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Review

Biochar-based materials as remediation strategy in petroleum hydrocarbon-contaminated soil and water: Performances, mechanisms, and environmental impact

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ABSTRACT

Petroleum contamination is considered as a major risk to the health of humans and environment. Biochars as low-cost and eco-friendly carbon materials, have been widely used for the removal of petroleum hydrocarbon in the environment. The purpose of this paper is to review the performance, mechanisms, and potential environmental toxicity of biochar, modified biochar and its integration use with other materials in petroleum contaminated soil and water. Specifically, the use of biochar in oil-contaminated water and soil as well as the factors that could influence the removal ability of biochar were systematically evaluated. In addition, the modification and integrated use of biochar for improving the removal efficiency were summarized from the aspects of sorption, biodegradation, chemical degradation, and reusability. Moreover, the functional impacts and associated ecotoxicity of pristine and modified biochars in various environments were demonstrated. Finally, some shortcoming of current approaches, and future research needs were provided for the future direction and challenges of modified biochar research. Overall, this paper gain insight into biochar application in petroleum remediation from the perspectives of performance enhancement and environmental sustainability.

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Introduction

Crude oil pollution has received great attention due to its catastrophic damage to the environment (Meng et al., 2019; Davoodi et al., 2020). Crude oil is the major source of energy, and the demand of crude oil in the U.S. was reported as 19.89 billion barrels per day in 2021 (USEIA, 2021). The processes of drilling, extraction, transport, and storage for oil generate huge volumes of oil-contaminated water (Strubinger et al., 2015). In addition, oil spill event has been frequently reported (ITOPF, 2018). Due to transport by wind and current, dispersion in water and accumulation in the sediments, an oil spill offshore can lead to significant petroleum hydrocarbons contamination in the environment, affecting the coastal area including beaches, marshes, and wetlands with serious ecological consequences on ecosystems (Cordes et al., 2016). Oil contamination is considered as a major risk to the health of animals, and humans, and environment (Pitchtel, 2016; Cocârtă et al., 2017).

Crude oil contains organic and non-organic, carcinogenic and growth-inhibiting chemicals, particularly polyaromatic hydrocarbons (PAHs), which are genotoxic and carcinogenic to human and animals by damaging normal function of liver and kidney (Wang et al., 2017). Aquatic systems with oil exposure also causes low dissolved oxygen due to water surface blockings and stimulated microbial degradation, which could further induce eutrophication. In addition, soil with oil pollution reduces agricultural productivity and leads to deterioration in soil health (Pernar et al., 2006; Borowik et al., 2019). Owing to its high viscosity, components of oil residue can be retained in soil pores, adsorbed onto the soil mineral's surface, or left a continuous cover on the surface of the soil (Trofimov and Razanova, 2003). Petroleum contamination generally decreases soil microbial diversity and total microbial populations while increases the proportion of hydrocarbon degraders, which consequently would affect soil nutrient cycling, natural balance of soil microorganisms, and ecosystem functions (Espenberg et al., 2018; Sun et al., 2018; Wei et al., 2020a). Therefore, US EPA has promoted the development of effective and environmentally friendly technologies to reduce the pollution and toxicity of oil hydrocarbons in soil and water (Cerqueira et al., 2011).

Meanwhile, as continued world population growth has demanded greater food consumption, there are massive amounts of crop residue produced resulted from crop harvesting (Turmal et al., 2015). Traditional "return to field" treatment of these crop residue has been shown to cause increased crop disease and add additional production cost (Soane et al., 2012; Arvidsson et al., 2014). Recently, there is increasing interest in using such easily biodegradable crop reside and other waste biomass to produce low-cost sorbents with higher petroleum hydrocarbon adsorption capacity that can be scaled up for the cleanup of an oil spill and other hazardous chemicals (Cheng et al., 2019). Biochar, as a pyrolyzed product of waste biomass, has high carbon stability and adsorption capacity for various organic pollutant compounds, including pesticides (Reguyal et al., 2017), antibiotics (Jeong et al., 2012), estrogen hormones (Wei et al., 2019), perfluorooctanoic acids (Kim et al., 2015), and PAHs (Rhodes et al., 2008; Beesley et al., 2010; Wu et al., 2013). Several review articles published in recent years extensively discussed the remediation of oil by carbon-based materials, including carbonaceous adsorbents (Li et al., 2020), polymer nanofibers (Sarbatly et al., 2016), bio-based materials (Doshi et al., 2018; Hu et al., 2020) in water. However, few reviews had focused on the applications of biochar-based materials for the removal of petroleum contaminants in soil and water environment. Available biochar reviews primarily focused on more PAHs pollutant or oil adsorption than degradations (Li et al., 2020; Madhubashani et al., 2021; Luo et al., 2022). Additionally, since mechanisms of biochar in oil-contaminated soil and water are complex and involve physical, chemical and biological process interactions, modification of pristine biochars could be required to facilitate the enhancement of oil removal capability for a specific environmental application. Moreover, the ecotoxicity assessments of biochar use in various environments are necessary, especially for surface modified biochar. A comprehensive understanding of these aspects could guide the development and application of biochar with high efficiency and low environmental impact for petroleum hydrocarbon remediation.

In this review, development and progresses of biochar-based materials in oil remediation in the environment are summarized. Additionally, mechanisms of pristine and modified biochars in oil remediation in water and soil environment are described. Finally, the potential environmental risks of biochar as well as the outlook and future study in this promising field are discussed.

1. Petroleum hydrocarbon remediation

1.1. *Remediation of oil in water*

1.1.1. *Traditional strategies*

The techniques to control and minimize the oil spill spread include physical, chemical and biological methods. Physical methods such as use of booms, skimmers and burning the oil on the surface of the water. Physical remediation techniques has advantage of low secondary contamination (Dhaka and Chattopadhyay, 2021), however showed some disadvantages such as ecosystem disruption by burning and time and energy consuming by boom (Michel et al., 1992; Prendergast and Gschwend, 2014; IOGP, 2016). Chemical methods including dispersants using, electrochemical treatment, and ultraviolet radiation. These strategies showed lower energy cost and easy to operate, however may have concerns of generating secondary pollution (Jamaly et al., 2015; Graham et al., 2016). Biological methods including the bioaugmentation (addition of oil degrading microbial strains) and biostimulation (addition of nutrients and oxygen) (Azubuike et al., 2016) were also used to enhance oil biodegradation, however, are low efficiency due to the long remediation period and environmental condition-dependent, and also potentially induce eutrophication or algae bloom due to nutrient stimulation and the change of microbial community structure (Okeke et al., 2022). Phytoremediation, such as phytovolatilization, rhizoremediation, and phytotransformation, are new techniques to clean up oil spill in water (Das and Chandran 2011). However, the limitations of the strategy are to select the proper plant that can effectively transform or degrade the specific PAHs and plant growth condition-dependent. Generally, these techniques are limited in removing oil spills that occur sporadically over a wide area and are insignificant in terms of economy, efficiency and safety. Therefore, new technologies and strategies that secure high efficiency and stability for oil spill removal are needed.

1.1.2. Biomass sorbents

For small-scale oil spills cleanup in water, sorbent materials are used to remove oil. In recent years, the use of adsorbents and particles with natural product origins are particularly gaining interest, due to the advantages of low cost, biodegradable, and low environmental impact compared to synthetic sorbents (Rubasinghe, 2013; Shahi, 2014; Yahya et al., 2015). For example, agricultural waste such as rice husk could treat crude oil with 3.0–9.2 g/g adsorption capacity (Angelova et al., 2011). Similarly, vegetable waste and fruit peels, such as luffa, orange, and banana, were also able to remove crude oil (Abdelwahab, 2014; Abdullah et al., 2016). Wood chips, ryegrass roots, bamboo leaves, and pine needles were able to adsorb PAHs (Chen et al., 2011). Akinpelu et al. (2021) reported that the maximum adsorption capacities of acenaphthylene, phenanthrene and fluoranthene by fallen leaves were 1.13, 2.12, and 2.25 mg/g, respectively. The sorption mechanism of PAHs onto crop residue were suggested as pore filling, hydrophobic effects, and π-π stacking interactions on the heterogeneous surface (Cheng et al., 2019). However, the application of these bulk materials on oil remediation in water are limited due to the low sorption capacity that associated with its poor hydrophobicity (Doshi et al., 2018).

1.1.3. Carbonaceous sorbents

Carbonaceous sorbents have been used to remove PAHs due to their high surface hydrophobicity, abundant porous structure, large surface area by suggested mechanisms including partitioning, hydrophobic interaction, pore filling, electrostatic interaction, and π–π electron donor-acceptor interaction (Canzano et al., 2014; Abbas et al., 2018). Typical carbon-based material that applied in PAHs adsorption include activated carbon (Ania et al., 2007; Brändli et al., 2008; Cabal et al., 2009; Cho et al., 2009; Rhodes et al., 2008; Xiao et al., 2015; Garcia-Delgado et al., 2019), graphene and its derivatives (Ghaffar and Younis, 2014; Wang et al., 2014; Eeshwarasinghe et al., 2018; Queiroz et al., 2022), and nanoparticles (Kalantari et al., 2019). The use of these carbon-based sorbents in oil spill treatment allows the recovery of the oil and reuse of sorbent material, which is considered economical and environmentally-friendly (Al-Majed et al., 2012).

Different from activated carbon which produced from carbon-rich materials such as coal, peat, petroleum pitch, and wood (Ravichandran et al., 2018), biochars are more sustainable materials that are produced from various agricultural waste biomass. Additionally, biochar is pyrolyzed under limited oxygen, and contains a variety of non-carbonized fractions (carboxyl, hydroxyl, and phenolic surface functional groups), which benefit forming chemical bonding with PAHs. Therefore, biochar has both properties of carbonaceous sorbents of high adsorption ability, and advantages of plant biomass of low cost and environmental impact, which make biochar a potential material in oil remediation in aqueous environment. Various studies have reported the high removal efficiency of biochar on individual PAH compounds in water (Beesley et al., 2010; Kong et al., 2011; Li et al., 2014; De Jesus et al., 2017; Hu et al., 2019).

1.2. Remediation of oil in soil

Microbial degradation is the major way for bioremediation of petroleum hydrocarbons (Haritash and Kaushik, 2009; Brzeszcz and Kaszycki, 2018). Many microorganisms in soil are capable utilizing PAHs under aerobic conditions, with ring-hydroxylating dioxygenases (RHD) as catalyst for the first step in the oxidative degradation of PAHs (Cerniglia, 1993). The enhancement of oil remediation in soil could be achieved by increasing both microbial degrading activity and bioavailability of oil (Okere and Semple, 2011). The microbial degrading activity could be enhanced by bioremediation, which is considered as an effective and inexpensive technology because of the high biodegradability of petroleum hydrocarbons and large oil degraders community (Azubuike et al., 2016; Varjani and Upasani, 2017). The effects of remediation materials are based on two opposite mechanisms that to reduce bioavailability by irreversible adsorption (e.g., sorbent), and to increase bioavailability by dispersing (e.g., surfactant). Recent studies reported the soil PAHs remediation by plant residue (Shahsavari et al., 2013) and biochar (Cao et al., 2016; Anyika et al., 2015; Kusmierz et al., 2016; Rao et al., 2017; Kong et al., 2018; Ni et al., 2018; Li et al., 2019a). Additionally, the application of bulking material such as sawdust into the soil to increase permeability or supplying electron acceptors in anoxic environments, such as NO_3^-, Fe(III), or Mn(II), to stimulate anaerobic microorganisms (Zedelius et al., 2011; Brown et al., 2017). On the other hand, surfactants are commonly used to enhance the processes of remediation due to the hydrophobicity property of many organic pollutants. They can reduce the surface and interfacial tension between organic phase and water phase and increase the bioavailability of PAHs (Leonardi et al., 2008; Uzoigwe et al., 2015). Chemical surfactants used in oil remediation were reported, such as dodecyl tetraethylene glycol ether (Brij30) (Masrat et al., 2013), and sodium dodecyl sulfate (SDS) (Couto et al., 2009). Biosurfactants that have been proved to greatly increase oil and PAHs removal from soil include rhamnolipid (Hickey et al., 2007; Santa Anna et al., 2007; Whang et al., 2008), glycolipid (Mulligan et al., 2001; Peng et al., 2006), and lipopeptides (Maneerat et al., 2006; Lee et al., 2007), which has been reviewed by Van Hamme and Urban (2009). Besides bioremediation, chemical oxidation using persulfate and ferrous ion was also explored for oil removal from soil (Yen et al., 2011).

On the other hand, phytoremediation has received more attention in recent years. Some plant species can effectively reduce oil contaminants by improving the soil properties such as increasing aggregate stability and enhancing microbial activities (Shirdam et al., 2022; Yavari et al., 2015). These plants are resistant to oil and can accumulate harmful organic compounds in their tissues (Singh and Jain, 2003). Particularly, rhizoremediation is known as an effective method for controlling the bioavailability of PAHs, and it has been reported that this effect exhibits specific functionality through the interaction between soil rhizosphere and microorganisms (Valizadeh et al., 2022).

2. Use of biochar in oil remediation

2.1. *Use of biochar as sorbent in oil-contaminated water*

Various studies investigated the adsorption of biochar for petroleum hydrocarbon compounds (Table 1) and the potential mechanisms were described in Fig. 1. For single ring aromatic contaminant, high sorption abilities of biochar were reported including toluene on pine wood biochar (Silvani et al., 2017), and nitrobenzene and m-dinitrobenzene on pine needles biochar (Chen et al., 2008). For low molecular weight polyaromatic hydrocarbon (LMW PAH) (2 or 3 ring), pine needles, wheat straw, and miscanthus biochar all showed high sorption on naphthalene (Chen et al., 2008; Jiang et al., 2014; Kim and Hyun, 2018). The sorption mechanisms were suggested as partitioning for low pyrolysis temperature biochar, and adsorption for high temperature biochar (Chen et al., 2008), physical and chemical adsorption (Jiang et al., 2014), and π-π interactions (Kim and Hyun, 2018). Another study showed that dissolved black carbon released from biochar can also adsorb naphthalene with 82% removal rate by hydrophobic partition (Fu et al., 2018). Similarly, the completely removal of phenanthrene were also found on poplar and wood chip biochar (Rao et al., 2017), and maize straw, wood dust, and swine manure biochar with the mechanisms of π–π interactions and pore filling (Wang et al., 2016). For 4 ring PAH, pyrene was reported adsorbed by biochar that derived from wood, corn stover, and soybean stalk were sink for in aqueous solution, with removal efficiency ranging 60%−99.5% by pore filling (Hale et al., 2011) and hydrophobic partition (Kong et al., 2011; Fu et al., 2018). Additionally, more complex PAHs, such as benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a, h)anthracene, were removed by coconut waste by 47%–83%, and orange waste biochar by 24%–84%, by π-π interactions and hydrophobic effects (de Jesus et al., 2017). Zhou et al. (2022) investigated the adsorption properties of 16 priority PAHs, 23 nitrated PAHs, 9 oxygenated PAHs by biochar derived from wood waste and reported that their adsorption capacities were dominated by pyrolysis temperature and functional group properties between biochar and PAHs. These studies showed the high potential with high efficiency of biochar with different mechanisms at work on remediation of petroleum hydrocarbon when applied in water.

2.2. *Use of biochar in oil-contaminated soil*

2.2.1. *Application of biochar*

Various studies reported that biochar significantly increased biodegradation efficiency of oil in soil, including PAHs (Zhang et al., 2010; Beesley et al., 2010; Qin et al., 2013; Liu et al., 2015; Kołtowski et al., 2016; Ni et al., 2018), volatile petroleum hydrocarbons (Bushnaf et al., 2011), and diesel oil (Jiang et al., 2016). The performances of biochar on petroleum removal are generally positive while show large variation (Table 2) and the potential mechanisms were described in Fig. 2. For oil remediation in soil, biochar's effect on oil remediation in soil are more complex than in water. The addition of biochars to soils reduced the freely dissolved hydrocarbons in the soil pore water, causing reductions in extractability and bioaccessibility to degrading microorganisms (Rhodes et al., 2008; Kołtowski et al., 2016). Besides the mechanisms of enhanced sorption (Zhang et al., 2010; Qin et al., 2013; Jiang et al., 2016) and increased soil-water partitioning (Bushnaf et al., 2011), the enhancement effect of biochar on biodegradation also attribute to the oil dissipation in soil (Beesley et al., 2010; Liu et al., 2015; Ni et al., 2018). However, the effect of biochar on the bioavailability of petroleum hydrocarbons are not in agreement. For example, Wang et al. (2017) found that the addition of biochar and nutrients caused increases in the microbial communities in petroleum contaminated soil. Liu et al. (2015) found that dairy manure biochar and rice hull biochar could stimulate the growth of PAH-degrading bacteria in paddy soil with increased 16S rRNA gene, PAH-RHDα

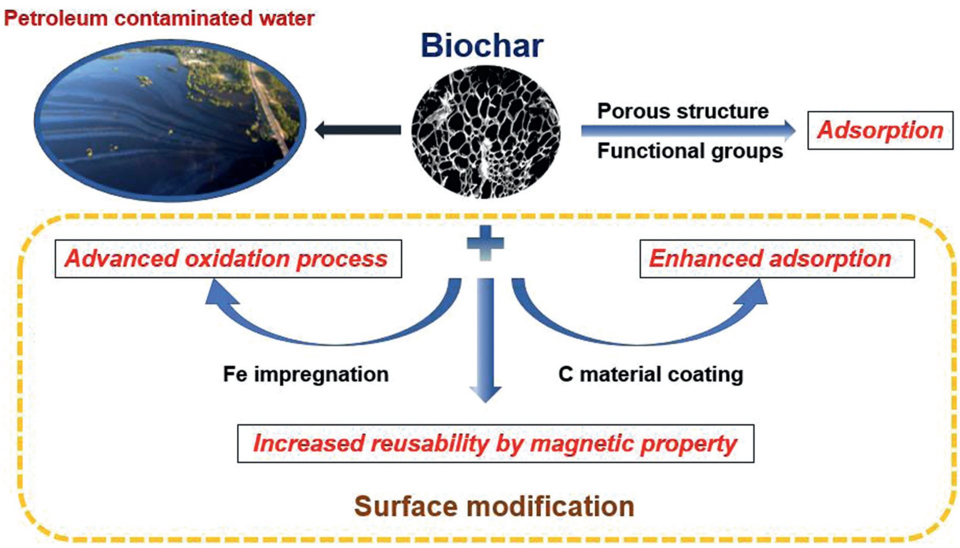

Fig. 1 – Application of biochar in petroleum contaminated water.

Table 1 – Biochar application in remediation of PAHs-contaminated water.

Biochar material	Production condition	Petroleum hydrocarbons	Effect	Mechanisms	References
Wood		Pyrene	Sorption strongly affected in presence of coexisting organic compounds	Sorption in low concentration, Partition in high concentration	Wang et al., 2006
		Competitive sorption with: (phenanthrene, MMBP, phenol, benzo[*a*]anthracene)			
Pine needle	100−700°C	Naphthalene	Sorption parameters (N and logK_f) are linearly related to sorbent aromaticity which increase with the pyrolytic temperature	Partitioning dominant at low pyrolysis temperature	Chen et al., 2008
		Nitrobenzene		Adsorption-dominant at high pyrolysis temperature	
		m-dinitrobenzene			
Corn stover	600°C	Pyrene		Adsorption by nano-porous	Hale et al., 2011
Soybean stalk	300–700°C	Phenanthrene	Removal: 96.6%–99.5%	Hydrophobic partition	Kong et al., 2011
Wheat straw	200, 400, 600°C	Naphthalene	Sorption capacity: 69.6 mg/g	Physical adsorption and chemical adsorption	Jiang et al., 2014
Maize straw	50–600°C	Phenanthrene	logK_{oc}: 5–6 mg/g	Biochar rise aromaticity, decrease H/C and polarity	Wang et al., 2016
Wood dust				π–π interactions	
Swine manure				Pore filling	
Coconut waste	350°C	Benzo(a)anthracene	Coconut biochar:47%–83%	Adsorption (π-π interactions, hydrophobic effects)	De Jesus et al., 2017
Orange waste		Benzo(b)fluoranthene	Orange waste biochar: 24%–84%		
		Benzo(k)fluoranthene			
		Benzo(a)pyrene			
		Dibenzo(a,h)anthracene			
Pine wood	850°C gasification	Toluene	Sorption ability of biochar is comparable with that of activated C	Interactions between toluene and the biochar surface	Silvani et al., 2017
Dissolved black carbon	400°C	Naphthalene, pyrene	Removal: 82%	Hydrophobic partition	Fu et al., 2018
Miscanthus	400°C	Naphthalene	Removal: 5%–30%	π–π interaction	Kim and Hyun, 2018
Rice husk	450, 550°C	Crude oil	Oil adsorption: 2–3 g oil/g biochar	Hydrophobic interactions	Kandanelli et al., 2018
Sawdust					

Table 2 – Biochar application in remediation of PAHs-contaminated soil.

Biochar-based material	Pyrolysis	Petroleum hydrocarbons	Matrix	Effect of biochar	Mechanisms	References
Hard wood biochar		PAHs	Soil	Reduce PAH by >50%	Sorption, biodegradation	Beesley et al., 2010
Pinus radiata biochar	350, 700°C	Phenanthrene	Soil	Increase sorption, decrease desorption	Irreversible sequestration	Zhang et al., 2010
Biochar		Volatile petroleum hydrocarbons	Aerobic sandy soil	Increase biodegradation rate of linear, cyclic and branched alkanes	Sorption	Bushnaf et al., 2011
				Biochar increase soil-water partitioning coefficient, retard petroleum hydrocarbon vapor migration	Biodegradation	
Sewage sludge biochar	500°C	16 US EPA PAHs	Soil	Reduce PAH by 58.0%-63.2%	Partitioning	Khan et al., 2013
Rice straw biochar		TPH	Soil	Increased TPH removal by 16.6%–23.6%, no negative impact on soil microbial community	Adsorption	Qin et al., 2013
Dairy manure biochar	350, 500°C	16 US EPA PAHs	Paddy soil	Removal efficiencies: 2–4 ring PAH >6 ring PAH	Adsorption	Liu et al., 2015
Rice hull biochar				Non-sterilized biochar: 35%–37% higher than sterilized biochar	Biodegradation	
				Stimulate the growth of PAH-degrader		
Pine biochar	450°C	16 US EPA PAHs	Agricultural soil	Biostimulation: insignificant effect on PAH degradation	Segregation of ligninolytic enzymes, PAH uptake	García-Delgado et al., 2015
Wheat straw						
Mycoremediation				Mycoremediation: reduce bioavailable PAH by 23%		
Willow biochar	700°C	PAHs	Coking plant	Wheat straw biochar: reduce free dissolved PAH by 44%–86%	Adsorption	Kołtowski et al., 2016
Wheat straw biochar				Willow biochar: reduce free dissolved PAH by 37%–68%		
Wheat straw biochar	300°C	Phenanthrene	Wheat field soil	Removal: Biochar: 93.4%–93.7% for Phe, 85.2%–88.5% for BaP	Adsorption and immobilization	Cao et al., 2016
Wheat straw		Benzo[*a*]pyrene		Biochar + wheat straw: 91.3%: enhance activity of dehydrogenase, polyphenol oxidase		
Surfactant Brij30				Biochar + wheat straw + surfactant: 90.1%: enhance activity of polyphenol oxidase, act as C source		
Wheat residue biochar	200–600°C	Diesel oil	Soil	600°C biochar increased adsorption capacity of soil from 13 to 18.2 mg/g	Sorption	Jiang et al., 2016
Bulrush straw biochar	300°C	Petroleum	Oilfield soil	Reduce saturated fraction by 80.82%	Improve biodegradation	Wang et al., 2017
Biochar		Light crude oil	Niger Delta soils	Nutrients, biochar and biosurfactant remove 23% more TPH than fertilizer alone	Reduce the hydrophobicity,	Brown et al., 2017
Nutrient					Provide safe harbor for microbial colonization	
Biosurfactant rhamnolipid					Increase the holding water capacity of the soil	
Fishmeal biochar	500°C	Diesel fuel	Landfarm field soil	Reduce *n*C10–C16 and *n*C16–C34	Increase PAH biodegradation	Karppinen et al., 2017
Bonemeal biochar	450°C			Bonemeal biochar selectively stimulated *n*C16-C34 degradation		
Wood biochar	450°C			Biochars increased NO_3^- availability,		

(*continued on next page*)

Table 2 (*continued*)

Biochar-based material	Pyrolysis	Petroleum hydrocarbons	Matrix	Effect of biochar	Mechanisms	References
Compost, fertilizer				Incorporation enhance aromatic functional gene abundance (C2,3O and *nahAc*)		
Poplar biochar, Conifer biochar	600°C	Phenanthrene	Soil	Soil: extractability after 21 day: reduced up to 2.7% for Phe	Hydrophobic interactions, entrapment in pore of biochar	Rao et al., 2017
Olive pomace manure						
Nut shell biochar	500°C	Phenanthrene	Anaerobic sediment	Removal increase: 11.5% for biochar	Sorption (π–π interaction)	Yang et al., 2018
Nitrate				4.4% for biochar immobilized bacteria	Catalyst, adsorbent	
Bioaugmentation				31.5% for biochar +nitrate	Encouraging the growth of nitrate reducing PAH-degraders	
				28.5% for biochar immobilized bacteria +nitrate		
Corn straw biochar	300°C	PAHs	Paddy soil	Biochar reduced the bioaccumulation of PAHs in rice root, especially high molecular weight polyaromatic hydrocarbon (HMW PAH)	Enhance biodegradation	Ni et al., 2018
Bamboo biochar	700°C			BB700 increased PAH, CB300 decreased PAH		
				CB300 increased bacteria, functional genes, and methanogens involved in PAH anaerobic degradation		
Walnut shell biochar	900°C	Light crude oil	Surface soil	Pine wood biochar: increases TPH biodegradation,	Improve biodegradation	Mukome et al., 2020
Pine wood chip biochar	900°C	Heavy crude oil		Walnut shell biochar: inhibit TPH biodegradation		
Fertilizers (N, P, K)						
Biochar (5%)		15 US EPA PAHs	Agricultural soil	Biochar: increase PAHs removal by 3.44%	Immobilization	Bao et al., 2020
Compost (5%)				Biochar+compost: reduce PAHs removal by 4.95%	Reduce PAH degradation genes	
Mushroom residue				Biochar+Mushroom residue: increase PAHs removal by 11.7%	Increase PAH degradation genes	
Corn straw				Biochar+corn straw: increase PAHs removal by 14.72%	Increase concentration of DOC	
Miscanthus biochars	400°C	8 PAHs	Agricultural soil	Biochars: decreased PAH availability by 60%–84%	Sorption	Janus et al., 2020
Ryegrass cultivation	600°C			Ryegrass cultivation: did not impact PAH availability		
Sugarcane biochar	550°C	Crude oil	Coastal wetland soil	Biochar+RL, biochar+N, and biochar+N+RL reduced 32.3%, 73.2%, 80.9% of TPH, respectively.	Biochar: Sorption	Wei et al., 2020a
Fertilizer (urea)				Biochar increased the sorption of aromatic compounds	RL and N: increase biodegradation	
Biosurfactant rhamnolipid				RL and N enhanced thedegradation of heavy and light aliphatic compounds		

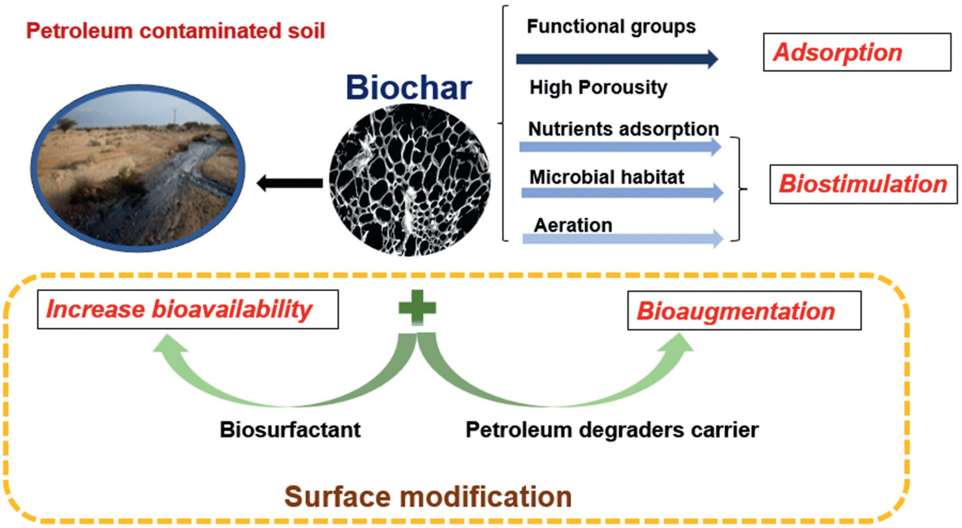

Fig. 2 – Application of biochar in petroleum contaminated soil.

GN genes, and *nirS* gene. Karppinen et al. (2017) reported that biochar enhanced abundance of functional gene for aromatic degradation (C2,3O and *nahAc*) but not for aliphatic compounds. Kong et al. (2018) reported the application of wheat straw biochar increased the relative abundances of PAHs degraders and increased biodegradation of PAHs in soils. Ye et al. (2019) indicated that the addition of activated biochar improved the microbe quantity and the copy number ratio of optimized fungi/bacteria gene in PAHs contaminated wetland soils. The promotion of biochar on the activity of oil degrading bacteria and relating enzymes were suggested due to providing habitats and substrates, increasing soil aeration, concentrating nutrients with the great surface area and high porosity (Chan and Xu, 2009; Thies and Rillig, 2009; Zhang et al., 2019b). Additionally, biochar produced in high temperature is recalcitrant C, which could stimulate microbial degradation of C substrates in oil, which is “positive priming effect”. Cometabolic stimulation of degradation on organic C of biochar has been reported (Kuzyakov et al., 2009; Keith et al., 2011; Luo et al., 2011). According to Zhang et al. (2020), when rice straw-derived biochar was treated with PAHs-contaminated soil, the biodegradation rate of PAHs increased from 40% to 59%, which is because the distribution rate of specific Acidobacteria-related genera in the soil increased compared to the control (soil without biochar).

In contrast, the controversial effect of biochar on oil biodegradation were also reported. Mukome et al. (2020) reported that 900°C pine wood biochar could increase crude oil biodegradation, while walnut shell biochar produced at same temperature inhibited Total petroleum hydrocarbons (TPH) biodegradation. Ni et al. (2018) found that 700°C bamboo biochar reduced PAH bioavailability in paddy soil primarily via immobilization, but 300°C corn straw biochar increased PAH biodegradation with increased bioavailability of immobilized PAH since LMW organic acids in biochar facilitated PAH release from soil. Wei et al. (2020a, 2020b) found that application of 550°C sugarcane cane residue biochar alone to oil-contaminated wetland soil did not stimulate microbial degradation of hydrocarbons or change the soil microbial community but enhanced petroleum dissipation from aqueous phase by adsorption. The different effects of biochar are likely depending on different characteristics of biochars and soils and their interactions. On the other hand, the negative effect of biochar on soil microbes was suggested due to sorption on organic C and physical protection (Zimmerman et al., 2011), disrupting soil microbial communication through sorption and hydrolysis of signal molecules, such as acyl-homoserine lactones (Gao et al., 2016), as well as depressed the activity of dehydrogenase and inhibit removal of phenanthrene (Cao et al., 2016).

2.2.2. *Integration use with bioremediation*

Recent investigations use biochar in accompany with bioremediation strategies to achieve higher petroleum remediation in soil. Availability of nutrients, especially N and P, has been reported to affect the persistence of petroleum hydrocarbons (Agarry and Ogunleye, 2012), therefore, natural fertilizers and organic waste for bioremediation efforts are usually applied to remove PAHs from contaminated soils (Colombo et al., 2011). Wang et al. (2017) reported that the addition of bulrush straw biochar and nutrients resulted in higher removal efficiency of TPH than biochar only (51.4% vs. 46.9%) and greater increases in the microbial communities in oilfield soil. Wei et al. (2020a) found that urea-N addition facilitated degradation of hydrocarbons, especially aliphatic, and enhanced the abundances of oil degraders. Particularly, in anaerobic environment such as wetland and sediment, NO_3^- is highly demanded for petroleum biodegradation as a thermodynamically priority as electron acceptor (Sarkar et al., 2016). Yang et al. (2018) found that biochar amendment combined with NO_3^- enhanced phenanthrene degradation compared to without biochar, and NO_3^- encouraged the growth of potential anaerobic PAH-degraders *Thiobacillus* and *Stenotrophomonas*.

The primary advantage of nutrient biostimulation is to facilitate the activity of those already present native microorganisms that are well-suited to the environment. As a biostimulation strategy, N addition has been shown to increase organic C utilization by preventing metabolic limitations and increasing degrading capacity of microbial community (Abed et al., 2015). Also, N addition is expected to stimulate oil biodegradation without causing adverse effect as it is retained by biochar. The utilization of organic C could also be associated with the increased available N and phosphorus (P), since biochar addition in soils has also been reported to support the activities of N cycling bacteria (*Bradyrhizobiaceae* and *Hyphomicrobiaceae*) as well as phosphate solubilizing bacteria (Major et al., 2009; Anderson et al., 2011).

However, biochar with plant residue as biostimulation showed inconsistent effect. Bao et al. (2020) found that addition of mushroom residue and corn straw could increase removal of 15 US EPA PAHs by 11.7%, and 14.7% comparing to biochar only, respectively, with increased PAH degradation genes, while compost addition reduce PAHs removal by 4.95% with reduced PAH degradation genes. García-Delgado et al. (2015) showed that both pine biochar and biostimulation showed insignificant effect on PAH degradation in creosote contaminated agricultural soil, while mycoremediation reduced bioavailable total PAH by 23% by segregation of ligninolytic enzymes and increased PAH uptake. In contrast, Janus et al. (2020) found that ryegrass cultivation with miscanthus biochar did not impact PAH availability in soil. These results may indicate that green manure is not as effective in stimulating oil biodegradation as fertilizer, which probably related to the lower N content and availability.

2.2.3. *Integrated use with surfactant*

Integration use of surfactant with biochar in oil adsorption is another approach to increase the mobility and biodegradability of oil. Misau et al. (2015) showed the eggshell modified by surfactant hexadecyl-trimethyl-ammonium-bromide (HDTMA-Br) for the effective removal of crude oil from water. Cao et al. (2016) reported that surfactant Brij30 is favorable to sustain soil microbiological activity and the removal of PAHs by enhancing activity of polyphenol oxidase and acting as C source. Compared with synthesized surfactants, biosurfactants (i.e. rhamnolipid, sophorolipid, surfactin) have the advantages of high biodegradability, lower toxicity, higher accessibility of raw material in production, and they can maintain activity under extreme conditions of temperatures, salinity and pH values (De Almeida et al., 2016). Brown et al. (2017) found that biochar + rhamnolipid could reduce 9% more TPH in bonny light crude oil contaminated soil comparing to biochar only by reducing hydrophobicity of oil and increasing degradation. Wei et al. (2020a) reported that TPH removal increased by 40.3% with biochar + rhamnolipid + N the comparing to biochar only in crude oil contaminated coastal wetland soil. While biochar reduced TPH only by sorption, integration use with rhamnolipid and fertilizer could enhance biodegradation with increased abundance of oil degrader.

2.2.4. *Integrated use with phytoremediation*

Biochar treatment in PAHs-contaminated soil can promote microbial activity and plant growth, thereby promoting immobilization and biodegradation of PAHs (Kong et al., 2021). Kong et al. (2018) reported that the biodegradation of PAHs could be accelerated due to biochar's ability to stimulate microbial activity and vegetative growth in soil. Barati et al. (2017) investigated the effect of poultry manure biochar and barley and oat on oil remediation in oilfield soil, and found that biochar could stimulate degradation of TPHs in the rhizosphere, and increased TPH reduction by 20%–45%. These authors also showed that the poultry manure biochar increased soil microbial respiration and TPHs degradation by 28.07% and 26.83%, respectively, over a petroleum-polluted soil applied with poultry manure (Barati et al., 2018). Jahantab et al. (2018) used urban waste biochar and *M. sativa, C. procera, S. plumosa* to treat oil impacted oilfield soil, and found that *C. procera* has better impact in reducing TPHs in soil than other plants. Biochar could increase TPH removal, and promote plant growth and soil microbial activity.

However, inconsistent results were also reported. For example, Saum et al. (2018) used biochar, compost, and mesquite tree to treat oil contaminated soil, and found that only application of biochar and compost could enhance oil degradation, but biochar cannot improve oil degradation when used as the sole amendment, and even showed negative effect with reduced the population size of the oil-degrading community. Similarly, Abbaspour et al. (2020) studied the effect of oak leave biochar and clover, mallow, mycorrhizae on oil removal in oilfield soil, and found that biochar significantly increased the degradation rate of TPH about 18.6% and 45.5% in clover and mallow. Biochar and mycorrhizae inoculation improved growth of plants and increased TPH degradation. However, biochar had insignificant effect on TPH degradation. The negative effect of biochar on oil biodegradation were suggested as decreased the bioavailability of petroleum hydrocarbons, but the biochar could still reduce the free TPH in soil with irreversible retaining by pore sorption. The above findings were summarized in Table 3.

2.3. *Factors influence biochar removal ability on petroleum hydrocarbons*

2.3.1. *Feedstock of biochar*

Biochar with the different source of feedstock may have different characters. Compared to crop residue and wood biomass, biochar produced from animal litter and solid waste feedstocks exhibits lower surface areas because of low C content and without cell wall structure of the biomass like plant feedstock (Ahmad et al., 2014). Raw materials with a high lignin content, such as woody biomass, are conducive to pyrolysis for the generation of biochar with a high fixed carbon content, high specific surface area, higher aromatic C contents, and fine aromatic structure (Jiang et al., 2020). Woody material-derived biochar showed high adsorption ability for petroleum hydrocarbon, for example, red gum biochar was reported to increase in the sorption of benzene and toluene (Bornemann et al., 2007) and pine needle biochar increase sorption of naphthalene and phenanthrene (Chen and Yuan, 2011). On the other hand, feedstock has strong influence on biochar-microbial interactions in the soil (Noyce et al., 2015). Greater priming effect was observed when soil was combined with the more labile biochar, which enhanced biodegradation of hydrocarbons. Promoted respiration with added grass biochar was found, while no increase of soil respiration was observed with pine

Table 3 – Biochar integrated use with phytoremediation in remediation of petroleum hydrocarbons.

Biochar	Pyrolysis	Petroleum hydrocarbons	Matrix	Effect	Mechanisms	References
Poultry manure biochar	400°C	TPH	Oilfield soil	Biochar increased TPH reduction by 20%–45%	Biochar stimulate degradation of TPHs in the rhizosphere	Barati et al., 2017
Barley and oat						
Urban waste biochar		Petroleum	Oilfield soil	C. procera has better impact in reducing TPHs in soil than other plant	Biochar promote plant growth, increase TPH remediation	Jahantab et al., 2018
M. sativa C. procera S. plumosa				Biochar increase TPH removal		
Biochar		Petroleum	Soil	Biochar +compost enhance oil degradation	Biochar showed egative effect	Saum et al., 2018
Mesquite tree Compost				Biochar cannot improve oil degradation when used as the sole amendment		
				Biochar generally reduced the population size of the oil-degrading community		
Poultry manure biochar		Petroleum	Soil	Biochar increased soil microbial respiration and TPHs degradation by 28.07% and 26.83%	Biochar promote plant growth, increase TPH remediation	Barati et al., 2018
Poultry manure Barley				Poultry manure increased soil microbial respiration and TPHs degradation by 15.64% and 12.74%	Biochar increased soil microbial activity	
Oak leave biochar Clover	500°C	Petroleum (TPH: 16.79 g/kg)	Oilfield soil	Biochar amendment increased significantly the degradation rate of TPH about 18.6% and 45.5% in clover and mallow	Biochar and/or mycorrhizae inoculation improve growth of plants, increase TPH degradation	Abbaspour et al., 2020
Mallow Mycorrhizae				Biochar had not significant effect on TPH degradation		
Green garden waste biochar Compost Bacteria Italian ryegrass	500°C	Crude oil (3.4%)	Agricultural fields soil	Biochar +compost+ consortia increase hydrocarbon removal by 85%	Biochar and compost amendment enhanced rhizosphere effect. Biochar and compost increase plant growth and TPH degraders	Hussain, et al., 2018

wood biochar in soil with substrates addition (Hilscher et al., 2009). Soil samples amended with pine wood biochar improved biodegradation of crude oil whereas the walnut shell biochar inhibited the biodegradation due to its high ash content and low surface area (Mukome et al., 2020). Additionally, the high content of ash in raw materials prevents the formation of polycyclic aromatic carbon, which leads to a decreased stability of biochar (Li et al., 2020).

2.3.2. *Temperature*

With increased pyrolysis temperature, the contents of hemicellulose, cellulose, lignin, protein, polysaccharide, and other macromolecules in the solid residue decrease, and the polarity and hydrophilicity of biochar also decreases, and separated aromatic rings begin to form (Jeong et al., 2016). Crombie et al. (2013) found that the pyrolysis temperature was negatively correlated with the (O + N)/C, O/C, and H/C ratios, and positively correlated with the C content, ash content. With losing H and O content, biochar surfaces become less polar and more aromatic, the surface area of the resultant biochar is highly microporous, which may further affect PAHs adsorption (Chen et al., 2008; Chen and Yuan, 2011). James et al. (2005) found that sorption of phenanthrene increased in wood char with increasing production pyrolysis temperatures. Bornemann et al. (2007) found that biochar prepared at higher temperatures from grass and wood had greater uptake of aromatic hydrocarbons in soil. Allen-King et al. (2002) suggested that with higher pyrolyzing temperature, more volatile matter can be removed, exposing more pores and allowing greater pore accessibility and thus larger surface area. On the other hand, application of biochar produced at low temperatures (250 and 400°C) resulted in greater soil respiration, because the present of more labile carbon, such as carbohydrates and cellulose which are bioavailable for microbes, and less aromatic compounds which are hard to utilize by microbes (Cross and Sohi, 2011). Additionally, low pyrolysis temperatures can promote formation of persistent free radicals (PFRs) in biochar, which could enhance oxidation of petroleum hydrocarbon in the presence of H_2O_2 through formation of •OH (Fang et al., 2015; Odinga et al., 2020).

The stability of biochar is also associated to biochar properties that depend on feedstock and pyrolysis conditions, which influence the composition of biochar and their extractability, as well as environmental factors such as precipitation, temperature, and soil properties which influence the fate, biodegradability, and interaction with soil minerals and organic contents of biochar (McBeath et al., 2015).

3. Modification and integrated use of biochar for efficiency improvement

Various modification methods have been used to improve the performance of biochar on petroleum hydrocarbon remediation and multifunction by engineering of biochar and/or integration use with other materials to enhance adsorption, biodegradation, and reusability. The current approaches have been summarized (Table 4).

3.1. *To enhance adsorption*

3.1.1. *Activation of biochar*

To enhance adsorption of PAHs onto biochar, appropriate modifications of the adsorbents are needed to either add or eliminate the surface functional groups (Kołtowski et al., 2017b). Oxygen-containing functional groups (e.g., carboxylic, lactonic) on biochar provide the possibility of hydrogen bonds with PAHs, and the functional groups could be introduced by oxidizing agents. Jin et al. (2017) investigated the sorption ability of phenanthrene by grass straw and animal waste biochars with HNO_3 oxidation, and found that HNO_3 treatment of biochars caused O enrichment, loss of alkyl C, and rise of aromaticity. Ye et al. (2019) reported that HCl activation decreased available PAHs in wetland soil by 2.44% compared with fresh biochar, with more excellent surface characteristics, served as co-substrate, providing better condition for microbial growth and activity, as well as optimized fungi/bacteria gene copy number ratio. However, high O content could reduce the hydrophobicity of biochar and inhibits the remediation of PAHs, and this is usually by thermal treatment or alkali modification (Wang and Wang, 2019). Additionally, Feng and Zhu (2018) found that base modification could increase the surface area and hydrophobicity of biochar by removal of base-soluble C, resulting the improved sorption of phenanthrene on biochars. Similarly, Cai et al. (2019) reported the KOH modification of crab shell waste biochar result high surface area, abundant functional groups, stratified surface structure, and increased the adsorption capacity of diesel oil from to 73.1 to 93.9 mg/g.

3.1.2. *Coating of biochar*

Carbonaceous materials modification was also used to increase the surface area of biochar. The coating of graphene on the surface of biochar resulted in greater surface area, pore size, pore volume, stronger vibration of $C = C$ bonds, an increasing negative surface charge, and more O-containing groups (Tang et al., 2015). The graphene-modified biochar increased removal efficiency of phenanthrene from 63.3% to 94.9% (Tang et al., 2015). Similarly, graphene-modification could improve the thermal stability of the biochar, and showed more than 20 times higher adsorption ability of PAHs methylene blue than unmodified biochar in another study (Zhang et al., 2012). Similarly, Ghaffar and Younis (2014) also found that the graphene covering on peanut shell biochar surface increased sorption ability towards phenol and methylene blue by up to 3.6 times than unmodified biochar, with increased surface area, pore volume, and carbon content.

3.2. *To facilitate biodegradation*

3.2.1. *Introducing surfactant on biochar*

Recently, biosurfactants have been found effective for improving the adsorption efficiency of organic contaminants by surface modification onto carbon-based materials (Wu et al., 2014; Zhen et al., 2018). For example, rhamnolipid-modified graphene oxide was found to increase adsorption of organic contaminants, and the adsorption mechanisms were attributed to increased functional groups, which improved electrostatic attraction, π–π interaction, and hydrogen bond

Table 4 – Performance and mechanisms of modified biochar in remediation of petroleum hydrocarbons.

Biochar-based material	Production condition	Modification	Petroleum hydrocarbons	Matrix	Effect	Mechanisms	References
Pinewood biochar	900°C; H_2O and CO_2	Fe-impregnation	Naphthalene	Water	94% removal	Catalyst, adsorbent	Abu El-Rub et al., 2008
Orange pell biochar	250–700°C	Fe-impregnation	Naphthalene p-nitrotoluene	Water	6.3%–74% for magnetic biochar; 92%–100% for biochar 29.6%–87% for magnetic biochar; 58%–99.7% for biochar	Adsorbent	Chen et al., 2011
Pine needle biochar Plant residue	100–700°C	Microorganism carrier *(Pseudomonas putida)*	15 PAHs	Soil	Enhance the removal of PAHs Effect: 4 and 5 ring PAH > 3 and 6 ring PAH	Sorption Degradation	Chen et al., 2012
Pine bark biochar	900°C; H_2O	Fe-impregnation	Toluene	Water	94% removal	Catalyst, adsorbent	Mani et al., 2013
Switchgrass biochar	800°C	Fe-impregnation	Toluene	Water	81% removal	Catalyst, adsorbent	Bhandari et al., 2014
Pine bark biochar	800°C; H_2O	Fe-impregnation	Toluene	Water	100% removal	Catalyst, adsorbent	Kastner et al., 2015
Biochar		Magnetic	PAHs	Sediment	Removal efficiency: Biochar> Organic Biochar, Magnetic Organic Biochar >Magnetic Biochar	Slower PAH uptake due to: magnetite or maghemite deposits on the surface and in macropores	Han et al., 2015
Wheat straw biochar		Graphene coated	Phenanthrene	Water	Greater thermal stability, and higher removal efficiency of phenanthrene	Partitioning and surface sorption Larger surface area, more functional groups	Tang et al., 2015
Birch wastes biochar Microbial immobilization	450°C	Microorganism carrier *(Pseudomonas aeruginosa, Acinetobacter radioresistens)*	Oil	Soil	Reduce PAH, altered bacterial community structure, reduce phytotoxicity	Biodegradation	Galitskaya et al., 2016
Fe_3O_4/ bamboo biochar composite/persulfate	800°C	Fe-coprecipitation in alkaline solution	16 US EPA PAHs	Marine sediment	86% removal	Adsorption Catalyst: Fe^{2+}–Fe^{3+} redox coupling, electron shuttling	Dong et al., 2018
Grass straw biochar Animal waste biochar	450°C	HNO_3 oxidation	Phenanthrene	Water	Biochar increase organic C-normalized surface area HNO_3 treatment caused O enrichment, loss of alkyl C, and rise of aromaticity HNO_3 oxidation effect on adsorption: Grass straw: increase; Animal waste biochar: decrease	Physical adsorption and chemical adsorption Pore filling π–π interaction Hydrophobic interaction	Jin et al., 2017

(continued on next page)

Table 4 (*continued*)

Biochar-based material	Production condition	Modification	Petroleum hydrocarbons	Matrix	Effect	Mechanisms	References
Willow biochar Coconut biochar Wheat straw biochar		Steam activated	PAHs	Soil	Biochar activation resulted in more pronounced reduction of C_{free} and C_{bioacc} PAHs		Kołtowski et al., 2017a
Rice straw biochar Wood biochar Bamboo biochar	300°C, 700°C	Base modification	Phenanthrene	Water	base modification improved the sorption of phenanthrene	Remove soluble C, increase surface area and hydrophobicity	Feng and Zhu, 2018
Mushroom biochar Microbial immobilization Nutrients	550°C	Microorganism carrier Oil-degrading bacteria	Oil (TPH 47.7 g/kg)	Oilfield soil	TPH removal: biochar (13.66 g/kg), biochar immobilized microbe (26.85 g/kg); biochar + microbe (20.74 g/kg)	Biodegradation	Zhang et al., 2019b
Crab shell biochar	700°C	KOH impregnation	Diesel oil	Water	Modification result high surface area, abundant functional groups, stratified surface structure	Multi-layer sorption	Cai et al., 2019
FeMn/biochar/H_2O_2		FeMn binary oxides modified	Naphthalene	Water	FeMn/biochar/H_2O_2 photo-Fenton system presented 80.7 and 2.18 higher naphthalene removal efficiency than biochar and Fe-Mn binary oxides	Synergistic effect of the combination of biochar and FeMn binary oxides Promoting adsorption capacity, increasing persistent free radicals	Li et al., 2019a
Biochar Biosurfactant rhamnolipid Phytoremediation (Spartina anglica)	700°C	Modification with rhamnolipid	TPH	Soil	Biochar: increase TPH removal by 19.1%; Biochar+hamnolpid: increase TPH removal by 23.8%; Rhamnolipid modified biochar: increase TPH removal by 26.5% Biochar alleviated the toxicity of petroleum hydrocarbons to plant	Biochar increases bacteria and plant mycorrhizal symbiotic fungus Biochar improves plant growth, assist phytoremediation	Zhen et al., 2018
Corn cob biochar Co-composting		HCl activation		Wetland soil	Available PAHs decreased by 6.44% for activated biochar, and 4% for fresh biochar Activated biochar increase microbial number and optimized fungi/bacteria gene copy number ratio	Biochar cause stronger adsorption and microbial activity	Ye et al., 2019
Corncob biochar Straw biochar Sawdust biochar Microbial immobilization	300–600°C	Microorganism carrier	Petroleum	Soil	Oil removal rate: corncob biochar (up to 70.7%), straw biochar (up to 58.3%), sawdust biochar (up to 57.4%),	Sorption Increase biodegradation	Ren et al., 2020

formation (Wu et al., 2014; Zhen et al., 2018). Particularly, Wei et al. (2020b) reported low ecotoxicity of rhamnolipids and the high efficiency of integrated use of biochar and rhamnolipid for petroleum hydrocarbon removal in wetland soils. With biochar as carrier, rhamnolipid could disperse oil in water while biochar assists the adsorption of oil. In addition, rhamnolipids can improve the mobility of biochar in water and increase the contact of organic contaminants with biochar. These results suggest that rhamnolipid-modified biochar could enhance the removal efficiency of oil and other organic contaminants from water. The hydrophilic-lipophilic property of biosurfactant rhamnolipids helped the dispersion of petroleum hydrocarbons and increased the contact of biochar with oil contaminants for removal (Karlapudi et al., 2018; Wei et al., 2020a, 2020b).

3.2.2. *Biochar as microorganism carrier*

Biochar was used as carrier material to immobilize the hydrocarbon-degrading bacteria strain to improve the bacterial density and competitive advantage of exogenous bacteria. This new approach assist biodegradation by involving bioaugmentation which conventionally has been used for treating contaminated soils by introducing pre-selected non-indigenous broad-type microorganisms (Mrozik and Piotrowska-Seget, 2010; Suja et al., 2014). Chen et al. (2012) demonstrated improved hydrocarbon degradation via an immobilized microorganism technique using biochar as the carrier of PAH-degrading bacteria. Galitskaya et al. (2016) immobilized bacterium isolated from PAH-contaminated soil (*Pseudomonas aeruginosa, Acinetobacter radioresistens*) on biochar to treat oil contaminated soil, and found that immobilization of microbes on biochar resulted in slightly increase of hydrocarbon decomposition rates. Ren et al. (2020) also reported the highest oil-removal rate of corncob biochar-immobilized microorganisms at 70.7% in petroleum-contaminated soil with functions of biodegradation and sorption. Biochar carried oil degraders was also applied with biostimulation. Further, Zhang et al. (2019c) investigated the petroleum-degrading bacteria immobilized mushroom biochar in presence of nutrients and showed an increase of TPH removal by 97% comparing to unmodified biochar in oil contaminated soil. In anaerobic environment, Yang et al. (2018) reported that biochar + nitrate, and biochar immobilized bacteria + nitrate increased PAHs removal in sediment by 20% and 17% comparing to biochar alone, respectively. The positive effect was suggested due to being nutrients as well as electron acceptor which encourage the growth of nitrate reducing PAH-degraders (*Thiobacillus* and *Stenotrophomonas*).

3.3. *To facilitate chemical degradation*

Chemical oxidation technology is a potent soil remedial option that can effectively eliminate oil contaminants. A new approach of biochar is to use in advanced oxidation processes (AOPs) for the degradation of recalcitrant organic pollutants by producing highly strong oxidizing species, such as $SO_4^{\bullet-}$, $\bullet OH$, $O_2^{\bullet-}$, and $^1O_2$. In recent studies, Fe impregnated biochar were used in AOPS to degrade single aromatics toluene in water with 81–100% removal rate (Mani et al., 2013; Bhandari et al., 2014; Kastner et al., 2015). Additionally, FeMn/biochar composite with H_2O_2 could degrade 82% naphthalene in water (Li et al., 2019a), and Fe_3O_4–biochar with persulfate exhibited degradation rate of 76%–90% of PAHs (pyrene, phenanthrene, and fluoranthene) in sediment (Dong et al., 2018). During AOPs, biochar promoted petroleum hydrocarbon dissipation with functions of supporting media, adsorbent, and catalyst. In Fenton/Fenton-like systems, Fe-modified biochar can replace iron catalysts due to pore structure, large surface area and highly active surface functional groups such as $C = O$ and C–OH, which could act as electron-donating groups which would activate oxidant to produce reactive species (Faheem et al., 2020).

To facilitate chemical degradation, a bioelectrochemical degradation combined with biochar in petroleum-contaminated soil remediation is potentially helpful. For example, Lu et al. (2014) observed the slight higher TPH removal efficiency (78.7%) of biochar anode compared to carbon cloth anode (73.1%) (initial TPH concentration: 11.46 g/kg soil) due to the higher sorption capabilities that could enhance hydrocarbon diffusion to the anode. In addition, Li et al. (2019b) found that biochar addition could improve the removal of PAHs and the electricity generation in microbial fuel cells (MFCs). Similarly, Rushimisha et al. (2023) demonstrated that non-oxidized and chemically oxidized biochar addition in bioelectrochemical systems (BES) could successfully remove the PAHs in soil by high electricity generation, reduction in mass transfer, and high abundances of functional genes.

3.4. *To improve recovery and reusability*

In recent years, the removal of oil by magnetism is attracting attention, as oil or emulsified oil can be easily separated from water by the external magnetic field after the magnetic biochar completely absorb oil (Li et al., 2017). Transition metals (Fe, Co, Ni, etc.) or their oxides have recently been introduced into biochar to form magnetic biochars by impregnation, co-precipitation, and hydrothermal carbonization during pyrolysis (Yi et al., 2020). A number of literatures have reported for the application of magnetic biochar with removal of various organic contaminants from environment by magnetic separation technique, which has been reviewed by Yi et al. (2020). In oil remediation approach, Gomes et al. (2010) used a magnetic polymer resin to recover oil from water. Several studies also reported the use of Fe-impregnated magnetic biochar in monoaromatic compound toluene/p-nitrotoluene adsorption in water (Chen et al., 2011; Mani et al., 2013; Bhandari et al., 2014; Kastner et al., 2015), as well as LMW naphthalene (Abu El-Rub et al., 2008; Chen et al., 2011). In soil, Han et al. (2015) reported high removal efficiency of Fe-magnetic activated carbon and biochar in PAHs (anthracene, pyrene, and chrysene) contaminated sediment with 77% recovery of magnetic activate carbon, although there was slower PAH uptake which caused by the magnetite or maghemite deposits on the surface and in macropores of adsorbents. Besides function of recovery, the incorporation of Fe oxides in biochar can also act as catalyst with persulfate in AOP (Dong et al., 2018), which has been discussed above.

4. Potential environmental impact of biochar in remediation of petroleum pollution

4.1. *Ecotoxicity of biochar*

Despite biochars are produced from biomass-based material, toxic substances from feedstock or generated products during pyrolysis still exist with potential environmental risk (Kusmierz and Oleszczuk, 2014). The toxic substances in biochar include organic fractions (VOC, PAHs, doxins), and inorganic fractions (heavy metals, persistent free radicals (PFRs) (Zheng et al., 2018). When biochar is applied in soil and water, the release of these substances from biochar to environment may cause secondary pollution which can impact plant and microorganism in water and soil (Zhang et al., 2019a). Particularly, the accumulated toxic substances in crops may further pass to consumer. Therefore, to reduce the environmental risk of biochar, removing undesired groups on biochar, and improving stability and recovery of biochar are essential. Regarding the risk of biochar and modified biochar to various ecosystems, agricultural crops as well as ocean and wetland ecology could be impacted.

4.1.1. *Impact of biochar on growth of plant and algae*

The application of biochar returns most of the plant nutrients back to the soil from where they were obtained, and thus plays a vital part in nutrient recycling (Laird, 2008). Previous studies have reported the promotion effect of biochar on the yield of plants (Jeffery et al., 2011, 2017). The positive effects of biochar on the growth of plants were suggested related to the liming effect, the increase of water retention capacity, the retention of heavy metals and organic compounds which are toxic to plant, and the enhancement on microbial activity (Bonanomi et al., 2017). However, differential responses of plants under different amendments of biochar were also reported. Wei et al. (2020b) reported that addition of biochar did not show any significant effect on biomass or soot/root ratio of saline marsh plant *Spartina Alterniflora*. Wang et al. (2018) found that application of biochar to soils may result in plant contamination and human cancer risk due to exposure of PAHs. Heavy metals in biochar (particularly in animal manure biochar) could hamper normal plant functioning and metabolic processes, resulting in the repression on photosynthesis, respiration, and enzymatic activities (Emamverdian et al., 2015). Persistent free radicals in biochar have been associated with inhibition on plant germination and survival and hindering the photosynthetic oxygen production in plants (Lian and Xing, 2017).

On the other hand, current literature on toxicity of biochar on algae is inconsistent. Magee et al. (2013) found that oil mallee biochar addition severely inhibited green algae (*Chlorella vulgaris*) growth and caused lipid accumulation, which is an environmental stress indicator of algae growth. Jia et al. (2018) also reported that the addition of apple tree biochar significantly inhibited the photosynthetic microorganism (*Oscillatoria* sp., *Phormidium* sp. and *Nostoc* sp.) growth in paddy soil, and the inhibition was suggested due to decrease of the light intensity by dissolved biochar, and further generated singlet oxygen and superoxide under sunlight exposure. Zhang et al. (2019a, 2019b) reported the toxicity of pine needles biochar on aquatic algae, and the effect is significantly correlated with PFRs in biochar. In contrast, Smith et al. (2013) showed that chicken litter and peanut shell biochar had no impact on growth inhibition while pinewood biochar was inhibitory on growth of aquatic photosynthetic microorganism blue–green alga (cyanobacteria *Synechococcus*) and eukaryotic green alga (*Desmodesmus*). The latter was attributed to water-extractable substances of biochar that contain carboxyl group (Smith et al., 2013). Wei et al. (2020b) found positive effect of sugarcane harvest residue biochar on wetland algae, and suggested that the promotion effect was due to the high silicon in sugarcane residue.

4.1.2. *Impact of biochar on soil microbial community*

Toxic components in biochar could impact activities of microbes, shift the soil microbial community, and alter the enzymatic actions in the soil that affect biogeochemical processes cycling (Spokas et al., 2011; Zhu et al., 2017). Various studies reported the change of microbial community of biochar addition in oil-contaminated soil. The ecotoxicity of biochar was showed by testing the relationship of soil biological activity and the content of PAHs and heavy metals (Hilber et al., 2012; Devi and Saroha, 2014). Cao et al. (2016) found that addition of biochar in phenanthrene and benzo[*a*]pyrene contaminated soil affected the microbial community and the intracellular process by reducing the activity of dehydrogenase as well as Shannon-Wiener index and evenness of community. Biochar addition in PAHs- contaminated upland soils could shift the bacterial community and increase abundance of oil degraders such as *Proteobacteria* which increased contact between phenanthrene and the PAH-degraders (Yang et al., 2018; Bao et al., 2020). Similarly, the promoted abundances of oil degraders (*Proteobacteria and Bacteroidetes*) were also reported in paddy (Liu et al., 2015) and coastal wetland soils (Wei et al., 2020a).

4.2. *Stability and toxicity of modified biochar*

4.2.1. *Toxicity of modification materials*

Besides the toxicity of biochar itself, modification materials on biochar also bring threat to the environment. Despite biosurfactants are generally considered ecofriendly, Wei et al. (2020b) found that application of rhamnolipid biosurfactant reduced plant biomass and inhibit algae growth in coastal environment. The negative effect of biosurfactants could be due to the triggered hypersensitive response-like defense and caused electrolyte leakage on plant (Monnier et al., 2018), and cell lysis and damaging to the plasma membrane and organelles on algae (Wang et al., 2005; Invally and Ju, 2017). Additionally, the integrated use of fertilizer could accumulate in natural water systems such as lakes and oceans, and subsequently, would cause eutrophication and stimulate growth of harmful algae (Bhateria and Jain, 2016). These pose risks to aquatic ecosystems by causing hypoxia and disrupting the growth of aquatic organisms since these harmful algae contain toxin and render the organic matters in water to be decomposed into harmful gasses, causing death of aquatic life, odor and taste problems (Yeoman et al., 1988). Besides consuming oxygen, algal decomposition produces dissolved inorganic carbon (DIC) and decreased the pH in bottom waters (Cai et al., 2011). These problems severely affect ecosystem functions and services (Rastogi et al., 2015). Moreover, addition of nutrients might also promote the growth of heterotrophic microorganisms which are not oil degraders, thereby creating competition between the microorganisms (Adams et al., 2015). On the other hand, introducing oil degrading bacteria bring in non-indigenous bacteria, and change soil microbial community in bioaugmentation approach with biochar as carrier.

4.3. *Possible ways to mitigate the contamination of biochar*

The toxic substances in biochar may be released from biochar in water, leading to secondary pollution and ecotoxicity. These substances, including PAHs, PFRs, and heavy metals, are generated during pyrolysis and closely depend on pyrolysis condition. Generally, organic pollutants are retained on biochar during its production from recondensation of pyrolysis vapors and liquids. For example, carboxyl groups are formed by breakage of bonds during moisture evaporation (Cárdenas-Aguiar et al., 2017). The PAHs and PFRs that are generated during carbonation of biomass and influenced by temperature. Specifically, low pyrolysis temperatures can promote formation of free radicals (Odinga et al., 2020). Temperatures of pyrolysis between 400 and 500°C, which are commonly used to produce biochar in industry, could result in maximum concentrations of PAHs in biochar (Keiluweit et al., 2012; Jeong et al., 2016). At temperatures higher than 650°C, lignin is converted into a PAHs and biochar becomes more hydrophobic (Buss et al., 2016). Additionally, heavy metal speciation in biochar was strongly impacted by pyrolysis temperature (Zeng et al., 2018). The internally introduced gas used in the biochar manufacturing process is closely related to the amount of PAHs generated. The content of PAHs generated during the biochar manufacturing process in nitrogen atmosphere was higher than that in CO_2 atmosphere (Hung et al., 2022). Moreover, increasing carrier gas flow and vaporization could decrease PAHs concentrations in biochar (Buss et al., 2016). On the other hand, feedstock selection influences the toxic substance content in biochar. For example, heavy metals in biochar were generally originated from the feedstock that contained heavy metals, which are left in biochar during pyrolysis. Buss et al. (2016) reported that straw-derived biochar contained 5.8 times higher PAHs concentration than softwood-derived biochar. Kołtowski and Oleszczuk (2015) reported that reduction of toxicity of biochars could be achieved by additional thermal treatment at 100–300°C. In addition, willow and wheat straw biochars showed lower toxicity of PAHs than miscanthus biochar. Clearly, selection of feedstock as well as pyrolysis conditions could largely reduce the toxicity of biochar. However, tolerant limit to biochar toxicity was different among diverse species, therefore when applied in a specific environment, an overall toxicity assessment of biochar is necessary.

5. Conclusion and outlook

Petroleum contamination in the environment causes serious environmental problem and requires remediation strategies with low impact to ecosystem as well as high removal efficiency. This review concludes the performance of biochar on oil remediation in water and soil environment, and reveal the different functions and mechanisms. Specifically, to remediate the oil contaminated soil, biochar enhanced the biodegradation by changing bioavailability of petroleum hydrocarbons and increasing the activity of oil degraders. Besides direct use, biochar-assisted bioremediation and bioaugmentation could be the main strategies in oil removal in soil. However, the potential ecotoxicity of biochar should also be considered as toxic substances such as PAHs and heavy metals from biochar could cause accumulation in plants, and disturbance on soil microbial community by reducing microbial diversity. While in oil contaminated water, biochar majorly acts as an adsorbent for recovering oil or as a catalyst to activate oxidant for chemical oxidation removal. The improvement in biochar oil removal efficiency could be achieved by feedstock selection, controlling pyrolysis condition, formation of composite with other adsorbents, as well as increase on mobility of oil by adding/attaching biosurfactant. Nonetheless, in aquatic environment, besides toxic substances in biochar, the attached materials in modified biochar may also show toxicity and hinder growth of aquatic plant and algae. Therefore, the stability and mitigation of potentially unwanted substances in biochar need to be further investigated.

Though biochar generally showed high remediation efficiency on petroleum hydrocarbons, the actual field performance effect of biochar application is still much limited as most studies were conducted in well controlled laboratory experiments. Future studies need to be carried out to validate these findings in a wide range of actual field scenarios. Additionally, the amount and type of raw materials that can contribute considerably to the favorable production cost and enhanced property of biochar, especially for specifically designed biochar with multifunction for pollutant remediation, need the further exploration. Overall, the future challenge and prospective is to develop efficient and recoverable biochar composites that can treat petroleum contaminated soil and water, while with minimized cost and environmental impact.

Declaration of Competing Interest

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

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