the inoculation of organic-amendment-borne microbial strains or consortia could be a possible solution for increasing TPH removal (Kästner and Miltner, 2016). Depending on the origin of organic amendments, specific microorganisms could be selectively enriched. For instance, the application of organic amendments containing a significant quantity of aromatic compounds (e.g., lignin-containing plant material in compost or in animal manure) contributes to selective enrichment of ligninolytic fungi in impacted soils (Adam et al., 2015). The presence of the ligninolytic fungi was essential, particularly when the degradation of recalcitrant hydrocarbon compounds (e.g., HMW PAHs) was intended (Cao et al., 2020). Of these fungi, white-rot fungi are ubiquitous in nature and could secrete extracellularly ligninolytic enzymes, mainly peroxidases and laccases (Ghosal et al., 2016). The enzymes can diffuse towards the adsorbed PAHs that bacterial intracellular enzymes cannot access to make initial oxidation steps on the PAHs in soil. The oxidation processes are carried out by various sorts of peroxidases and laccases and considered to be crucial for the metabolism of HMW PAHs (Ghosal et al., 2016). Microbial consortia consisting of bacteria and fungi were shown to degrade effectively HMW PAHs in soil (Zhu et al., 2020). Indeed, microbes frequently exchange metabolites with other microorganisms, and, as a result, the cooperative cross-feeding could increase microbes' fitness and survival in soil (Pande et al., 2014). Several examples of effective degradation of PAHs by white-rot fungi in consortia with other soil microbes have been reported in the literature, which suggests that, along with bacteria, organic amendment-derived fungi could be exploited as ideal candidates for the bioremediation of highly recalcitrant fractions of TPH contaminants (Acevedo et al., 2011, Lee et al., 2014).

5. Factors influencing the remediation of TPH-contaminated soils with the organic amendments

Whether organic amendments immobilize or mobilize TPH in contaminated soils depends on many interactive factors involving soil properties, TPH characteristics and concentrations, the nature and quantity of organic amendments, and the environmental factors (Figure 4).

5.1. Soil properties

The amount of natural SOM is important in determining the effect that exogenous organic matter added to soil has on TPH availability (Yu et al., 2018). Moreover, different components of SOM (e.g., humic substances; black C, also known as soot) show distinct TPH sorption potentials due to differences in molecular weights, chemical structure, and polarity (Bejger et al., 2018, Chen et al., 2017, Ukalska-Jaruga et al., 2019). Generally, a high proportion of moderately aged humic compounds and the presence of black C (which are characterized by relatively high aromaticity and low polarity) in soils could cause strong sorption of hydrocarbon compounds, leading to sequestration of the pollutants (Ren et al., 2018). However, the sorption of phenanthrene to an aliphatic-rich SOM was higher than that of the highly aromatic humic acid (Salloum et al., 2002). The authors concluded that aliphatic SOM domains have a stronger affinity for phenanthrene and other PAHs compared to that of aromatic counterparts. Therefore, other characteristics of SOM such as functional groups might be the determining factor for soil TPH sorption, and, consequently the efficacy of added organic amendments in TPH remediation (Ehlers et al., 2010). These results highlight the importance of the joint consideration of functionalities and surface domain distribution of SOM on bioavailability of TPH.

Soil type plays an essential role in the bioremediation process. Reduced porosity in clay-rich soil coupled with low oxygen availability was shown to reduce remediation efficiency (Haghollahi et al., 2016). In general, biodegradation rates are restricted by poor bioavailability of TPH and limited diffusion of oxygen and nutrients to hydrocarbon-degrading aerobes in such soils (Masy et al., 2016). Incorporation of maize residue into an industrially PAH-contaminated soil led to rapid degradation of the contaminant in the large size sand fraction, while it became stabilized and accumulated in the fine size silt fraction (Pernot et al., 2014). Residue incorporation in the sand led to increased availability of PAH, whereas silts stabilized the added organic matter and associated PAH, protecting the complexes from biodegradation (Pernot et al., 2014). However, in a few field observations, fine-textured soils showed notably higher TPH remediation efficacy than medium- and coarse-textured soils following the application of organic amendments. For example, soil with a higher sand content (pore size: 2000-50 µm) showed lower loss of HMW PAHs compared to soil with higher silt content (pore size: 50-2 µm) after compost addition (Wu et al., 2013). This phenomenon was attributed to the low N content in the soil with a high proportion of sand, depicting a more prominent effect of nutrient addition via compost in the sandy than silty soil. In this study, the lower bioremediation of PAHs in the sandy soil compared to the silty soil was attributed to the high C: N: P ratio, which may potentially reduce the catabolic activity of hydrocarbon-degrading microorganisms (Wu et al., 2013). The TPH degradation rate could also depend on the pore size of soil particles that decide whether microorganisms can enter and access the contaminant molecules residing inside the soil pores (SI Figure 4). The large pore size in light-textured soils could also facilitate aeration (supply of oxygen) to TPH-degrading microorganisms inhabiting inside the pores, thereby enhancing their growth and TPH degradation rate. For example, the PAH degradation rate was slower in a clay aggregate than a sand aggregate because of higher volumes of inaccessible pores in the former (Akbari and Ghoshal, 2015). Living plants and their root systems, however, could act as extracting agents of PAH, thereby locally enhancing the contaminant availability from within the inaccessible pores making it liable to microbial degradation over a long term (Akbari and Ghoshal, 2015).

5.2. TPH characteristics and concentrations

Varjani (2017) showed that bioavailability and biodegradability of TPH were negatively correlated with the K_{ow} of the hydrocarbon compounds following organic amendment application. The LMW TPH (lower K_{ow}) are more likely to be biodegraded by microorganisms than the HMW TPH (higher K_{ow}) (Das and Chandran, 2011). For example, fresh sludge compost incorporated to PAH-contaminated soil resulted in higher removal of 2-3 ring PAHs compared to 4-6 ring PAHs (Feng et al., 2014). Saturate (*n*-alkane) fractions were efficiently removed compared to aromatic fractions from an TPH-contaminated soil after amending the soil with 5% (w/w) bulrush straw powder or biochar (Wang et al., 2017) (Figure 5). Similarly, the removal of three-ring and four-ring PAHs in all organic treatments was higher than five-ring PAHs (Lukić et al., 2016). However, an opposite observation was reported in the case of lignin-rich organic amendments. For instance, the addition of buffalo manure resulted in higher removal of HMW PAHs compared to LMW PAHs (Lukić et al., 2016). Presumably, lignin-containing material in buffalo manure promoted ligninolytic fungi and enzymes to degrade lignin. As HMW PAHs have structural similarity with lignin, the highly recalcitrant PAH pollutants were also co-metabolized along with lignin (Lukić et al., 2016).

In addition to TPH characteristics, the sorption and desorption processes could be influenced by TPH concentrations and co-contaminants through "conditioning effects" on the soil matrices and by displacement of TPH from the adsorption sites, which could result in enhanced TPH desorption and bioavailability (Yu et al., 2018). Similarly, the TPH adsorption capacity of soil could be decreased by an increase in the initial TPH concentration when a gradual saturation of adsorption sites takes place, and excess TPH exits in available form. For example, higher PAH removal was observed at higher initial concentration during the incubation of contaminated soil with compost amendment (Wu et al., 2013).

5.3. Nature and quantity of organic amendments

High diversity of the origin and processing procedure of organic amendments could result in differences in their nature and quality, thereby affecting TPH remediation efficacy. For instance, maize stover biochar showed higher sorption capacity to PAHs in comparison to paper mill waste biochar (Oleszczuk et al., 2012) (Figure 6-A), whereas lignin-containing organic amendments were more effective in benz[*a*]anthracene mineralization (Zhu et al., 2020) (Figure 6-B). Various humification stages of organic amendments could lead to the change or formation of distinct redox-active functional groups of exogenous organic matter, affecting the sorption and desorption of TPH (He et al., 2019). Furthermore, different decomposition stages of organic amendments might be accompanied with specific microbial population sizes and taxonomic composition, which are expected to have different effects on TPH bioremediation in contaminated soils (Lin et al., 2012, Sayara and Sánchez, 2020). A change in the processing procedure of one specific type of organic amendment even led to varying remediation efficiencies (SI Table 2).

The soluble fraction of fresh exogenous organic matter, such as free sugars, lipids, starch, and soluble protein, is the primary energy source for soil microorganisms due to its readily biodegradable nature (Bolan et al., 2011). Therefore, if the organic amendment contains a high proportion of soluble fractions, it would lead to a high native microbial abundance and activity, at least over a short-term, and this would favour the bioremediation of TPH-contaminated soils (Lukić et al., 2016). In addition, a high DOC content in soils facilitates the formation of soluble metal-organic complexes, thus increasing TPH mobility via cation bridging interactions (Yu et al., 2018). Consequently, the efficacy of added organic amendments with a high content of DOC on TPH biodegradation in TPH-contaminated soils might be greater than that in a soil with a low DOC content (Farzadkia et al., 2019). However, soils with the same DOC content, but with different DOC compositions, might degrade the TPH differently. Differences in DOC's chemical composition among organic amendments could explain the different sorption behaviours observed for TPH in amended, contaminated soils (Gigliotti et al., 2002). However, the mechanisms in which diversified DOC compositions influence soil TPH remediation warrant future research.

In addition to supplying an energy source, fresh organic amendments (e.g., sewage sludge, animal manures) often contain significant amounts of available nutrients essential for plants and soil microorganisms that enhance their growth and activity, in addition to affecting TPH bioavailability (Gómez-Sagasti et al., 2018, Ros et al., 2010). However, with increasing study time, the observed changes in TPH bioavailability and remediation efficiency of the amended soil become less apparent, because of the dominance of aromatic organic fractions towards the end of the experiment. Moreover, the use of unstable or immature organic amendments may adversely affect soil quality, the growth of plants and microbes, and the surrounding environment (Gómez-Sagasti et al., 2018). Conversely, increased humification of exogenous organic matter might not have the above issues, but it could decrease TPH bioavailability by increasing less-readily available organic fractions to plants and microorganisms. The reduction of TPH bioavailability could also be associated with the formation of TPH-humic complexes (Ren et al., 2018).

The quantity and quality of humic substance-like components in an organic amendment are considered to be important indicators of its maturity and stability. Indeed, stabilized organic matter could affect soil physical status via enhancing soil aggregate formation and stability. Additionally, the organic matter can protect and preserve the extracellular enzymatic activity via the formation of humus-enzyme complexes (Lamichhane et al., 2016). Stabilized organic matter, both exogenous and endogenous, could stimulate TPH incorporation into inaccessible soil-aggregate compartments (Ni et al., 2020). In the immobilization approach of TPH remediation, the greater the amount of humic substance-like fractions present in the organic amendment, the more TPH sorption could be expected, which could be an environmentally safe and economically viable option of TPH remediation.

Finally, the remediation efficiency is associated with the quantity or dosage of the organic amendment. Generally, soil microorganisms prefer easily degradable organic materials from organic amendments, and, thus, the selection of the amendment type could alter the degradation of pollutants (Jones et al., 2019). Therefore, surplus application of readily available C might overtake the utilization of TPH, and act as the primary substrate for the metabolism of hydrocarbon-degrading microorganisms. Additionally, application of high levels of organic amendments also could lead to negative impacts on soil properties and soil organisms, which would decrease the remediation efficiency (Ezenne et al., 2014, Haderlein et al., 2001).

However, low mixing ratios of organic amendments and impacted soils were shown to be inefficient in promoting microbial growth and activity (Bastida et al., 2016). Hence, the optimal application rate of an organic amendment should be determined in a pilot-scale test prior to applying it to any TPH-contaminated site.

5.4. Environmental factors

Little information exists concerning the influence of environmental conditions on the remediation of TPH-contaminated soil treated with organic amendments. Given the significant effects of SOM conditions and decomposition rates of organic amendments on TPH in the soil (Neina, 2019, Tonon et al., 2010), it could be assumed that environmental factors (e.g., pH, temperature, and moisture) could influence the remediation efficiency after the addition of organic-amendment. The influences could occur through changes in SOM properties or in TPH bioavailability in the contaminated soil. Yu et al. (2018) showed that, under different pH conditions, SOM existed in different forms, which exhibited different sorptive capacities for TPH contaminants (SI Figure 5). For example, increasing the soil pH in the range of pH 3-6 promoted desorption of SOM, and, consequently, the solubility of the associated phenanthrene increased in the aqueous phase (Yu et al., 2016). Similarly, sorption of naphthalene and phenanthrene in a number of soils was reported to increase with decreasing soil pH in the range of approximately pH 2.5-7 (Zhu et al., 2004). The authors proposed that varying the sorption capacities under different soil pH was due to changes in π-acceptor sites of SOM.

Variation in temperature during the remediation process could result in different availabilities of TPH (Kästner and Miltner, 2016). Perfumo et al. (2007) showed that increased temperature (from 7 to 23 ^{o}C) could enhance desorption of 3-4 ring PAHs by up to 28 times through reduced interfacial surface resistance in NAPL in the contaminated soil. Likewise, high temperature (60 ^{o}C) showed a higher hexadecane degradation rate than that at room temperature (18 ^{o}C) in an incubation experiment (Perfumo et al., 2007). Temperature variations brought about by freeze-thaw cycles, in contrast, have been reported to reduce soil aggregate stability and promote sequestration of PAHs, thereby decreasing PAH biodegradation rates (Wang et al., 2016). In addition, Kim and Kwon (2020) found that, after drying, SOM became more hydrophobic and the surface area and total pore volume increased, which led to an increase in the sorption capacity of TPH. Soil moisture content might also have a profound effect on soil structure, erodability, and aeration and, consequently, TPH availability and its remediation (Akbari and Ghoshal, 2015, Kværnø and Øygarden, 2006). The role of environmental factors including soil mositre and temperature on soil TPH remediation following organic amendment application needs more research.

6. Final remarks and future research needs

Organic amendments have been recognized as carbon- and nutrient-rich materials with a great potential to remediate soils contaminated with TPH. However, as a soil amendment, some potential risks resulting from the application of organic amendments to soil need to be investigated prior to land application. In addition, dual effects on TPH fate and behaviour have been reported following the incorporation of organic amendments to TPH-contaminated soils. Indeed, both net immobilization and mobilization of TPH have been observed to occur with the addition of organic amendments. Such contrasting observations result from the variations in soil properties, nature and concentration of organic amendments, TPH characteristics, and environmental factors. Immobilization processes may reduce short-term toxicity, leaching, and volatilization, particularly for PAHs. However, TPH degradation, which results after mobilization, is generally the preferred remediation pathway for compounds that are not highly resistant to biodegradation. Plant-microbe interactions can favour the microbiome involved in degrading TPH compounds, and they benefit from organic-amendment applications. Specially, combined immobilization and mobilization and biodegradation approaches would be an effective strategy to enhance TPH remediation whereby immobilization acts as a pre-treatment to reduce oxidative stress in plants and microorganisms. Following that, an enhancement of microbial growth and activity coupled with a gradual TPH mobilization through desorption by addition of organic amendments would complete the final step of the remediation process (i.e., biodegradation). The present review highlights the interactive factors influencing the remediation of TPH-conaminated soils to make some guidance for future organic-amendment application.

Some key knowledge gaps regarding TPH remediation after introducing organic amendments in contaminated soils which require future research include: (i) What is the long-term stability of immobilized TPH contaminants? (ii) What are the interactions between exogenous organic matter and the receiving TPH-contaminated soil? (iii) What are the soil microbial communities that can remediate TPH-contaminated soils amended with organic materials? and (iv) What are the appropriate application times and amounts of organic amendments that need to be added to TPH-contaminated soils for maximum remediation? In addition, until date, very few studies aimed to ascertain the predominant immobilization and mobilization mechanisms involved in the organic amendment-induced remediation of TPH under realistic field scenarios. Hence, future research is warranted to confirm results from lab-scale studies, allowing not only to determine the sorption and desorption capacities of organic amendments with TPH compounds, but also to understand their interactions, fate, and ultimate remediation efficacy of organic amendments.

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Tables

Table 1. Selected studies on increased immobilization of total petroleum hydrocarbons (TPH) using organic amendments

Organic amendments	Hydrocarbon compounds	Observations	References
Poultry dung	PAHs	85% sequestration was obtained. The organic amendment rendered PAHs unavailable for plant uptake	Nwaichi et al. (2015)
Wood chip biochar at 800ºC	Monoaromatic petroleum hydrocarbons	Soil adsorption coefficient (K_d) was increased up to a factor 36 after adding the biochar. Volatilization of monoaromatic hydrocarbons was retarded due to increased sorption.	Bushnaf et al. (2017)
Softwood chip at 500ºC	Phenanthrene	Adding biochar enhanced the soil K_d by a factor of 2. The addition reduced pore-water flow rate up to 80%, reducing pollutant leaching risks.	Trinh et al. (2017)
Poultry manure (PM) and poultry manure biochar (PMB)	TPH	PM and PMB amendments reduced toxicity of TPH to barley (*Hordeum vulgare*)	Barati et al. (2018)
Pin needle biochar at 400 and 700ºC	Naphthalene, Acenaphthene, Phenanthrene, and Pyrene	Sorption of the PAHs to biochar reduced lowered freely dissolved PAHs, thus reducing plant uptake of PAHs	Zhu et al. (2017)
Sugarcane residue biochar at 550ºC, then grounded to pass through a 200 µm sieve	TPH	Biochar enhanced TPH dissipation mainly due to irreversible sorption into its non-recoverable domain. Biochar application had little effect on TPH biodegradation.	Wei et al. (2020)
Sewage sludge biochar at 700ºC	Phenanthrene and Pyrene	The addition of biochar enhanced the sorption of phenanthrene and pyrene from 8.3 to 20.3%, and 14.5 to 31.7%, respectively. Dissolved organic carbon and clay minerals in the soil reduced the sorption efficiency of biochar.	Zielińska and Oleszczuk (2016a)
Compost and silver grass biochar at 550ºC	PAHs	Sorption coefficients for PAHs increased tenfold after adding compost and up to a hundredfold with further biochar addition.	Sigmund et al. (2018)
Vermicompost	Naphthalene, Anthracene, and Benzo[a]pyrene	Vermicompost enhanced sorption of the PAHs, and the sorption effect increased with the number of benzene ring of the pollutants.	Dores-Silva et al. (2018)
Green waste and meat waste compost	TPH	Compost addition reduced TPH bioavailability; however, the effect was only observed during the first 3 months.	Wu et al. (2013)

Table 2. Selected studies on increased mobilization and biodegradation of total petroleum hydrocarbons (TPH) using organic amendments

Organic amendments	Hydrocarbon compounds	Observations	References
Wheat straw	TPH	Adding wheat straw to crude oil-contaminated soil enhanced soil microbial respiration and organic C mineralization, and hence, increased the biodegradability of the crude oil.	Alotaibi et al. (2018)
Rice straw and sawdust	TPH and PAHs	The abundance of hydrocarbon-degrading taxa enhanced after rice straw and sawdust addition leading to a significant decline in TPH and PAHs.	Huang et al. (2019)
Compost	Pyrene	Amended compost in rhizosphere soils facilitated bioavailability of pyrene. Compost addition enhanced plant biomass and the concentration of butyric and iso-butyric acid in the rhizosphere. Increased biodegradation of pyrene was observed with the addition of compost.	Wang et al. (2012)
Ramial chipped wood, horse manure, and brewer spent grain	TPH	Higher TPH reduction was reported only in horse manure treatment compared to inorganic fertilizer-treated soil. A greater diversity of microorganisms in horse manure treatment resulted in enhanced TPH remediation.	Robichaud et al. (2019)
Sawdust and wheat straw biochar at 300 and 500ºC	PAHs	For two different feedstock, lower PAH contents in soil was detected when adding biochar produced at 500ºC. Wheat straw biochar resulted in higher specific bacterial abundance and evenness compared to sawdust and hence higher PAH biodegradation.	Kong et al. (2018)
Compost	PAHs and alkanes	Increased abundance of catabolic enzymes was observed in compost-assisted bioremediation. The addition of compost resulted in higher removal of PAHs and alkanes up to 88% after 50 days.	Bastida et al. (2016)
Compost	PAHs	PAH dissipation was higher in non-mature than in mature compost due to higher PAH availability and co-metabolic microbial activity.	Brimo et al. (2018)
Grass straw compost	TPH	Compost addition was effective in supply nutrients for the biodegradation of TPH. TPH removal was highest in compost-treated soil (93%) compared to inorganic fertilizer and control treatments.	Nwankwegu et al. (2016)

		Highest germination percentage (89%) was obtained in compost-amended treatment	
Sawdust	TPH	Sawdust addition at 5% increased microbial activity and degradation rate of TPH.	Alvim and Pontes (2018)
Pea straw	PAHs	The addition of plan residue resulted in enhanced degradation of PAHs (66%) at the beginning of the experiment. The pea straw-amended soil led to the largest shift in bacterial communities with increased abundance of hydrocarbon-degrading microorganisms.	Koshlaf et al. (2019)
Cotton stalks	TPH	Oil sludge-contaminated soil treated with cotton stalks showed increased removal efficiency of TPH, saturated, and aromatic fraction because of improved soil physicochemical properties and enhanced soil microbial quantity and diversity.	Wang et al. (2016)
Cow manure, lignin, straw, and mushroom culture waste	Benz[a]anthracene	Cow manure had no significant effect on the dissipation of benz[a]anthracene. Lignin-containing materials (lignin, straw, and mushroom culture waste) enhanced the dissipation of the pollutant due to increased degrading bacterial and cytochrome P450 metabolic pathway.	Zhu et al. (2020)
Pea straw	TPH	Pea straw-treated soil showed an increase in the number of 16S rRNA and *alkB* genes compared to natural attenuation-treated soil. Higher TPH removal and significant reduction in soil toxicity were obtained in pea straw-treated soil than in natural attenuation.	Koshlaf et al. (2020)
Spent mushroom substrate	PAHs	The mixing of spent mushroom substrate to the contaminated soil resulted in enhanced PAH-degrading microorganisms and degradation of PAHs. The organic amendment reduced PAH toxicity to *Vicia faba* L. through germination test	Di Gregorio et al. (2016)
Fruit waste biochar, sewage sludge biochar, and cow dung	TPH	The addition of biochar significantly enhanced soil microbial abundance and diversity, and hence, biodegradation of TPH. Application of biochar bioaugmented with cow dung showed further decline in soil TPH concentration.	Aziz et al. (2020)
Spent mushroom compost	PAHs	The addition of spent mushroom compost enhanced the development of	Asemoloye et al. (2017)

		guinea grass in a black oil hydrocarbon polluted soil. The combination of organic amendment and plant showed improved remediation efficiency compared to the plant alone and natural attenuation.	
Poultry manure biochar at 400ºC	TPH	Biochar addition increased microbial respiration in the rhizosphere of barley (*Hordum* vulgare) and oat (*Avena sativa*) compared to non-biochar treatments. Plants in biochar-treated soil showed higher capability to stimulated degradation of TPH in the rhizosphere than non-biochar-treated soil.	Barati et al. (2017)

Figures

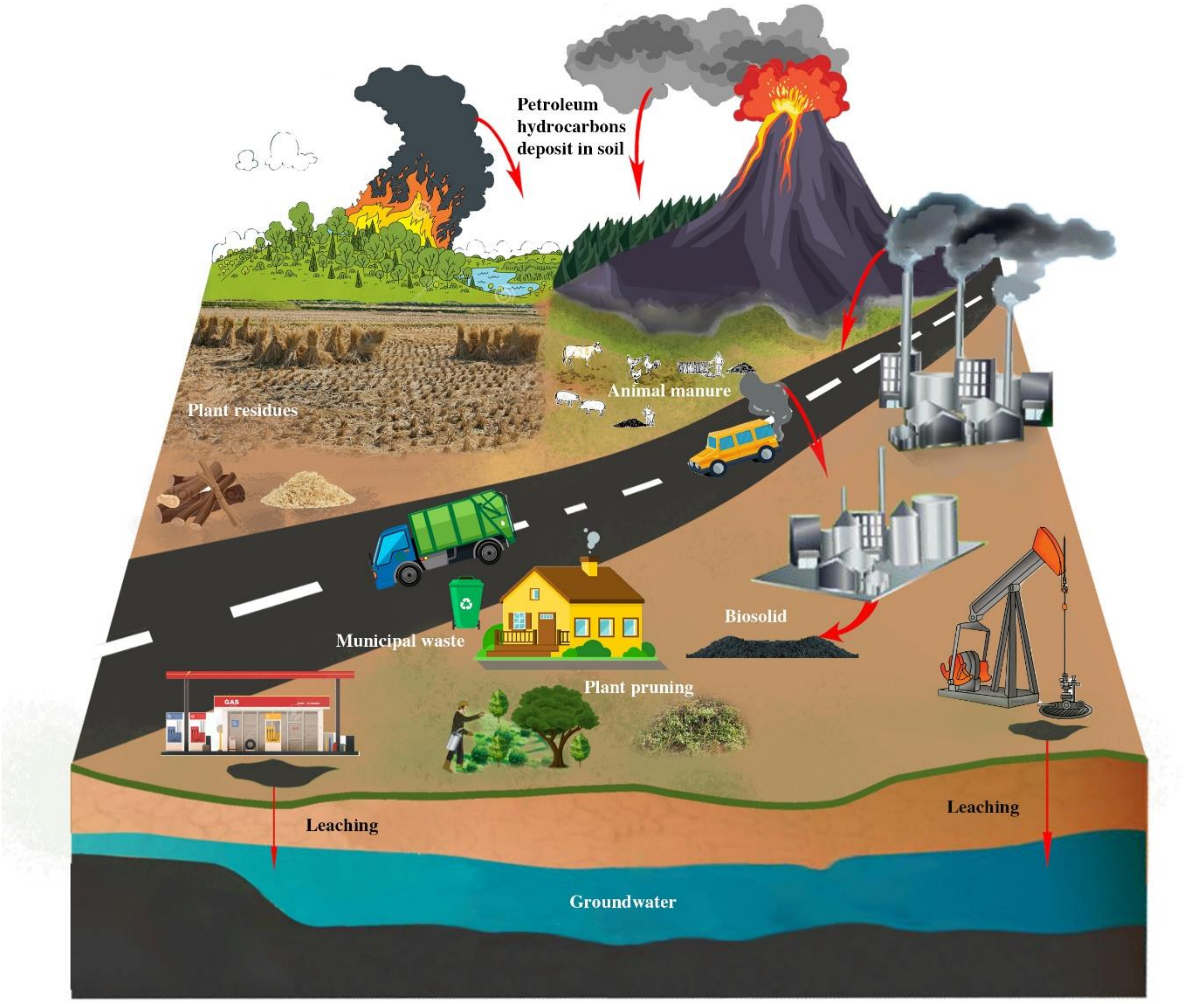


Figure 1. Potential sources of petroleum hydrocarbons and organic wastes in the soil environment

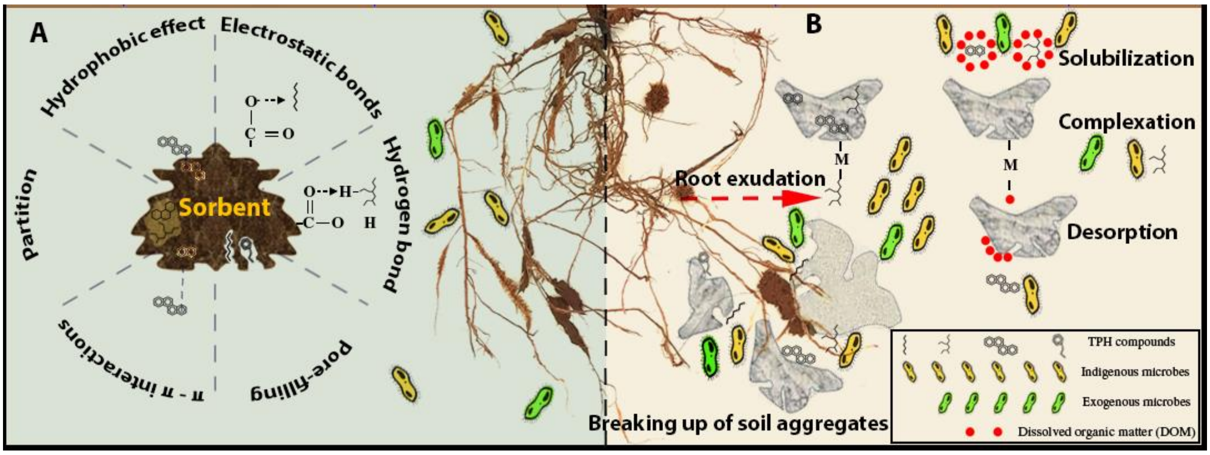
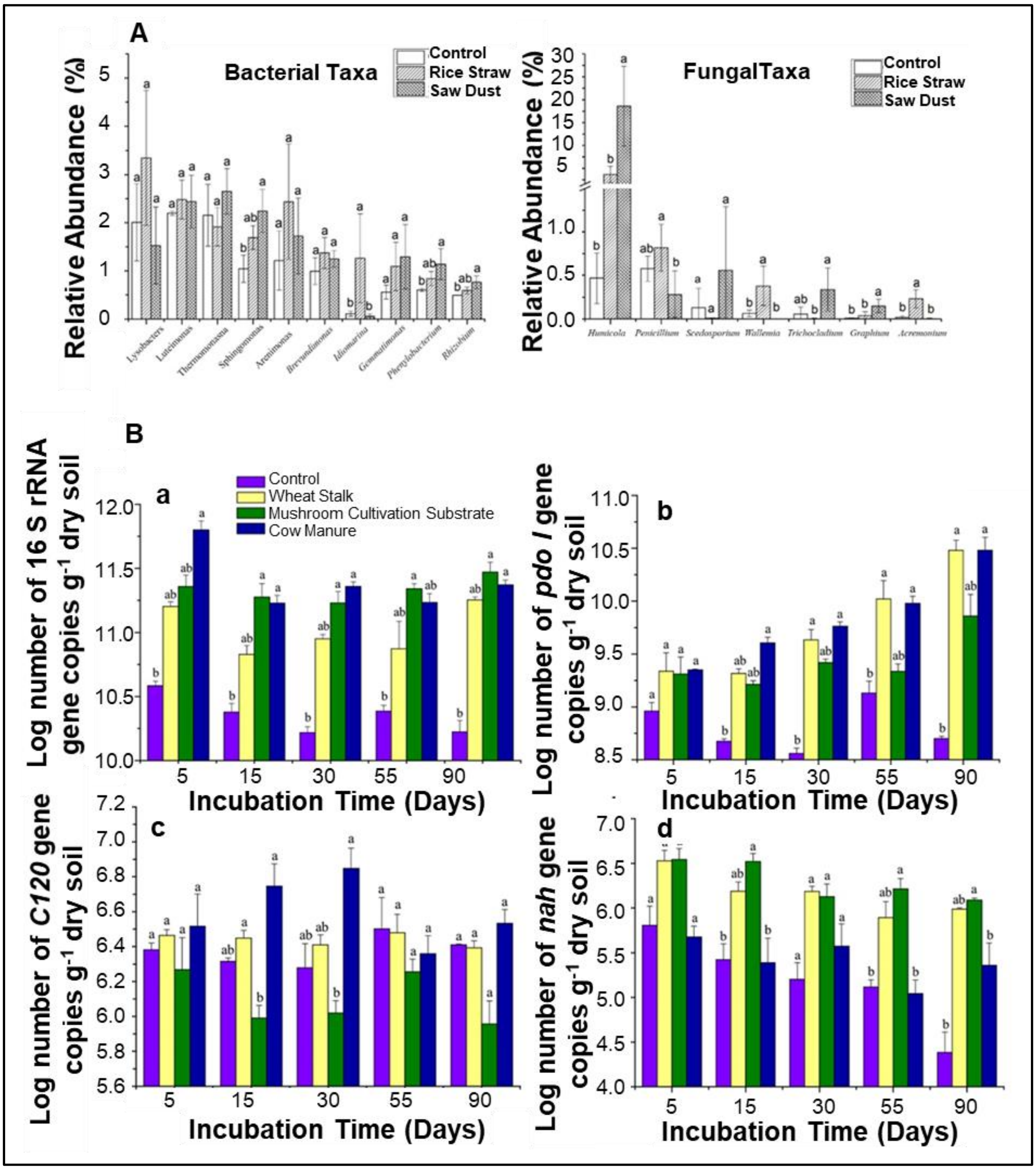


Figure 2. The dual effects of organic amendments on TPH in soil (A – Reducing TPH mobility, and B – Enhancing TPH availability and microbial degradation)



1512 **Figure 3**. Application of organic amendments enhance microbial abundance (A) and catabolic
1513 activity (B) in soil contaminated with TPH, reprinted from Huang et al. (2019) and Han et al.
1514 (2017)
1515

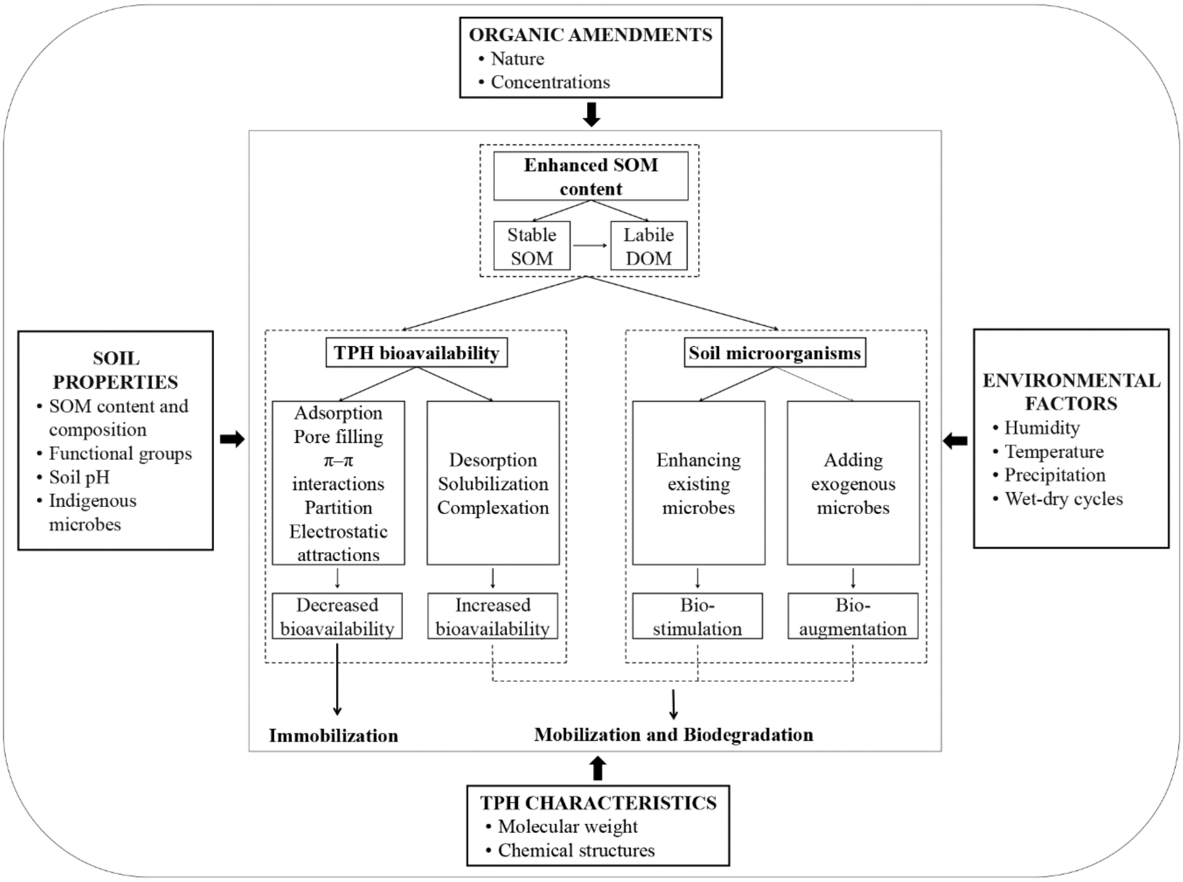


Figure 4. Factors influencing the remediation of TPH-contaminated soils with organic amendments

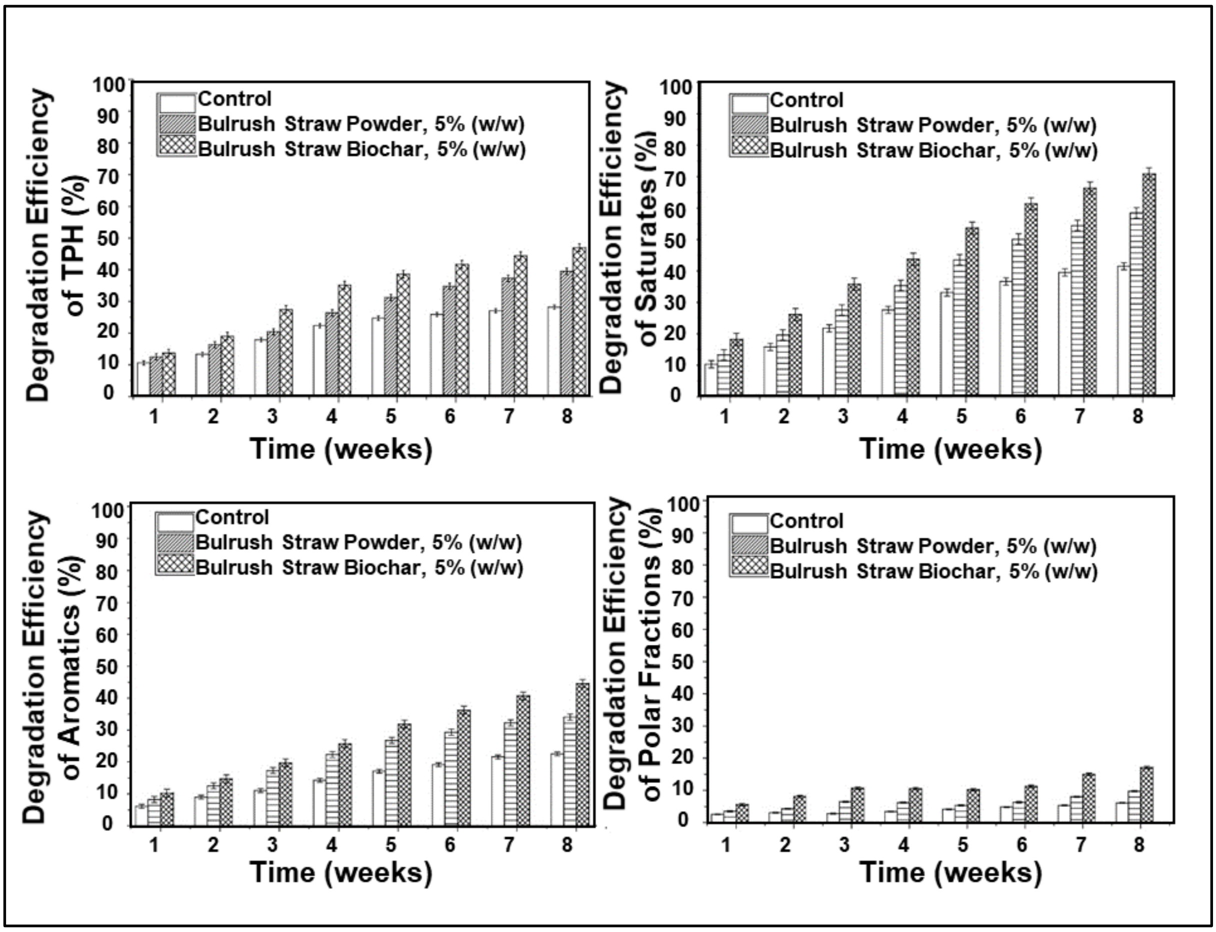


Figure 5. Degradation efficiency of different fractions of TPH after adding organic amendments, reprinted from Wang et al. (2017)

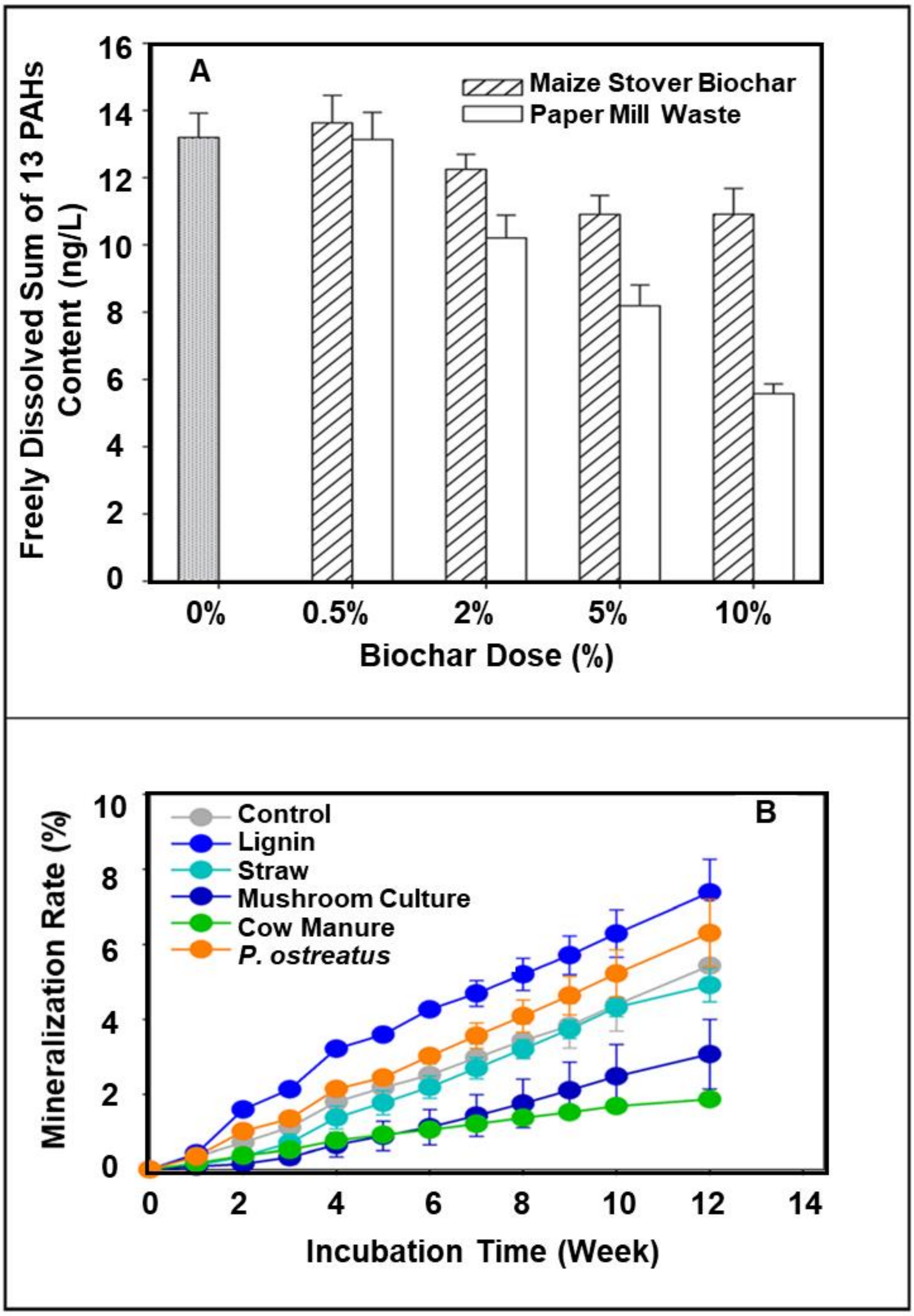


Figure 6. Effects of application rate and nature of organic amendments on bioavailability of TPH (A), and mineralization efficiency of TPH (B) in soils, reprinted from Oleszczuk et al. (2012) and Zhu et al. (2020)

Supplementary information (SI)

SI Table 1. Estimated quantity of organic amendments generated in selected countries and their carbon and nutrients value, adapted from Thangarajan et al. (2013)

Category	**Type of organic waste**	**Typical proportion (%) of carbon and nutrients (C: N: P: K: S)**	**Country**	**Quantity 10^3 Mg year^{-1}**	**Potential carbon and nutrient value (10^3 Mg year^{-1})**				
					C	**N**	**P**	**K**	**S**
Crop residues and green manures	Wheat residue	41.6: 0.38: 0.19: 1.5: 0.14	Australia	108,056	165	4	1	7	1
			United States	154,167	64,134	586	293	2313	216
			United Kingdom	15,512	6453	59	29	233	22
			New Zealand	438	182	2	1	7	1
			China	194,049	80,724	737	369	2911	272
			India	227,680	94,715	865	433	3415	319
			Germany	26,382	10,975	100	50	396	37
			Russia	173,118	72,017	658	329	2597	242
	Maize residue	42.7: 0.66: 0.29: 1.57: 0.1	Australia	814	348	5	2	13	1
			United States	454,854	194,222	3002	1319	7141	455
			New Zealand	242	103	2	1	4	0
			China	448,747	191,615	2962	1301	7045	449
			Bangladesh	2098	896	14	6	33	2
			S. Africa	37,840	16,158	250	110	594	38
			India	99,084	42,309	654	287	1556	99
			Germany	6398	2732	42	19	100	6
			Russia	14,148	6041	93	41	222	14
	Rice residue	34.8: 0.67: 0.8: 1.15: 0.08	Australia	576	143	4	5	7	0
			United States	44,620	11,066	299	357	513	36
			China	918,576	227,807	6154	7349	10,564	735
			Bangladesh	356,850	88,499	2391	2855	4104	285
			S. Africa	34	8	0	0	0	0
			India	1,298,080	321,924	8697	10,385	14,928	1038
			Russia	6127	1520	41	49	70	5
	Barley residue	45: 0.63: 0.21: 2.05: 0.14	Australia	53,144	23,915	335	112	1089	74

			United States	12,968	5836	82	27	266	18
			United Kingdom	11,973	5388	75	25	245	17
			New Zealand	680	306	4	1	14	1
			China	8450	3803	53	18	173	12
			Bangladesh	8	4	0	0	0	0
			Japan	763	344	5	2	16	1
			S. Africa	1075	484	7	2	22	2
			India	8060	3627	51	17	165	11
			Germany	21,492	9671	135	45	441	30
			Russia	64,215	28,897	405	135	1316	90
Animal manures	Chicken manure	22.4: 3.2: 2.7: 1.6: 0.7	Australia	893	200	29	24	14	6
			United States	22,599	5062	723	610	362	158
			United Kingdom	1765	395	56	48	28	12
			New Zealand	145	33	5	4	2	1
			China	51,684	11,577	1654	1395	827	362
			Bangladesh	2454	550	79	66	39	17
			Japan	3078	689	98	83	49	22
			S. Africa	1722	386	55	46	28	12
			India	8328	1865	266	225	133	58
			Germany	1228	275	39	33	20	9
	Cattle manure	30.1: 2.15: 0.2: 0.8: 0.15	Australia	21,953	6608	472	44	176	33
			United States	77,094	23,205	1658	154	617	116
			United Kingdom	8304	2499	179	17	66	12
			New Zealand	8100	2438	174	16	65	12
			China	68,814	20,713	1479	138	551	103
			Bangladesh	18,929	5698	407	38	151	28
			Japan	3593	1082	77	7	29	5
			S. Africa	11,276	3394	242	23	90	17
			India	172,613	51,956	3711	345	1381	259
			Germany	10,519	3166	226	21	84	16
	Pig manure	36.6: 3.25: 0.48: 0.41: 0.09	Australia	396	145	13	2	2	0
			United States	11,233	4111	365	54	46	10

			United Kingdom	772	282	25	4	3	1
			New Zealand	58	21	2	0	0	0
			China	82,400	30,158	2678	396	338	74
			Japan	1696	621	55	8	7	2
			S. Africa	276	101	9	1	1	0
			Germany	4586	1679	149	22	19	4
	Sheep manure	28.75: 2.5: 0.5: 1.2: 0.3	Australia	11,272	3241	282	56	135	34
			United States	930	268	23	5	11	3
			United Kingdom	5146	1480	129	26	62	15
			New Zealand	5391	1550	135	27	65	16
			China	22,189	6379	555	111	266	67
			Bangladesh	301	87	8	2	4	1
			Japan	2	1	0	0	0	0
			S. Africa	4056	1166	101	20	49	12
			India	12,250	3522	306	61	147	37
			Germany	346	99	9	2	4	1
Biosolids		40: 3: 2.23: 0.31: 1.6	Australia	407	163	12	9	1	7
			Austria	153	61	5	3	0	2
			United States	5645	2258	169	127	18	90
			United Kingdom	1120	448	34	25	3	18
			New Zealand	80	32	2	2	0	1
			China	24,691	9876	741	556	77	395
			Bangladesh	2940	1176	88	66	9	47
			Japan	2319	928	70	52	7	37
			India	22,086	8834	663	497	68	353
			Germany	1491	597	45	34	5	24
			Russia	2590	1036	78	58	8	41
Municipal solid waste			United States	100					
			France	20,500	-	-	-	-	-
			Poland	12,300	-	-	-	-	-
			India	48,000	-	-	-	-	-

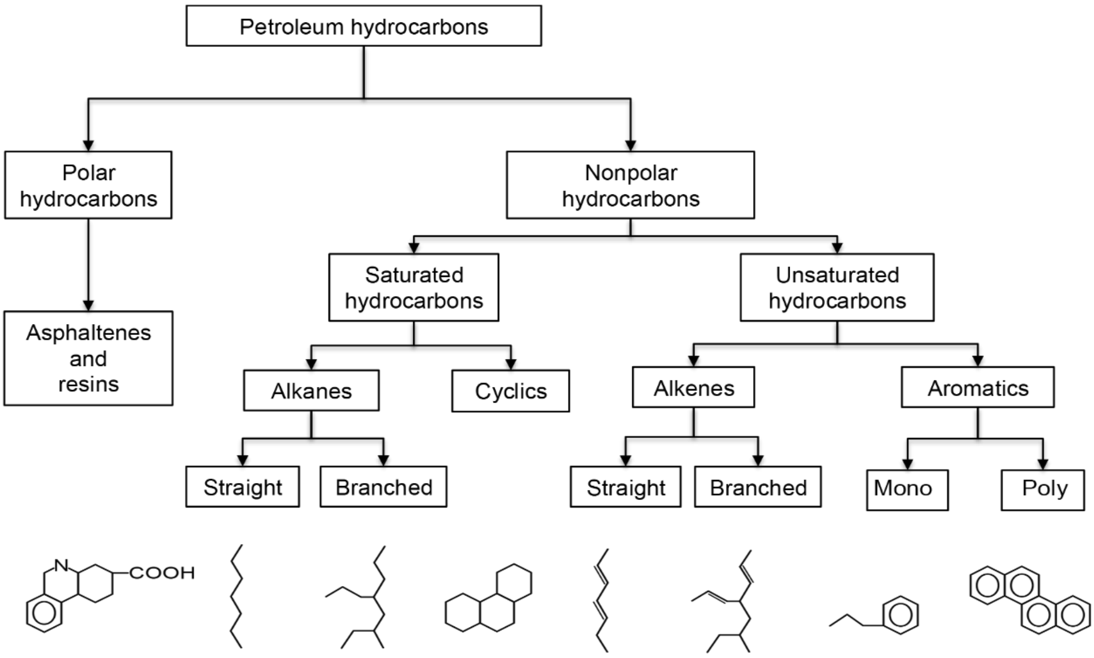
	United Kingdom	28,000	-	-	-	-	-

- *not available*

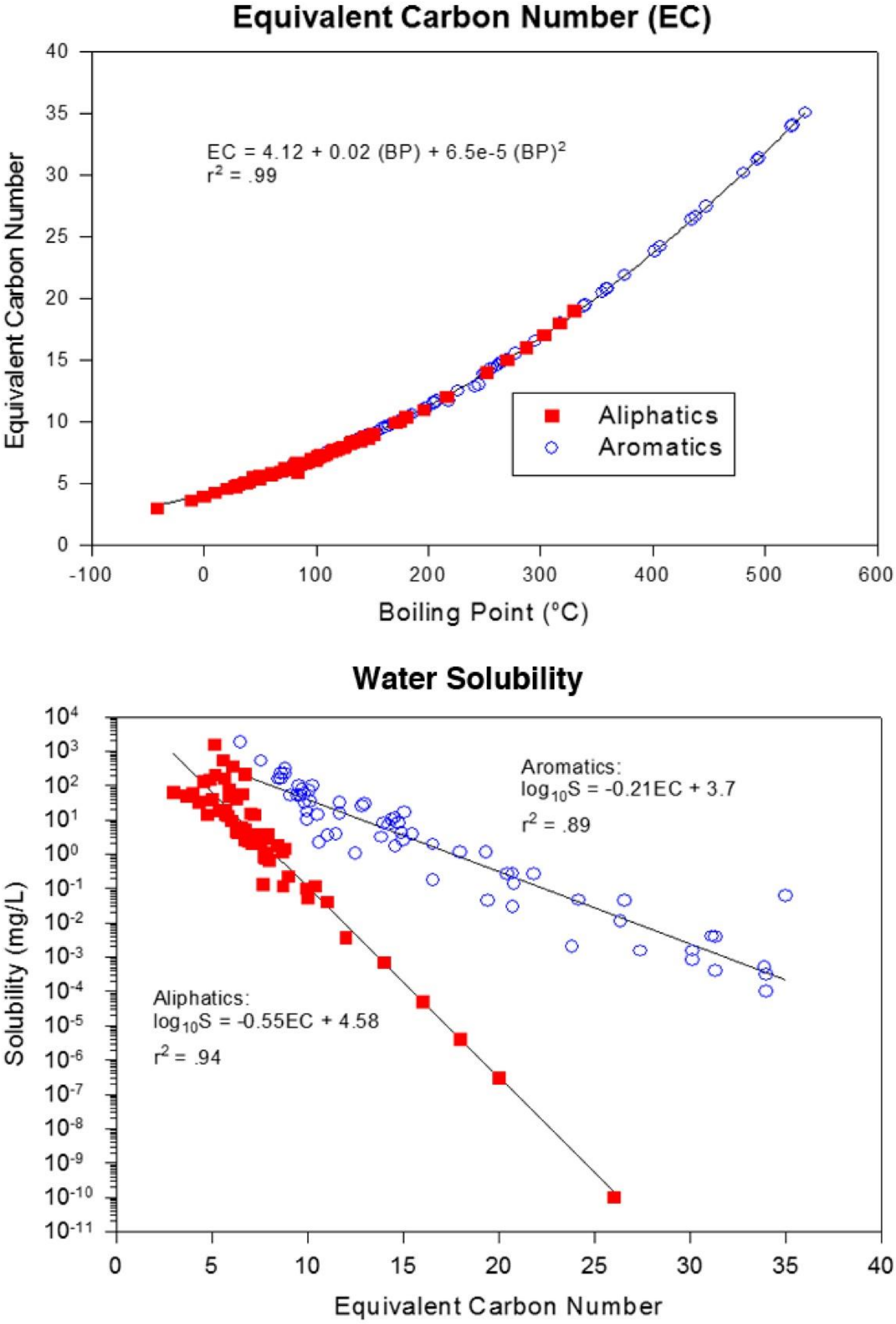
SI Table 2. Characteristics of biochars, produced from different feedstock and at different temperatures, and their remediation efficiency in total petroleum hydrocarbon (TPH)-contaminated soils

Feed stock	Temperature (ºC)	BET surface area (m^2/g)	Total pore volume (cm^3/g)	Petroleum hydrocarbon compounds	Removal efficiency/ Sorption capacity	Reference
Orange peel	200	7.75	0.0098	naphthalene	7.1 %	Chen and Chen (2009)
	300	32.3	0.0313		13.5%	
	400	34	0.0099		33.8%	
	500	42.4	0.0191		50.7%	
	600	7.78	0.0083		53.7%	
	700	201	0.0350		100%	
Wheat straw	400	427	0.526	phenanthrene, fluoranthene, pyrene	71 – 88%	Li et al. (2014)
	600	537	0.574		82.4 – 93.4%	
	800	652	0.634		95.8 – 98.6%	
Sewage sludge	500	31.81	0.0738	PAHs	31.6%	Zielińska and Oleszczuk (2016)
	700	54.05	0.0894		25.1%	
Pine needle	250	9.52	-	naphthalene	1.724 mg/g	Chen et al. (2008)
	400	112.4	0.0442		25.69 mg/g	
	500	236.4	0.0952		27.40 mg/g	
	600	206.7	0.0764		15.14 mg/g	
	700	490.8	0.186		136.8 mg/g	
Pine wood	150	1.83	0.0017	naphthalene	1.83 mg/g	Chen et al. (2012)
	250	5.89	0.0023		11.8 mg/g	
	350	166	0.0112		44.4 mg/g	
	500	434	0.0228		96.5 mg/g	
	700	637	0.0378		208 mg/g	

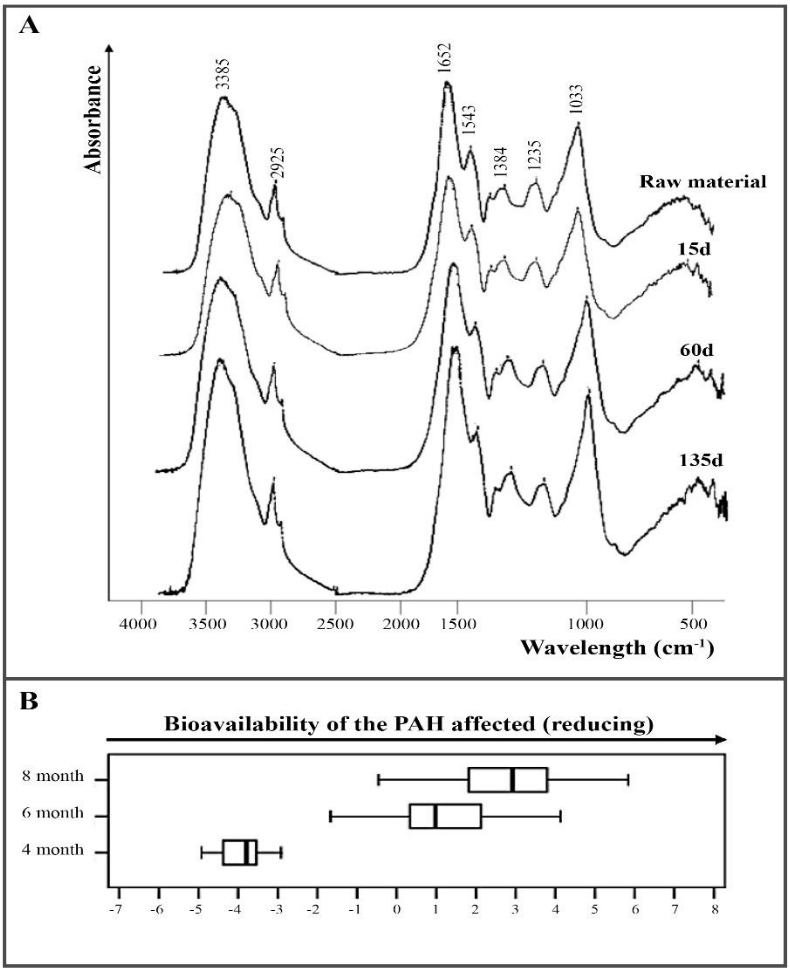
SI figures



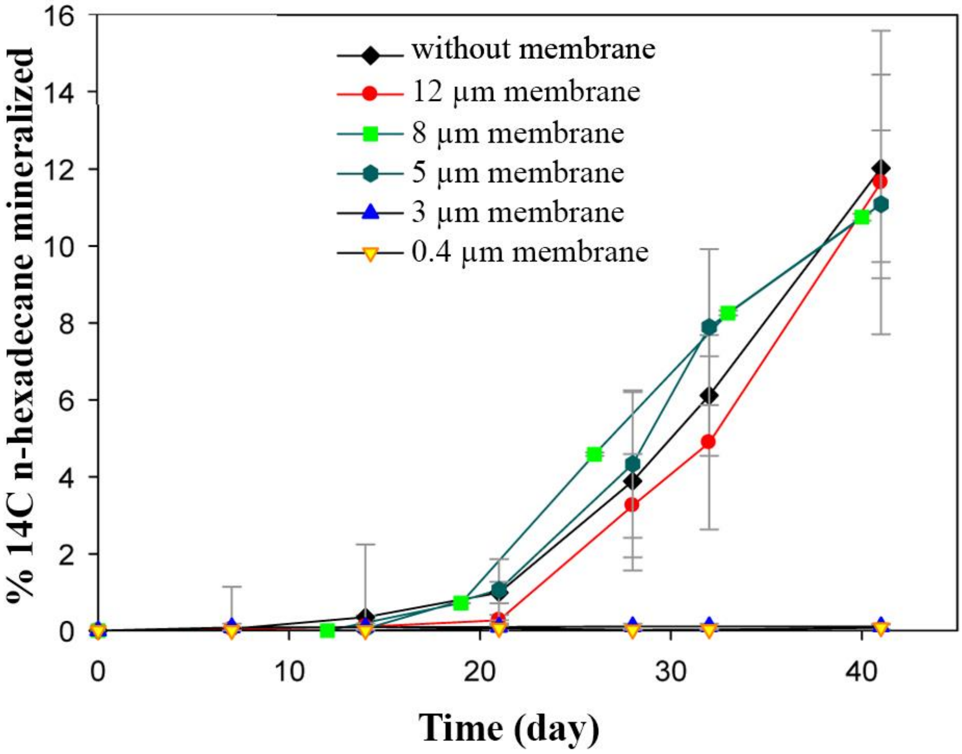
SI Figure 1. Chemical classification of petroleum hydrocarbons (Coulon and Wu, 2014)



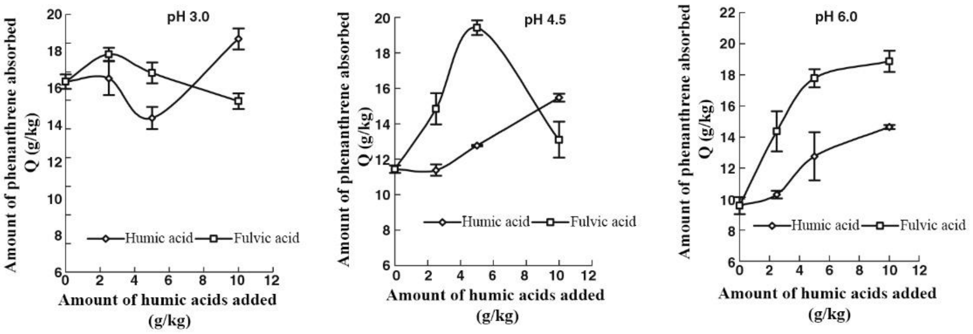
SI Figure 2. Relationship between equivalent carbon number and boiling point (°C) and water solubility (mg L^{-1}) of petroleum hydrocarbon compounds (https://tphrisk-1.itrcweb.org/4-tph-fundamentals/)



SI Figure 3. Characteristics of raw materials and composts at different time scales (A), and effects on PAH bioavailability over time (B), reprinted from Amir et al. (2010)



SI Figure 4. Effect of soil pore size on total petroleum hydrocarbon (TPH) mineralization, reprinted from Akbari and Ghoshal (2015)



SI Figure 5. Soil pH and total petroleum hydrocarbon (TPH) sorption efficiency after adding organic amendments, reprinted from Ping et al. (2006)