



Polycyclic aromatic hydrocarbons: soil pollution and remediation

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Abstract

Soil is an important environmental matrix to support the life of all organisms directly or indirectly. Despite being the ultimate sink for all pollutants, it has been neglected for long, which has negatively affected the quality of the soil. Disposal of pollutants has resulted in changes in properties of soils and introduction of toxicity into it. The presence of heavy metals, pesticides, polychlorinated biphenyls and polycyclic aromatic hydrocarbons (PAHs) affects all forms of life since these chemicals have associated toxicity, mutagenicity, and carcinogenicity. PAHs are typical pollutants of soil which result in alteration in grain size, porosity and water-holding capacity of soil and affect diversity/population of microbes adversely. Significant changes in permeability, volume, plasticity, etc., are also brought about resulting in poor quality of contaminated soils. Considering the toxicity and global prevalence of PAHs, remediation of contaminated soils has become a challenge. Therefore, it is important to understand the detailed mechanism of physical, chemical or biological changes in soil. Simultaneously, it becomes pertinent to identify the environmentally sustainable treatment options for remediation of contaminated sites. Whereas physical and chemical treatment methods are either cost, chemical, or energy prohibitive, the biological treatment is emerging as an efficient and effective option which employs microorganisms for mitigation. Microorganisms are known for their enzyme-catalyzed catabolic activity when degradation/mineralization of a pollutant is aimed at and can prove useful in degradation of PAHs. Therefore, the present study reviews the effects of PAHs on soil properties, different remediation techniques and the role of microorganisms in remediating contaminated sites.

Keywords Advanced oxidation process · Bioremediation · Geotechnical properties · Toxicity · Treatment

Introduction

With increasing awareness of the adverse effects of hazardous chemicals on human health and environment, the remediation of such chemicals has received more attention internationally. Environmental pollution caused by dumping of solid waste, untreated industrial effluents, persistent organic pollutants (POPs) like PAHs, PCBs, and pesticides, is a critical problem. These compounds, directly or indirectly released in the environment, are found to be very toxic and lead to contamination of water, soil, and air. Soils contaminated with organic pollutants and inorganic contaminants like heavy metals have high potential health risk because

such contaminants can enter food chain (Xiaojun et al. 2007). Contamination due to leakage/spillage of crude oil can adversely affect exposed organisms since it is an intricate mixture of toxic aliphatic and complex aromatic hydrocarbons. Studies have established that toxicity increases with an increase in complexity of chemical structure (Patnaik 1999). Moreover, since complex/branched structure results in chemical stability, such pollutants are stable, resistant to biodegradation and as a result accumulate in environment which leads to accumulation of toxicity. When hazardous chemicals are introduced into soil, it may lead to soil contamination and can affect agricultural production too (Ibrahim 2004). Whereas contamination of soil can directly affect human health, the presence of PAHs in marine ecosystem may affect humans indirectly (Pathak et al. 2011). Soil contaminated with hazardous petroleum hydrocarbons can adversely affect human health, as it gets absorbed through direct dermal contact. Therefore, it is high time to adopt effective measures to reclaim contaminated sites and control other sites from being getting contaminated. If no efforts

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are made to clean the contaminated sites, then the harmful effects of such sites will persist in environment, and add up to the Brownfield list.

Organic pollutants associated with petroleum such as PAHs are considered as the hazardous pollutants due to their toxic, mutagenic, and carcinogenic effects. They have varying physicochemical, and toxicological characteristics according to their molecular weight (Table 1). PAHs are composed of fused benzene rings consisting carbon and hydrogen arranged in a simple to complex structural configuration, i.e., in linear, angular, or cluster arrangements. PAHs are classified according to their aromatic benzene ring number, i.e., consisting two or three benzene rings are classified as low molecular weight (LMW) PAHs such as naphthalene, anthracene, and phenanthrene, and PAHs consisting more than three benzene rings are classified as high molecular weight (HMW) PAHs (pyrene, chrysene, benzo(a) pyrene, coronene, etc.). Usually, these are produced from anthropogenic as well as natural activities (Table 2). Forest fires, oil seeps, volcanic eruptions and exudates from trees are some natural sources; and burning of fossil fuel, solid biomass fuel (SBF) such as wood, crop residue or dung cake, coal tar, crude oil or petroleum spill (oil spillage and leakage), high-temperature industrial processes, and petroleum refinery effluent are some anthropogenic sources (Haritash and Kaushik 2009; Sharma and Jain 2019). There are many other natural as well as anthropogenic sources of PAHs and in addition viz. smoke from wood-burning stoves, automotive emissions, creosote waste materials and manufactured gas plants (coal gasification) are some other anthropogenic sources of PAHs (Abdel-Shafy and Mansour 2016). More than 100 unique PAHs are widely distributed and ubiquitously found in the environment, 16 PAHs among them have been classified and identified as priority pollutants, and some of them are classified as possible human carcinogen by U.S. EPA and European Union (Potin et al. 2004). PAHs are ubiquitously present around the globe and have contaminated the soil in different geographical regions of the world (Table 3), and they are hydrophobic with low water solubility so they have tendency to bind with organic matter present in soil/sediments. PAHs are relatively stable contaminants showing recalcitrant nature in soils, and it is difficult to degrade them as compared to many other organic contaminants. Considering the toxic properties and persistence of PAHs in soil, reclamation of PAH-contaminated soils become imperative. There are a number of physical (solvent extraction, air sparging, thermal desorption, microwave heating, vitrification, electrokinetic treatments, etc.); chemical (oxidation using Fenton's reagent, ozone, etc., photocatalytic degradation); and biological (microbial, landfarming, composting, phytoremediation, etc.) methods being used to remediate the contaminated site. The selection of treatment method is subject to type of contaminant, soil type, interferences present,

and risk analysis associated with the techniques. It has been studied that during PAH degradation some epoxides and dihydrodiols are formed which are found to be more toxic than the parent PAHs (Cerniglia and Sutherland 2010). So, identification of intermediate PAH metabolites is also essential during its degradation. Although physical and chemical methods are effective and efficient, most of them are energy, cost, or chemical intensive. Moreover, these methods are responsible for generation of secondary pollutants. Keeping in view, the limitations associated with physicochemical treatment, the biological methods are gaining popularity owing to their ecofriendly nature and bioconversion of toxic pollutants into harmless/nontoxic chemical species.

Properties of contaminated soils


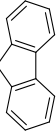
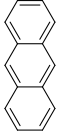
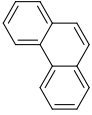
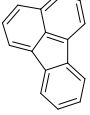

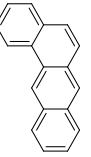

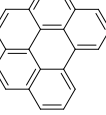
Physicochemical properties

Soil is the major sink for a broad range of harmful contaminants, and these contaminants may affect the physical, chemical, and biological properties of soil. Crude oil is the major source of PAHs which may easily diffuse and get absorbed inside soil particles. Soil is extremely sensitive to different contaminants, and its behavior changes under different environmental conditions. Properties of soil are affected by different activities occurring on land and different types of contaminant, crude oil accidental spillage, and leakage, etc. Longer contamination may affect geotechnical properties such as Atterberg limits, permeability, hydraulic conductivity, strength parameters, consolidation, compaction and shear strength as well as the biological properties (biomass and enzyme activity) of soils. Since microbes have the ability to degrade hazardous contaminants, they are involved in remediation and soil self-purification processes. PAHs are toxic even to microbes and contaminated soil has lower self-purification capacity due to a decrease in microbial activity of soil (Hreniuc et al. 2015). Effects on properties of soil may be due to partial or full replacement of the soil pore liquid with PAH, and choking of soil pores with contaminant due to which soil aeration and water infiltration could reduce.

The degree of effects of PAH is regulated by certain important parameters of soil viz. grain size of soil particles, organic carbon associated, and to some extent pH of the soil. A number of studies have confirmed that binding of PAHs takes place preferentially on finer grain size, i.e., silt and clay (Magi et al. 2002). The clay particles have more surface area, and as a result, more bonding sites resulting in adsorption of PAHs firmly on finer fraction of soil. Fine particles also result in less porosity and hence lesser movement of adsorbed contaminants over the period of time which results in persistent toxicity and long-term effects.



Table 1 Physicochemical properties of major PAHs

S. no.	Name	Mol. formula	Chemical structure	CAS registry no. ^a	B.Pt. (°C) ^a	M.Pt. (°C) ^a	V.P. (Pa at 25 °C) ^a	Aq. Sol. (mg/l) ^b	Log K_{ow}^c	IARC group ^a	Half-life (days) ^d
1.	Naphthalene	C ₁₀ H ₈		91-20-3	218	80.2	11	31.7 ± 0.26	3.35	NE	5.66
2.	Fluorene	C ₁₃ H ₁₀		86-73-7	295	116.7	0.08	1.98 ± 0.04	4.18	3	15.14
3.	Anthracene	C ₁₄ H ₁₀		120-12-7	342	216.4	1 × 10 ⁻³	0.073 ± 0.005	4.54	3	123
4.	Phenanthrene	C ₁₄ H ₁₀		85-01-8	340	100.5	2 × 10 ⁻²	1.29 ± 0.07	4.57	3	14.97
5.	Fluoranthene	C ₁₆ H ₁₀		206-44-0	375	108.8	1.2 × 10 ⁻³	0.26 ± 0.002	5.22	3	191.4
6.	Pyrene	C ₁₆ H ₁₀		129-00-0	150.4	393	6.0 × 10 ⁻⁴	0.135 ± 0.005	5.18	3	283.4
7.	Benzo[a]anthracene	C ₁₈ H ₁₂		56-55-3	438	158	0.548	0.014 ± 0.0002	5.91	2A	343.8
8.	Benzo[a]pyrene	C ₂₀ H ₁₂		50-32-8	495	179	4 × 10 ⁻⁷	0.0038 ± 0.00031	5.98	2A	421.6
9.	Benzo[ghi]perylene	C ₂₂ H ₁₂		191-24-2	500	277	6 × 10 ⁻⁸	0.00026 ± 0.00001	7.10	3	517.1

Mol. formula, molecular formula; CAS, chemical abstracts service; B.Pt., boiling point; M.Pt., melting point; V.P., vapor pressure; Aq. Sol., aqueous solubility; Log K_{ow} , octanol/water partition coefficients; IARC, International Agency for Research on Cancer

^aWGPAH (2001)

^bMackay and Shiu (1977)

^cMichele et al. (1985)

^dHoward et al. (2005)



Table 2 Sources of PAHs in contaminated soil

S. no.	Type of sources	Activities	Process	References
1.	Pyrogenic (associated with combustion of wood, petroleum product, coal)	Industrial (anthropogenic) activities	Wood burning Burning of tires Burning of fossil fuel Burning of tobacco Burning of agricultural waste Combustion of oil, diesel, coal and oil products	Wilson and Jones (1993) Downard et al. (2015) McRae et al. (2000) Hausmann (2012) Lai et al. (2009) Kaushik et al. (2012)
		Natural activities	Volcanic eruption Forest fire	Kozak et al. (2017) Denis et al. (2012)
2.	Petrogenic (associated with substances originate from crude oil/petroleum)	Industrial (anthropogenic) activities	Oil spill	Soriano et al. (2006)
			Outlets from oil refinery	Pettersen et al. (1997)
			Petroleum handling facilities like kerosene tank, generating plant, petrol stations, mechanic workshops, leaking pipeline, and airport fuel dump	Nganje et al. (2007)
			Petroleum oil industries and industrial activities	Varjani et al. (2017)
3.	Biogenic	Natural activities	Creosote, asphalt production	Abdel-Shafy and Mansour (2016)
			Used engine oil, jet fuel, kerosene	Kaushik and Haritash (2006), Kaushik et al. (2012)
		Natural activities	Manufactured gas plants	Abdel-Shafy and Mansour (2016)
			Oil seeps	Pampanin and Sydnes (2013)
		Natural activities	Surface waxes of leaves and insect cuticles	Oleszczuk and Baran (2005) Krauss et al. (2005)
			Wood of tropical forests	Krauss et al. (2005)
			PAH synthesis in termite organisms	Ratola et al. (2006)
			Pine needles	

Some of the studies have also confirmed that HMW-PAHs bind firmly to the soil compared to LMW-PAHs (García-Alonso et al. 2008). Since the inter-particle spaces in fine soil are less, the porosity-mediated effects are minimum resulting in nontransfer of HMW-PAHs from one location to another due to their hydrophobic nature. Some of the studies have also reported that within-the-particle movement of PAHs from the surface to core also takes place (Bogan and Trbovic 2003). The intra-particle movement is ascribed to be the function of organic carbon associated with the soil particles. The two types of domains which interact with contaminant are surface layer of humic acid and fulvic acid (HA–FA), and core formed of hard layer of humin. The HA–FA layer is porous, flexible, and lipophilic, and humin layer is rigid, hard, and glassy (Xing and Pignatello 1997). The humin bound PAHs are firmly bonded to the soil which have been subjected to diagenetic alteration (derived from weathering of sedimentary rocks) (Lueking et al. 2000). On the other hand, soils which have not been subjected to diagenetic alterations have organic matter with soft carbon fraction (HA–FA). Therefore, the effect as well as treatment strategy of contaminated soil significantly depends upon the grain size, its paedogenetic profile and the type/fraction of

organic carbon associated with the soil. The pH of soil also, to some extent, regulates the degree of contamination. The HA–FA-rich acidic soils bind preferably with PAHs resulting in higher and persistent contamination, whereas high/basic pH results in frequent dissociation of soil particles which does not favor the binding of PAHs (Saba et al. 2010). Thus, organic carbon-rich fine-grained acidic soil is more prone to PAH contamination and its effect and is difficult to remediate.

Biological properties

The biological activities, i.e., microbial biomass and enzymatic activities of soil, are highly sensitive to environmental pressure/parameters (Labud et al. 2007). Soil contamination may affect the microbial community/population and microbial activity/enzymatic activities of the soil. Various experiments have been done under controlled environments to investigate the adverse effect of petroleum hydrocarbon (PAHs) contamination on soil microorganisms and metabolic activities. Microbial activity may be inhibited due to the presence of high concentration of organic contaminants. Soil contamination with crude oil may develop anaerobic



Table 3 Polyaromatic hydrocarbons in soils and sediments of different geographical regions of world

Country	Location	PAHs ^a	Total PAH range (ng g ⁻¹)	Soil pH; TOC/OC (%)	References
India (Asia)	Agra (urban soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)F, CHRY, B(b)F, B(k)F, B(a)P, and B(ghi)P	3190–28,540	NM; NM	Masih and Taneja (2006)
	Delhi (vehicular traffic soil)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	1062–9652	NM; NM	Agarwal (2009)
	Assam and Arunachal Pradesh (soil from Indian Himalayan Region)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	15.3–4762	NM; NM	Devi et al. (2016)
China (Asia)	Beijing (urban soils, < 10 m from roads)	NAP, ACEY, ACE, FLU, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, F, B(a)P, IPYR, D(ah)A, and B(ghi)P	1470–6610	NM; NM	Chu et al. (2003)
	Macao (coastal sediments)	PHE, ANT, PYR, MPHE, FLA, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, B(e)P, IPYR, D(ah)A, and B(ghi)P	294–12,741	NM; 0.42–1.7	Mai et al. (2003)
	Hong Kong (urban soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	7.0–410	4.62–7.52; 0.532–2.92	Zhang et al. (2006)
	Hong Kong (Mangrove swamps sediments)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, and IPYR	356–11,098	NM; 1.23–7.50	Tam et al. (2001)
	North and tropical forests in South China (forest soils)	ACEY, ACE, FLU, PHE, ANT, FLA, PYR, CHRY, B(a)A, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	24.81–4539	NM; 2.007–52.375	Syed et al. (2017)
Russia (Asia)	Moscow and the south Taiga near Moscow (grassland and forest soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(j)F, B(k)F, B(a)P, B(e)P, PERY, D(ah)A, B(ghi)P and COR	59–1350	3.8–5.9; 1–6.8	Wilcke et al. (2005)
Thailand (Asia)	Bangkok (urban soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(j)F, B(k)F, B(a)P, B(e)P, PERY, D(ah)A, B(ghi)P, and IPYR	12–380	3.6–7.4; 1.04 ± 0.69–2.03 ± 1.94	Wilcke et al. (1999b)



Table 3 (continued)

Country	Location	PAHs ^a	Total PAH range (ng g ⁻¹)	Soil pH; TOC/OC (%)	References
South Korea (Asia)	Korean peninsula (paddy soils and upland soils)	NAP, ACEY, ACE, FLU, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	23.3–2834	NM; NM	Nam et al. (2003)
Korea (Asia)	Kyeonggi Bay (sediments)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, PERY, D(ab)A, B(ghi)P, and IPYR	9.1–1400	NM; 0.08–1.2	Kim et al. (1999)
Taiwan (Asia)	Gao-ping River (sediments)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	8 to 356	NM; NM	Doong and Lin (2003)
Germany (Europe)	Stephanskirchen (urban soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, PERY, and B(e)P	400–470,000	4.82–12.21; 1.2–44.5	Wilcke et al. (1997)
Switzerland (Europe)	Swiss soil monitoring network (NABO)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	50–619	3.9–7.2; 1.0–22.7	Bucheli et al. (2004)
Poland (Europe)	Upper Silesia (agricultural soil)	FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	28–2447	3.8–6.3; 1.45–17.20	Maliszewska-Kordybach (1996)
Sweden (Europe)	Karlstad (gasworks plant soil), and Stockholm (wood tar production site)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, and B(ghi)P	270–2,601,000	4.7–8.63; 2.0±0.3 to 49.1–0.4	Arp et al. (2014)
France (Europe)	Coke oven plant and metal-lurgy site	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	78,000–2,651,000	7.5–8.35; 7.7±2.1 to 33±0.6	Arp et al. (2014)
Belgium (Europe)	Gasworks site	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	296,880	7.57; 2.5	Arp et al. (2014)
Slovak Republic (Europe)	Ziar/Central Slovakia (forest soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	40,000–200,000	3.85–4.30; 0.81–2.46	Wilcke et al. (1996)



Table 3 (continued)

Country	Location	PAHs ^a	Total PAH range (ng g ⁻¹)	Soil pH; TOC/OC (%)	References
Czech Republic (Europe)	Bohemian mountains (forest soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(j)F, B(k)F, B(a)P, B(e)P, PERY, B(ghi)P, and IPYR	2000–30,000	NM; 4.3–46.5	Wilcke and Zech (1997)
Spain (Europe)	Valasske Mezirici (agricultural and forest soil)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	860–10,840 (agricultural) 7660–79,390 (forest soil)	4.62–7.12; 1.78–3.82 3.24–4.03; 3.45–19.20	Plachá et al. (2009)
Spain (Europe)	Tarragona County, Catalonia (soil and vegetation samples)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, PERY, D(ab)A, B(ghi)P, and IPYR	112–1002	7.08–8.22; 2.6–9.5	Nadal et al. (2004)
Italy (Europe)	Chioggia and Ancona (sediments of Adriatic Sea)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	24.1–501.1	NM; NM	Magi et al. (2002)
Tunisia (Africa)	Sfax (Sfax–Kerkennah coastal zone sediments)	NAP, PHE, ANT, FLA, PYR, B(k)F, CHRY, B(a)P, PERY, B(a)A, and B(ghi)P	110–10,720	NM; NM	Zaghden et al. (2007)
South Africa (Africa)	South Africa (soils from industrial, residential, and agricultural areas)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(j)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	44–39,000	NM; 0.0000001–0.00000049	Nieuwoudt et al. (2011)
Nigeria (Africa)	Okobo (soil from the vicinity of coal mine)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	0–10	NM; 4–8	Ugwu and Ukooha (2016)
USA (North America)	New Orleans, Louisiana (urban soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(j)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	731–2927	NM; NM	Mielke et al. (2004)
Canada (North America)	Kitimat fjord (sediments)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	1000–10,000,000	NM; 1.4–30	Simpson et al. (1996)
Cuba (North America)	Havana (soils of Mayabeque)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ab)A, B(ghi)P, and IPYR	20–106	5.3–8.2; 1.26–4.70	Sosa et al. (2017)



Table 3 (continued)

Country	Location	PAHs ^a	Total PAH range (ng g ⁻¹)	Soil pH; TOC/OC (%)	References
Mexico (North America)	San Nicolás (soils from brick manufacturing site)	NAP, ACEY, FLU, ANT, PHE, B(a)A, CHRY, FLA, PYR, B(b)F, B(k)F, B(a)P, and D(ah)A	7–1384	NM; NM	Barrán-Berdón et al. (2012)
	Uberlândia (urban soils)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(j)F, B(k)F, B(a)P, PERY, D(ah)A, B(ghi)P, and IPYR	7–390	4.6–8.4; 0.37–2.34	Wilcke et al. (1999a)
Brazil (South America)	Argentinean Pampas to the Patagonian Steppe (uppermost topsoil and the first subsoil horizons)	NAP, 2-MNAP, 1-MNAP, 1,3-DMNAP, ACEY, ACE, FLU, PHE, ANT, 1-MPHE, 3,6-DMPHE, FLA, PYR, B(a)A, CHRY, B(b)F, B(j)F, B(k)F, B(a)P, B(e)P, PERY, D(ah)A, B(ghi)P, COR, and IPYR	2.4–38	NM; NM	Wilcke et al. (2014)
Argentina (South America)	South Central Chile (Laja River Basin soil)	NAP, FLU, PHE, ANT, FLA, PYR, B(a)A, and CHRY	600–4243	NM; NM	Barra et al. (2005)
Chile (South America)	Sydney Harbor (surface sediment)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, PERY, D(ah)A, B(ghi)P, and IPYR	100–380,000	NM; NM	McCready et al. (2000)
Australia	Kogarah, Sydney (surface natural soils of Urban area) (NS-surface natural soil, RDS-road deposited sediments, and WS-water sediments)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, PERY, D(ah)A, B(ghi)P, and IPYR	NS-400–7490; RDS-1650–4000; WS-490–5190	NM; 1.8–3.8 NM; 0.8–13.0 NM; 0.3–3.0	Nguyen et al. (2014)
	McMurdo Station (surface soil)	NAP, ACEY, ACE, PHE, FLU, ANT, FLA, PYR, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(e)P, D(ah)P, D(ac)P, B(b)C, and COR	1000,000–27,000,000	NM; NM	Mazzera et al. (1999)
Ross Island (Antarctica)	Scott Base, Marble Point and the Wright Valley (surface and subsurface soil)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	41–8105	NM; NM	Aislabie et al. (1999)
McMurdo Sound-Dry Valley Region (Antarctica)	James Ross Island (soil)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	34.9–171	NM; 0.09–0.34	Klánová et al. (2008)



Table 3 (continued)

Country	Location	PAHs ^a	Total PAH range (ng g ⁻¹)	Soil pH; TOC/OC (%)	References
Antarctic Peninsula (Antarctica)	James Ross Island (sediments)	NAP, ACEY, ACE, FLU, PHE, ANT, FLA, PYR, B(a)A, CHRY, B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, and IPYR	1.4–205	NM; 0.04–1.71	Klánová et al. (2008)

^aNAP-Naphthalene, MNAP-Methyl-naphthalene, 2-MNAP-2-methylnaphthalene, 1-MNAP-1-methylnaphthalene, 1,3-DMNAP-1,3-dimethylnaphthalene, ACEY-Acenaphthylene, ACE-Acenaphthene, FLU-Fluorene, PHE-Phenanthrene, 1-MPHE-1-methylphenanthrene, 3,6-DMPHE-3,6-dimethylphenanthrene, ANT-Anthracene, FLA-Fluoranthene, B(j)F-benzo(j)fluoranthene, PYR-Pyrene, B(a)A-Benzo(a)anthracene, CHRY-Chrysene, B(b)C-Benzo(b)chrysene, B(a)F-Benzo(a)fluoranthene, B(b)F-Benzo(b)fluoranthene, B(k)F-Benzo(k)fluoranthene, B(a)P-Benzo(a)pyrene, D(ah)A-Dibenz(ah)anthracene, D(ah)P-Dibenz(ah)pyrene, D(ac)P-Dibenzo(ah)pyrene, B(ghi)P-Benzo(g,h,i)perylene, IPYR-Indeno(1,2,3-cd)pyrene, B(e)P-Benzo(e)pyrene, PERY-Perylene, and COR-Coronene

NM not mentioned

conditions in soil by blocking soil pore with consequent effects on microbial communities of soil (Sutton et al. 2013). By long-term contamination of soil by petroleum hydrocarbon and at high concentration of total petroleum hydrocarbons, there is decline in microbial biomass and soil enzyme activity owing to toxicity induced by high molecular weight hydrocarbons, i.e., PAHs (soil enzymes—dehydrogenase and urease), whereas at low petroleum hydrocarbon concentration, there is no effect on microbial biomass and enzymes (Verrhiest et al. 2002; Lipińska et al. 2014). Soil microbial diversity may be affected by PAHs contamination as PAHs may have toxic effects toward microorganisms present in soil. In an investigation, it was observed that PAH contamination has a significant effect on soil bacterial community structure (Khomarbaghi et al. 2019). In certain cases, PAH contamination may lead to complete loss of specific microbial species and if any particular species from contaminated soil have been damaged then certain essential soil function may be lost (Muckian et al. 2009). A study carried out to investigate the effect of pyrene on bacterial richness and microbial diversity in soil. It has been reported that after pyrene contamination microorganism's population belonging to phyla *Chlorflexi*, *Alphaproteobacteria*, *Actinobacteria*, *Deltaproteobacteria*, and *Crenarchaeota* were extensively reduced (Ren et al. 2015). The effect of three PAH mixture (phenanthrene, fluoranthene, and benzo(k)fluoranthene) on the indigenous microbial species of natural freshwater sediment was investigated. It was reported that due to heavy dose of PAHs, microbial activity was adversely affected. At low PAH concentration, i.e., 30 mg PAH/kg no negative effect on microbes was observed but at 300 mg PAH/kg harmful effects of PAHs, i.e., reduction in bacterial density and partial inhibition of the enzyme activity were observed as compared to the control 0 mg PAH/kg (Verrhiest et al. 2002). It has been studied that there is a significant effect of oil contamination on biological activity of soil, a strong decline in dehydrogenase and urease enzyme activity was observed in chainsaw oil (containing a complex mixture of PAHs, highly toxic.)-contaminated soil. It was also found that there is major influence of chainsaw oil on earthworm biomass and density (Klamerus-Iwan et al. 2015).

Remediation of PAH-contaminated soils

Today, remediation of polyaromatic hydrocarbon (diesel/crude oil)-contaminated soils is a global concern due to adverse risk to public health. Reclamation/remediation of PAH-polluted soils is essential and it can be done by different methods which involve removal/isolation or alteration of the contaminant. Various physical, chemical, thermal and biological remediation techniques (ex situ and in situ) have been developed for soil reclamation (Fig. 1). In situ



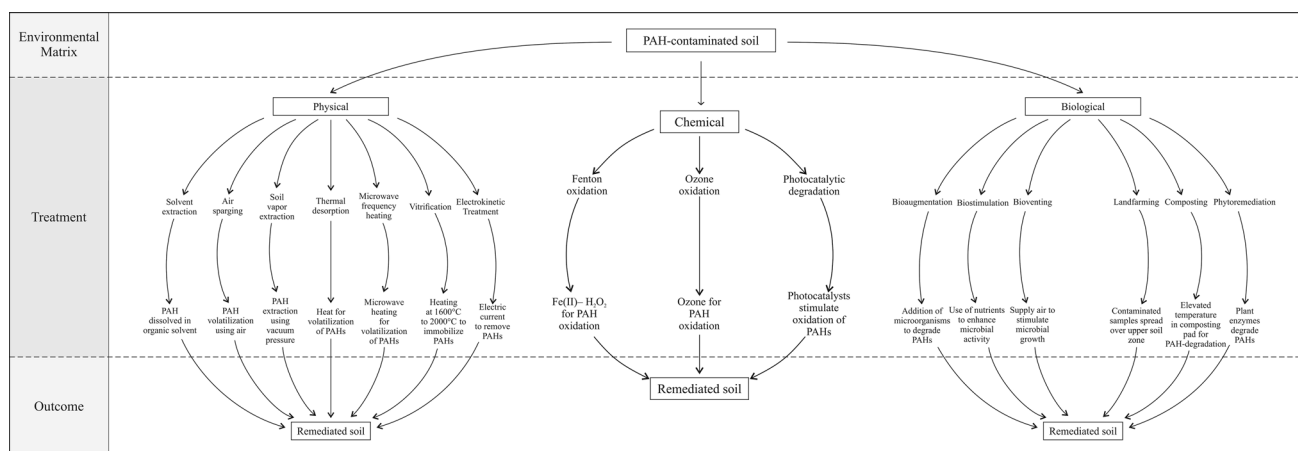


Fig. 1 Physical, chemical, and biological methods for treatment of hydrocarbon-contaminated soil

remediation process takes place at the contaminated site, whereas ex situ remediation is an alternative to in situ remediation where contaminants are treated off-site. There are some advantages of in situ remediation. It has lower cost, lower risk factor, limited involvement of human, and environmental surroundings can help in remediation process to transform the contaminants. For ex situ remediation, contaminated soil is excavated (dig and haul) from the site and transfer to another location for treatment so it requires mechanical as well as civil work. It is costly, time-consuming, more human involvement, direct exposure to contaminant, and due to digging it can harm underground utilities. There are some regulatory constraints also for ex situ remediation. Some of these remediation techniques are solvent extraction, UV oxidation, photochemical or photocatalytic degradation, bioremediation, and phytoremediation. The selection of suitable remediation technique for contaminated soils depends on several factors such as type of contaminant, future use of contaminated soil, type and properties of soil, budget, etc.

Physical treatment

Solvent extraction/soil washing

PAHs have high tendency to get absorb on organic matter present in soil due to their hydrophobic nature. Solvent extraction/soil washing is a separation or cleanup technique which is used to separate compounds based on their solubility. This technique can be used for PAH removal from contaminated soils. Different organic solvents (individual solvent or mixture of solvents), cyclodextrins (β -cyclodextrin (BCD), hydroxypropyl- β -cyclodextrin (HPCD) and methyl- β -cyclodextrin (MCD)), and vegetable oils can be used for extraction of PAHs from soils. The extraction of PAHs from soils is a two-step process. The first step is desorption of

compound from soil and the second step involves leaching (elution of compound into extraction fluid) of desorbed compound. Efficiency of solvent extraction is influenced by the nature of solvent used in extraction process and the ratio of the mass of contaminated soil to the volume of solvent (Silva et al. 2005; Viglianti et al. 2006). PAHs from extremely contaminated soils obtained from manufactured gas site can be extracted effectively using a solvent mixture containing ethanol or 2-propanol with 1-pentanol and water. The solvent mixture 1-pentanol (5%), water (10%), and ethanol (85%) is highly efficient and more effective than single solvent in removal of extractable PAH (19 PAH having two to six aromatic rings, e.g., naphthalene, 2-methyl naphthalene, acenaphthene, fluorene, phenanthrene, fluoranthene, pyrene, benzo(e)pyrene, benzo(a)pyrene, benzo(ghi)perylene, etc.) from soil. The extraction efficiency of this mixture is up to 95% when 4 ml solvent mixture is used for 1 g soil for one hour in three crosscurrent wash (extraction) stages (Khodadoust et al. 2000). Organic solvent mixture of Cyclohexane and ethanol (3:1) is suitable for fluoranthene extraction from contaminated soil. Its extraction efficiency is approximately 93% (Rababah and Matsuzawa 2002a). Similarly, it has been documented that solvent mixture of ethyl acetate (50%), acetone (40%), and water (10%) is suitable for extraction of hydrocarbons from soil (Silva et al. 2005). A major concern in this technique is the toxic nature of solvents and the liquid phase, i.e., the solvent containing desorbed PAHs. Therefore, selection of solvents is crucial and solvent containing desorbed PAHs exposed to other treatment for complete degradation of extracted PAHs.

Organic and mineral soil amendments

The organic and mineral soil amendment is an important process for reclamation of contaminated soils. These amendments in the contaminated soils help in attaining high

remediation efficiency as many biological, chemical and physical processes start after these amendments. Compost, manures, organic by-products, etc., are organic amendments and foundry sand, gypsum, coal combustion products, volcanic ashes, etc., are mineral amendments which are found to be useful for pollutant degradation (Fernández-Luqueño et al. 2017). In an investigation, activated carbon and olive mill waste compost were used as amendments for PAH-contaminated soil and enhanced degradation of total PAHs was observed (García-Delgado et al. 2019). Similarly, it was studied that sand can be used as an efficient amendment as sand amendment increases both oxygen and proton passage which increase soil porosity, reduce Ohmic resistance, and increase charge output. Sand amendment was found to be an effective method to accelerate degradation of PAHs by bio-electrochemical treatment (Li et al. 2015). In order to allow soil amendments effectively improve remediation of contaminated soils, it is essential to find different amendments which can improve soil porosity, increase microbial activity, and increase pollutant mineralization in different soils while retaining soil functions.

Thermal treatment

Thermal treatment for soil remediation uses heat to destroy contaminants. Organic chemicals such as PAHs can be destroyed or volatilize by heat, these contaminants changes into gases which results in increased mobility and these gases can be collected in wells for ex situ treatment. Different techniques have been used under thermal remediation such as thermal desorption, microwave frequency heating and vitrification.

Thermal desorption

Thermal desorption is the process in which heat is applied to increase vapor pressure of organic contaminants results in volatilization of contaminants and release of them from contaminated sites such as soil (Rushton et al. 2007). In this process, volatilized contaminants carried away or sweep by gas for secondary treatment or removal. High PAHs removal efficiencies could be obtained using thermal desorption. Soil contaminated with different PAHs from a manufacturing gas plant treated by thermal desorption process at laboratory scale. After thermal treatment at maximum temperatures above 450 °C, the concentration of different PAHs is reduced to below 0.05 mg/kg dry weight. For dibenzo(a,h)anthracene, efficiency is around 87% at temperature 250 °C, and for fluoranthene and pyrene, efficiency is nearly 100% with temperature above 350 °C (Renoldi et al. 2003). The subsurface soil of a wood treatment plant contaminated with benzo(a)pyrene equivalents (B(a)P-E) was treated by thermal desorption at field scale. Approximately 12,385 m³

(16,200 cubic yards) of predominantly silty soil containing 30.6 mg/Kg B(a)P-E was treated to a maximum depth of 32 m. After 130 days of post-treatment the remaining B(a)P-E concentration was 0.059 mg/Kg, which is equivalent to 99.8% PAHs removal. Therefore, thermal desorption is highly efficient at field scale for PAHs remediation (Baker et al. 2007). Temperature increase from 100 to 250 °C, results in an increase in removal efficiency of diesel from 47 to 100% from sand (Falciglia et al. 2011). Similar laboratory studies also showed that the efficiency of diesel removal from soil is increased during thermal desorption and removal efficiency depends on soil composition, temperature, temperature time, concentration of contaminants (Piña et al. 2002; Tatàno et al. 2013).

Microwave frequency heating

Microwave frequency heating is an effective thermal remediation technique in which microwave energy converted into thermal energy to eliminate contaminants through volatilization via heating (Rushton et al. 2007). This technique has been effectively proposed for soil remediation (Falciglia et al. 2013). 99% diesel oil could be removed within 10 min from contaminated soil using microwave induced thermal treatment in a modified domestic microwave oven with power 800 W and frequency 2.45 GHz (Li et al. 2008). A domestic microwave oven with a power 700 W and frequency of 2.45 GHz was used to treat heavy oil (diesel fuels and marine fuels)-contaminated soil, and it was found that up to 92.5% diesel and 89.5% marine fuel removed from soil in 20–150 s (Chang et al. 2011). 75.6–98.4% petroleum removal was achieved within 3.5 h at field-scale remediation of petroleum hydrocarbon-contaminated soil using on-site microwave heating system using antenna of 4 m, with power 2 kW and frequency of 2450 MHz (Chien 2012). On the other hand, using a modified domestic microwave with a power 1000 W and frequency of 2450 MHz, maximum of 95% diesel removed from soil in 5–60 min (Falciglia et al. 2013). Other similar studies also investigated the efficiency of this technique for oil removal from soil. Using this technique up to 100% PAHs could be removed in 60 min from soil artificially contaminated with different PAHs (Bph, Flu, Phe, Ant, Flt, Pyr, BaA, Chr and Per) when microwave frequency 2.45 GHz applied at 1000 W for remediation process. Total removal was achieved for biphenyl and fluorene whereas up to 90% removal was achieved for phenanthrene and anthracene and 50–80% removal was achieved for other PAHs (Falciglia et al. 2017).

Vitrification

Vitrification is a thermal technique which can be used for in situ soil remediation. This technique uses very high



temperatures, i.e., 1600–2000 °C to melt and immobilize contaminants in soil. Heat is delivered to soil through electric current via molybdenum electrodes. In this process, high-temperature melts the contaminants as well as the soil. After melting process, electrodes are turned off, allowing fusion of contaminants with soil and then after cooling both convert into a glass-like solid (Shearer 1991). Vitrification product, i.e., glass-like solid is a chemically stable, leach-resistant and crystalline material. Vitrification can be used to treat various organic contaminants such as petroleum products (Hinchee and Smith 1992). Fly ash is a major source of PAHs, after vitrification process in a coke bed furnace total 21 PAHs (naphthalene (NaP), acenaphthylene (AcPy), acenaphthene (Ant), fluoranthene (FL), pyrene (Pyr), cyclopenta(c,d)pyrene (CYC), benz(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(e)pyrene (BeP), benzo(a)pyrene (BaP), perylene (PER), indeno(1,2,3,-cd)pyrene (IND), dibenz-(a,h)anthracene (DBA), benzo(b)chrysene (BbC), benzo(ghi)perylene (BghiP) and coronene (COR)) present in fly ash significantly dropped from 69.6 µg to 3.46 ng, which results in high efficiency of PAH depletion. The lowest percent removal of individual PAH is higher than 99.9%. Therefore, PAHs in fly ash fully destroyed in vitrification process due to very high temperature (Kuo et al. 2003).

The thermal methods as mentioned above, i.e., thermal desorption, microwave frequency heating, and vitrification are essentially taking place under anaerobic conditions since oxygen in subsurface is absent. The added advantage of thermal processes is the absence of formation of secondary toxic pollutants that are formed sometimes during PAH-oxidation. Some toxic oxidized products such as epoxides and dihydrodiols are found to be formed and have higher toxicity than the parent PAHs (Cerniglia and Sutherland 2010).

Electrokinetic treatment

Electrokinetic technique is an in situ remediation technique where direct electric current is used to remove organic and other contaminants (inorganic and heavy metal) from the soil (Huang et al. 2012; Karaca et al. 2016). During this method, low-voltage direct current electric potential is applied through electrodes (anode and cathode). Low-voltage electric current causes mobilization of contaminants and their transportation toward electrodes placed inside contaminated soil matrix. Contaminants collected on these electrodes are pumped out for further treatment. Different transport mechanisms are induced by electric current such as Electromigration, electro-osmosis, electrophoresis, and diffusion (Acar et al. 1995). It has been found that using upward electrokinetic remediation process 67% of phenanthrene could be removed after 6 days from contaminated soil (Wang et al. 2007). Electrokinetic treatment combined with

different surfactants and complexing agents has been used to increase desorption and solubility of contaminant. It is observed that 70% phenanthrene was removed after 30 days from kaolin clay using electrokinetic treatment combined with 1% Tween 80 and 0.1 M EDTA (Alcántara et al. 2012).

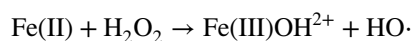
Chemical treatment

Chemical oxidation

Chemical oxidation treatment involves redox (oxidation/reduction) reactions that involve electron transfer from one chemical to other chemicals. This treatment converts hazardous contaminants into less toxic or nonhazardous compounds (Sharma et al. 2016; Verma and Haritash 2019). In recent days, advanced oxidation processes (AOPs) involving different oxidants have been used for treatment of PAH-contaminated soils. Different types of oxidants such as hydrogen peroxide, persulfate, ozone, Fenton's reagent, persulfate, peroxymonosulfate, and potassium permanganate have been used for the remediation of oil-contaminated soil via chemical oxidation reactions (Goi et al. 2006; Do et al. 2009, 2010; Yen et al. 2011). Chemical oxidation reactions have been used for treatment of oil or PAH-contaminated soils, different oxidants can be added to soil to oxidize contaminants (Tsai and Kao 2009; Rivas 2006).

Fenton's reagent

Fenton's reagent (Fe(II)–H₂O₂) for Chemical oxidation remediation uses hydrogen peroxide as oxidant in the presence of ferrous sulfate to generate free radicals, i.e., hydroxyl radicals (OH·).



These radicals are powerful oxidants (Fenton 1894). The use of the Fenton's reagent for chemical oxidation process has been found effective for the remediation of soil contaminated by polycyclic aromatic hydrocarbons (PAHs). Fenton oxidation treatment efficiency on PAH-contaminated soils has been found in different laboratory scale experiments. Soil samples contaminated with fluoranthene, benzo(b)fluoranthene, and benzo(a)pyrene were treated using Fenton oxidation process. Soil samples were mixed with water to form suspension (2 g soil sample in 10 ml water) in which 0.01 M Fe(II) was added with successive addition of H₂O₂. The suspension was then magnetically stirred for 24 h process to allow Fenton oxidation at room temperature with no pH adjustment. After 24 h oxidation process, high efficiency for PAH removal was observed, with removal of 85.7% fluoranthene, 87.4% benzo(b)fluoranthene, and 88.6% benzo(a)pyrene (Flotron et al. 2005). 24 different PAHs (2–6 ring) in different nine

contaminated soil samples were effectively degraded using Fenton's reagent. PAH degradation efficiency was 40–86% (20 g soil in 10 ml water) in the presence of 30% H_2O_2 and 4 mM Fe(II) at 70 °C and 3 pH (Jonsson et al. 2006). So, Fenton oxidation process could be used as an efficient remediation technique for PAH-contaminated sites.

Ozone

Ozone is one of the stronger oxidizing agents for chemical oxidation technique, which can be used for remediation of PAH-contaminated soil. During ozonation, the ozone molecule may directly attack double bonds and can form reactive hydroxyl radicals. Different intermediates such as quinone and biphenyl-type products are formed in ring cleavage radical oxidation process (Yao et al. 1998). In situ ozone treatment for soils can be done by injecting gaseous ozone which is the most advanced method and by injecting aqueous ozone because gaseous ozone is more effective than aqueous ozone. It was found that 20% PAHs remain in soil after gaseous ozone treatment and 40% PAHs remain in soil after aqueous ozone treatment (Masten and Davies 1997). This is due to easy and more diffusivity of gaseous ozone, resulting in more contact between contaminants and ozone (oxidizing agent). In an investigation, it has been shown that ozone could be used to remove phenanthrene from phenanthrene-spiked farm soils. At least 50% of phenanthrene level reduction in air-dried soils was achieved when soil samples were exposed to ozone at 20 ppm for 6 h (O'Mahony et al. 2006). From PAH-contaminated soil 95% of phenanthrene removal was achieved with ozonation for 2.3 h at an ozone flux of 250 mg h^{-1} , 91% of pyrene and 50% of chrysene were removed using ozone flux of 600 mg h^{-1} for 4 h (Masten and Davies 1997). Other similar studies also find the efficiency of ozone around 90–95% in removal of total petroleum hydrocarbons or diesel fuel from sand or unsaturated soils (Shin et al. 2005; Yu et al. 2007).

Other oxidants

Several studies have investigated that other alternative oxidants like Persulfate/Fe(II), Peroxymonosulfate (PMS), Persulfate, H_2O_2 , and permanganate can also be used for chemical oxidation treatment for diesel and fuel oil (Do et al. 2009, 2010; Yen et al. 2011). The feasibility of KMnO_4 (potassium permanganate) as an oxidant for removal of PAH from contaminated soil has been investigated and it has been found that there is significant reduction in PAHs (benzo(a)pyrene-72.1%, pyrene-64.2%, phenanthrene-56.2% and anthracene-53.8%) present in soil (Brown et al. 2003).

Photocatalytic degradation

Photocatalytic degradation is a process in which photocatalysts are used to stimulate oxidizing reactions, i.e., photo-reaction. This process is used for treatment of petroleum-contaminated soils to destroy organic contaminants in the presence of the light radiation. To enhance the degradation rate of fluoranthene, a photocatalytic solar reactor was developed. During photocatalysis process, both TiO_2 and H_2O_2 were used for fluoranthene degradation. Fluoranthene degradation efficiency was found 99% in the presence of both TiO_2 and H_2O_2 . However, lower fluoranthene degradation (83%) was observed when only TiO_2 was present as catalyst (Rababah and Matsuzawa 2002b). The photocatalytic degradation of phenanthrene, pyrene, and benzo(a)pyrene on soil surfaces using titanium dioxide TiO_2 in the presence of ultraviolet (UV) light was investigated in a photodegradation chamber at 30 °C. Photocatalyst TiO_2 (0.5%) accelerated the photodegradation process of phenanthrene, pyrene and benzo(a)pyrene significantly as compared to degradation in absence of TiO_2 catalyst, their half-lives being reduced from 533.15 to 130.77 h, 630.09 to 192.53 h and 363.22 to 103.26 h, respectively. There is synergistic effect of ultraviolet light and TiO_2 catalyst for PAHs degradation in soil (Zhang et al. 2008). Phenanthrene degradation on soil surface using photocatalysis under UV-irradiation was investigated where nanometer anatase TiO_2 was used as photocatalyst. Soil samples spiked with phenanthrene, loaded with TiO_2 (0 wt%, 1 wt%, 2 wt%, 3 wt%, and 4 wt%) exposed to UV light for 25 h. It has been observed that TiO_2 significantly increase the degradation rate of phenanthrene with half-life reduced from 45.90 to 31.36 h for 0 wt% and 4 wt%, respectively (Gu et al. 2012). So, TiO_2 is an efficient photocatalyst which has been used for oxidation of PAHs through photocatalytic degradation for treatment of oil polluted soils.

Biological treatment

Bioremediation: role of microbes in PAH degradation

Over the last two decades, another method in which microorganisms are used to degrade PAHs (bioremediation) is more accepted. Bioremediation is considered an ecofriendly and sustainable remediation technique and has recently gained considerable interest all around the globe (Sharma and Reddy 2004; Reddy and Adams 2015). The natural degradative potential of microorganisms, i.e., bacteria, yeasts, fungi, and algae is used in bioremediation of contaminants to convert them into less toxic compounds or into water and carbon dioxide (Alexander 1994). The microorganisms can produce a number of enzymes to detoxify and mineralize PAHs eventually leading to its degradation. The microorganisms

isolated from contaminated sites show more degradation ability as these microorganisms have been adapted to the polluted environment and can survive in the presence of the pollutant. A very challenging task in bioremediation processes is the confirmation of the first step, i.e., which microorganism should be used for degradation and endpoint, i.e., the end products. For efficient contaminant degradation the microbe must be present in favorable environmental conditions, there are various factors affecting biodegradation are pH, temperature, nutrients, and metabolites (Boopathy 2000). Degradation of PAHs could be done by using various naturally occurring soil microorganisms, i.e., aerobic and anaerobic microorganisms belong to genera *Pseudomonas*, *Alcanivorax*, *Microbulbifer*, *Mycobacteria*, *Sphingomonas*, *Micrococcus*, *Alcaligenes*, *Ralstonia*, *Paenibacillus*, *Bacillus*, *Aeromonas*, *Xanthomonas*, *Arthrobacter*, *Acinetobacter*, *Corynebacterium*, *Enterobacter* and others (Bayoumi 2009; Haritash and Kaushik 2016). Basidiomycetes *Pleurotus ostreatus* and *Irpex lacteus* were used for fungal bioremediation of soil contaminated with creosote (50–200 mg kg⁻¹ PAH) from a wood-preserving plant. 55–67% PAH removal was observed in *P. ostreatus* treatments (86–96% of 2-rings PAHs, 63–72% of 3-rings PAHs, 32–49% of 4-rings PAHs and 31–38% of 5–6-rings PAHs) and 27–36% PAH removal was observed in *I. lacteus* treatments (47–59% of 2-rings PAHs, 33–45% of 3-rings PAHs, 9–14% of 4-rings PAHs and 11–13% of 5–6-rings PAHs) in 120 days (Byss et al. 2008). There are some other fungal species, mainly *Aspergillus* and White Rot Fungi, such as *Phanerochaete chrysosporium*, *Bjerkandera adusta*, and *Pleurotus ostreatus*, have been documented for efficient biodegradation of PAHs (Haritash and Kaushik 2009, 2016). From various studies, several PAH catabolic genes encoding PAH catabolic enzymes have been characterized. Mono-oxygenases and PAH ring-hydroxylating dioxygenase enzymes were identified in *Novosphingobium pentaromativorans* proteome analysis. These enzymes are involved in the degradation of PAHs including phenanthrene, pyrene, and benzo(a)pyrene (Lyu et al. 2014). *Serratia marcescens* L-11 strain has an ability to produce lipases and aromatic ring cleavage enzyme (catechol 1, 2-dioxygenase). It can degrade phenanthrene, anthracene, fluorene, and pyrene (Pandey et al. 2012). Similarly, *Pseudomonas aeruginosa* PSA5 and *Rhodococcus* sp. NJ2 isolated from petroleum sludge were investigated for B(a)P degradation. It was observed that various PAH catabolic enzymes such as salicylate hydroxylase, 2-carboxy-benzaldehyde dehydrogenase, catechol 1,2-dioxygenase, and catechol 2,3-dioxygenase were differentially expressed in both bacterial species involved in B(a)P degradation (88% and 47% by *Pseudomonas aeruginosa* PSA5 and *Rhodococcus* sp. NJ2, respectively) (Mishra and Singh 2014). Various genes are involved in PAHs degradation and these were upregulated after exposing the microbe to PAHs (Lyu et al.

2014). In order to develop an effective sustainable strategy of bioremediation of PAHs, the characteristics, as well as the metabolic potential of the microbes need to be better understood in PAH-contaminated areas.

In situ bioremediation

Bioaugmentation Bioaugmentation is a technique which involves the addition of microorganisms (indigenous or exogenous) to the contaminated sites/soils, these microorganisms degrade the contaminants. This bioremediation technique is used where natural degrading microbes are present in low number or absent. Microbes are selected on the basis of their metabolic capacity to degrade contaminant (Boopathy 2000). Various researches have been done to study the effects of microorganism addition into contaminated soil (bioaugmentation) for in situ PAH degradation. It has been studied that microbial communities are efficient for PAH degradation under aerobic as well as anaerobic conditions (Kiamarsi et al. 2018). Pollutant bioavailability, survival of microorganism and their enzymatic catabolic activities are important for bioaugmentation (Heinaru et al. 2005). PAH degradation in soil by augmentation with specific isolated bacteria or fungus has been documented in various studies. *Scopulariopsis brevicaulis* PZ-4 isolated from an aged polycyclic aromatic hydrocarbon (PAH)-contaminated soil was found to have the ability to degrade PAHs. In a PAH-contaminated soil, *Scopulariopsis brevicaulis* PZ-4 removed 77% of total PAHs and the highest removal of PAHs occurred for phenanthrene (89%) and benzo(a)pyrene (75%) after incubation for 28 days (Mao and Guan 2016). Fungal isolate, *Penicillium* sp. 06, was effective at oxidizing a range of PAH in petroleum-contaminated soils. After 28 days of incubation, 89% of the phenanthrene presents in oily waste residues from the petrochemical refining industry in Singapore oxidized by *Penicillium* sp. 06. This isolate could also oxidize more than 75% of the acenaphthene, fluorene, and fluoranthene after 30 days of incubation (Zheng and Obbard 2003). Enhanced fluorene degradation in soil slurry system by augmenting with *Absidia Cylindrospora* a fungal isolate has been reported. In soil slurry system augmented with *A. Cylindrospora* more than 90% of the fluorene was removed after 288 h, while in nonfungal soil slurry 576 h contact time required for 90% removal of the fluorene (Garon et al. 2004). This is an efficient bioremediation technique which can also be used in ex situ remediation processes.

Biostimulation The process of environmental modification through addition of nutrients (nitrogen, phosphorous, and carbon, organic biostimulants), and oxygen (electron acceptor) to stimulate the activity of contaminant/oil-degrading indigenous microorganisms is known as biostimulation. These nutrients are the building blocks of life, therefore

these nutrients allow microbe to synthesize necessary enzymes to degrade the contaminant. It is one of the important strategies for increasing the efficiency of bioremediation of crude oil/PAHs in soil (Garon et al. 2004).

Addition of nutrients to PAH-contaminated soil has been shown increased microbial biomass and activity, therefore results in enhanced degradation efficiency in soils. Inorganic nitrogen and phosphorous were added for in situ bioremediation of PAHs in creosote-contaminated soil of wood-preserving plant in Norway. The addition of nutrients stimulated PAH degradation rate in the topsoil and the aquifer sand (Breedveld and Sparrevik 2000). The influence of addition or amendments of nutrients in crude oil-contaminated soil has been investigated where crude oil biodegradation in soil was observed for fertilized (soil added with nitrogen, phosphorus, and potassium) and unfertilized soil for 150 days. It was observed that fertilized soil has high biodegradation efficiency (62%) as compared to unfertilized soil (47%) (Chaineau et al. 2005). Biostimulation of hydrocarbon-degrading bacterial community with mineral nutrient and surfactant solution in crude oil-contaminated soil has been investigated and, found 39.5% reduction of total hydrocarbon content (Zucchi et al. 2003). Similarly, Abed et al. (2015) investigated that addition of NH_4Cl and NaH_2PO_4 as nitrogen and phosphorus sources for biostimulation of oil-contaminated desert soil. They observed that after addition of nutrients oil removal efficiency has been increased by 20%. Organic biostimulants such as phycocyanin (a proteic emulsifier extracted from the *Spirulina platensis* biomass) and inactive biomass of *S. platensis*, or ammonium sulfate are effective in bioremediation application. The soil contaminated with 4% of diesel or biodiesel was biostimulated for 60 days with these organic biostimulants. The biomass of *S. platensis* was found most effective biostimulant for diesel removal as after 60 days of biostimulation 63.89% of 4% diesel was degraded and the extracted phycocyanin of *Spirulina platensis* was found most effective biostimulant for biodiesel removal as after 60 days of biostimulation a biodegradation value of 88.75% for biodiesel was obtained (Decesaro et al. 2017). Similar to bioaugmentation, this technique can also be used in in situ or ex situ remediation processes.

Bioventing It is the most common in situ remediation treatment that involves supplying air or oxygen through wells to contaminated soil (in the unsaturated zone) to stimulate growth of the indigenous microorganisms. Soils contaminated by petroleum hydrocarbons have been successfully remediated using this technique (Hinchee 1993). A pilot-scale bioventing for remediation treatment of polycyclic aromatic hydrocarbons including high-ring PAHs such as pyrene, fluoranthene, and benzo(a)anthracene was done to treat a 15.2 m² area by The Reilly Tar and Chemical Corporation site in St. Louis Park, Minnesota. Preliminary

results of this study showed 62% reduction in 2-ring PAHs; 50% reduction in 3-ring PAHs; 31% reduction in 4-ring PAHs; 20% reduction in 5-ring PAHs; and 24% reduction in 6-ring PAHs (Alleman et al. 1995). 93% of phenanthrene was removed after 7 months of bioventing treatment of artificially phenanthrene-contaminated soil (1000 mg/kg soil) under Optimum conditions of mineralization (humidity=60% WHC; C/N/P=100:20:1) (García Frutos et al. 2010). Maximum degradation effectiveness of 85% was observed for remediation of 4 wt% of B20 (blend of diesel and biodiesel fuel)-contaminated soil (clay) through bioventing (Thomé et al. 2014).

Ex situ bioremediation

Landfarming Landfarming is a simple bioremediation technique in which contaminated soil is excavated, transported to the landfarming site and spread over a prepared bed and periodically tilled (turned over that provides aeration) until pollutants are degraded. Contaminants are degraded and transformed by microbiological metabolic processes and by oxidation (Riser-Roberts 1998). After 3 months of landfarming treatment, there is 63% reduction in total PAH concentration in a field contaminated mainly due to high concentrations of PAHs (1140 mg/Kg dry weight). There is up to 79% reduction in PAHs concentration with 2, 3 and 4 rings (Picado et al. 2001). Similarly, landfarming was used for bioremediation of creosote-contaminated soil in South Africa. After 6 months of treatment low molecular weight PAHs, i.e., 2–3 ring PAHs (naphthalene, anthracene, phenanthrene, and fluorene) were removed from soil but high molecular weight PAHs were still found in soil after 6 months treatment. At the end of treatment, i.e., after 10 months 76–87% of 4–5 ring PAHs (86.8% pyrene, 78.64% B(a)P, 76% chrysene and 85.5% fluoranthene) were removed (Atagana 2004). Effective remediation of diesel-contaminated soils at former military base at Resolution Island, Nunavut was done using landfarming for 3-year period with rototilling. 80% diesel fuel level was reduced after 3 years of landfarming treatment (Paudyn et al. 2008). Apart from PAHs, other petroleum hydrocarbons such as diesel-range organics, trimethylbenzenes, gasoline-range organics and BTEX compounds can also be degraded using landfarming (McCarthy et al. 2004).

Composting The process in which microorganisms (mesophilic and thermophilic) degrade organic contaminants at elevated temperature, i.e., 55–66 °C, is known as composting. During this process, microorganisms release heat results in an increase in temperature which further results in more solubility of contaminants and higher microbial activity in compost. For composting, contaminated soil is transferred to the composting pad (Namkoong et al. 2002).



Various studies have been done to investigate the efficiency of composting to degrade PAHs. An investigation on spent mushroom compost was done for bioremediation of soil contaminated with PAHs and it was observed that naphthalene, phenanthrene, benzo(a)pyrene and benzo(g,h,i)perylene was completely degraded after 48 h at 80 °C (Lau et al. 2003). Remediation of soil contaminated with PAHs through composting has been investigated in thermally insulated composting chamber. Mushroom compost, which consists wheat straw, chicken manure, and gypsum, has been used for this investigation. Substantial PAH removal or degradation has been observed during composting. At the end of 54 days of composting 20–60%, PAHs were removed, and after another 100 days 37–80%, PAHs were removed (Sasek et al. 2003). Diesel-, coal tar- and coal ash-contaminated soil mixed with compost results in enhancement of PAHs bioavailability and its increased removal rate up to 90% (Wu et al. 2013).

Phytoremediation Phytoremediation or plant-assisted bioremediation can be defined as an in situ technique that uses green plants and associated microorganisms to remove (extract, degrade, or immobilize) contaminants (PAHs) from environment (soil). It is an ecofriendly and cheaper alternative to other physical and chemical treatments, which prevents excavation of soil from contaminated sites. Different phytoremediation techniques such as phytoextraction, phytotransformation, phytostabilization, phytodegradation rhizodegradation can be used for treatment of oil-contaminated soils (Germida et al. 2002; McCutcheon, and Schnoor 2003). Plant species secrete various enzymes such as monooxygenase, dioxygenase, dehydrogenase, hydrolase, peroxidases, and dehalogenase into the soil which have ability to transform or degrade aromatic contaminants (Campos et al. 2008). Various factors such as contaminant nature, soil properties, bioavailability of contaminant and type of plant affect phytoremediation efficiency (Sreelal and Jayanthi 2017). However, few studies have investigated efficiency of plants for PAH remediation in soils, different types of plants and grasses such as *Festuca arundinacea*, ryegrass (*Lolium multiflorum*, *Lolium perenne*), *Dactylis glomerata*, *Festuca rubra*, yellow sweet clover (*Melilotus officinalis*), *Lotus corniculatus* (birdsfoot-trefoil), *Trifolium pratense* (red clover) and *Trifolium repens* (white clover) are found to degrade different PAHs like naphthalene, acenaphthylene, acenaphthene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, benzo(g,h,i)perylene, dibenz(a,h)anthracene (Parrish et al. 2005; Smith et al. 2006; Rezek et al. 2008). It has been observed that ryegrasses could effectively reduce the amount of mixture of hydrocarbons included n-alkanes, pristane, hexadecane, phenanthrene, anthracene, fluoran-

thene and pyrene (Gunther et al. 1996). In another study, it has been found that after 6 months of phytoremediation using three plant species *Medicago sativa* (alfalfa), *Panicum virgatum* (switch grass), and *Schizachyrium scoparium* (little bluestem grass) there is 57% reduction of total PAHs in soils collected from a former MGP (manufactured gas plant) in Newark, New Jersey (Pradhan et al. 1998). Similarly, phytoremediation efficiency of Arctared red fescue and annual ryegrass was examined after planted together in crude oil- or diesel-contaminated soil. Results showed lower concentrations of TPH in contaminated soils planted with both plants (Reynolds and Wolf 1999). A pot-culture experiment for reduction in petroleum hydrocarbons was done using five different plant species, *Echinacea purpurea*, Fawn (*Festuca arundinacea*), Fire Phoenix, *Gaillardia aristata* and Alfalfa (*Medicago sativa*). These plants significantly remove TPH including saturated hydrocarbon, aromatic hydrocarbon, asphaltene, and polar compound (Liu et al. 2012). Phytoremediation of contaminated soils can be enhanced along with different technologies such as bioremediation or electrokinetic treatment. It was observed that it is possible to enhance removal of phenanthrene and anthracene in soils by electrophytoremediation with *Brassica rapa* (Cameselle and Gouveia 2019). 47% TPH removal was observed using *Medicago sativa* and the TPH removal rate was found to increase up to 68% for bioaugmentation assisted phytoremediation where *Pseudomonas aeruginosa* was used for bioaugmentation (Agnello et al. 2016). There is synergistic effect of bioremediation and phytoremediation for TPH removal (Chaudry et al. 2005).

The remediation technologies as discussed above, i.e., physical, chemical, and biological treatment are effective for PAH degradation at contaminated sites. In order to achieve high efficiency, one has to choose the best remediation technologies as efficiency of these technologies depend on various factors such as type of soil, toxicity associated with contaminant, and environmental conditions. Each of these remediation technologies is associated with their advantage and disadvantage (Lim et al. 2016) with some opportunities and challenges and discussed in Table 4.

Future outlook and perspective

The contamination of soils with PAHs is inescapable in some sectors such as petroleum industry. A number of treatment methods for PAH-contaminated soils have been developed to remediate such sites. Some of these treatment methods have high efficiency in field-scale application. The understanding of contaminated site, i.e., site-specific conditions such as contaminant nature, soil properties, and weather is important to choose better remediation treatment method as some of them are not efficient for sites

Table 4 Analysis of strengths, weaknesses, opportunities, and challenges (SWOC) of techniques for soil remediation

Classification	Treatment method	Strength	Weakness	Opportunities	Challenges	Cost
Physical treatment	Solvent extraction/soil washing	Volume reduction or minimization of contaminants, implement easily and quick	Contaminant transfer to another phase, large amount of solvent required, high cost	Investigate effective and environmental friendly solvent, recycling/regeneration of solvent	May not be effective in treatment of clayey or silty soils, adsorption of solvent into soil particles	Moderate
Thermal treatment	Thermal desorption	Volatile contaminants treated over a broad range of moisture contents, high removal efficiency, no excavation required for in situ treatment	Cost-ineffective for large volumes of contaminated soils, volatilization and release of contaminants from the soil into the air at high temperatures, less efficiency at low temperature	Investigate different factors such as temperature, soil matrix, contaminant nature and concentration, and moisture content, which effect thermal desorption	Limited to volatile contaminants	Moderate
	Microwave frequency heating	High efficiency, quick, treat significant volumes of contaminated soil	High costs, affected with high moisture content	Investigate geochemical changes after treatment	Microwave exposure may cause health problem	High
	Vitrification	Easily applied for remediation of highly contaminated soils, destroys contaminants quickly	High cost, transforms treated soil into glassy composition	Investigate vitrified end products which may be recycled or used as other reusable materials	Collection and treatment of off-gases, utility of end product	High
	Electrokinetic treatment	Minimal requirement of equipment, safe, suitable for clays with low permeability and heterogeneous soil, used for in situ or ex situ treatment, cost-effective, low electrical energy	Time-consuming, microbial activity may be affected as a result of soil nutrients mobilization	Investigate environmental friendly electrolytes, interaction between electrokinetic parameters and contaminants	Not efficient in low contaminant concentration	Moderate
Chemical treatment	Chemical oxidation (AOPs oxidants, i.e., Fenton's reagent, ozone)	Oxidizes organic contaminants simply, efficiency not modified due to toxicity of contaminant	High treatment cost, treatments of extracts, excavation, pH dependency, natural microorganisms destroyed	Investigate other oxidants with in situ application and optimization of parameters such as pH and oxidant concentration.	Restricted by low soil permeability, oxidant high reactivity with soil components	High
	Photocatalytic degradation	Pollutant removal at very low concentration, complete degradation of organic contaminant within few hours, efficiency not modified due to toxicity of contaminant	High cost, highly dependent on various parameters such as pH, temperature, amount and nature of photocatalyst, intensity and time of light irradiation,	Investigate active and cheap photocatalyst, and optimization of parameters to achieve high efficiency	Not suitable for commercial application for volatile contaminants removal	High
Biological treatment	Bioaugmentation	Natural process and perceived by public, cheaper cost, environmental friendly, contaminants transform into nonhazardous substance, safe	Time-consuming, contaminant toxicity and bioavailability, exogenous microbes decline later addition to a site, i.e., failure of bioaugmentation	Investigate conditions which are required to make bioaugmentation an effective bioremediation tool	Competition between added exogenous microbes and indigenous microbes for limited nutrients	Low



Table 4 (continued)

Classification	Treatment method	Strength	Weakness	Opportunities	Challenges	Cost
	Biostimulation	Natural process and perceived by public, environmental friendly, contaminants transform into nonhazardous substance, safe, native microbes well adapted to environment	Time-consuming, contaminant toxicity and bioavailability, additives for biostimulation not readily available to native microbes	Investigate effects of biostimulation on the native microbial community structure and metabolic activities	Additives might promote growth of other microbes which are not involved in degradation and create competition with microbes involve in degradation	Low
	Bioventing	Natural process and perceived by public, cost-effective, environmental friendly, high efficiency, safe, contaminants transform into nonhazardous substance	Not suitable for volatile contaminants, cannot be used for surficial soils, time-consuming, contaminant toxicity and bioavailability	Off-gases treatment in case of volatile contaminant	Suitable only for less volatile contaminant	Moderate
	Landfarming	Natural process and perceived by public, environmental friendly, extensive primary assessment of contaminated site prior to treatment not required, contaminants transform into nonhazardous substance, safe	Require large operating space, unfavorable environmental conditions for microbial activities, additional cost due to excavation, time-consuming, contaminant toxicity and bioavailability, low efficiency as compared to other bioremediation treatment methods	Investigate the capability in complex contaminant degradation	Contaminated soil and dust particles spread during excavation and transportation, weather sensitive, ground water contamination	Moderate
	Composting	Natural process and perceived by public, low cost, simple operation, high microbial diversity high efficiency, environmental friendly, parameters such as moisture content, nutrients, and pH can be controlled, no surface mixing, contaminants transform into nonhazardous substance, safe	Require large area, time-consuming, contaminant toxicity and bioavailability, capture off-gases is difficult	Field studies with optimized environmental conditions and microbial consortia for contaminant removal	Controlling moisture, nutrients and pH during treatment	Low
	Phytoremediation	Low cost, environmental friendly, large-scale operation, low installation and maintenance cost, soil structure preservation, safe, plants can be easily monitored	Time-consuming, slow plant growth rate, contaminant toxicity and bioavailability	Treatment of sites contaminated with complex mixture of different contaminants	Plant growth requirement (climate, altitude and temperature)	Low



comprising soils with low permeability or having mixed contaminants. In addition to these site-specific conditions, the selection of remediation technology also depends on their advantages, limitations, cost of alternative remediation method, implementation applicability, and probable environmental impact. So, selection of the best remediation method for field-scale application is very crucial step for PAH removal from contaminated sites. However, each remediation method has its own advantages and constraints, and any single treatment method cannot be used universally for PAH removal. Therefore, two or more remediation methods could be integrated/combined and investigated in future research for different type of soils as enhanced PAH degradation/removal efficiency could be achieved with integrated treatment technologies/methods. Further, comprehensive research is essential to find precisely the existing condition of PAH-contaminated sites.

Conclusion

Soil pollution arises from different industrial activities, open burning of solid waste, combustion of fossil fuels, etc., and is a critical challenge due to emission of PAHs and its harmful effects on human health and soil ecosystem. This review is an attempt to overview the effects of PAHs on physicochemical and microbial properties of soil, as well as the remediation techniques for reclamation of soils contaminated with PAH. The PAH-contaminated soils may hold the persistent pollutants for long period of time depending on its physicochemical properties (pH, grain size, porosity and organic carbon) and its microbial diversity. Considering the toxic, mutagenic, and carcinogenic effects of PAHs, it is imperative to devise a sustainable, cost-effective, and universally adopted treatment method. Whereas the physical and chemical methods are cost and energy intensive, biological methods for reclamation of PAH-contaminated soils are gaining popularity. Landfarming, bioventing, composting, phytoremediation are some of the biotreatment technologies in addition to microbial degradation. Factors like availability of nutrients, favorable environment, microbial adaptation, and their catabolic activity are important regulators of PAH biodegradation. Depending on the chemical structure of PAH, its binding affinity to soil, toxicity profile, characteristics of soil substrate, possible future use of soil, availability of infrastructure, and risk analysis of different treatment options, a suitable method or an integrated approach of two or more methods may be adopted for reclamation of PAH-contaminated soils sustainably. Screening of genetic makeup of native flora and fauna for stimulated microbial activity may be foreseen as a potential tool for remediation of PAH contamination.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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