



Remediation of soils and sediments polluted with polycyclic aromatic hydrocarbons: To immobilize, mobilize, or degrade?

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are generated due to incomplete burning of organic substances. Use of fossil fuels is the primary anthropogenic cause of PAHs emission in natural settings. Although several PAH compounds exist in the natural environmental setting, only 16 of these compounds are considered priority pollutants. PAHs imposes several health impacts on humans and other living organisms due to their carcinogenic, mutagenic, or teratogenic properties. The specific characteristics of PAHs, such as their high hydrophobicity and low water solubility, influence their active adsorption onto soils and sediments, affecting their bioavailability and subsequent degradation. Therefore, this review first discusses various sources of PAHs, including source identification techniques, bioavailability, and interactions of PAHs with soils and sediments. Then this review addresses the remediation technologies adopted so far of PAHs in soils and sediments using immobilization

Abbreviations: AC, Activated carbon; Ace, Acenaphthene; Acy, Acenaphthylene; Ant, Anthracene; AOP, Advance oxidation process; BAF, Bioaccumulation factor; BaP, Benzo(a)pyrene; BbF, Benzo(b)fluoranthene; BghiP, Benzo(g,h,i)perylene; BkF, Benzo(k)fluoranthene; C, Carbon; CBRs, Critical body residues; CD, Cyclodextrins; CE, Capping efficiency; CEC, Cation exchange capacity; CFU, Colony forming unit; Chr, Chrysene; CMC, Critical micelle concentration; DBaH, Dibenz(a,h)anthracene; DGGG, Denaturing gradient gel electrophoresis; DNA, Deoxyribonucleic acid; DO, Dissolved oxygen; DOM, Dissolve organic matter; EDTA, Ethylenediaminetetraacetic acid; FAME, Fatty acid methyl ester; Fla, Fluoranthene; Flu, Fluorene; HAS, Humic acids; HMW, High molecular weight; HPCD, Hydroxypropyl-β-cyclodextrin; InP, Indeno(1,2,3-c,d) pyrene; K_{oc} , Organic carbon-water partition coefficient; K_{ow} , Octanol-water partition coefficient; LMW, Low molecular weight; N, Nitrogen; Nap, Naphthalene; nZVI, Nano-zerovalent iron; OMs, Organic matters; P, Phosphorous; PAHs, Polycyclic aromatic hydrocarbons; PCA-MLR, Principal component analysis-multiple linear regression; PCBs, Polychlorinated biphenyls; PDMS, Polydimethylsiloxane; PGPR, Plant growth-promoting rhizobacteria; Phe, Phenanthrene; PMF, Positive matrix factorization; PPPE, Poly-2,6-diphenyl-p-phenylene ether; Pyr, Pyrene; SDS, Sodium dodecylsulphate; SOM, Soil organic matter; SPMD, Semi-permeable membrane devices; SPME, Solid phase microextraction; STA, Sites of toxicity; TD, Thermal desorption; TX100, Triton X100; US EPA, United State Environmental Protection Agency.

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techniques (capping, stabilization, dredging, and excavation), mobilization techniques (thermal desorption, washing, electrokinetics, and surfactant assisted), and biological degradation techniques. The pros and cons of each technology are discussed. A detailed systematic compilation of eco-friendly approaches used to degrade PAHs, such as phytoremediation, microbial remediation, and emerging hybrid or integrated technologies are reviewed along with case studies and provided prospects for future research.

Table 1
Physical and chemical properties of the 16 priority PAHs.

PAHs compounds	Structure	No. of benzene rings	MW (g/mol)	Water solubility (mg/L)	Boiling point (C)	Melting point (C)	Vapor pressure at 25 °C, Pa	Log K _{ow}	Carcinogenicity
Naphthalene (Nap)		2	128.17	3.93	218	80.2	10.4	3.37	3
Acenaphthylene (Acy)		3	152.2	3.93	265.280	92–93	9×10^{-1}	4	3
Acenaphthene (Ace)		3	154.2	1.93	278–279	90–96	3×10^{-1}	4	3
Fluorene (Flu)		3	166.2	1.68–1.98	293–295	116–118	9×10^{-2}	4.18	3
Phenanthrene (Phe)		3	178.2	1.2	339–340	96–101	2×10^{-2}	4.57	3
Anthracene (Ant)		3	178.2	0.076	340	216–219	1×10^{-3}	4.54	3
Fluoranthene (Fla)		4	202.26	0.2–2.6	375–393	107–111	1.2×10^{-3}	5.22	3
Pyrene (Pyr)		4	202.25	0.077	360–404	150–156	6×10^{-4}	5.18	3
Benzo(a)anthracene (BaA)		4	228.3	0.01	435	157–167	2.8×10^{-5}	5.91	2B
Chrysene (Chr)		4	228.3	0.0028	441–448	252–256	5.7×10^{-7}	1.65	2B
Benzo(b)fluoranthene (BbF)		5	252.31	0.0012	481	167–168	NA	5.80	2B
Benzo(k)fluoranthene (BkF)		5	252.31	0.00076	480–471	198–217	5.2×10^{-8}	6	2B
Benzo(a)pyrene (BaP)		5	252.31	0.0023	493–496	177–179	7×10^{-7}	6.04	1
Dibenz(a,h)anthracene (DBaA)		5	278.4	0.0005	524	266–270	3.7×10^{-10}	6.75	2A
Indeno(1,2,3-c,d)pyrene (InP)		6	276.33	0.062	530	162–163	NA	6.58	2B
Benzo(g,h,i)perylene (BghiP)		6	276.33	0.00026	525	275–278	1.4×10^{-8}	6.5	3

1: Carcinogenic to humans.

2A: Probably carcinogenic to humans.

2B: Possibly carcinogenic to humans.

3: Not classifiable as carcinogenic to humans.

1. Introduction

PAHs are a category of organic chemicals with two, or more than two, bonded benzene rings, and 16 of them have been cited as priority contaminants by the Environmental Protection Agency of the United States (US EPA) owing to their oncogenic, teratogenic, and mutagenic characteristics (Zhang et al., 2021b; Lukić et al., 2016). PAHs in the natural environment originate from incomplete combustion of organic substances, including fossil fuel and biomass (Patel et al., 2020; Balmer et al., 2019), agricultural, mining, and industrial activities (Yuan et al., 2021; Bao et al., 2020; Tarafdar and Sinha, 2019), or from natural geogenic inputs (McGrath et al., 2019). Soils and sediments are contaminated with PAHs with concentrations greater than those that are a risk to humans and ecosystems (Xing et al., 2020; Yavar Ashayeri et al., 2018). PAHs pollution can cause adverse health impacts on humans and other living organisms (Yuan et al., 2021; Hussain et al., 2018) (Table 1). Low molecular weight (LMW) PAH compounds, consisting of 2–3 rings, have been reported to cause acute toxicity but are not carcinogenic (Olayinka et al., 2019). In contrast, high molecular weight (HMW) PAHs, consisting of 4–7 rings are relatively lower in toxicity but have carcinogenic, mutagenic, or teratogenic properties (Bauer et al., 2018; Ghosal et al., 2016).

PAHs are ubiquitous contaminants, and, due to their lipophilicity, they can easily sorb onto soils and sediments and persist over there for a long duration (Wu et al., 2019; McGrath et al., 2019). Thus, soils and sediments are considered to be the ultimate sinks of PAHs in terrestrial and aquatic ecosystems (Barhoumi et al., 2019; Alegbeleye et al., 2017). The level of PAHs in soils depends on the distance from the emission source, with higher quantities in industrial and urban areas than in suburban and rural areas (Yuan et al., 2015). The spatial dispersal of PAH compounds in soils is also affected by dissipation processes, such as sorption-desorption, abiotic degradation, volatilization, biodegradation, leaching, and bioaccumulation (Li et al., 2010; Su and Zhu, 2008). These processes are influenced by physicochemical properties of PAHs (e.g., molecular weight, the octanol-water partition coefficient (K_{ow}), and the organic carbon-water partition coefficient (K_{oc})), along with soil characteristics, such as pH, soil carbon concentrations, texture, and moisture content (Omores et al., 2017; Zhang et al., 2013). Environmental conditions (e.g., temperature and precipitation) are also factors that disturb PAHs distribution in the soil (Hong et al., 2020; Zhao et al., 2015). Similar to soils, sediments are considered as sinks for PAHs in aquatic ecosystems. When PAHs enter the riverine system, they are adsorbed onto particulate matter and immobilized on the surface sediments (Gong et al., 2018). Sediment-adsorbed PAHs in overlying waters are a threat for aquatic organisms (Idowu et al., 2020a; Soukarieh et al., 2018). PAHs enter in the flora and fauna and can be transferred to humans via the food chain and display biomagnification (Honda and Suzuki, 2020).

Increased concentration of PAHs in the environmental, along with their eco-toxicity and health impact on humans, have resulted in several investigations concerning removing them from the environment using various physical, chemical, biological, and integrated or hybrid technologies (Zhang et al., 2021a; Cui et al., 2020; Idowu et al., 2020b; Kuppasamy et al., 2017). Due to the heterogeneity of soils and other factors, a huge percentage of the overall existing PAH compounds are non-bioavailable to living beings. Bioavailability is reduced as PAHs age within a complex soil and sediment matrix (Maletić et al., 2019; Yang et al., 2016). For instance, the remediation rate of PAHs can be either rapid or slow reliant on the soil and sediment properties, bioavailability, and the time it takes to generate derivatives (oxygenated derivatives) that are more noxious than the initial PAH compounds (Zang et al., 2021; Idowu et al., 2020a; Kuppasamy et al., 2017). Specific characteristics of PAHs, such as their high hydrophobicity and low water solubility, reduce their bioavailability and subsequent degradation (Ranjbar Jafarabadi et al., 2020; Idowu et al., 2020b; Zang et al., 2020). Because of these constraints, the need for evolving methods to remediate PAHs is evident. Avenues have developed to design novel remediation

technologies that can override the prevailing technological limitations. Assessment built on bioavailability is considered as an important tool in risk-based tactics for remediation of PAHs polluted sites (Duan et al., 2015).

There have been a number of publications on PAH contamination and its remediation technologies (Dai et al., 2020; Lu et al., 2019; Ma et al., 2018; Kuppasamy, 2017; Lawal, 2017) (Fig. 1). However, only a limited number of reviews have reported contamination of soils and sediments by PAHs and remediation approaches based on their bioavailability. This review discusses methods to remediate soils and sediments contaminated with PAHs using conventional and advanced immobilization, mobilization, and degradation techniques. In addition, case studies are presented giving the source of PAHs and how the PAHs interact with soils and sediments. Also, this review covers the limitations of current technologies. Finally, future research is discussed, which is needed to evolve innovative approaches to improve the remediation of soils and sediments polluted with PAHs.

2. Sources, source identification techniques, and interaction of PAHs in soils and sediments

PAHs sources can be categorized as pyrogenic, petrogenic, and biological or natural (Abdel-Shafy and Mansour, 2016). When biomass is processed at high temperatures in low or no oxygen conditions, pyrogenic PAHs are emitted (Balmer et al., 2019). The transformation of coal into coke/coal tar, or the pyrolytic refining of petroleum residues into lower hydrocarbons, leads to emission of pyrogenic PAHs (Guarino et al., 2019; Abdel-Shafy and Mansour, 2016). Petrogenic sources of PAHs include transportation, stowage, and utilization of crude liquid fuel and its products. Major petrogenic PAHs sources include oceanic and riverine water oil spills, above and belowground oil storage tank seepages (Guarino et al., 2019), and the accretion of large quantities of minor discharges of gasoline, diesel oil, and interrelated constituents linked with transportation (Hussain et al., 2018). Volcanic eruptions, forest fires, and biological syntheses are considered as natural PAHs sources in the environmental setting (Jiao et al., 2017; Hiller et al., 2015) (Fig. 2). PAHs in soil can accumulate in biota and plants via the food chain, resulting in direct or indirect human exposure (Bortey-Sam et al., 2014). For outdoor environments, most human exposure to PAHs is via the soil rather than air and water (Kamal et al., 2015). As a result, it is critical to recognize the sources and extent of PAHs contamination in soils and sediments to avoid or reduce ecological impact and animal health risks.

2.1. Sources of PAHs in soils

Many researchers have investigated the concentrations and various PAHs sources in soils and/or sediments from various regions worldwide (Table 2). Due to differences in industries, fuel consumption, and combustion and incineration processes, the level and extent of pollution

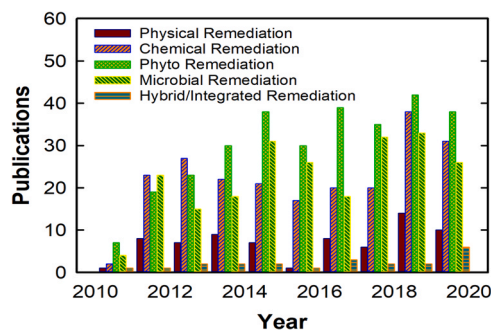


Fig. 1. : Publications over the past 10 years related to various technologies used to remediate PAHs.

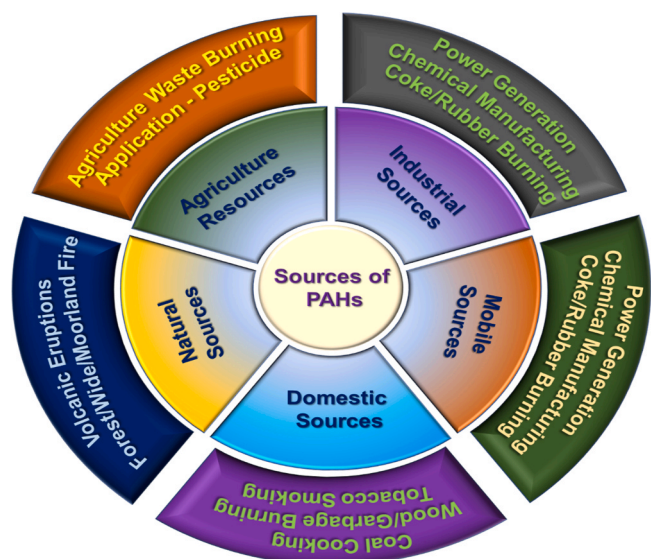


Fig. 2. Sources of PAHs that pollute soils and sediments.

from PAHs are different in developed and urban areas compared to undeveloped and rural areas (Zhang and Chen, 2017). Among different PAHs sources, emissions from industries are the primary source of PAHs identified in soils and sediments (Han et al., 2020b; Liu et al., 2020b; Idowu et al., 2020b). Elevated levels of PAHs have been reported in locations with rapid intensification of industrial activities (Wang et al., 2017b). For instance, the source identification of PAHs concentration in samples of soil collected from Ulsan, an industrial region in South Korea, exhibited much higher levels of PAHs at industrial sites than in urban and rural areas (Kwon and Choi, 2014). Likewise, a higher level of PAHs was detected in the soil from industrial areas of Shandong than in rural regions around it (Chai et al., 2017). The sources of PAHs in urban centered soils in two Portuguese cities revealed that PAHs in the larger and more industrialized city of Lisbon came from industry, traffic, and incineration processes. In contrast, in the smaller city of Viseu, atmospheric transport was the principal source of PAHs contamination in soils (Cachada et al., 2012).

The emitted PAHs from an industrial area could also be deposited on agricultural soils through wastewater irrigation and atmospheric transport (Wang et al., 2017b). Two major rivers serving as water sources for agricultural activities in the Huanghuai region of China were reported to be severely polluted with PAHs along with other contaminants (Chen et al., 2005). A high concentration of PAHs was noticed in the topsoil of a paddy field, where wastewater was used for irrigation (Bu et al., 2009). Parks in urban areas are recreational places for tourists and residents and have been studied to assess the PAHs levels in their soils due to the potential risk of direct contact with humans. The primary sources of PAHs in the topsoils from parks in Beijing were found to be pyrogenic, resulting from the combustion of coal and oil fuels (gasoline and diesel) (Qu et al., 2020). The PAHs found in urban soils obtained from public parks, residential areas, commercial areas, roadside areas, and cultural and educational areas in Beijing were studied, and they were found to have originated from vehicle emission (54%) and coal burning (46%) (Liu et al., 2010). In these studies, atmospheric transport was found to be a potential source of PAHs in soils.

Apart from coal combustion, traffic emission is an imperative PAHs source in soils of industrial and urban sites (Kwon and Choi, 2014). For example, emissions from traffic were found as the key source of PAHs contamination in industrial area soils of Shandong, China (Chai et al., 2017). Due to a consequence of rapid urbanization, a considerable increase in vehicles on roads has occurred. PAHs can be released from tire abrasion, vehicle emissions, and asphalt pavements and, subsequently, be transferred to roadside agricultural soils through runoff, dust, splash,

spray, and airborne particles (Kibblewhite, 2018; Wickle, 2000). As a significant PAHs repository, roadside agricultural soils present a threat to food safety, because PAHs can be taken up by crops and transferred through the food chain (Yang et al., 2021). Hence, it becomes critical to measure the potential risks of roadside agricultural soils to human health due to their proximity to heavy vehicular traffic (Cachada et al., 2012).

For a long time, Antarctica was well thought-out to be an un-polluted environment. But nowadays it is affected by anthropogenic activities, such as military installations, research activities, and tourism, which are responsible for a rise in traffic, fuel usage and storage, sewage discharge, and scientific drilling activities (Deelman et al., 2020). Fifteen PAHs were measured in the soil at the Fildes Peninsula in the west of Antarctica, which has regular human activities. The results demonstrated that local sources, such as fossil fuel burning and spilling, were the foremost PAHs sources. The results also showed that, in polar areas, PAHs mainly originate from long-range atmospheric circulation and “grasshopper” effects (evaporate with warm air and return to earth with rain and snow) from release sources (Na et al., 2020). Similarly, the PAHs in field soils of King George Island in Antarctica were found to have petrogenic release, such as oil spills and fuel leaks. The pyrogenic origins of PAHs in this region were from incomplete burning of fossil fuels, crude liquid fuel, diesel, and, coal (Deelman et al., 2020).

Apart from regional variations, seasonal variations also influence the level of PAHs contamination in soils. For instance, throughout summer, the key PAHs sources in the soil of Dalian, a coastal city in northeast China, were traffic exhaust (54%). However, PAHs were primarily emitted by coal-fired boilers (72%) during the winter. The difference was explained because there is a need for heating in winter, with greater consumption of coal than in summer (Wang et al., 2009). PAHs in soil of Kutahya, Turkey, located near coal-fired power plants, showed higher levels during winter than summer. The key PAHs sources in this region were coal combustion for residential heating and power generation and from diesel and gasoline exhaust emissions (Dumanoglu et al., 2017). At low temperatures, gaseous PAHs condense and become less volatile (Manzetti, 2013), which may account for the high occurrence of PAHs in Antarctic soils.

2.2. Sources of PAHs in sediments

Due to their physicochemical properties, PAHs are likely to come in contact with suspended particles present in the marine environment and eventually get deposited as sediments (de Almeida et al., 2018; Srogi, 2007). The deposition of PAHs in sediments is controlled by environmental conditions; biological, physicochemical, and textural properties of sediments; and the physicochemical properties of PAHs (Darilmaz et al., 2019). The re-release of PAHs into the aquatic environment from sediments is possible owing to external environmental changes and bioturbation (Du et al., 2018). The occurrence of PAHs in sediments thus imposes a significant risk to living being health due to the sediments' direct contact with aquatic organisms and transfer of PAHs via the food chain (Meng et al., 2019). To control sediments pollution from PAHs, it is essential to recognize the sources and their distribution in sediments (Meng et al., 2019). Several investigations have examined the concentrations of PAHs, and identified their origins in marine and freshwater sediments, as summarized in (Table 2).

PAHs are being discharged into the marine ecosystem as a result of hasty industrialization and urbanization, and they pose a threat to this environment (Meng et al., 2019; Yuan et al., 2017). PAHs in oceanic sediments can be generated from a combination of petrogenic and pyrogenic sources. The sources of 16 PAHs detected in an intertidal sediment at the estuary of the Shuangtaizi river, one of the most contaminated rivers in China, were primarily attributed to aquacultural activities, agricultural activities, and effluents from nearby oilfields. Although petrogenic sources (63%) of PAHs were dominant in the estuary sediments, a significant portion (22%) of PAHs were found to have

Table 2
Selected references on sources of PAHs in soils and sediments.

Region	PAHs source	Number of PAHs	PAHs content range (mean±STD) (ng g ⁻¹)	Reference
Soil				
China				
Surface soils of Yellow River Delta	Pyrogenic (coal and biomass combustion, vehicular emission), petrogenic (petroleum spills)	16	27–753 (118 ± 132)	(Yuan et al., 2014)
Soils near an e-waste recycling site in South China	Gaseous emission and particle deposition from open burning of e-waste	16	127–10,600	(Wang et al., 2012a)
Urban soils of Shanghai	Pyrogenic (coal and biomass combustion, creosote, coke tar related sources, vehicular emissions), petrogenic	26	Σ26PAHs = 133–8650 (2420) Σ16PAHs = 83.3–7220 (1970)	(Wang et al., 2013)
Urban soils of Beijing	<i>Primary sources:</i> Pyrogenic (vehicle exhaust, coal combustion) <i>Secondary source:</i> atmospheric deposition of long-range transported PAHs	16	93–13141 (1228)	(Peng et al., 2011)
Soils from Chengdu Economic Region, Sichuan, western China	Pyrogenic (fossil fuels combustion in the mountain region, incomplete petroleum combustion in the developed plain area)	16	12.52–75,432 (3233.92)	(Xing et al., 2011)
Soils of Huanghuai Plain	Pyrogenic (combustion of wood/biomass, fossil fuel, traffic emission)	15	14–1246 (128)	(Yang et al., 2013)
Soils of vegetable greenhouses in Shandong	Coal combustion, traffic coke source, biomass combustion, petroleum source, or mixed sources	16	152.2–1317.7 (407.4)	(Chai et al., 2017)
Soils in industrial areas of the Yangtze River Delta region	Pyrogenic (coal and petroleum combustion)	16	Surface soil = 189.5–1070.4 (471.30) Subsoil = 103.8–743.7 (341.40)	(Wang et al., 2017b)
Agricultural soils of Huanghuai plain	Pyrogenic (coal combustion, low-temperature combustion, traffic emissions)	16	15.7–1247.6 (129.5)	(Yang et al., 2012)
Surface soil near a large steel-smelting manufacturer in northern China	Pyrogenic (a mixture of industrial and domestic combustion of coal and biomass, traffic exhaust)	15	689.6	(Liu et al., 2017)
Agricultural soils around a chemical plant region	Diesel emissions, coke ovens, coal and fuel combustion, gasoline emissions	16	250.49–9387.26 (2780.42)	(Liu et al., 2016)
Surface soil from rural and urban areas of Hong Kong, China	<i>Rural area:</i> biomass combustion <i>Urban area:</i> vehicular emission around the heavy traffic roads	16	7–410 (54.6)	(Zhang et al., 2006)
Urban soils in Beijing	Pyrogenic (motor vehicle exhausts, industrial activities, coal combustion), petrogenic	16	366–27,825 (3917)	(Tang et al., 2005)
Agricultural soils from Shunde, Guangdong	Combustion of grass, wood, coal and fossil fuel, petroleum	16	33.7–350	(Li et al., 2008)
- Soil of Dalian	- Pyrogenic (coal combustion, traffic exhaust)	19	1220.48 ± 316.04–35,003.38 ± 3086.63	(Wang et al., 2009)
Surface soil of urban parks in Beijing	Pyrogenic (coal combustion, oil fuels such as gasoline and diesel)	16	66–6867 (460)	(Qu et al., 2020)
Surface soils from Dajiu Sub-alpine Wetland	Petroleum, coal/biomass combustion emission	15	7.3–191.48 (42.4 ± 38.5)	(Xing et al., 2020)
Roadside agricultural soils, Shanghai	Pyrogenic (vehicle emission, combustion of coal, biomass, and natural gas)	16	17.2–3775 (339 ± 594)	(Yang et al., 2021)
Biomass ashes disposed of in the urban landfill, Tibetan Plateau	Domestic consumption of biomass fuels	16	Surrounding soils: 23.7–293 (103) landfill cover soils: (1009)	(Li et al., 2020)
Surface soil, Tibetan Plateau	Pyrogenic (combustion of yak dung)	15	5.54–389 (59.9)	(Wang et al., 2014)
South Korea				
Industrial, urban, and rural soil of Ulsan	Pyrogenic sources and traffic emission	16	65–12000 (960)	(Kwon and Choi, 2014)
India				
Agricultural soils of Delhi	Pyrogenic (biomass and fossil fuel combustion)	16	830–3880 (1910 ± 1020)	(Agarwal et al., 2009)
Urban traffic soil of Dhanbad	Pyrogenic (vehicular emission, coal and biomass combustion)	13	1.02–10.9 (3.488)	(Suman et al., 2016)
Traffic soil of Dhanbad heavy mining area	Pyrogenic (fossil fuel combustion, vehicular pollution)	13	8256–12562	(Tarafdar and Sinha, 2019)
Pakistan				
Soil along Jhelum riverine system of lesser Himalayan	Pyrogenic (biomass combustion, vehicular emissions)	15	62.79–1080 (433)	(Riaz et al., 2019)
Iran				
- Urban soil of Isfahan metropolis	Geogenic and pyrogenic	16	57.7–11,730 (2000)	(Moore et al., 2015)
Turkey				
Soil samples of Aliaga industrial region in Izmir	Iron–steel plants, petroleum refinery, ship dismantling plants	16	1–1166	(Kaya et al., 2012)
France				
Soils in the Seine River basin	Automobile traffic, domestic heating, industrial emissions	14	450–5650 (2510)	(Motelay-Massei et al., 2004)
Germany				
Bank soils along Saar and Mosel Rivers	Petrogenic and pyrogenic	45	Σ16PAHs = <200–128000 Σ45PAHs = 18,000–197,000	(Pies et al., 2008)
Portugal				
Urban soils in Lisbon and Viseu	<i>Lisbon:</i> traffic, industry and incineration processes <i>Viseu:</i> atmospheric transport	16	<i>Lisbon:</i> 6.3–22,670 (1544) <i>Viseu:</i> 6–790 (169)	(Cachada et al., 2012)
Australia				
Soils of Newcastle, Australia	Pyrogenic (combustion of liquid fossil fuel and coal of power plants)	13	<i>Recreational site:</i> 5567–100,931 <i>Industrial site:</i> 2509–392,932	(Idowu et al., 2020a)

(continued on next page)

Table 2 (continued)

Region	PAHs source	Number of PAHs	PAHs content range (mean±STD) (ng g ⁻¹)	Reference
Ghana			<i>Smoking area:</i> 1395.4–8741.3 <i>Residential site:</i> 201.3–11,176	
Surface soils from the Kumasi Metropolis	Pyrogenic (fuel combustion)	22	14.78–2084 (442.5 ± 527.2)	(Bortey-Sam et al., 2014)
Antarctica				(Na et al., 2020)
Soil at the Fildes Peninsula	Pyrogenic (fossil fuel combustion), petrogenic (spilling)	15	20.9–2197 (155)	
Terrestrial soils of King George Island	Petrogenic (fuel leaks and oil spills), pyrogenic (incomplete combustion of petroleum vehicle fuels and crude oil, coal combustion, diesel emissions)	12	1.83–32.9 (10.8 ± 8.22)	(Deelman et al., 2020)
Sediment				
China				(Deng et al., 2013)
Surface sediments of mud areas from the East China Sea	Pyrogenic (Biomass, coal, and petroleum combustion residues)	34	<i>Inner shelf sites:</i> 294–1381 (703 ± 332) <i>Outer shelf sites:</i> 257–779 (481 ± 204)	
Surface sediments of Fenhe reservoir and watershed	Pyrogenic (coal combustion, diesel, and gasoline emissions)	16	539–6282 (2214.8)	(Li et al., 2012b)
Sediments of Zhanjiang and Leizhou Bays, South China	Petroleum and its combustion	16	<i>Zhanjiang bay:</i> 41.96–934 (315.98) <i>Leizhou bay:</i> 21.72–319.61 (103.9)	(Huang et al., 2012)
Surface sediments of rivers (Huangpu, Suzhou) and an estuary (Yangtze), Shanghai	Petrogenic and pyrogenic (incomplete combustion of coal, grass, and wood, vehicle, and vessel exhaust)	17	107–1707 (823) (excluding perylene)	(Liu et al., 2008)
Surface sediments of Huangpu River, Shanghai	Petrogenic (spills of oil products), pyrogenic (coal combustion, traffic-related pollution)	18	NA	(Liu et al., 2009)
Sediments near wetland plants in the Chongming wetland, Shanghai	Pyrogenic (incomplete combustion of coal and biomass)	16	38.7–136.2	(Wang et al., 2012b)
Sediment from Lake Chaohu	Pyrogenic (coal and wood combustion, vehicle emission)	16	109.7–6245.8 (908.5 ± 1878.1)	(Qin et al., 2014)
Surface sediments from Guan River Estuary	Pyrolytic (exhaust of diesel engines, heavy traffic, coal heating), petrogenic (gas, oil, or diesel spills, oily sewage discharge)	21	90–218 (132.7)	(He et al., 2014)
Surface sediments from Nan'ao Island, South China	Petroleum and combustion of coal and biomass	16	265.5–951 (516)	(Gu et al., 2013)
Surface sediments of Yellow River	Pyrogenic (traffic-related pollution, coal and biomass burning, coke oven)	16	~ 20–250	(Feng et al., 2014)
Sediments from Taihu Lake	Pyrogenic (vehicular emission, combustion of coal and wood)	15	209–1003 (476)	(Zhang et al., 2012)
Surface sediments of the Bohai Sea	Petrogenic (spilled oil products), pyrogenic (coal combustion, traffic-related pollution)	16	97–300.7 (175.7 ± 37.3)	(Hu et al., 2013)
Surface sediments from Taihu Lake body (THL) and Taihu Lake basin (THB)	Petrogenic and pyrogenic (grass, wood, coal combustion)	16	<i>THB:</i> 12–2281 (348.7 ± 399.4) <i>THL:</i> 11.4–209.9 (63.6 ± 40.9)	(Zhao et al., 2017a)
Surface sediments from Lake Qinghai, northeast Qinghai–Tibet plateau	Pyrogenic (combustion of biomass and coal-based fossil fuels)	16	366–966 (590)	(Wu et al., 2014)
Intertidal sediment at Shuangtaizi Estuary, Bohai Sea of China	Pyrogenic and petrogenic	16	28.79–281.97 (115.92)	(Yuan et al., 2017)
Sediments of Chinese Bohai, Yellow Seas, European Baltic, and North Seas	Pyrogenic (coal combustion, vehicular emission, coke plant and petroleum residue)	18	<i>European Baltic:</i> 0.91–5361 (549 ± 1124) <i>North Seas:</i> 0.46–227 (30.2 ± 57.2) <i>Chinese Bohai:</i> 25.0–308 (184 ± 109) <i>Yellow Seas:</i> 4.3–659 (138 ± 133)	(Wang et al., 2020b)
Sediments along Yangtze River Estuary Deepwater Channel	High-temperature combustion of local fossil fuels (vehicular exhaust, anthropogenic combustion, pyrogenic sources)	15	89.52–208.02 ng/g (140.48)	(Liu et al., 2020b)
Sediment in Shilaoren Bay, Qingdao	Incomplete combustion of petroleum, coal, and biomass affected by petroleum sources	16	11.78–129.21 (57.99)	(Han et al., 2020a)
Sediments of mouth bar of the Yangtze River Estuary	Vehicle emissions, natural gas combustion, biomass and coal combustion	17	34.94–580.26 (146.31)	(Liu et al., 2020a)
Surface sediments of East China Sea	Pyrogenic	16	57.5–364.5 (166.2)	(Zhao et al., 2020)
Sediments of Maba River, South China	Pyrogenic (coal, grass, wood, and petroleum combustion)	16	<i>Maba River:</i> 47.61–25480.98 (4382.98) <i>Meihua River:</i> 60.30–15956.62 (3664.32)	(Chen et al., 2020)
Sediments of Lanzhou Reach of Yellow River	Petroleum and combustion of petroleum, biomass, and coal	18	638–1620 (901)	(Jiang et al., 2020)
Sediments from laizhou bay	Pyrogenic (fossil fuel combustion, biomass burning, coke oven)	16	268.97–895.37 (612.52)	(Han et al., 2020b)
Sediment of Jiaozhou Bay	Petrogenic and pyrogenic (vehicle emission, biomass and coal combustion)	15	37.7–290.9 (125.8)	(Cao et al., 2020)
Sediments of Taihu Lake	Coal, petroleum, coke, and biomass combustion	12	NA	(Wang et al., 2020a)
Lake sediment cores from Songnen Plain, Northeast China	Straw burning, forest fire, combustion of gasoline, coal, coke, and diesel	13	59.7–506	(Bao et al., 2020)
Surficial sediments of Yangtze Estuary	Petroleum, biomass, and coal combustion	16	90.14–502.12 (221.18)	(Li et al., 2012a)
Sediments from Pearl River Delta and Northern South China Sea	Petrogenic and pyrogenic (combustion of coal and wood, petroleum spills, vehicle emissions, nature sources)	25	138–6793	(Luo et al., 2008)
Surface and core sediments from Daya Bay, South China	Petrogenic and pyrogenic (coal/grass, and wood combustion)	16	<i>Surface sediment:</i> 42.5–158.2 (126.2) <i>Core 8 sediment:</i> 77.4–305.7 (92.1) <i>Core 10 sediment:</i> 118.1–319.9 (210.2)	(Yan et al., 2009)

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Table 2 (continued)

Region	PAHs source	Number of PAHs	PAHs content range (mean±STD) (ng g ⁻¹)	Reference
Surface sediments from Caofeidian Long Island	Pyrogenic (coal, biomass, and petroleum combustion)	16	51.55–805.86 (403.50)	(Han et al., 2019)
Surface sediments of the Subei Shoal	Coal combustion, transportation, coke processing, petroleum sources	16	<DL-25.24 (5.88)	(Zhang et al., 2019a)
Qinhuai River, Nanjing	Pyrogenic (biomass and coal combustion, vehicular emission), petrogenic	14	796.2–10470 (2713.8)	(Zhao et al., 2017b)
Maozhou River, Shenzhen, South China	Petrogenic (oil pollution), pyrogenic (fossil fuel and biomass combustion)	16	28–1051 (458)	(Zhang et al., 2017)
Korea				(Yim et al., 2005)
Surface and core sediments from Masan Bay	Pyrogenic with little influence of petrogenic sources	24	207–2670 (680)	
Iran				(Fakhradini et al., 2019)
Sediment of Hoor Al-Azim wetland	Petrogenic and pyrogenic (oil leakage, unburned and combusted fossil fuels from fishing boats and vehicle engines, incomplete combustion, auto emission, fossil fuels and biomass combustion)	16	15.78–410.2 (51.71 ± 94.86)	
Surface sediment of Musa Estuary, the largest Estuary in the Persian Gulf	petrogenic and pyrogenic	16	9.48–1514.3 (148.76)	(Mehr et al., 2020)
Surface sediment from Hormozgan coastline	Petrogenic and pyrogenic	16	14.54–85.00 (34.92)	(Jahromi et al., 2020)
Shadegan wetland	Petrogenic and pyrogenic (incomplete combustion, bulrush combustion, vehicular exhaust, fishing boat emissions, oil spill)	16	10 ± 0.5–317 ± 14.3 (36.5 ± 1.6)	(Yavar Ashayeri et al., 2018)
India				(Dudhagara et al., 2016)
Surface sediments at Bhavnagar coast, Gujarat	Petrogenic and pyrogenic (burning of coal, gas, oil, production of cement, petrochemicals, and rubber tires)	16	5020–981180 (345,000)	
Surface sediments of Hooghly estuary	Pyrolytic and petrogenic	16	3.3–630 (137.7 ± 226)	(Mitra et al., 2019)
Pakistan				(Riaz et al., 2019)
- Sediments along Jhelum riverine system, lesser Himalayan	- Pyrogenic (biomass combustion, vehicular emissions)	15	14.54–437.43 (492)	
Turkey				(Darilmaz et al., 2019)
Surficial Sediments of Edremit Bay (Aegean Sea)	Pyrogenic (wood-coal combustion, vehicle emissions)	18	0.65–175 (43.1)	
Thailand				(Boonyatumanond et al., 2006)
Riverine, estuarine, and marine sediments	Petrogenic and pyrogenic	17	6–8399	
France				(Gardes et al., 2020)
Sediment cores from the major Seine estuary tributary	Pyrogenic	16	<i>Martot pond</i> : 2930–38200 (12,700 ± 0.51) <i>Les Damps pond</i> : 2270–28200 (11,100 ± 0.89)	
Arc River and Berre lagoon	Biogenic and pyrolytic	16	151–1257 (585.3)	(Kanzari et al., 2012)
Ireland				(Guinan et al., 2001)
Sediments of two Northern Irish Sea-loughs, east coast of Northern Ireland	Pyrogenic	13	83–23,000	
USA				(Christensen and Bzdusek, 2005)
Sediments of Black River and Ashtabula River, Ohio	Pyrogenic (traffic, coke oven, wood burning/coal tar)	16	0–46,600	
Sediments of Lake Calumet, Chicago	Pyrogenic (coke ovens and traffic)	17	4900–20,000	(Li et al., 2003)
Mexico				(Macias-Zamora et al., 2002)
Surface marine sediments in Todos Santos Bay, B.C.	Pyrogenic (a combination of urban air, wood and brush fires with a slight influence of oil)	16	7.6–813 (96)	
Canada				(Davis et al., 2019)
Small craft harbor surficial sediments in Nova Scotia	Pyrogenic (coal and biomass combustion, automobile exhaust), petrogenic	16	NA	
Surface and dated sediment cores from Saskatchewan lakes, east of the Athabasca oil sands	Softwood combustion from boreal forest fires	15	9.3–92.1	(Ahad et al., 2015)
Brazil				(de Almeida et al., 2018)
Surface sediment of Todos os Santos Bay,	Pyrogenic (pyrolysis of fossil fuel, biomass, and coal)	16	<DL-533	
Australia				(Duodu et al., 2017)
Sediments from Brisbane River,	Pyrogenic (gasoline and diesel emissions, biomass and natural gas combustion)	15	148–3079 (849 ± 646)	
Australia				
Sediments of Lake Macquarie	Pyrogenic (atmospheric deposition, coal combustion, vehicular emissions)	13	29–1059.7 (264.8)	(Idowu et al., 2020b)
Egypt				(Barakat et al., 2013)
Surface Sediments of Lake Manzala	Petrogenic (near-shore urban hotspots), pyrolytic (in coastal and offshore areas), biogenic	39	246–9910 (1480)	
Nigeria				(Anyanwu et al., 2020)
Sediment cores from Bonny Estuary, Niger Delta	Petrogenic and pyrogenic	14	8699–22,528	
Sediments from Calabar River, SE Niger Delta	Petrogenic and pyrogenic	17	1670–20,100 (9370 ± 579)	(Oyo-Ita et al., 2013)
		26, 18, 14		(Saha et al., 2009)

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Table 2 (continued)

Region	PAHs source	Number of PAHs	PAHs content range (mean±STD) (ng g ⁻¹)	Reference
Surface sediment from tropical Asian waters (India, Indonesia, Malaysia, Thailand, Vietnam, Cambodia, Laos, and the Philippines)	Petrogenic and pyrogenic (crude oil, automobile exhaust, coal, and wood combustion products)		Σ26PAHs = 4–42900 Σ18PAHs = 4–40500 Σ14PAHs = 4–38,100	

NA: Not available

originated from fossil fuel and biofuel combustion and were transported through freshwater runoff and atmospheric deposition (Yuan et al., 2017).

Aside from industrialization and urbanization, overexploitation of sea areas also increase the PAHs level in the marine environment through direct release into water via oil spills and fuel discharge (Meng et al., 2019; Zheng et al., 2016). Bao et al. (2020) reported that oil processing from the Daqing Oil Field, the largest base of petroleum processing and production in the Songnen Plain (China), was accountable for the release of PAHs in this area, either through direct oil spill during transportation, drilling, and extraction or by indirect fuel burning. Moreover, the origin of PAHs in surface sediments of the Pearl River Delta and the Northern South China Sea was found to be mainly from petroleum spills (36%), followed by coal or wood burning (27%), vehicle exhaust (25%), and natural origin (12%) (Luo et al., 2008). Similarly, PAHs in sediments of the Shuangtaizi Estuary were found to have originated predominantly from petrogenic sources, like petroleum spills and gasoline emissions (Yuan et al., 2017).

In addition to emissions from land vehicles, marine vehicles also can contribute to sediment PAHs (Darilmaz et al., 2019). PAHs from vehicular emissions in aquatic sediments are expected to be mostly from marine vehicles, such as motorboats, fishing vessels, and ferries. For example, in surficial sediments in Nova Scotia, Canada, PAHs from two sources were found: from regular boat traffic by fishing vessels and from automobile exhaust that came from diesel engines (Davis et al., 2019). Furthermore, the source distribution of 15 PAHs in Taihu Lake sediments in China revealed that most PAHs came from vehicular emission (~54%). The high contribution of vehicular emission was due to fishing activities using motorboats that emitted exhaust around the lake (Zhang et al., 2012). Surface sediments of the Huangpu River showed that 40% of the PAHs came from traffic-related sources, which was attributed to a rapid rise in the number of automobiles in Shanghai and exhaust from passenger ferries and cargo vessels in the Huangpu River (Liu et al., 2009). Also, petrogenic PAHs sources, such as crude and refined oil leaks, contributed 24% of the PAHs in outward sediments of the Huangpu River (Liu et al., 2009). Similarly, a significant portion (15%) of PAHs in sediments of the Shuangtaizi Estuary was found to have originated from diesel engine emissions by ship traffic (Yuan et al., 2017).

2.3. Source identification techniques

Accurate source identification of the PAHs is essential to mitigate the cause of pollution and reduce the risk of human exposure (Yang et al., 2021). Several investigations have focused on the distribution of PAHs in soils and sediments, due to different sources, using various techniques. Some of the PAH-source-identification techniques that are extensively used are diagnostic ratios, a model based on positive matrix factorization (PMF) receptors, and principal component analysis-multiple linear regression (PCA-MLR) (Qu et al., 2020; Yang et al., 2021). The PAHs sources can be recognized using diagnostic ratios. This is accomplished by a relative comparison among the individual PAHs (e.g., phenanthrene (Phe), naphthalene (Nap)), and they are categorized into low molecular weight (LMW) or high molecular weight (HMW) PAHs, which leads to a qualitative classification between pyrolytic and petrogenic sources (Wang et al., 2017a). The sources affect

the ratios. For instance, LMW/HMW ratio is <1 suggests pyrogenic sources of PAHs, while ratio > 1 indicates petrogenic sources. Therefore, the LMW/HMW ratio can be useful in identification of the probable sources of PAHs (Zheng et al., 2016).

While the PAH ratios provide qualitative information, receptor models allow quantitative assessments (Zhang et al., 2012). Using statistical analysis, the PMF model is based on putting converging multiple variables into two matrices, i.e., factor characteristics/profiles and factor contributions. The interpretation of the factor matrices is carried out by the user with the help of already measured emissions or discharges and source-information inventories to understand the possible source of a sample (Wang et al., 2017a). This model, based on a point-by-point estimation of uncertainty errors in the dataset, can provide reliable results (Yang et al., 2013).

Like the PMF model, PCA-MLR is also based on multivariate analysis, which is extensively applied in source identification of PAHs in the environment. Through this model, multiple variables are combined to latent factors (factors which not directly observed), which facilitate quantitative estimation of potential sources (Duodu et al., 2017). PCA-MLR is a valuable model for investigating the complex characteristics of sources, and the one that highlights primary sources and describes their relative contribution (Zheng et al., 2016).

Various physical, chemical, and biological progressions that occur in soils and sediments disturb the molecular composition of PAHs, introducing errors in the accurate source identification of PAHs (Yang et al., 2021). Therefore, applying multiple methods is recommended to determine the precise source of PAHs. Combining results from several techniques minimizes the weaknesses of individual techniques and strengthens the overall conclusions about the sources of PAHs (Yang et al., 2013).

2.4. Interaction of PAHs in soil and sediments

PAHs undergo a number of reactions, including adsorption, volatilization, photolysis, redox reactions, and sometimes only transportation and translocation. Microbial transformation is the primary natural attenuation method of PAH-polluted sites (Haritash, 2020). The physico-chemical characteristics of PAHs govern their interactions with the environment. The most important physical and chemical characteristics of the 16 priority PAH pollutants are given in Table 1. These properties control the interactions of PAH with soil components and subsequent bioavailability and biodegradation processes. For example, the water solubility of PAHs is likely to decrease as the number of attached benzene rings increases, indicating that HMW PAHs are more gradually mobilized from solid substrates and dissolved into aquatic media than LMW PAHs and, as a result, they are less susceptible to biodegradation (Yamada et al., 2003).

Similarly, partitioning based on octanol and water (K_{ow}) is usually used to predict the affinity of organic contaminants that are retained on organic substrates (Moses et al., 2015). Higher K_{ow} of an organic compound indicates its lower biodegradability and higher bioaccumulation potency (Jonker and van der Heijden, 2007). In soils, the partition value of PAHs, normalized by soil organic matter (SOM), indicates the extent of sorption and mobility of PAHs. The higher the K_{oc} , the stronger the partition onto SOM rather than mobilization in the aqueous phase (Zhang et al., 2009). The electrochemical stability, resistance toward

biodegradation, persistency in the environmental, and carcinogenic index of PAHs magnify with an increase in number of aromatic rings and their hydrophobicity, while the volatility of PAHs is likely to decrease with increase in molecular weight (Ghosal et al., 2016).

Most PAHs are hydrophobic, which leads to high adsorption onto SOM and consequent persistency in soils and sediments (Zhang et al., 2017). The main pathways of PAHs exposure include ingestion, inhalation, and dermal contact, thereby impacting human and animal health (Abdel-Shafy and Mansour, 2016). The bioavailability of PAHs in environmental substrates, such as soils and sediments, is influenced by both the extent of adsorption and the contact period between substrates and contaminants (Lawal, 2017). The residence period in soils (i.e., aging) allows the diffusion of pollutants into soil micropores, resulting in their amalgamation into steady phases and reduction in their mobility and bioavailability (Ukalska-Jaruga and Smreczak, 2020). Bioaccumulation refers to the propensity of PAHs to become deposited in tissue of a living being resulting from exposure to a contaminated medium or by ingestion of contaminated food sources (Olayinka et al., 2019). The bioaccumulation factor (BAF), which refers to the ratio of the concentration of a pollutant in a living organism and the natural environmental, such as soils or sediments, is usually used to predict uptake, accumulation and subsequent monitoring as a hazardous to human and ecosystem health (Arnot and Gobas, 2006).

3. Bioavailability of PAHs in soil and sediments

The bioavailability of PAHs in soils represents the bioavailable fraction of PAHs, when they are in a bound state (Beckles et al., 2007). Compared with the total amount of pollutants, the bioavailable fraction of pollutants is considered as the fraction that is accumulated in the food chain. Therefore, bioavailability is a reasonable assessment in determining environmental risks of PAHs (Yang et al., 2016). Bioavailability of PAHs varies in different environments (Peng et al., 2013; Lanno et al., 2004). The bioavailability of pollutants in soil consists of three categories: environmental availability, environmental bioavailability, and toxicological bioavailability (Lanno et al., 2004). Some soil pollutants are adsorbed by soil colloids and organic matters (OMs) (Ehlers and Luthy, 2003). Some are combined with solid soil particles, hindering their contact with biological receptors. The unbound part of the pollutant can migrate and become transformed in soil, and this part is regarded as available (Lanno et al., 2004). Simultaneously, these pollutants may interact with animals, plants, and microorganisms or move with water in the environment (Ehlers and Luthy, 2003). Soil fauna will absorb bioavailable pollutants through the epidermis and gastrointestinal tract, and the absorbed pollutants may be metabolized, excreted, or deposited in various tissues or transferred to sites of toxicity (STA) (organs/tissue where toxic impact appeared) (Lanno et al., 2004). Pollutants interacting with STA are considered to have toxicological bioavailability. Hence, evaluation based on pollutants bioavailability is well thought-out approach for pollutants remediation (Zhang et al., 2021b; Yang et al., 2016).

Several biological and chemical approaches have been reported that evaluate bioavailability of PAHs in the environment, as discussed below.

3.1. Biological assays

Biological methods apply model organisms involving animals, plants, and microorganisms to measure the bioavailability of PAHs fraction in the soil (Zhang et al., 2021b).

3.1.1. Animal indication methods

Animal indication methods refer to in vivo experiments done with soil fauna. Earthworms are the most commonly used soil fauna to evaluate the bioavailability of PAHs in soil, due to their large body size, sensitivity to pollutants, and ease of monitoring (Shi et al., 2020). There are two methods to study the bioavailability of PAHs using earthworms:

the direct measurement method (Ma et al., 2012) and the indirect measurement method (Yong-Ping et al., 2013; Shi et al., 2020). Direct measurement determines the concentration of PAHs in earthworms, and it measures bioaccumulation and critical body residues (CBRs) (Hankard et al., 2004). Bioaccumulation of PAHs in earthworms is measured when the concentration of PAHs in soil is in equilibrium (Hankard et al., 2004). CBRs are associated with PAHs at lethal or semi-lethal concentrations in soil, and the total concentration of PAHs in earthworms is determined when the concentration of pollutants at the toxicity site is higher than the toxicity threshold (Saint-Denis et al., 1999). Ma et al. (2012) investigated the impacts of pyrene (Pyr) concentrations on earthworms. They found that the concentration of Pyr in earthworms increased with accumulating content of Pyr in soil. Aging of PAHs is a particular problem and must be considered in experiments dealing with soil fauna (Alexander, 2000).

The indirect measurement method is determined by the reaction of earthworms to exposure to pollutants (Shi et al., 2020). Yong-Ping et al. (2013) explored the impact of fluoranthene (Fla) on the growth of earthworms, and they found that growth inhibition of earthworms was directly proportional to the content of Fla and exposure time. There was a significant dose-effect relationship between concentration of the pollutant and mortality of earthworms (Li et al., 2015). The results indirectly reflected the effect of PAHs on the earthworms, but the bioavailability of actual pollutants could not be quantitatively estimated. Since PAHs are lipophilic in nature, therefore its bioaccumulation in living organisms is associated with the lipid contents (Honda and Suzuki, 2020). Jafarabadi et al. (2019) and Yu et al. (2019) spotted out positive correlations between fatty acid contents and PAHs concentrations in marine fishes, which imitated that PAHs bioaccumulation is highly depends upon fatty acid contents. Although, Frapiccini et al. (2018) and Soltani et al. (2019) found out very weak correlations between fatty acid content and PAHs bioaccumulation. Therefore, these studies may direct that, along with fatty acid other factors was also responsible for PAHs bioaccumulation in the marine fishes.

Commonly, compare to freshwater fishes, marine fishes are more prone to PAHs contamination due to higher concentration of PAHs in marine sediments than freshwater sediments (Honda and Suzuki, 2020). In comparison to invertebrates, fishes showed higher metabolic and excretory rate for PAHs; hence, PAH concentrations found in fishes are low (Dsikowitzky et al., 2016). Therefore, invertebrates are considered as good indicators for PAHs biomonitoring in comparison to fishes (Soltani et al., 2019; Honda et al., 2018). Animals applied in biomonitoring of PAHs in costal environment requires various specific characteristics such as its widespread distribution, easy to sampled, higher salinity tolerance capability, and higher bioaccumulation capacity (Chandurvelan et al., 2015). According to the above mention requirements, bivalves (oysters and mussels) are preferably applied as bioindicators for various PAHs and its derivatives (Idowu et al., 2020a; Jafarabadi et al., 2019; Honda and Suzuki, 2020).

3.1.2. Plant indication methods

Bioavailability of PAHs in terrestrial plants is affected by soil properties, plant species, chemical properties, and other factors (Wei et al., 2014). The plant-indication method evaluates the uptake of PAHs into plants (Chen et al., 2003). Therefore, plant indication method is effective and environmentally sustainable to evaluate the bioavailability and toxicological effects of cocktail of pollutants in the environment (Zhang et al., 2019b). In response of biotic and abiotic environmental stresses, plants acquire several complex protective mechanisms, such as disabling oxidative environmental by releasing the various osmo-protectants and antioxidant enzymes (Fuke et al., 2021; Allah et al., 2019; Georgiadou et al., 2018; Jia et al., 2020). When plants exposed to PAHs it can accumulate PAHs in their biomass, after plants death, PAHs release in the soil and consequently impose an adverse impact on soil organisms (Wei et al., 2014; Zhan et al., 2015). Wei et al. (2014) found that

phenanthrene (Phe) led to decreased wheat growth rate. Zhan et al. (2015) reported that the wheat roots absorbed and accumulate benzo(a) pyrene (BaP). The quantity of PAHs captivated by a plant is proportional to the time that the plant has been exposed to the PAHs.

More recently Zhang et al. (2021b) applied wheat plant to access the human health risk of PAHs contaminated soil obtained from chemical industry park, China. Statistical investigation between polluted soil and its toxic impacts in wheat plant obtained a R value < 0.05 for chlorophyll content, as well as for enzymes such as peroxidase and amylase activity. This study revealed that the chlorophyll content, peroxidase and amylase activity could be a suitable indicator for assessment of the toxicity and bioavailability of the contaminants. Still further studies are required to established plant indicator method as environmentally sustainable and economical method to estimate the bioavailability of environmental contaminate like PAHs.

3.1.3. Microbiological indicator methods

The microbial indicator method of bioavailability evaluates degradation of pollutants by microorganisms and the impacts that the pollutants imposed on microorganisms (Lawal, 2017; Peng et al., 2013). Bioavailability of PAHs is measured by variation in enzyme activity and mineralization of contaminants (Gong and Chen, 2010a; Márquez-Rocha et al. 2000). Márquez-Rocha et al. (2000) confirmed that mineralization rates of phenanthrene (Phe) and pyrene (Pyr) were 63% and 50%, respectively, after 21 d cultivation of white-rot fungi in the soil. Gong and Chen (2010a) found that urease and dehydrogenase activities in soil were first inhibited and then activated with Pyr concentrations higher than 50 $\mu\text{g kg}^{-1}$. The inhibition effect was due to the binding of Pyr with active sites in enzyme molecules, resulting in a competitive inhibition effect with the substrate. The later activation effect was because the secreted enzymes by microorganisms in soil adapted to the new environment, and more carbon sources could be utilized by the organism, leading to the restoration in enzyme activity.

Bezza and Chirwa (2014) examined a bacterial rhamnolipid biosurfactant produced by *Pseudomonas aeruginosa* BP9 strain and its application in improving the bioavailability and subsequent degradation of Phe. Bioavailability of Phe in aqueous medium was improved by introduction of 400 mg L^{-1} of the rhamnolipid in microcosm. Microcosm study exhibited higher degradation of Phe (92%) in surfactant assisted setup in comparison to control (27%) within 6 days with initial concentration 200 mg L^{-1} . Similarly, biosurfactant-assisted improved bioavailability and degradation of PAHs was achieved 86.5% and 57% in treated and control sample respectively, within 45 days by Bezza and Chirwa (2016). The inclusive biodegradation of PAHs was achieved after biosurfactant introduction due to improved bioavailability of PHAs to bacteria and reduced lag phase of bacterial growth (Bezza and Chirwa, 2016, 2014). Similarly, several fungal strains (Gupta and Kumar, 2020; Zhang et al., 2020) can improve bioavailability of PHAs to bacteria resulting enhance biodegradation of PAHs in microbial consortia. Due to deficiency of appropriate enzymes, usually fungal strains cannot able to entirely degrade HMW PAHs, but can transmute them into relatively more polar metabolite(s) via extracellular enzymatic mechanism, on which bacteria and others microbes can act and finalize the biodegradation PAHs (Ghosal et al., 2016).

3.2. Chemical methods

To evaluate the bioavailability and bio-accessibility of PAHs, a series of extraction methods have been applied, such as extraction with cyclodextrin (hydroxypropyl- β -cyclodextrin or HPCD) (Rufeng et al., 2016), methyl- β -cyclodextrin, Tenax (2,6-diphenyleneoxide polymer), or Amberlite XAD (polymeric adsorbent) (Nutile et al., 2020). Researchers also use co-solvents, addition of surfactants, extraction via supercritical fluids, persulphate oxidation, and thermal desorption (Maletić et al., 2019). Cui et al. (2013) concluded that, for PAHs having K_{ow} values lower than 6.0, supercritical fluid extraction, XAD-2

desorption, persulphate oxidation, and HPCD extraction were preferable methods. For PAHs having K_{ow} values higher than 6.0, these methods cannot estimate the bioavailability of PAHs precisely, but they may estimate the potentially available fraction of PAHs that affects living organisms.

3.2.1. Semi-permeable membrane devices (SPMDs)

Semi-permeable membrane devices, or SPMDs, are a passive sampling instrument applied to screen trace quantities of organic compounds. It is a biological simulation sampling method and has the advantages of relatively simple sample pre-treatment, easy storage, convenient carrying, and good reproducibility (Zhu et al., 2013). It can be used to evaluate bioavailability of pollutants in environmental media. Many studies have confirmed that SPMDs can predict the bioavailability of organic pollutants. Vinturella et al. (2004) successfully used SPMDs to simulate the absorption of PAHs by a deep-sea polychaete (annelid worm). This finding exhibited that the bioavailability of PAHs in marine sediments can be estimated via SPMDs. Sun et al. (2008) used SPMDs to analyze the desorption behavior of Phe, Pyr, and BaP in three categories of soils, and the results showed that SPMDs are an excellent way to illustrate the desorption and bioavailability of PAHs in soil. Desorption of PAHs is affected by soil characteristics and the initial concentration of the PAHs. A higher content of SOM and a higher initial concentration of PAHs result in a lower and higher desorption rate, respectively. Vrana et al. (2005) demonstrated that, after pollutants entered the SPMDs, no obvious biodegradation was found. This is another advantage for its use in biological monitoring to evaluate the effects of PAHs.

3.2.2. Solid phase microextraction (SPME)

Solid phase microextraction (SPME) was first proposed by Arthur and Pawliszyn (1990). The most widely used coating in SPME is polydimethylsiloxane (PDMS) (Beiranvand and Ghiasvand, 2020). It has good hydrophobicity and lipophilicity, stable structure, high mechanical strength, strong corrosion resistance, low environmental impact, and can be reused. In the study of Heijden and Jonker (2009), SPME predicted that the ratio of the bioaccumulation of PAHs in farmland soil to the actual bioaccumulation was close to 1. Li et al. (2006) used SPME technology to sense the content of PAHs in marine sediments. The detection of PAHs in the samples and the standard recovery test results showed that the linear correlation coefficients and recovery rates of multiple PAHs were relatively high, which indicated that, compared with traditional methods, the method has the advantages of faster analysis and use of less solvent. The results verified the feasibility of predicting the content of PAHs in sediment system with the method (Li et al., 2006). Lu et al. (2011a), (2011b) found a good correlation between the concentration of PAHs, as indicated by SPME, and the actual concentration of PAHs in organisms, which showed the possibility of using SPME for the detection of the bioavailability of PAHs.

3.2.3. TENAX extraction

The Tenax extraction method, as a biological simulation method, has been extensively applied to simulate the bioavailability and bio-accessibility of persistent organic pollutants in environmental media (Nutile et al., 2020). The main material used during Tenax extraction is poly-2,6-diphenyl-p-phenylene ether (PPPE). Because organisms can only use water-soluble OM, this technology evaluates the bioavailability of the OM by extracting part of the OM that is quickly desorbed in the environmental medium. This technology can also simulate the absorption and utilization of pollutants in the organism (Cornelissen et al., 2001). Due to the long time required for continuous extraction, many environmental researchers use two points-in-time Tenax extraction and believe that the components obtained at 6 h and 24 h can be used to characterize the rapid desorption components (Cornelissen et al., 2001). The rapid desorption component of PAHs is an indicator of bioavailability, and, as it changes, it can reflect the change of bioavailability of PAHs. A study of 10 types of PAHs in 6 kinds of sediments showed that

the extraction rate after 6 h with Tenax is about 0.5 times of its rapid desorption rate, and the rapidly desorbed part can represent the adsorbed part of PAHs (Cornelissen et al., 2001). According to Cornelissen et al. (2001), results from Tenax (6 h) are more accurate than measurement of the total concentration of PAHs, which verified the feasibility of the Tenax extraction method in estimating the bioavailability of PAHs adsorbed onto sediments.

3.2.4. Hydroxypropyl- β -cyclodextrin (HPCD) extraction

Hydroxypropyl- β -cyclodextrin (HPCD), as a mild extractant, is often applied to evaluate the bioavailability of pollutants in soils or sediments (Rufeng et al., 2016). It has a nanoscale cavity with a hydrophilic exterior and a hydrophobic interior, and the molecule can be used to contain pollutants selectively and to generate an inclusion compound of an aqueous solution, which increases the solubility and stability of the pollutants (Rufeng et al., 2016). Reid et al. (2000) demonstrated that HPCD extraction predicted the bioavailability of PAHs. Gao et al. (2014) used HPCD to extract five categories of PAHs from the soil. The results exhibited that the addition of HPCD improved the solubility of PAHs in the water phase. In soil contaminated with PAHs, the amount extracted by HPCD of five PAHs was linearly related to the amount of biodegradation, indicating that the HPCD extraction method can predict bioavailability of PAHs (Zhang et al., 2010).

Reid et al. (2000) demonstrated that the content of Phe in soil extracted by HPCD is approximately equal to the content of Phe degraded by viable bacteria. Adding HPCD can increase the solubility of Pyr, Phe, and BaP by nearly 6.25 times. Therefore, the cyclodextrin solution can be applied to extract PAHs in polluted soil, and the content of desorbed PAHs can be utilized to predict the bioavailability of the PAHs (Reid et al., 1998). Research on soil contaminated by PAHs for many years has shown that the concentration of tricyclic PAHs extracted

using HPCD and the PAHs accumulated in earthworms have a reasonable correlation ($R^2 = 0.77-0.79$). Therefore, HPCD may be applied to sense the bioavailability of PAHs in soil that has been contaminated for a long time (Jiang et al., 2002). Moreover, a single method either biological or chemical would not be good enough to estimate the bioavailability of PAHs. Therefore, coupling of biological methods with chemical methods and vice-versa could be a best way to evaluate bioavailability of PHAs (Guo et al., 2017).

4. Remediation of PAH in soils and sediments

Information on techniques used to remediate soils and sediments contaminated with PAHs is critical to limit environmental and living organism health risks (Guarino et al., 2019; Gitipour et al., 2018). Various methodologies have been adopted to remediate PAHs in soils and sediments (Fig. 3). They can be immobilized, mobilized, and degraded from a solid medium (e.g., soils, sediments, or wastes) via selective chemicals or materials, or their concentration can be decreased by plant uptake and by chemical or microbial degradation (Kramer et al., 2019; Cui et al., 2020) (Table 3).

4.1. Immobilization based remediation techniques for PAHs

To check the leaching of the PAH compounds from contaminated soils and sediments various remediation approaches have been adopted such as capping (Maletić et al., 2019), stabilization and solidification, (Walker, 2014; Wang et al., 2018), and dredging and excavation (Walker et al., 2013) which have been discussed in details below.

4.1.1. Capping

Capping immobilizes and reduces the discharge of pollutants into the

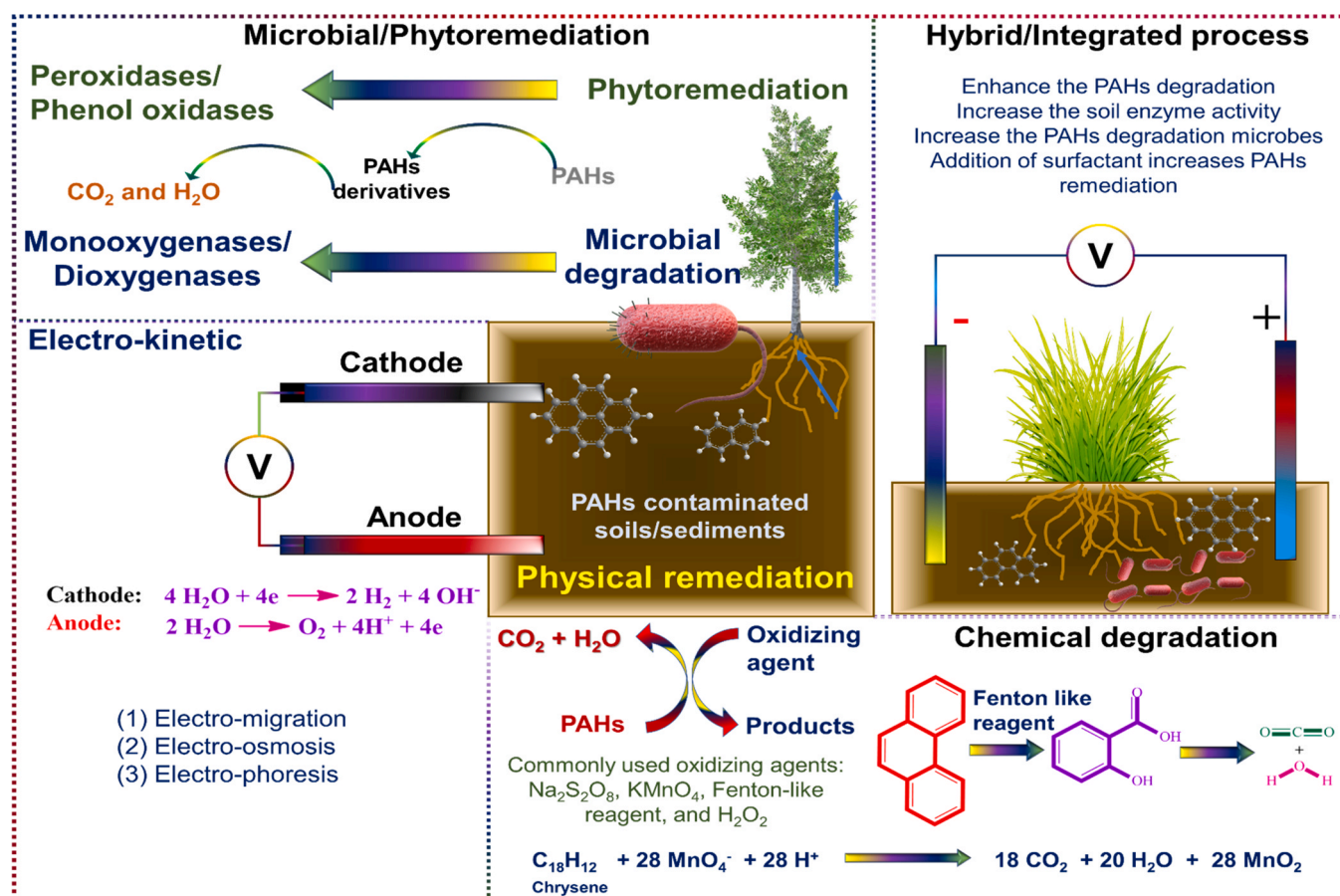


Fig. 3. : Schematic representation of various technologies used to remediate PAHs.

Table 3
Approaches for the remediation of PAHs in soils and sediments.

Physical approach						
PAHs	PAHs concentration (mg kg ⁻¹)	Methods	Soil/Sediment	Duration (days)	% removal	References
Nap, Flu, Flt, Pyr, BbF, BkF, BaP, IcdP, BghiP	11,600	Soil washing/solvent extraction	1 g	1	96.30	(Khodadoust et al., 2000)
Nap, Phe, Flt, Pyr, BaA, Chr, BbF, BkF, BaP, BghiP, IcdP	1000	Soil venting thermal desorption	70 kg	35	90.00	(Hosseini, 2006)
BaP	30.6	Thermal desorption	29,800,000 kg	130	99.90	(Baker et al., 2007)
Nap, Ace, Acy, Flu, Phe, Ant, Flt, Pyr, BaA, Chr, BbF	1000	Thermal desorption and incineration	142,000,000 kg	480	90.00	(Acharya and Ives, 1994)
Chemical degradation approach						
PAH	Soil/Sediment	Oxidizing agent (OA)	Dosage of OA	Soil/sediment Properties	Degradation efficiency	References
16 PAHs Nap, Acn, Acp, Flu Phe, Ant, Flo Pyr, Ban, Chr, Bbn Bkn, Bke, Dib, Inc, Bgh	Old coking plant soil	Fenton-like reagent	14.5 mL/5 g soil and 500 mg magnetite	TOC: 6.5% EOM:	7.03%	(Biache et al., 2015)
		H ₂ O ₂ (30%)	14.5 mL/5 g soil	10.3 mg/g	5.32%	
		KMnO ₄	100 mL (16.7 g/L)/ 5 g soil	Total PAHs: 1.127 mg/g	56.34%	
	Soil near old gas plant located	Fenton-like reagent	14.5 mL/5 g soil and 500 mg magnetite	TOC: 3.3% EOM:	80.82%	(Biache et al., 2015)
		H ₂ O ₂	14.5 mL/5 g soil	11.5 mg/g	90.32%	
		KMnO ₄	100 mL (16.7 g/L)/ 5 g soil	Total PAHs: 1.757 mg/g	88.73%	
Soil at near active wood treating plant	Fenton-like reagent	14.5 mL/5 g soil and 500 mg magnetite	TOC: 17.2% EOM:	47.32%	(Biache et al., 2015)	
	H ₂ O ₂	14.5 mL/5 g soil	81.94 mg/g	52.22%		
	KMnO ₄	100 mL (16.7 g/L)/ 5 g soil	Total PAHs: 11.962 mg/g	58.70%		
2 PAHs Phe Py	Aged contaminated soil	Na ₂ S ₂ O ₈ /24 °C	267 g/kg	pH: 6.6	*Phe: 0%	(Peluffo et al., 2018)
		Na ₂ S ₂ O ₈ /EDTA/FeSO ₄	(267/16.2/14.6 g)/kg	OC: 4.67%	*Py: 62.8%	
		Na ₂ S ₂ O ₈ /Buffer	267 g/kg with phosphate buffer	SOM: 8.05%	*Phe: 6.3%	
		Na ₂ S ₂ O ₈ /NaOH	267 g/kg (pH = 10)	TN: 3.89 mg/kg	*Py: 48%	
		Na ₂ S ₂ O ₈ /65 °C	267 g/kg	EC: 3.3 dS/m	*Phe: 33.3%	
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Py, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Soil from coking plant field	Fenton reagent	(H ₂ O ₂ 30%) 1 mmol/g (FeSO ₄ 0.5 M) 0.2 mmol/g	pH: 7.57 TOC: 2.29%		(Liao et al., 2019)
		Modified Fenton reagent	(H ₂ O ₂ 30%) 2 mmol/g (FeSO ₄ 0.5 M) 0.4 mmol/g	Total PAHs: 385.2 mg/kg	92.5%	
		KMnO ₄	(0.4 M) 1.5 mmol/g		95.4%	
		Activated Na ₂ S ₂ O ₈	(Na ₂ S ₂ O ₈ 1 M) 2 mmol/g FeSO ₄ (0.5 M) 0.2 mmol/g Citric acid: 0.04 mmol/g		93.6%	
		Fenton reagent	(H ₂ O ₂ 30%) 1 mmol/g (FeSO ₄ 0.5 M) 0.2 mmol/g	pH: 7.41 TOC: 0.27	74.2%	
		Modified Fenton reagent	(H ₂ O ₂ 30%) 2 mmol/g (FeSO ₄ 0.5 M) 0.4 mmol/g Citric acid 0.4 mmol/g	Total PAHs: 263.6 mg/kg	94.5%	
	Soil from coal gas field	Fenton reagent	(H ₂ O ₂ 30%) 1 mmol/g (FeSO ₄ 0.5 M) 0.2 mmol/g	pH: 7.41 TOC: 0.27	74.2%	(Liao et al., 2019)
		Modified Fenton reagent	(H ₂ O ₂ 30%) 2 mmol/g (FeSO ₄ 0.5 M) 0.4 mmol/g Citric acid 0.4 mmol/g	Total PAHs: 263.6 mg/kg	94.5%	
		KMnO ₄	(0.4 M) 1.5 mmol/g		97.9%	
		Activated Na ₂ S ₂ O ₈	(Na ₂ S ₂ O ₈ 1 M) 2 mmol/g FeSO ₄ (0.5 M) 0.2 mmol/g Citric acid: 0.04 mmol/g		95.6%	
		Fenton-like reagent	(Low dose) H ₂ O ₂ : 1200 mL (0.975 w %) and magnetite (6 g)/ 60 g soil (High dose) H ₂ O ₂ : 1200 mL (4.35 w %) and magnetite (6 g)/ 60 g soil	TOC: 33 g/kg EOC: 11.9 mg/g	28.8% 79.9%	
		KMnO ₄	(Low dose) 3.34 g/L (1200 mL) (High dose) 16.7 g/L (1200 mL)	Total PAHs: 1.21 mg/g	37.4% 83.7%	
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Py, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Soil from gas plant	Fenton-like reagent	(Low dose) H ₂ O ₂ : 1200 mL (0.975 w %) and magnetite (6 g)/ 60 g soil (High dose) H ₂ O ₂ : 1200 mL (4.35 w %) and magnetite (6 g)/ 60 g soil	TOC: 65 g/kg EOC: 10.7 mg/g	7% 14.3%	(Boulangé et al., 2019)
		KMnO ₄	(Low dose) 3.34 g/L (1200 mL) (High dose) 16.7 g/L (1200 mL)	Total PAHs: 1.136 mg/g	41.2% 58.2%	
	Soil from coking plant	Fenton-like reagent	(Low dose) H ₂ O ₂ : 1200 mL (0.975 w %) and magnetite (6 g)/ 60 g soil (High dose) H ₂ O ₂ : 1200 mL (4.35 w %) and magnetite (6 g)/ 60 g soil	TOC: 65 g/kg EOC: 10.7 mg/g	7% 14.3%	
		KMnO ₄	(Low dose) 3.34 g/L (1200 mL) (High dose) 16.7 g/L (1200 mL)	Total PAHs: 1.136 mg/g	41.2% 58.2%	
Electrokinetic approach						

(continued on next page)

Table 3 (continued)

Physical approach						
PAHs	PAHs concentration (mg kg ⁻¹)	Methods	Soil/Sediment	Duration (days)	% removal	References
PAHs	Soil/Sediment	Electrodes (voltage gradient)	Electrolytes (cathode/anode)	Soil/sediment properties	Removal efficiency	References
Ban, Bbn, Bkn, Bke, Dib, Inc	Coal gas plant soil	Ti/RuO ₂ alloy sheet (1 V/cm)	NaCl (0.01 M)/ NaCl (0.01 M) NaCl (0.01 M)/ NaCl (0.01 M) and Na ₂ S ₂ O ₈ (20%) NaCl (0.01 M, pH: 4)/ NaCl (0.01 M) and Na ₂ S ₂ O ₈ (20%) NaCl (0.01 M)/ NaCl (0.01 M) and Na ₂ S ₂ O ₈ (20%)	pH: 9.1 EC: 800 µS/cm Total PAHs: 4.88 mg/kg	18.4% 33.1% 93.1%	(Xu et al., 2020)
Pyr	Artificially contaminated soil	Ti/RuO ₂ alloy sheet with cation exchange membrane (1 V/cm) Graphite (1 V/cm)	–	pH: 7.64 EC: 245 µS/cm OM: 58.6 g/kg CEC: 15.9 cmol/kg Total PAHs: 102.4 mg/kg	52–56%	(Lu, 2020)
Phe	Contaminated laboratory synthetic soil	Graphite (1.5 V/cm)	Distilled water	CEC: 20.33 mq PAHs: 1000 mg/kg	40–42%	(Adhami et al., 2020)
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Pyr, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Sediment	Graphite sheet (1 V/cm)	Rhamnolipids (1.1 g/L) and citric acid (0.2 M) in deionized water Saponin (0.85 g/L) and citric acid (0.2 M) in deionized water Saponin (0.85 g/L) and citric acid (0.1 M) in deionized water	pH: 8.3 EC: 2200 µS/cm OM: 11% Total PAHs: 2.114 mg/kg	17.4% 29.2% 13.1%	(Tian et al., 2017)
Nap Pyr Ban	Marine sediment	Stainless steel (1.2 V/cm)	EDTA (0.1 M)/ methylglycinediacetic acid: tween 80 (1: 1) 5%	pH: 8.1 OM: 7.6% Total PAHs: 55 mg/kg	59.45%	(Falciglia et al., 2017)
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Pyr, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Marine sediment	Graphite plate (1 V/cm)	Citric acid (0.1 M)/tween 20 (4.94 g/L) Citric acid (0.5 M)/tween 20 (4.94 g/L) Citric acid (1 M)/tween 20 (4.94 g/L) Citric acid (0.1 M)/rhamnolipid (0.275 g/L) Citric acid (0.1 M)/vicosin-like (0.25 g/L)	pH: 8.4 EC: 1.82 mS/cm OM: 2.6% Total PAHs: 4.044 mg/kg	35.3% 43.1% 54.4% 24.7%	(Ammami et al., 2015)
Pyr	Red soil	Ti/RuO ₂ /IrO ₂ alloy (2 V/cm)	NaNO ₃ (0.1 M) and KMnO ₄ (9 g/L)/ NaNO ₃ (0.1 M)	pH: 4.8 EC: 28 µS/cm CEC: 24.8 cmol/kg SOM: 3.75 g/kg Total PAHs: 485 and 475 mg/kg	70.7%	(Fan et al., 2021)
Phytoremediation and microbial remediation						
PAHs	Matrixes	Matrixes properties	Remediator	Time	Efficiency	References
8 PAHs Flo, Py, Ban, Chr, Bbn, Bkn, Bke, Dib	Soil collected at oil field	pH: 7.66 C: 45.77 g/kg P:0.65 g/kg N:0.73 g/kg Total PAHs: 34.17–122.46 mg/kg	<i>Echinacea purpurea</i> (purple coneflower) <i>Callistephus chinensis</i> (Aster Callistephus) <i>Festuca arundinacea</i> (fawn) (Fire Phoenix) <i>Medicago sativa</i> (alfalfa) (Fire phoenix) mixture of <i>Festuca arundinacea</i> , <i>Festuca elata</i> , and <i>Festuca gigantea</i>	120 days	92.41% 84.09% 87.30% 96.18% 94.21% 64.57%	(Xiao et al., 2015)
4 PAHs Pyr, Chr, Bbn, Bkn	Soil collected at oil field (low contamination)	pH: 7.64 C: 7.13% P: 0.16% N: 0.37% Total PAHs: ≈150 mg/kg		150 days	68.29%	(Dai et al., 2020)
	Soil collected at oil field (high contamination)	pH: 7.23 C: 16.83% P: 0.26% N: 0.82% Total PAHs: ≈200 mg/kg				
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Pyr, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Aged polluted soil at agricultural field	pH: 7.5 CEC: 7.6 mmol/g C: 36.7 g/kg N: 2.9 g/kg Total PAHs: 1.132 mg/kg	<i>Zea mays</i> (maize) <i>Crucibulum laeve</i>	180 days	45.3% 48%	(García-Sánchez et al., 2018)

(continued on next page)

Table 3 (continued)

Physical approach						
PAHs	PAHs concentration (mg kg ⁻¹)	Methods	Soil/Sediment	Duration (days)	% removal	References
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Pyr, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Mangrove sediment contaminated with crude oil	pH:4.9 OM: 5% DO: 5.7 mg/L Total PAHs: 10540.33 ng/g	<i>Rhizophora mangle</i> (red mangrove)	90 days	60.76%	(Veràne et al., 2020)
8 PAHs Flo, Py, Ban, Chr, Bbn, Bkn, Bke, Dib	Soil from oil field	pH: 7.66 C: 45.77 g/kg P:0.65 g/kg N:0.73 g/kg Total PAHs: 228–398 mg/kg (diluted to 70.8–79.81 mg/kg)	<i>Festuca arundinacea</i> , <i>Festuca elata</i> , and <i>Festuca gigantea</i>	120 days	96.18%	(Liu et al., 2014)
16 PAHs Nap, Acn, Acp, Flu, Phe, Ant, Flo, Pyr, Ban, Chr, Bbn, Bkn, Bke, Dib, Inc, Bgh	Aged contaminated yellow-brown soil	pH: 7.21 N: 1254 mg/kg C: 43.2 g/kg P: 860 mg/kg Total PAHs: 97.63 mg/kg	Bacterial community, including <i>Methylobacillus</i> , <i>Caulobacter</i> , <i>Achromobacter</i> , <i>Pseudomonas</i> , and <i>Pseudoxanthomonas</i>	35 days	75.42%	(Lu et al., 2019)

natural environment and is well-thought-out to be an economical physical remediation method (Maletić et al., 2019; Lamb et al., 2014). The capping material can be sand or sediment (passive capping), or it may be a carbonaceous material that is added to immobilize the contaminants (Silvani et al., 2017). Compared to dredging, capping is cost-effective, but requires continuous monitoring (Silvani et al., 2017), and imposes a possible risk because contaminants are left out in the open at certain places (Maletić et al., 2019). Based on the condition and level of pollutants in sediments, a cap is fabricated to minimize the risk posed through leaching of the contaminants (US EPA, 2005). Capping can minimize contact of organisms to pollutants and has the potential to immobilize pollutants for a longer duration. Also, this method does not necessitate an expensive set-up. Nevertheless, it has shortfalls. The pollutants remain in the environment, and capping constituents may change the biotic community. Consequently, capped sites require continuous monitoring (Reis et al., 2007). Assessment of contaminated sites, along with an understanding about capping materials and their layering, is crucial in estimating the viability and efficiency of sites that are capped (US EPA, 2005).

Generally, capping is done with various carbonaceous materials like activated carbon (AC) (Janssen and Beckingham, 2013), biochar (Silvani et al., 2017), apatite, zeolites, organoclay, and nano-zerovalent iron (nZVI) (Lofrano et al., 2017; Patmont et al., 2015; US EPA, 2013). Amendments improve contaminant binding ability and stability to minimize their release in the surrounding environment. The preferable carbonaceous material used in capping is AC (Janssen and Beckingham, 2013). Basically, AC is charcoal, which is fabricated in oxygen to improve its specific surface area and micro-porosity (Maletić et al., 2019). The application of biochar is gaining attention these days as capping material. Biochar is also a carbonaceous material, synthesized via pyrolysis, and has a medium-to-large surface area, and it is considered to be cost-effective (Kumar et al., 2021; Bolan et al., 2021; Ahmad et al., 2014). Various investigations have established that biochar is a greener sorbent than others and has diverse environmental applications (Bolan et al., 2021; Kumar et al., 2020b).

4.1.2. Stabilization and solidification

The solidification and stabilization methods are applied to check the migration of pollutants from polluted media via addition of cementing materials and/or various chemicals (Maletić et al., 2019; Bolan et al., 2014). Stabilization is a process in which chemicals are used to minimize the leachability of a contaminated material and, simultaneously, it reduces the solubility of the contaminants. It makes them less detrimental, because they are immobilized. Solidification is a method that binds a

polluted media with the help of reagent. The physical characteristics of the media are changed by improving its strength, reducing its permeability, and facilitating immobilization of the pollutants (US EPA, 2009; Walker, 2014). For solidification, the cement-based method has been primarily applied in the immobilization of environmental contaminants (Wang et al., 2018). But it is used along with other materials such as organophilic clay, phosphate, soluble silicate, AC, fly ash, and lime. Wang et al. (2018) applied innovative and green technology to remove contaminated sediment via calcium-rich materials obtained from industry by-products. The key parameters that influence solidification include water/binder ratio, pH, moisture, and temperature. (Lofrano et al., 2017). Comprehensive information about immobilization of contaminants by stabilization or solidification at polluted sites has been provided by the US EPA (2009).

4.1.3. Dredging and excavation

Dredging and excavation are physical remediation methods that apply for the removal of contaminants from submerged or drained conditions. These methods typically required the transport of sediments from their original place to where they are treated and disposed. (Megharaj et al., 2011; US EPA, 2005). This technology permanently remediates the pollutants in the sediment, but it is not considered cost-effective (Maletić et al., 2019). The method requires a substantial amount of money for transportation. Another constraint of this method is the leaching of other contaminants from disposed sediments (EPA, 2005; Patmont et al., 2018). Walker et al. (2013) discussed economical sediment dredging, discarding, and disposal opportunities for sites in Canada. They recommended various approaches to manage the polluted sites and innovative disposal methods of contaminants, along with how to improve the economy of the processes. The US EPA also issued a report about the advantages, disadvantages, and application of excavation and dredging methods (US EPA, 2005). This report describes how, after extracting the sediment and immobilizing the contaminants, the sediment can be further exposed to treatment and finally disposed of or reutilized with proper monitoring. If it is not possible to further use the treated sediments, then disposal of them in upland sanitary dumping site is recommended. In few cases, polluted sediments can be dumped without pre-treatment or treatment, but, in those cases, an advanced management approach to monitor the landfill site is necessary (US EPA, 2005).

4.2. Mobilization based remediation techniques for PAHs

Mobilization based approaches generally applied to remediate the

PAHs from soils and sediments and transform PAHs and its derivatives in to more soluble and volatile form via heat treatment (Song et al., 2019), washing (Maletić et al., 2019), surfactant treatment (Dell'Anno et al., 2018), and electrokinetic (Adhami et al., 2020).

4.2.1. Thermal desorption

Thermal desorption (TD) or conductive heating is one of the effective methods applied to mobilize, and subsequently degrade, PAHs present in polluted soils (Gan et al., 2009). Incineration degrades PAHs in the polluted plume at high temperatures (900–1200 °C) (Chen et al., 2013). In 1986, U.S. Environmental Protection Agency performed a remediation study of a superfund site situated in Louisiana that was severely polluted with creosote (Acharya and Ives, 1994). Remediation involved excavation along with incineration of polluted solids (soil + sediment + waste piles). Incineration demolished almost 90% of the contaminants within 40 months and was shown to be an effective method.

Like incineration, a novel pyrolytic PAHs remediation technology has been developed by Gao et al. (2020). This method can hastily and consistently remediate PAHs from soil with lower energy consumptions and improved post-treatment fertility of soil in comparison to other thermal treatment methods (Vidonish et al., 2016, 2018; Song et al., 2019). For instance, pyrolytic remediation of PAHs contaminated soils at 420 °C for 15-min in a continuous batch reactor diminished the total PAH level by nearly 95%, and re-established fertility of soil by 98% in comparison to unpolluted soil (Song et al., 2019). Results of this finding undoubtedly established that pyrolysis treatment has the capacity to enhanced ecosystem rebuilding along with remediation of PAHs contaminated soils in comparison to other conventional thermal treatment methods (Gao et al., 2020). Like incineration and pyrolysis, TD also utilizes energy to isolate PAHs physically from soils and sediments, but it is relatively safer and releases no, or at least fewer, PAHs to the surroundings (Kuppusamy et al., 2016). This is due to applying a carrier gas and vacuum system in TD, which separates the volatilized fraction of PAHs from the system for offsite dumping. Amongst several PAHs remediation technologies for soils, TD can be best suited technologies due to its comparatively shorter duration (Choi et al., 2020).

Hosseini (2006) performed a pilot-scale demonstration to show the practicability of a TD operation for remediating soils that contained PAHs along with lampblack residues. TD mobilized PAHs and simultaneously brought down the concentration at moderate temperatures (250–300 °C) within 35 days. Similarly, Renoldi et al. (2003) performed a lab-scale demonstration to remediate the PAHs (2700 mg/kg) contaminated soil via TD at temperature ranging from 300° to 350 °C. The experimental finding revealed that 99.7% PAHs remediation can be achievable via TD at 300 °C, which showed the feasibility of TD at minimum temperature. Recently, TD is gaining attention because of its several advantages such as appropriateness to diverse range of environmental contaminants, shorter duration, high competence, and potentiality to recover soil and pollutants (Choi et al., 2020; Zhao et al., 2017c). However, owing to the heterogeneous nature and varying moisture content in the soils and sediments, the TD method posed a major challenge when it was applied at the pilot and field-scale, which needs further consideration in future demonstration work.

4.2.2. Washing

This technology is considered economical (Fig. 4) and feasible to remediate soils and sediments polluted with HMW PAHs that are not quickly remediated owing to their higher hydrophobicity, lower bioavailability, and slower rate of desorption (Gong et al., 2010; Maletić et al., 2019). The mechanism is built on PAHs desorption from the solid matrix via the action of water, organics, surface-active agents, complexing agents, plant oils, or humic acids (HAs). Afterward, the PAHs are eluted from the sample (Gan et al., 2009; Mousset et al., 2014). The eluted phase is further disposed of or treated with biochemicals or chemicals for absolute detoxification. Conte et al. (2005) reported that HAs were a better agent than water or surfactants, such as sodium

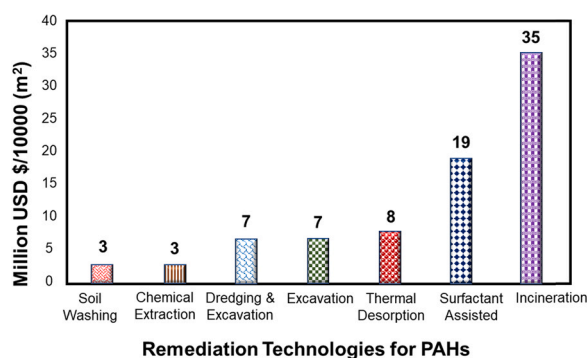


Fig. 4. Representation of cost involved in various remediation technologies for PAHs (adapted and modified from US EPA, 2000).

dodecylsulphate (SDS) and triton X100 (TX100), for washing soil having a high concentration of PAHs. Additionally, HAs facilitate growth of the natural microbial community, which further helps in the degradation of bioavailable PAHs present in washed soils. Moreover, fatty acid methyl ester (FAME) with biodiesel were more effective in the remediation of PAHs than their parent constituents (methanol and lipid) and other surfactants, such as cyclodextrins (CD), HPCD, and TX100 (Gong et al., 2010). The authors recommended that a petro-plant (biodiesel/lipids producing plants species) could be a potential candidate to remediate soils highly contaminated with PAHs. Moreover, vegetable fats can offer several options in the remediation of PAHs. They can be used as a green solvent for extraction of PAHs or as a soil amendment to increase microbial activity (Kuppusamy et al., 2017).

4.2.3. Surfactant-assisted remediation

Surfactants can improve the solubility of PAHs in the water and simultaneously enhance their bioavailability (Li and Chen, 2009; Liang et al., 2017). Surfactants are natural and synthetic organic and may be grouped as cationic, anionic, non-ionic, or zwitterionic according to the ionic charge present on their hydrophilic groups (Lamichhane et al., 2017). When the aqueous concentration of a surfactant surpasses its own critical micelle concentration (CMC), formation of micelles occurs. These micelles act as a hydrocarbon-like phase and increase the separation of PAHs into aquatic media (Shah et al., 2016). The use of surfactants at sites contaminated with PAHs is considered a double-edged sword. They enhance the solubility and dissolution, but simultaneously, they reduce the bioavailability of PAHs and are toxic to endogenous microbes (Li and Chen, 2009). Dell'Anno et al. (2018) provided comprehensive information about applying biosurfactants in removing PAHs from contaminated sediments. In a study of biodegradation induced by surfactants, Tsomides et al. (1995) used seven different non-ionic surfactants and showed that only TX100 induced the mineralization of Phe above the CMC. The results indicated that several surfactants are lethal to microbes that degrade PAHs and the application of surfactants may not be necessary to accomplish complete pollutant remediation.

The effectiveness of the treatment process is governed by various factors, such as surfactant concentration, its hydrophilic-lipophilic equilibrium, the K_{ow} of the PAHs, temperature, pH, dissolved organic matter (DOM), salinity, and co-solutes (Lamichhane et al., 2017). Only a few field trials have been performed using biosurfactants to remediate PAHs due to the high cost. Consideration of the bioavailability of PAHs by applying surfactants, their physicochemical and biological interaction, and their economical production is essential for use of surfactants in the field (Bezza and Chirwa, 2016). In some studies, treatment with surfactants is coupled with the electro-Fenton method (Iglesias et al., 2014) or the electrokinetic method (Falciglia et al., 2017). The electrokinetic method is discussed below.

4.2.4. Electrokinetic remediation

Electrokinetic remediation is an in-situ method that influences the movement of contaminants (Asadollahfardi et al., 2018; Istrate et al., 2018). Alterations are essential to enable the electrokinetic process to remove contaminants with inadequate solubility. Co-solvents, cyclodextrins, and surfactants are the usually applied solubilizing agents in electro-remediation, and they enhance the desorption of PAHs. Various types of electrodes are used, including a Ti/RuO₂ alloy sheet (Xu et al., 2020), graphite (Adhami et al., 2020; Lu, 2020), and a Ti/RuO₂/IrO₂ alloy (Fan et al., 2021), along with different electrolytes (Table 3). Commonly, water hydrolysis generates H⁺ at the anode and OH⁻ at the cathode, leading to PAH degradation (Adhami et al., 2020; Falciglia et al., 2017). However, the degradation efficiency of PAHs is enhanced with the addition of chelators, such as citric acid and ethylenediaminetetraacetic acid (EDTA), as well as Tween 20 and oxidizing agents (Table 3). This technique is effective for saturated sediments having high clay or OM contents and small capillary pores. In compressed sediments having an overlapped diffuse dual layer, electroosmosis technology requires adequate ionic strength to maintain the flow of electrons (Gan et al., 2009).

4.3. Degradation of PAHs

Removal and adsorption methods of environmental contaminants only can transfer pollutants from one media to another media. Therefore, degradation or mineralization of organic pollutants are considered as safe and effective approach (Kumar et al., 2020a). Remediation of PAHs contaminated soils and sediments are gaining attention these days, which have been discussed below compressively.

4.3.1. Chemical degradation

Chemical degradation is well-known to have beneficial outcomes on the remediation of HMW PAHs fraction, which are hard to eliminate from soils and sediments. The practice can be performed via two methods: conventional and advanced (Gitipour et al., 2018). Conventional methods apply hydrogen peroxide (H₂O₂), ozone (O₃), and permanganate. In the advanced oxidation process (AOP), two or more chemicals are used to generate extremely reactive free radicals (Cheng et al., 2016). Among several oxidants used for soil remediation, O₃ (gas) and Fenton's reagent (liquid) are generally acceptable and effective (Kramer et al., 2019; Zhao et al., 2019). Also, the formation of secondary compounds during this process, their leachability, and environmental toxicities need to be considered. Most studies have used the Fenton reagent with and without modification. The Fenton oxidizing agent exhibited 7–94.5% removal of PAHs from soils and sediments, whereas potassium permanganate exhibited 37.4–97.9% removal (Table 3). Sodium persulfate with and without modification showed 6.3–95.6% removal efficiency, and that for hydrogen peroxide was 5.32–90.32% (Table 3). Generally, the PAHs are oxidized into carbon dioxide and water by the oxidizing agents, and this takes place via a fission reaction at the PAHs. Kanel et al. (2004) said that oxidation of Phe by H₂O₂ occurred via salicylic acid formation and ended with mineralization.

4.3.2. Phytoremediation

Phytoremediation is an in-situ remediation process that does not interrupt the soil profile. This approach is economical and a green way to remediate sites polluted with PAHs (Arora, 2018). Phytoremediators (plants that take up pollutants) exist in most environmental settings, and they are beneficial for phytoremediation (Fuke et al., 2021; Guarino et al., 2019; Gitipour et al., 2018). Generally, once the pollutants are taken up by plants, enzymatic degradation breaks the PAHs into simple molecules (CO₂ and H₂O) (Fig. 3) (Turkovskaya and Muratova, 2019). Numerous studies have applied phytoremediation and obtained high removals of PAHs (Table 3). To increase phytoremediation of soils contaminated with PAHs, Sun et al. (2011) recommended intercropping with alfalfa (*Medicago sativa*) and tall fescue. Intercropping may improve

the microbial community diversity resulting in better phytoremediation efficiency. Their 7-month field trial showed that, via intercropping, 31% of PAHs have been removed from contaminated soil, significantly better than monoculture and unplanted soils which was 20% and 0%, respectively. Meng et al. (2011) also said that some other plant species, when intercropped, such as ryegrass (*Lolium* sp.), white clover (*Trifolium repens*), and celery (*Apium graveolens*), enable better remediation of soils polluted with PAHs in comparison to monocultures. Moreover, the key route for phytoremediation of PAHs is plant-induced biodegradation in the soil, while removal via plant uptake is minimal (Sun and Zhou, 2016).

A way to increase the efficiency of phytoremediation is to apply plant growth-promoting rhizobacteria (PGPR), which usually boost the capacity of plants to tolerate adverse environments (Fuke et al., 2021; de Boer and Wagelmans, 2016). The PGPR induce plant root growth that results in improved water absorption by the plant. Also, the PGPR facilitate N-fixation, production of siderophores (a siderophore is a molecule that binds and transports iron in microorganisms) and phytohormones, enhance the availability of mineral elements, and improve soil fertility. Huang et al. (2004a), (2004b) established that PGPR (*Pseudomonas putida*, *Azospirillum brasilense*, and *Enterobacter cloacae*) enhanced plant growth, specifically that of roots, in profoundly polluted soils, reducing the lethal impacts of PAHs on plant growth. The improved root biomass of wild rye (*Elymus canadensis*) in PGPR-amended soil resulted in increased removal of PAHs up to 90%. Teng et al. (2011) saw a synergistic relation between alfalfa and *Rhizobium* that increased growth of the rhizospheric microfloral community, resulting in the degradation of PAHs of up to 51% within 90 days.

4.3.3. Microbial degradation

Microbial degradation is a process used to remediate polluted media, including water, soils, and sediments, by optimizing environmental parameters to stimulate the growth of microbes, thereby degrading targeted pollutants (Patel et al., 2020; Lu et al., 2011a, 2011b). Bioremediation of a contaminant can be achieved directly through enzymatic reactions by stimulating and bioaugmenting the microorganism (Sun et al., 2020; Kong et al., 2018) or indirectly through non-enzymatic reactions, which result in microbially-induced changes in soil properties (Patel et al., 2020). The enzymatic degradation of PAHs is mediated mainly by monooxygenase and dioxygenase enzymes by various types of bacteria that degrade PAHs (Ghosal et al., 2016; Dhar et al., 2020), as well as by the manganese peroxidase enzyme that is produced by ligninolytic fungi, which are commonly found in the environmental matrixes (Aydin et al., 2017). The degradation of PAHs by *Stenotrophomonas* sp. was investigated in a culture medium, and the results showed that the bacteria degraded 48.4% of the PAHs (Zang et al., 2020). Similarly, 60% of PAHs were degraded by *Pseudomonas mendocina* and *Brevundimonas olei* isolated from creosote oil (Smulek et al., 2020). The fungus *Cladosporium* sp. was shown to remediate PAHs (Birolli et al., 2018). Lu et al. (2019) stated that the degradation of LMW PAHs was mediated by strains of *Pseudoxanthomonas*, *Sphingomonas*, *Pseudomonas*, *Pseudorhodofera*, and *Sphingobium*, whereas degradation of HMW PAHs was mediated by *Nocardioidea* and *Methylophaga*.

The actual implementation of microbial degradation is inadequate by the ability of microbes to degrade PAHs, and the process is slow and unpredictable (Patel et al., 2020). Several approaches fall under microbial degradation, like biostimulation and bioaugmentation (Maletić et al., 2019). Bamfort and Singleton (2005) discussed the role of various fungi and bacteria, which showed the capability to degrade PAHs via aerobic and anaerobic degradation. They highlighted the few microbial degradation methods that are presently useable. Microbial break down of PAHs leads to the generation of the simple molecules of CO₂ and H₂O (Fig. 3). This process is called natural attenuation (Megharaj et al., 2011). The time needed for natural attenuation depends on the environmental condition of the contaminated site as well as the microbes present. If contaminated sites do not need a fast, destructive approach,

natural attenuation is an effective and economical approach. When exogenous microbes with the ability to degrade PAHs are added to a site contaminated with PAHs, that process is called bioaugmentation (Kong et al., 2018). When indigenous microbes are not competent enough to degrade contaminants, investigators have induced microbial action with the supplementation of various nutrients in the form of fertilizers, such as KNO_3 , NaNO_3 , NH_3NO_3 , K_2HPO_4 , and MgNH_4PO_4 (Megharaj et al., 2011). This amendment process is called biostimulation (Haleyur et al., 2019).

Sivaram et al. (2019) described that, biostimulation via a low molecular weight organic acid (LMWOA) mixture, composed of succinic, oxalic, citric, and malic acid, encouraged the rhizospheric microbial degradation of BaP. More recently, Zhang et al. (2021a) thoroughly analyzed the significant roles of LMWOAs in the rhizospheric microbial degradation of Phe using potential bacteria strain *B. subtilis* ZL09–26 and their mechanisms. This finding exhibited that biostimulation using LMWOAs can enhance the solubility and degradation of Phe, improve bacterial cell biomass and degradation potency, and provide protection from environmental stress. Citric acid boosted Phe degradation primarily by inducing the biomass growth and action, whereas glutaric and oxalic acid enhanced Phe degradation mostly by inducing enzymatic expression and acting as a co-substrate and energy sources. Therefore, this finding offers a new avenue for rhizospheric microbial degradation of PAHs via biostimulation.

4.3.4. Enzyme-mediated degradation

Enzyme-mediated remediation comprises the application of enzymes isolated from living organisms that can degrade PAHs. The action of enzymes is efficient and selective, and they have the potency to catalyze reactions over a inclusive range of temperatures and pH (Kuppusamy et al., 2017). Several enzymes are well-known for the degradation of PAHs, such as oxygenase, dehydrogenase, manganese peroxidase, lignin peroxidase, laccases, and phenol oxidases (Patel et al., 2020; Kumar et al., 2020c). Oxidative enzymes secreted by fungi are considered more competent due to their low substrate-specificity (Gupta and Kumar, 2020). Zhang et al. (2020) isolated a manganese peroxidase gene from *Cerrena unicolor* BBP6 (a wood-rotting fungus) and transferred it into *Pichia pastoris* (a yeast), which degraded 80% and 91% of Flu and Phe, respectively, within 24 h. Laccase isolated from *Trametes*, a fungal species, was employed to estimate its ability to degrade 15 priority PAHs-pollutants in field soils. The PAHs were the carbon source for the fungus (Wu et al., 2008). Outcomes showed the enzymatic conversion of PAHs into relatively fewer toxic intermediates, such as Ant to anthraquinone and BaP to benzo[a] pyrenyl acetate. With a 10 U g^{-1} dose of laccase, degradation of 24% of the PAHs was reported within 24 h, and, on the 14th day, more than 80% of BaP was degraded. But still, this enzymatic degradation of PAHs is not economical due to the cost involved in the production of enzymes and their purification (Kuppusamy et al., 2017).

4.3.5. Compost-mediated degradation

Composting is a biochemical method built on the ability of microbes to decompose organic substrates, with the consequential release of heat, CO_2 , and water along with organic material (i.e., compost) (Rastogi et al., 2020; Ayilara et al., 2020). Composting is of interest due to the potential application of compost as an amendment in soils and sediments to hasten the degradation of organic pollutants, including PAHs (Lukić et al., 2016). A number of methods are deployed to hasten the remediation of organic pollutants, including PAHs, via co-composting of polluted samples with organic materials (Lukić et al., 2016). These comprise: (i) an increase in temperature during composting; (ii) supplementation of additional carbon and nutrient ingredients (biostimulation); and (iii) inoculation with of a diverse group of microbes having the capability to degrade PAHs (bioaugmentation). An increase in temperature during composting can hasten biodegradation, such as increasing the solubility, thus enabling the pollutants to become

accessible and bioavailable to microbes as a carbon source (Haritash and Kaushik, 2009).

The compost-integrated degradation of PAHs in soils and sediments is governed by the physical, chemical, and biological characteristics of the contaminated matrix, the organic compost substrate, and environmental parameters (Lukić et al., 2016; Poluszyńska et al., 2017). The composting substrate enriches the contaminated soil with microorganisms derived from the composting substrate, and it also increases the moisture retention capacity of the soil, thereby facilitating the bioremediation of PAHs (Lukić et al., 2016). The compost-integrated bioremediation approach also facilitates the eco-friendly disposal of organic waste used as a composting substrate, because the waste is simultaneously decomposed (Lukić et al., 2016). Organic amendments used as a compost substrate are likely to improve soil structure and rate of oxygen transfer and supply additional nutrients and carbon to microbes. Addition of an organic-compost substrate increases the capability of microorganisms to degrade PAHs. Antizar-Ladislao et al. (2005) showed that PAH degradation in soil during composting is more effective with the amendment of fresh organic substrates in comparison to mature compost, which may be attributed to the active composting of the fresh organic substrates, thereby facilitating the degradation of the contaminant. Ma et al. (2018) utilized modified peanut shell powder as a bio-agent for *Mycobacterium gilvum*, which promoted the biodegradation of PAHs in farmland soils. Application of the bio-agent to the farmland decreased bioaccumulation of PAHs in mustard plants grown on the farmland polluted with PAHs (Ma et al., 2018).

4.4. Hybrid/integrated technology

The coupling of two or more remediation techniques to remove PAHs is referred to as a hybrid-remediation technology. Various physical-chemical, chemical-biological, biological-physical, physical-chemical-biological, and biological-biological techniques are practiced (Kuppusamy et al., 2017). In soil polluted with different PAHs, Posada-Baquero et al. (2019) investigated their biodegradation using *Mycobacterium gilvum* bacteria in the presence and absence of rhamnolipid, an enhancer of the solubility of PAHs. The biodegradation of PAHs was 73.53–98.06% in the presence of rhamnolipid, whereas 72.22–97.43% was obtained in the absence of rhamnolipid (Posada-Baquero et al., 2019). Similarly, biodegradation of PAHs by *Stenotrophomonas* sp. was investigated with the assistance of a surfactant and the surfactant with phenol, which were applied to enhance the bioavailability of the PAHs (Zang et al., 2020). Zang et al. (2020) demonstrated that, compared to natural degradation, the total degradation of PAHs was increased by 9.9% with assistance from the surfactant, whereas it was increased by 14.84% with the support of surfactant with the phenol.

Hybrid technology (electrokinetic-oxidation-bioremediation technique) was applied to remediate PAHs from a highly polluted coking-plant soil (3605 mg/kg PAHs). The technology removed 95.2% of PAHs (Liu et al., 2020c). This observation might be because of the partial oxidation of PAHs during the electro-oxidation process, which enhanced the biodegradation of PAHs. Gou et al. (2020) reported that bacteria from the phyla *Firmicutes* and *Proteobacteria* carried out the anoxic biodegradation of PAHs. After 180 days, the remediation efficiency was approximately 25–95% for PAHs with 3–4 rings, whereas it was around 30–75% for PAHs with 5 rings PAHs (Gou et al., 2020).

5. Case studies of field application of remediation technologies

Several studies using laboratory-based, batch-scale, and pilot-scale remediation technologies have demonstrated their ability to remediate soils and sediments polluted with PAHs (Table 3). Nevertheless, few field experiments have been carried out to show the importance of the above-discussed remediation technologies. Below, field trials using these remediation technologies are discussed.

5.1. A case study at the Bagnoli (Italy) industrial area

Bagnoli, an abandoned manufacturing industrial site, was a leading steel plant in Italy before the mid-1990s. The products it produced and the side products from the manufacturing processes altered the natural environment (De Vivo and Lima, 2018). It became a priority site for remediation. Among the various remediation strategies that were applied was an in-situ bioremediation strategy, which was adopted and it is currently being used at the Bagnoli site (Guarino et al., 2019). This strategy uses plants and endogenous microbial communities that naturally occupied, and adapted to, the contaminated soil. The diverse bacterial communities that naturally prevail in the soil environment, including those with PAHs, are α , β , and γ -Proteobacteria, Bacteroidetes, Actinobacteria, and Firmicutes (Crampon et al., 2018).

The families of plants that have been identified at the Bagnoli site are Poaceae, Fabaceae, Asteraceae, and Apiaceae. These plant families are well adapted to grow under the PAH-polluted soil conditions (Guarino et al., 2019). The concentrations of PAHs in various plant parts, such as the roots, were analyzed for 16 endogenous plant species. The concentrations of PAHs in the plant roots and rhizospheres were estimated to be in the range of 0.2–30 mg/kg and 1.60–190 mg/kg, respectively. Varying concentrations of PAHs in plant parts showed that the LMW PAHs (4-ring and 5-ring) were typically found in the plant's roots and leaves. In this case study, rhizosphere soil samples collected from three different plant species (*Piptatherum miliaceum* L., *Lotus corniculatus* L., and *Plantago lanceolata* L.,) were investigated and characterized for biodiversity. It was found to be high, around 93–97% of the total biodiversity in each sample.

A metagenomic investigation of soil samples showed that, within the Proteobacteria, α -Proteobacteria and γ -Proteobacteria were the dominant taxa, comprising well known PAH-degrader bacterial species, which occurred along with an abundance of enriched Actinobacteria populations. Firmicutes and γ -Proteobacteria were shown to be less abundant in the samples. All three samples investigated displayed the fact that the γ -Proteobacteria were the most copious class and they are in the Pseudomonadaceae family. A previous report also discussed the application of the genus *Pseudomonas* to remediate soils polluted with PAHs (Sun et al., 2020). This real, field-case study addressed the application of microbial and phytoremediation technologies to remove PAHs from contaminated sites, although, in this study, the investigators did not mention the removal potency, removal rate, and quantity of PAHs removed, which need to be discussed in future research.

5.2. A case study at the Oviedo, Spain, industrial area

For a preliminary test at the laboratory-scale and pilot-scale, field samples were obtained from a site (25,000 m²) near Oviedo, Spain, polluted with PAHs. A chemical manufacturing plant had been functioning there since the 1970s. This manufacturing plant was producing chemicals, such as naphthalene and phenols, from coal tar processing. To start the “on site” bioremediation at the field scale, an experimental area was designated before putting the polluted soil (around 14,000 m³ soil volume) on it (Pelaez et al., 2013). The polluted soil was in biopiles. Each biopile consisted of 900 m³ of PAH-polluted soil.

Bioremediation treatment was performed at the field-scale using specific sets of conditions, such as total initial concentration of PAHs (637.8 mg/kg), biopile characteristics (polluted water + Bioversal™ + Ivey-sol®) (Bioversal™ is the generic term for a wide range of cleaning agents that accelerate biological degradation and Ivey-sol® is a surfactant remediation technology), average relative humidity (79.2%), average temperature (13.7 °C), and average total sun hours (162.3 h month⁻¹) (Pelaez et al., 2013). The initial concentrations of PAHs at the field scale were almost five times less than those at the pilot-scale, but they were comparable to the concentrations used at the laboratory-scale. A considerable decrease in the concentration of PAHs was estimated at 48 days and 161 days (end day). It was challenging to

establish the degradation rate due to sample heterogeneity. A GC-MS qualitative analysis demonstrated that lighter PAHs (more simply biodegradable) were reduced to a better extent than the heavier ones. After 161 days, the degradation of naphthalene (2-ring) and PAHs was 98.2% and 94.4%, respectively. These decreases were adequate to get the concentrations of the PAHs under the target level, which was defined in a risk evaluation for this site. Compared to microcosm- and pilot-scale demonstrations, the field experiment showed an almost 2-times decrease in microbial diversity after 161 days. The number of colony forming unit (CFU) increased for 48 days, and, after that, they decreased to day 100 of the experiment. Denaturing gradient gel electrophoresis (DGGE) analysis revealed that the deoxyribonucleic acid (DNA) band intensities were highest at 48 days and were reduced noticeably at 161 days after treatment. This field trial showed in-situ microbial remediation of PAHs at a contaminated site. But the economic and environmental impacts were not included in this trial, and they need to be considered in the future.

5.3. A case study at Trondheim harbor (Norway)

At the Trondheim harbor (Norway) in April 2008, in-situ, thin-layer capping using AC was used to remediate sediment polluted with PAHs and polychlorinated biphenyls (PCBs) (Samuelsson et al., 2015). Briefly, for 12 months, three sets of combinations, which were AC only, AC + clay, and AC + sand, were tried on a 10 × 10 m experimental plot with a depth of 4–6 m, which had a salinity of 32 parts per trillion (ppt). Two model organisms, the omnivorous polychaete worm, *Hediste diversicolor*, and the surface deposit-feeding clam, *Abra nitida*, were selected as naturally appropriate species in the North Sea and best for laboratory trials. The concentrations of pollutants within the organisms were estimated after one month. Four treated sediment samples were sampled from the experimental site (AC-only, AC + clay, AC + sand, control) and applied in the bioaccumulation trial with *H. diversicolor*. For *A. nitida*, only two samples were compared (AC + clay and control).

The two organisms were exposed to 9 PAHs and 9 PCBs in the uncapped sediment (reference sample) to determine their bioaccumulation. In comparison to background levels, both organisms tested in the reference sample accumulated up to 8 times more PAHs and PCBs. After one month of exposure, normalized PAH concentrations, based on the wet weight of the organisms (ng g⁻¹), were found to be 1.8–30.9 times higher in *A. nitida* than *H. diversicolor*. Treatments of sediments with AC + clay reduced the concentrations of PAHs and PCBs in *H. diversicolor*. Also, a decrease in uptake of PAHs was seen in *A. nitida* for 9 PAHs. Moreover, AC + clay displayed a reduction of PCB levels in *A. nitida*, although the experimental results were unclear because of a fewer samples number and great deviation in the analyzed samples. More replications and longer contact times could have provided better information about the bioaccumulation of PCBs in *A. nitida*. The estimated capping efficiency (CE) was in the range of 69–86%. The CE for *A. nitida* (PAHs 81% and PCBs 79%) was comparable to the CE for *H. diversicolor* (PAHs 79% and PCBs 59%), despite the fact that *A. nitida* was in the vicinity of the AC-cap on the sediments. In comparison, *H. diversicolor* developed a burrow network to deep depths in the sediment layer (García-Arberas and Rallo, 2002). Bioturbation enables mixing of AC sediments and simultaneously sequesters pollutants in deeper layers of the sediment (Lin et al., 2014). In this trial, AC was mixed 20–30 mm under the original cap, possibly because of the bioturbation (Cornelissen et al., 2011). Improved CE can be expected over time due to continuous bioturbation by benthonic organisms. The results of the trial showed that the amendment with AC + clay could deliver a high CE and decreased bio-uptake in *A. nitida*, which was in the top layer of sediment, as well as in *H. diversicolor*, which foraged from the surface to deep layers. AC-only led to a lessening in the bioaccumulation of PCBs. There was no significant impact observed for the bioaccumulation of the PAHs. Also, in the AC + sand amendment, no substantial decrease in bioaccumulation efficiency of both organisms for PAHs or PCBs

was detected.

In a marine environmental setting, thin-capping with AC without mechanical mixing was effective in decreasing the concentrations of PAHs and PCBs in the pore water of sediments and in surface water, as well as in two benthonic species, a polychaete and a bivalve. AC-amended clay (AC + clay) displayed better retention of pollutants and fewer negative impacts on biota than AC or AC amended with sand (AC + sand).

6. Knowledge gaps and prospective

These days several technologies have been emerged out to remediate PAHs contaminated soils and sediments and transmute PAHs compounds in to less toxic, and/or nontoxic forms as discussed in this review comprehensively. These technologies are successfully adopted to eliminate or diminish PAHs from the contaminated media at minimum environmental cost. These technologies, also minimizes the carcinogenic, mutagenic and teratogenic properties of PAHs prevailing in soils and sediments. Still, the most serious challenge associated with use of advanced remediation technologies of PAHs contaminated soils and sediments is their unaffordability as it is represented in Fig. 4. Cost-effective technologies are being developed at a slow pace.

Future research should be devoted to the following challenges.

- Trophic transfer and impacts on human health of individually PAH compound, individually and in the mixed form, have not been fully explored. In particular, the extent to which PAHs presence in the natural environment as solid crystals should be studied. Future investigations should consider not only the toxicities of distinct PAHs and groups of PAHs but also mixtures of PAHs along with other environmental contaminants.
- Several factors influence remediation of PAHs in soils and sediments, such as site characteristics and the surrounding environment; microorganisms and co-contaminants present; and remediation cost. Proper optimization of these factors is key to improve the efficacy of remediation technologies and their successful execution at the field-scale.
- Bioremediation has been established to be an effective and environmentally friendly technology that can be deployed to control the impact of pollution from PAHs. However, the application of this technology is still in its infancy due to insufficient monitoring tools and technologies that can certify the persistence of effective biodegradation methods.
- Recent, innovative remedial methods include transgenic, nano-remedial, and photo-hetero microbial methods. These methods have remediated various organic and inorganic contaminants. However, their efficiency in removing PAHs from soils and sediments is still unknown, and, henceforward, research should focus on these technologies
- Optimization and commercialization of procedures to monitor PAHs in polluted soils and sediments are needed. These optimized procedures should be put into legislation and laws. Also, investigators must deploy these technologies in field studies, because few field trials have been carried out. Moreover, after remediation, investigations need to assess the possibility of re-contamination of the soils and sediments by PAHs.
- Investigations have shown that there are various ways to remediate sites polluted with PAHs. Nevertheless, the cost of the remedial technologies and monitoring of polluted sites is high. Future investigations must focus on advanced, sustainable, cost-effective, and energy-efficient remediation strategies.
- Advanced molecular tools (metagenomic) are being deployed to understand the microbial diversity of PAH-polluted sites. However, the biodegradable potential of culture-independent microbes remains unexplored. They need to be studied under field conditions.

7. Conclusions

Worldwide, soils and sediments continue to be polluted with PAHs. This review has discussed the advantages and challenges associated with immobilization, mobilization and degradation-based remediation approaches of soils and sediments polluted with PAHs. Many technologies and methods are available for remediation of polluted sites. The adoption of a technology depends upon the type of contamination, level of contamination and their bioavailability, environmental conditions, duration of the remediation, cost, and risk assessment. Recently several bioremediation, biodegradation, and advanced approaches are emerged out to transmute PAHs to less toxic or nonhazardous compounds in greener and more cost-effective way. They also minimize the carcinogenicity, mutagenicity and teratogenicity of PAH in polluted soils and sediments.

CRediT authorship contribution statement

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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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