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Review

Comprehensive review of polycyclic aromatic hydrocarbons in water sources, their effects and treatments



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- PAHs ranged from 0.03 ng/L to 8,310,000 ng/L in aquatic environments.
- PAHs impact microorganisms, plants, animals and humans.
- · GC/MS and HPLC have been widely used in PAHs analysis.
- · Adsorption and combined treatment techniques are most effective at PAHs removal.



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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are principally derived from the incomplete combustion of fossil fuels. This study investigated the occurrence of PAHs in aquatic environments around the world, their effects on the environment and humans, and methods for their removal. Polycyclic aromatic hydrocarbons have a great negative impact on the humans and environment, and can even cause cancer in humans. Use of good methods and equipment are essential to monitoring PAHs, and GC/MS and HPLC are usually used for their analysis in aqueous solutions. In aquatic environments, the PAHs concentrations range widely from 0.03 ng/L (seawater; Southeastern Japan Sea, Japan) to 8,310,000 ng/L (Domestic Wastewater Treatment Plant, Siloam, South Africa). Moreover, bioaccumulation of \sum 16PAHs in fish has been reported to range from 11.2 ng/L (*Cynoscion guatucupa*, South Africa) to 4207.5 ng/L (Saurida undosquamis, Egypt). Several biological, physical and chemical and biological techniques have been reported to treat water contaminated by PAHs, but adsorption and combined treatment methods have shown better removal performance, with some methods removing up to 99.99% of PAHs.

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1. Introduction

Water pollution and the lack of access to clean water are general global problems that result from the expansion of industrial and agricultural activities (Wang et al., 2019). In recent decades, organic compounds such as polycyclic aromatic hydrocarbons (PAHs) have commonly been observed in aquatic environments. Moreover, the number of new organic compounds arriving the worldwide market is increasing remarkably every year, and most of these compounds, including pharmaceuticals, pesticides, personal care products, and PAHs surfactants are used worldwide in high amounts in industrial activities, after which they are discharged in to various water bodies, where they can persist, causing severe health and environmental problems (Zambianchi et al., 2017). Recent studies have stated the occurrence of organic pollutants, such as PAHs, in various aquatic systems such as influent and effluent from wastewater treatment plants. groundwater, surface waters or seawater (Grandclement et al., 2017). Researchers have reported the presence of organic compounds in America (Gilliom, 2007), Africa (Edokpayi et al., 2017), Asia (Lin et al., 2017), Europe (Wen et al., 2017) and Oceania (Tremblay et al., 2016). In the current study, PAHs, which are toxic to living organisms, were investigated as organic pollutants. Chief sources of PAHs in the atmosphere comprise coal and wood combustion for automobile gases and domestic heating. Polycyclic aromatic hydrocarbons are also present as dry deposits on municipal surfaces (Walaszek et al., 2018).

Biological, Physical and chemical ways have been used for the elimination of organic contaminants from water and wastewater (J. He et al., 2017). In previous studies, physical/chemical techniques such as adsorption (Altmann et al., 2015), advanced oxidation processes (AOP), and membrane methods (Paredes et al., 2018) as well as biological techniques such as activated sludge procedures (Falås et al., 2016) and anaerobic and aerobic processes (Torresi et al., 2019) have been reported for abatement of various organic pollutants, such as PAHs.

However, a comprehensive description of PAHs, including their characteristics, effects and treatment methods is currently lacking; therefore, this review was conducted to provide this information.

2. Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are organic pollutants produced by anthropogenic activities associated with industrialization and urbanization, as well as through natural activities (Fig. 1) (Qiao et al., 2018). To date, over 400 kinds of PAHs and their ramifications have been identified (Pan et al., 2006). PAHs are a group of hazardous organic compounds comprising of two or more benzene rings bonded in linear, angular of cluster arrangements. Most PAHs are colorless, white or paleyellow solids (Pogorzelec and Piekarska, 2018). PAHs in the environment are primarily of pyrogenic, petrogenic, and biologic origin (Hac-Wydro et al., 2019). Most PAHs are believed to originate from pyrogenic sources such as volcanoes and the combustion of petroleum products and plant materials (Gennadiev and Tsibart, 2013). PAHs of diagenetic or biogenic origin include those formed by plants, algae, microorganisms and phytoplankton or during slow alterations of organic matter (Rocha and Palma, 2019). PAHs are derived from the incomplete combustion of organic matter, such as transportation fuel, emissions from power plants and petroleum spills, coal mining, and other anthropogenic sources. Mostly PAHs are hydrophobic and lipophilic and therefore very difficult to biodegrade (Kronenberg et al., 2017). Table 1 shows PAH pollutants and their characteristics. There are several PAHs, although most regulations, analyses, and research typically focus on only 14 to 20 individual PAHs (Abdel-Shafy and Mansour, 2016). In comparing with high molecular weight PAHs (four or more rings), low molecular weight PAHs (two or three rings) are more degradable because of the fairly higher volatility and solubility of the former (Behera et al., 2018).

2.1. 16 PAHs

The USEPA has categorized 16 of the PAHs (Table 1) as prioritycontaminants based on their possible for human exposure, toxicity, frequency of occurrence at hazardous waste sites, and the extent of information available (Bojes and Pope, 2007). These 16 PAHs are including acenaphthene, benzo[*ghi*]perylene, chrysene, acenaphthylene, benz[*a*] anthracene, benzo[*b*]fluoranthene, anthracene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, fluoranthene, Indeno[1,2,3- cd]pyrene, naphthalene, phenanthrene, dibenz[*a*,*h*]anthracene, fluorene, and pyrene.

Acenaphthene (ACE) is obtained from creosote oil by distillation, which has various drawbacks such as a long production route, high energy consumption and strict operation conditions (Ye et al., 2016). Acenaphthene (ACE) is widely employed in different industries for the manufacture of dyes, pharmaceuticals, plastics, fungicides and insecticides. Because of the wide use of materials including ACE, its release and accumulation in the environment is currently posing a threat to many areas (Mallick, 2019). Acenaphthene ultimately settles on the ground or into ponds, lakes, and rivers. When acenaphthene is attached to particles in water or soil it might be taken up by plants or swallowed by animals (MDH, 2015a,b).



Fig. 1. PAHs sources.

Acenaphthylene (ACY) is a simple and stable aromatic hydrocarbon containing of naphthalene with an ethylene bridge (Fukumoto et al., 2011). Acenaphthylene is a vital intermediate material to many organic synthesis processes that is commonly applied during the production of advanced pigments, polymers and dyes (He and Liu, 2007). Riva et al. (2017) reported that ACE and ACY are objectively unique among PAHs as they include a carbon carbon double bond in their structure that allows them to rapidly react with all atmospheric oxidants containing OH as well as NO₃ radicals, Cl atoms and O₃.

Anthracene (ANT), which is mainly generated during the incomplete combustion of organic materials, is a common organic contaminant in water bodies that has been classified among priority contaminants owing to its mutagenicity, carcinogenicity, toxicity and bioaccumulation (Kalantari et al., 2019). Furthermore, anthracene is an extremely hydrophobic compound with low biodegradability because of its chemical stability (Rubio-Clemente et al., 2014).

Benz[*a*]anthracene (BaA) is not synthesized commercially. The chief source of BaA, in air, is the combustion of fuels and wood. BaA discharged into the atmosphere may be deposited onto water or soil. In surface water, benz[*a*]anthracene may volatilize, bind to suspended particles, or accumulate in aquatic organisms (Gray and Hall, 2014). Benz(a)anthracene, which has four rings, is remarked to be a human carcinogen and one of the most aggressive PAHs (Othman et al., 2012). Benz[*a*]anthracene (BaA) is hydrophobic (log K_{ow} = 5.6–5.9), with a high sorption capacity on particles and organic matter and a high tendency for accumulation in lipid-rich tissues (Bihanic et al., 2015).

Benzo[b]fluoranthene (BbF) is a common constituent of PAH complexes generated from the fossil fuels and tobacco, and is defined as a possible human carcinogen (Kim et al., 2011). The WHO (1998) has reported the occurrence of BbF in rainwater, snow and fog.

Benzo[k]fluoranthene (BkF), which is found in smoke from tobacco and polluted air, is a dangerous carcinogenic pollutant that appears to be increasing in aquatic systems (Pan et al., 2005). Moreover, BkF has been identified as a key toxicant impacting aquatic organisms (Kim et al., 2014). Benzo[*a*]pyrene (BaP) is a high molecular weight PAH that is produced as a consequence of incomplete combustion of organic substrates at temperatures between 300 °C and 600 °C and is found in products varying from coal tar to many foods, especially smoked and grilled meats, and tobacco smoke (Lee et al., 2019).

Benzo[*ghi*]perylene (BghiP) exemplifies a fascinating class of highly conjugated polyaromatic compounds formed by condensing benzenoid units and a vital group of fluorescent perylene dyes (Raouafi and Aloui, 2019). BghiP is a high molecular weight PAH compound with six benzene rings that is highly recalcitrant to degradation (Mandal and Das, 2018).

Chrysene (CHY) is lipophilic, slightly soluble in polar solvents such as alcohol and ether and moderately soluble in benzene and toluene. Because of its poor water solubility and low vapor pressure, chrysene is not easily removed from the environment (Biswas and Ghosh, 2014). Indeed, Diamante et al. (2017) reported that chrysene is one of the most persistent PAHs in the water column.

Dibenz[a,h]anthracene (DahA) is soluble in organic solvents like petroleum ether, ether, toluene and benzene, but insoluble in aqueous media. Dibenz[a,h]anthracene is adsorbed very intensely into sediments and particulate matter if discharged into water. As previously illustrated, it will not hydrolyze and volatilize. Additionally, it shows some bioconcentration in aquatic organisms that lack microsomal oxidase (Bhattacharya et al., 2014).

Fluorene (FL) is frequently derived from gas turbine engines, dieselfueled and gasoline-fueled engines, roofing tar, coke ovens, kerosenefueled stoves and oil flames (Ding et al., 2019). The fluorene scaffold comprises a unique structure comprising of a five-membered ring stacked in between two benzene rings. Therefore, fluorene has properties typical of cyclopentadienes as well as of benzenes. Fluorene is widely used as a ligand for the formation of metallocene-like complexes in the organometallic chemistry (Kaiser et al., 2019).

Fluoranthene (FLU), which is the most ubiquitous and abundant pyrogenic PAH (Lei et al., 2007), has a low water solubility of 0.25 mg/L that strongly decreases its bioavailability (Patel et al., 2019). Fluoranthene is produced from an activity such as wood burns or gasoline.

Table 1

Most reported polycyclic aromatic hydrocarbons (PAHs).

PHAs	Benzene rings	Chemical formula	Molecular weight (g/mol)	CAS number	References
Acenaphthene, (ACE) ^a		$C_{12}H_{10}$	154.2	83-32-9	Lerda (2011); Zelinkova and Wenzl (2015)
Acenaphthylene, (ACY) ^a		C ₁₂ H ₈	152.1	208-96-8	PubChem (2005); Zelinkova and Wenzl (2015)
Anthracene, (ANT) ^a		$C_{14}H_{10}$	178.2	120-12-7	Lawal (2017); Lerda (2011);
Benz[a]anthracene, (BaA) ^a		C ₁₈ H ₁₂	228.3	56-55-3	Gen and Hartwig (2012); Yan et al. (2004)
Benzo[b]fluoranthene, (BbF) ^a		$C_{20}H_{12}$	252.3	205-99-2	ILO and WHO (2017)
Benzo[<i>j</i>]fluoranthene, (BjF)		$C_{20}H_{12}$	252.3	205-82-3	Zhang et al. (2014)
Benzo[k]fluoranthene, (BkF) ^a		$C_{20}H_{12}$	252.3	207-08-9	Abdel-Shafy and Mansour (2016)
Benzo[<i>a</i>]pyrene, (BaP) ^a		$C_{20}H_{12}$	252.3	50-32-8	Lerda (2011)
Benzo[<i>ghi</i>]perylene, (BghiP) ^a		$C_{22}H_{12}$	276.3	191-24-2	ECHA (2018)
Chrysene, (CHY) ^a		C ₁₈ H ₁₂	228.2	218-01-9	Rachna et al. (2018)
Cyclopenta[cd]pyrene, (CcdP)		C ₁₈ H ₁₀	226.2	27208-27-3	Fabbri et al. (2006)
Dibenz[<i>a,h</i>]anthracene, (DahA) ^a		C ₂₂ H ₁₄	278.3	53-70-3	Zelinkova and Wenzl (2015)
Dibenzo[<i>a,e</i>]pyrene, (DaeP)		C ₂₄ H ₁₄	302.3	192-65-4	Lerda (2011)
Dibenzo[<i>a</i> , <i>h</i>]pyrene, (DahP)		C ₂₄ H ₁₄	302.3	189-64-0	Zelinkova and Wenzl (2015)

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Table 1 (continued)

PHAs	Benzene rings	Chemical formula	Molecular weight (g/mol)	CAS number	References
Dibenzo[<i>a</i> , <i>i</i>]pyrene, (DaiP)		C ₂₄ H ₁₄	302.3	189-55-9	Lerda (2011)
Dibenzo[<i>a</i> , <i>l</i>]pyrene, (DalP)		C ₂₄ H ₁₄	302.3	191-30-0	DeMarini et al. (2011)
Fluoranthene, (FLU) ^a		C ₁₆ H ₁₀	202.2	206-44-0	Abdel-Shafy and Mansour (2016)
Fluorene, (FL) ^a		C ₁₃ H ₁₀	166.2	86-73-7	Abdel-Shafy and Mansour (2016)
Indeno[1,2,3-cd]pyrene, (IcdP) ^a		$C_{22}H_{12}$	276.3	193-39-5	Lerda (2011)
5-Methylchrysene (5-Met)		$C_{19}H_{14}$	242.3	3697-24-3	PubChem (2005)
Naphthalene, (NAP) ^a		C ₁₀ H8	128.1	91-20-3	Lawal (2017)
Perylene, (PER)		$C_{20}H_{12}$	252.3	198-55-0	Zelinkova and Wenzl (2015)
Phenanthrene, (PHE) ^a		C ₁₄ H ₁₀	174.2	85-01-8	Lerda (2011)
Pyrene, (PYR) ^a		C ₁₆ H ₁₀	202-2	129-00-0	Lawal (2017)
^a Listed as 16 PAHs					

Fluoranthene sticks to small airborne particles that can be inhaled by people and animals or ultimately settle back onto the ground or into ponds, rivers or lakes (MDH, 2015a,b).

Naphthalene (NAP) is generated from coal tar, which is formed from heavy petroleum fractions during petroleum refining. Naphthalene is widely employed in pigments, 2-naphthol synthesis, and precursors for several dyestuffs (Sharma and Lee, 2015).

Phenanthrene (PHE) is a typical low molecular weight PAH with three fused benzene rings that is present at high levels in PAH-contaminated environments (Fu et al., 2018). Wang et al. (2019) stated that, in some heavily polluted waters such as petroleum wastewater, the concentrations of phenanthrene might be as high as 7.6–9.9 μ g/L. Mahvi and Mardani (2005) reported that PHE was present in all monitored street runoff samples in Tehran-Iran.

Pyrene (PYR) consists of four fused benzene rings made by incomplete combustion of fossil fuels such as low rank coal or biomass combusted at high temperature (800–1000 °C), especially in gasification or pyrolysis procedures (Wang et al., 2018). Pyrene is a key component of the PAH compounds broadly present in the environment (Makelane et al., 2019). Zhou et al. (2018) stated that pyrene is widely distributed in municipal sewage sludge from different countries.

2.2. Substituted PAHs

Most studies of polycyclic aromatic hydrocarbons (PAHs) have focused on homocyclic compounds. Nevertheless, two-thirds of the known aromatic compounds are heterocyclic with sulfur, oxygen, and/ or nitrogen in-ring substitutions of one or more carbon atoms. Ringuet et al. (2012) reported that, once in the atmosphere, PAHs can react with oxidants such as NO_x, O₃, and OH to procedure nitrated or oxygenated PAHs, as substituted PAHs. Substituted PAHs may be added in aquatic environments directly from atmospheric particulate matter or as fallout from rain (Idowu et al., 2019). The environmental and toxicological significance of nitrogen heterocyclic derivatives of PAHs (NPAHs) has been known in previous studies. Large differences in biological reactivity and chemical characteristics are possible to exist among PAHs and their NPAHs (Table 2). Nitrated PAHs were discovered in several environmental compartments with PAHs. While they are generally found in concentrations far lower than their parent PAHs, they may have important toxicity (Ozaki et al., 2010). The substitution of a carbon atom by a nitrogen atom makes the substances more polar and increases their water solubility (Pašková et al., 2009). Pašková et al. (2009) reported the presence of NPAHs in air, both freshwater and marine environments and groundwater. Another type of PAH is formed in the presence of oxygen. Specifically, oxygenated polycyclic aromatic hydrocarbons (OPAHs, Table 2) are organic compounds present in the atmosphere that are extremely toxic. Because OPAHs are semi-volatile compounds with lower vapor pressures than their parent PAHs, they are commonly adsorbed on the surface of airborne particulate matter (PM) (Filippo et al., 2015). As urban areas grow, undeveloped land and forests are being replaced by impervious types of surface cover such as commercial parking lots, roads, and residential driveways, which increases surface runoff during snow events and rain and works as a vital transport pathway for substituted PAHs (Sulfur-PAHs) in addition to entry of PAHs to urban streams (Witter and Nguyen, 2016).

2.3. Presence of PAHs in water and wastewater

PAHs have been detected in sediments, water sources, wastewater and crustaceans as mixtures and typically co-occur with other contaminants (Jaward et al., 2012; Ozaki et al., 2019). PAHs are released to the environment mostly as by-products of the combustion of fuels, but agricultural fires, industrial wastes, and cooking can rise the discharge of these toxic chemicals. The solubility of PAHs in water commonly diminishes as the molecular weight increases, while their boiling and melting

Table 2

Most reported substituted PAHs.

Compounds	Chemical formula	Molecular weight (g/mol)	CAS number
NPHAs 1-Nitronaphthalene (1-NNaph) 2-Nitronaphthalene (2-NNaph)	C ₁₀ H ₇ NO ₂ C ₁₀ H ₇ NO ₂	173.171 173.171	86-57-7 581-89-5
2-Nitrofluorene (2-NFluo)	C ₁₃ H ₉ NO ₂	211.22	607-57-8
9-Nitroanthracene (9-NA)	$C_{14}H_9NO_2$	223.231	602-60-8
9-Nitrophenanthrene (9-NPhen)	$C_{14}H_9NO_2$	223.231	954-46-1
3-Nitrophenanthrene (3-NPhen)	$C_{14}H_9NO_2$	223.231	17024-19-0
2 + 3-Nitrofluoranthene ($2 + 3$ -NFlt)	$C_{16}H_9NO_2$	247.253	892-21-7
1-Nitropyrene (1-NP)	$C_{16}H_9NO_2$	247.253	5522-43-0
2-Nitropyrene (2-NP)	C ₁₆ H ₉ NO ₂	247.253	789-07-1
4-Nitropyrene (4-NP)	$C_{16}H_9NO_2$	247.253	57835-92-4
7-Nitrobenzo[a]anthracene (7-NB[a]A)	$C_{18}H_{11}NO_2$	273.291	20268-51-3
6-Nitrochrysene (6-NChr)	$C_{18}H_{11}NO_2$	2/3.291	7496-02-8
1,3-Dinitropyrene (1,3-DNP)	$C_{16}H_8N_2O_4$	292.25	/5321-20-9
1,6-DINITROPYFENE (1,6-DNP)	$C_{16}H_8N_2O_4$	292.25	42397-64-8
1,0-DIHILIOPYTEHE (1,0-DINP)	$C_{16}\Pi_8N_2O_4$	292.23	42597-05-9
2 Nitrobonzo[a]pyrene (2 NP[a]P)	$C_{20}\Pi_{11}NO_2$	297.515	70021-42-0
6-Nitrobenzo[<i>a</i>]pyrene (6 -NB[<i>a</i>]P)	$C_{20}H_1NO_2$	297.313	630/1-90-7
o-minobelizolujpyrene (o-mbluji)	C201111102	237.313	05041-50-7
OPHAs			
1-Naphthaldehyde (1-Naph)	$C_{11}H_8O$	156.184	66-77-3
9-Fluorenone (9-Fluo)	C ₁₃ H ₈ O	180.206	486-25-9
9-Phenanthrenecarboxaldehyde (9-Phen)	C ₁₅ H ₁₀ O	206.24	4707-71-5
9,10-Anthraquinone (9,10-Ant)	$C_{14}H_8O_2$	208.21	84-65-1
1,4-Anthraquinone (1,4-Ant)	$C_{14}H_8O_2$	208.216	635-12-1
Benzo[a]fluorenone (B[a]Fone)	$C_{17}H_{10}O$	230.26	479-79-8
Benzo[b]fluorenone (B[b]Fone)	$C_{20}H_{12}$	252.31	205-99-2
Benzanthrone (Benz-one)	$C_{17}H_{10}O$	230.266	82-05-3
Benz[<i>a</i>]anthracene-7,12-dione (B[<i>a</i>] A-7,12-dione)	$C_{18}H_{10}O_2$	258.276	2498-66-0
Others			
Dibenzothiophene (DBT)	$C_{12}H_8S$	184.256	132-65-0
Benzo[b]naphtho[2,1-d]thiophene (BNT)	$C_{16}H_{10}S$	234.316	239-35-0

point increases (Adeniji et al., 2018). Adeniji et al. (2018) reported that four-ring and five-ring PAHs such as chrysene and benzo[*a*]pyrene are almost insoluble in water. Because of this characteristic, they can attach to the surface of particulate matter, and this mechanism is remarked the main transport pathway of PAHs from land and air to aquatic systems, as well as from the sea surface to lower depths (Vagge et al., 2018). Karyab et al. (2013) stated that PAHs generally enter water sources through dry and wet deposition, road runoff, leaching from creosoteimpregnated wood, industrial wastewater, petroleum spills, and fossil fuels combustion. Many researchers have reported PAHs in aquatic environments (Table 3); for example for drinking water PAHs at concentrations between 1.33 ng/L (for BaP in treated drinking water in Tehran, Iran) to 139,000 ng/L (for PHE in untreated drinking water in Lagos, Nigeria). Moreover, the concentrations of the PAHs ranged between 0.5 ng/L (for BaP in Northeastern China) and 1,138,000 ng/L (for PYR in South Africa) in rivers and lakes. In wastewater, PAHs have been found at levels ranging from 14 ng/L (for FLU in domestic wastewater in Jordan) to 8,310,000 ng/L (for BbF in domestic wastewater in South Africa). Finally, PAHs concentrations in seawater and groundwater ranged between 0.02 ng/L (for CHY in the Persian Gulf) and 46,600 ng/L (IcdP in the Timor Sea, Indonesia), and 0.1 ng/L (for BaP in North China) and 739.1 ng/L (for NAP in Near Huai River China), respectively.

2.4. Effects of PAHs on the environment

PAHs are widespread organic contaminants in the environment that are recognized to have carcinogenic and mutagenic effects, and to bioaccumulate in human and animal tissue (Adjiboye et al., 2011).

Table 3 Most reported PAHs in water source

PAH	Water sources	Concentration (ranges; ng/L or ng/g)
ACE	Drinking water	3.8 to 478.0
	Rivers and lakes	2.6 to 579,000.0
	Groundwater	0.4 to 148.7
	Wastewater	28.8 to 100.0
	Seawater	2.6 to 4200.0
ACV	Drinking water	0.6 to 1821.0
ACI	Rivers and lakes	2.7 to 537.000.0
	Groundwater	0.8 to 12.5
	Wastewater	16.6 to 65.9
	Seawater	4.5 to 4100
	Sediments	1.7 to 12.7
ANT	Drinking water	1.4 to 71.0
	Croundwater	0.1 to 195.6
	Wastewater	42.0 to 294.9
	Seawater	0.1 to 3350.0
	Sediments	2.0 to 658.0
	Drinking water	2.29 to 9.7
	Rivers and lakes	0.6 to 3200.0
BaA	Groundwater	0.1 to 5.8
	Segwater	45.5 to 0.0 to 17.490.0
	Sediments	0.2 to 152.0
	Drinking water	1.3 to 8.0
	Rivers and lakes	0.5 to 1,239,000.0
BaP	Groundwater	3.0 to 12.5
Dui	Wastewater	71.6 to 1,447,000.0
	Seawater	0.2 to 28,490.0
	Drinking water	2 1 to 24 0
	Rivers and lakes	1.2 to 7,800,000.0
DLC	Groundwater	1.9 to 39.3
BDF	Wastewater	82.0 to 8,310,000.0
	Seawater	0.0 to 32,050.0
	Sediments	<1 to 932.0
	Drinking water Rivers and lakes	4.6 to 24.0
	Groundwater	5.1 to 29.8
Bkf	Wastewater	100.0 to 203.8
	Seawater	0.0 to 1290.0
	Sediments	3.8 to 17,486.0
	Drinking water	2.0 to 8.0
	Rivers and lakes	0 to 11,700.0
BghiP	Wastewater	^a to 92 0
	Seawater	0.2 to 14.790.0
	Sediments	8.9 to 5153.0
	Drinking water	1.8 to 27.3
	Rivers and lakes	1.8 to 4300.0
CHY	Groundwater	0.1 to 71.2
	Seawater	20.7 to 112.3 0.1 to 42 710.0
	Sediments	0.9 to 193.0
	Drinking water	2.0 to 8.5
	Rivers and lakes	4.0 to 11,400.0
DahA	Groundwater	0.1 to 4.2
Duini	Wastewater	a 0.0.1.20.040.0
	Seawater	0.0 to 32,340.0
	Drinking water	4 0 to 41 000 0
	Rivers and lakes	5.6 to 2.480.000
E1	Groundwater	0.4 to 167.7
FL	Wastewater	20.0 to 234,000.0
	Seawater	0.2 to 1520.0
	Sediments	<1 to 52.0
	Drinking water	6.5 to 143,000.0
	Groundwater	4.2 to 2,490,000.0 2 0 to 50 6
FLU	Wastewater	14.0 to 2.340.000.0
	Seawater	0.0 to 6610.0
	Sediments	<1 to 24,857.0
	Drinking water	1.6 to 3.0
IcdP	Rivers and lakes	1.0 to 7200.0
	Groundwater	3.6 to 12.1

Table 3 (continued)

PAH	Water sources	Concentration (ranges; ng/L or ng/g)
	Wastewater	21.0 to ^a
	Seawater	0.0 to 46,600.0
	Sediments	0.4 to 552.0
	Drinking water	4.6 to 14,000
	Rivers and lakes	52.5 to 6900.0
NAD	Groundwater	2.1 to 281.1
INAF	Wastewater	40.0 to 47,000.0
	Seawater	75.9 to 7800.0
	Sediments	<1 to 68.7
	Drinking water	13.1 to 139,000
	Rivers and lakes	13.3 to 126,000.0
DUE	Groundwater	2.0 to 179.2
FHE	Wastewater	33.0 to 6,495,000.0
	Seawater	0.2 to 1080.0
	Sediments	5.7 to 410.0
	Drinking water	4.2 to 92,000
	Rivers and lakes	2.9 to 1,138,000.0
DVD	Groundwater	0.3 to 41.9
FIK	Wastewater	19.1 to 1,186,600.0
	Seawater	0.0 to 9870.0
	Sediments	2.8 to 27.1

^a Not Reported.

^b This table was extracted from Table A.1 in the Appendix.

PAHs also have detrimental impacts on the fauna and flora of affected habitats, ensuing in the uptake and accumulation of toxic chemicals via food chains (biomagnification), and in some instances, serious health issues and/or genetic defects in humans (Chauhan et al., 2008).

2.4.1. PAHs impacts on microorganisms

Abdel-Shafy and Mansour (2016) stated that PAHs and their epoxides are greatly toxic, carcinogenic and/or mutagenic to microorganisms. Al-Turki (2009) reported that high levels of PAHs can inhibit all microbial growth. Yan et al. (2019) investigated the co-occurrence patterns of the microbial community in PAH-polluted riverine sediments and found that microbes in heavier PAH-contaminated sediment had stronger relationships and were more centrally clustered within the network than those in the lower PAH-polluted sediment. Johnsen et al. (2002) reported that growth of bacteria on various PAHs led to slow bacterial growth and low cell yields.

2.4.2. PAHs impacts on fishes and aquatic animals

Several studies of wild fish have connected the occurrence of hepatic neoplasms and neoplasia-related toxicopathic liver lesions to PAH exposure as defined by SPAHs in sediments, PAH metabolites or fluorescent aromatic compounds (FACs) in fish bile, PAH-DNA adducts in liver, or components of the natural diet of these species (Collier et al., 2013). The bioaccumulation of PAHs in fish is described in the appendix (Table A.2). The PAHs in fish samples ranged from 0.0 ng/L (for ANT accumulation in Tilapia queneesis, Nigeria) to 4207.5 ng/L (for 16PAHs accumulation in Saurida undosquamis, Egypt). The acquired immune defense mechanisms of fish are the same as those for mammals and include cell- and humoral-mediated responses (Reynaud and Deschaux, 2006). Reynaud and Deschaux (2006) stated that, among innate immune parameters, several authors have focused on macrophage activities in fish exposed to polycyclic aromatic hydrocarbons. Macrophage respiratory bursts appear to be particularly sensitive to polycyclic aromatic hydrocarbons. Among acquired immune parameters, lymphocyte proliferation is extremely sensitive to polycyclic aromatic hydrocarbon exposure. Vignet et al. (2016) found that PAHs might disrupt fish reproduction. Hayakawa et al. (2016) stated that teleosts converted PAHs into monohydroxylated polycyclic aromatic hydrocarbons (OHPAHs) via cytochrome P4501A1; thus, OHPAH could have a toxic effect on teleosts. Paruk et al. (2014) detected polycyclic aromatic hydrocarbons in Common Loons (Gavia immer) wintering off coastal Louisiana.

3. Methods of measuring PAHs in aqueous solutions

PAHs are generally identified using analytical techniques that have been approved by organizations such as the United States Environmental Protection Agency (USEPA) or International Organization for Standardization (ISO). There are mainly three types of techniques used for their identification: chromatographic, immunoassay and spectrometric (Adeniji et al., 2018). Immunoassay techniques (EPA 4030 and 4035, Update III), which exist frequently as kits, are not popular due to their tendency to introduce strong biases to the final results (Adeniji et al., 2017). Among spectrometric methods, ultraviolet (UV) and infrared (IR) techniques are the most common; however, UV techniques (absorption and fluorescence), which are remarked sensitive and selective to aromatic compounds such as PAHs, are more frequently affected by interference caused by the presence of other compounds such as lipids. Moreover, the IR spectrometric technique, which is rapid and inexpensive, needs the sample to undergo a mandatory cleanup step after extraction before analytical determination (Adeniii et al., 2018). Chromatographic techniques for testing PAHs in environmental media have also been established and widely applied over the past few decades, with liquid and/or gas chromatography (LC and GC) being the prominent methods utilized (Poster et al., 2006). The most commonly reported methods for PAHs analysis in water sources are listed in Table 4.

Detector

Table 4

DAHe

Methods used to measure PAHs in aqueous solutions Method

Simplicity of operation, reduction in volume of solvents used, and the possibility of automation are some advantages of using gas chromatography (GC) and high-performance liquid chromatography (HPLC) to analyze PAHs (Gilgenast et al., 2011). Varlet et al. (2007) reported that one advantage of using GC/MS was its specificity allows the transitions of PAHs to be focused on. Moreover, use of GC/MS allows for the extraction to be optimized to improve the signal. Gilgenast et al. (2011) listed advantages of HPLC including the (1) prospect of observing the range of fraction collection by using an HPLC detector (refractive index, fluorescence or UV), (2) probability of using backflushing in the HPLC column to elute highly polar components, resulting in a substantial reduction in analysis time, and (3) amended recovery of PAHs and lower relative standard deviation values.

4. Removal of PAHs from aqueous solutions

During the last few decades, several researchers have concentrated on effective sequestering of organic contaminants from aqueous solution. A diversity of methods have been adopted including coagulation, chemical oxidation, membrane filtration, photocatalytic degradation, and adsorption (Khairy et al., 2018). All organic pollutants removal may be divided into biological and physical/chemical methods. Some of the utmost common

IOD^a or IOO

References

11110	method	Detector		(ng/mL, ng/g)	hererences
16 PAHs	GC/MS	Mass selective (MSD)	Gas carrier: Helium	LOD = 0.02 to 0.05	Soares et al. (2015)
ACE, ANT, FL, NAP, PHE	HPLC/FL	Fluorescence	Mobile phase: mixture of pure water, methanol and acetonitrile	NR ^b	Smol et al. (2014)
ACE, ANT, FL, FLU, NAP, PHE, PYR	HPLC/FL	Fluorescence	NR	LOD = 0.10 to 12.20	Zhang et al. (2018)
ACE, NAP, PHE	UV–Vis spectrophotometer	-	Maximum wavelengths (λ_{max}) were 220.5, 225.4 and 250.0 nm for ACE, NAP and PHE, respectively.	-	Balati et al. (2015)
16PHAs and 5NPAHs	GC/MS	Mass selective (MSD)	Mass Spectrometry: Selective ion monitoring (SIM)	LOD = $2(10^{-5})$ to 0.008 LOQ = $1(10^{-5})$ to 0.012	J. Zhao et al. (2019) and Q. Zhao et al. (2019)
NAP, PYR BaP, FLU, PYR	HPLC/FL HPLC/UV	Fluorescence UV	Mobile phase: ultrapure water and acetonitrile Maximum wavelengths (λ_{max}) were 236, 240 and 285 nm for BaP, FLU and PYR Mobile phase: ultrapure water and methanol	NR NR	Li et al. (2019) J. Zhao et al. (2019) and Q. Zhao et al. (2019)
BaP, NAP, PHE	ACQUITY UPLC	Fluorescence	Mobile phase: water and acetonitrile	LOD = 0.3, 0.3 and 0.1 for PHE, NAP and BaP	Zhang et al. (2019)
ANT, FL, FLU, PHE, PYR	UV-Vis spectrophotometer	-	Maximum wavelengths (λ max) were 200 to 800 nm	-	Topuz and Uyar (2017)
ACE, ANT, FL, FLU, PHE	GC/MS	Mass selective (MSD)	Gas carrier: Helium	NR	Gong et al. (2017)
ACE, NAP, PHE	HPLC/FL	Fluorescence	Mobile phase: ultra-pure water and acetonitrile	NR	Cheng et al. (2019)
ACE, NAP, PHE	GPLC/FL	Fluorescence	Mobile phase: ultra-pure water and acetonitrile	NR	Xi and Chen (2014)
ACE, FL, FLU, NAP, PYR	GC/MS	Mass selective (MSD)	Gas carrier: Helium	NR	Vidal et al. (2011)
PAHS and substituted PAHs	GC/MS	Mass selective (MSD)	Gas carrier: Helium	NR	Adhikari et al. (2019)
16 PAHs	GC/MS	Mass selective (MSD)	Gas carrier: Helium; Mass Spectrometry: Electron ionization (El) and selective ion monitoring (SIM) modes	LOD = 0.60 to 5.40	Dong et al. (2012)
16 PAHs	UHPLC (ultra-high performance liquid chromatography)/PDA and UHPLC/FL	Photo Diode Array (PDA) and Fluorescence	Mobile phase: ultra-pure water and acetonitrile	LOD = 4 to 179 (for PDA detector) LOD = 1 to 68	Layton et al. (2018)
NAP	GC/FID	Flame ionization detector (FID)	NR	NR	Shang and Sun (2019)

Romarke

^a Limit of Detection (LOD) and Limit of Qualification (LOQ).

^b NR: Not Reported.

physical/chemical and biological treatment methods for PAHs removal from aqueous solutions are listed in Table 5

4.1. Biological treatment methods

4.1.1. Bioreactor

Municipal wastewater treatment plants (MWTPs) accept urban and industrial sewage and eliminate solids, nutrients and organic matter by biological, physical and chemical treatment ways to get a significant reduction in contaminants and ecotoxicity in the obtaining surface or groundwater (Han et al., 2018). Giordano et al. (2005) reported that PAHs biological treatment methods, activated sludge processes, sequencing batch reactors and membrane bioreactors have been most commonly applied in organic pollutants removal. Qiao et al. (2016) reported that the abatement efficiency of lower molecular weight organic pollutants was much higher than high molecular weight organic pollutants because the lower molecular weight organic pollutmore easily biodegraded/biotrans formed during biological treatment.

J. Zhao et al. (2019) and Q. Zhao et al. (2019) reported that the total abatement of Σ NPAHs in summer reached 63.22% to 63.58% in a

Table 5

PAHs removal by physical, chemical and biological methods.

municipal biological wastewater treatment plant. Qiao et al. (2016) reported 83% to 97% removal efficiency during PHE treatment with aerobic activated sludge treatment. Giordano et al. (2005) reported that about 55% of PAHs were removed by a sequencing batch reactor (SBR), while 0.0% to 73.5% of PAHs (mostly BghiP, NAP and PYR) were removed from wastewater by an anaerobic-anoxic-oxic treatment process (Sun et al., 2013).

4.1.2. Phytoremediation and bioremediation

Plants have been applied to remediate polluted soil and water because phytoremediation is an inexpensive and non-invasive method. Phytoremediation is also an approach that provides more ecological benefits than current techniques (Mojiri et al., 2016). Tian et al. (2019) stated that plants may function as contaminant bioaccumulators and bioindicators because of their extensive surface distribution and specific responses. Tree leaves are so effective at trapping PAHs and thus play the unique role in diminishing the level of respirable fine particulates that cause serious human diseases. N. Li et al. (2017) and L. Li et al. (2017) found that plant lipids are the key chemical compounds responsible for the assimilation of organic contaminants. Alagić et al. (2015) reported that assimilation of PAHs from matrices into plants

PAHs	Methods	Efficiency (%)	Sources	References
ACE, ANT, FL, FLU. PHE	Electrocoagulation	Up to 86	Industrial wastewater	Gong et al. (2017)
BaA PHE PYR	Bioreactor in combining with Pseudomonas stutzeri CECT 930	100 100 98	Aqueous solution	Moscoso et al. (2015)
PHE PYR	Adsorption by stearoyl grafted cellulose	97.6 96.9	Aqueous solution	Kim et al. (2018)
FL, PHE, PYR	Biochar	71.8 to 98.6	Aqueous solution (soil washing effluents)	Li et al. (2014)
7 PAHs	Filter-activated sludge technology, and rotating biological contractors with extended aeration technologies	28.0 to 71.6	Wastewater	Alawi et al. (2017)
PHE	Under hypersaline and hyperalkaline condition in a membrane bioreactor system with <i>Pseudomonas</i> sp. LZ-Q	96.0	Crude oil-contaminated wastewater	Jiang et al. (2016)
18 PAHs	O ₃ /ultraviolet fluidized bed reactor	41.0 to 75.0	Coking wastewater	Lin et al. (2014)
6 PAHs	Chemical precipitation	6.0 to 40.0	Landfill leachate	Ates and Argun (2018)
6 PAHs	Chemical precipitation (CP), Fenton oxidation (FO) and ozone oxidation	80.0 to 100	Landfill leachate	Ates and Argun (2018)
BaP	Sequential and Simultaneous Ozonation and Biotreatment	91.0 to 100.0	Aqueous solution	Yerushalmi et al. (2006)
16 PAHs	Electrocoagulation and membrane filtration	90.0	Industrial wastewater	Gong et al. (2017)
16 PAHs	Electrochemical oxidation Ti/RuO(2)	80.0 to 82.0	Aqueous solution	Tran et al. (2009)
16 PAHs	Electrochemical Oxidation	Around 87.0	Aqueous solution	Yaqub et al. (2013)
16 PAHs	Modified electrochemical process	Around 95.2 at optimum process	Wastewater separated from petroleum crude oil	Yaqub et al. (2017)
16 PAHs	Advanced oxidation processes and electrooxidation	90.0 to 95.0	River sediments	Andreottola and Ferrarese (2008)
ACE, FL, NAP, PHE, PYR	Plant residue materials as a biosorbent	Up to 90	Aqueous solution	Chen et al. (2011)
BaA	Bioremediation (lactic acid bacteria)	38.4 to 56.0	From phosphate buffer	Yousefi et al.
BaF		36.3 to 54.2	solution	(2019)
BaP		50.0 to 66.6		
CHY		43.7 to 56.0		
16 PAHs	Fenton	83.5	Textile dying	Lin et al. (2016)
	Ultrasound-Fenton	75.5		
	Ultrasound	45.5		
BbF, BkF, BghiP, IcdP	Modified coagulation method (several coagulants: coagulants Al ₂ (SO ₄) ₃ ·18H ₂ O, hydrolysed salts, and polyaluminum chlorides (PAX1910 and PAX19F)	93.8 to 95.8	Drinking water	Rosi'nska and Dabrowska
ACE	Combining UV radiation, ozone and hydrogen peroxide	33.0 to 95.0	Aqueous solution	Rivas et al. (2000)
PHE	Anaerobic Reactor	52.3	Sewage	Lin et al. (2019)
BaP	Microbacterium maritypicum CB7 (biodegradation)	69.0	Water	Mansouri et al. (2017)
CHY	Photodegradation in presence of carbon-modified $n-TiO_2$ nanoparticles	High removal efficiency (>90%)	Seawater	Shaban (2018)
NAP	Magnesium peroxide (MgO_2) nanoparticles plus <i>P. putida</i> and <i>P. mendocina</i>	100	Groundwater	Gholami et al. (2019)
PAHs (FLU	Selenastrum capricornutum	78	Aqueous solution	Lei et al. (2007)
and PYR)	Chlorella vulgaris	48		
,				

may be treated as an equilibrium procedure in which the accumulated PAHs are in equilibrium with PAHs available in the matrix. Plant uptake of PAHs is supplemented by water flow from the transpiration stream and water transfers into the root system across apoplastic and symplastic pathways. For hydrophobic substances, such as PAHs, the root uptake is evident, and is strongly correlated with the root lipid content (Alagić et al., 2015). The uptake of PAH is affected by the characteristics of both plant species and organic chemicals (Li et al., 2002). Once the organic pollutant gets the plant system, it is apportioned to several plant parts over translocation; after that, any number of reactions may occur within the following series: oxidations, reductions or hydrolysis (Reynoso-Cuevas et al., 2010). Lower molecular PAHs are main in plants (Tao et al., 2006) in comparing with high molecular PAHs. Reynoso-Cuevas et al. (2010) investigated PAHs removal by phytoremediation methods using F. arundinacea and found that they were able to transform 40.40% of the initial PHE, while they accumulated 6.99% in their stems and almost three times as much in their roots (20.66%). They also reported that the efficiency of removal of organic pollutants, such as PAHs, by phytoremediation is limited because of their low water solubility. Therefore, using bioremediation in conjunction with phytoremediation would improve the removal efficiency. Rhizoremediation, which consists of both phytostimulation and rhizodegradation, provides the beneficial interaction of both the plant and the rhizobacteria. Many studies have investigated the rhizoremediation of PAH to date (Bisht et al., 2015), and some of the plants most often used for removal of PAHs are listed in Table 6.

Bioremediation is the partial or complete conversion of a pollutant of interest to its elemental constituents by microorganisms such as bacteria or fungi (Eevers et al., 2017). One of the problems associated with bioremediation of PAHs is the toxicity of these compounds to cells because these lipophilic substances have a direct impact on cellular membranes (Hąc-Wydro et al., 2019).

Zhang et al. (2010) and Mallick (2019) reported that biodegradation of acenaphthene has gained significant interest and various bacterial species can be used in removing PAHs, such as *Pseudomonas fluorescens*, *Pseudomonas putida*, *Burkholderia cepacia*, *Cycloclasticus* sp., *Alcaligenes eutrophus*, *Neptunomonas naphthovorans*, *Alcaligenes paradoxus*, *Pseudomonas* sp., *Sphingomonas* sp. A4, and *Beijerinckia* sp. Fu et al. (2018) investigated PHE removal by the endophytic fungus *Phomopsis liquidambari*. de Llasera et al. (2018) stated that BaP could be removed by the microalgae *Selenastrum capricornutum*. Mansouri et al. (2017) reported that some bacteria, such as *Alcaligenes denitrificans*, *Mycobacterium* sp., and *Bacillus subtilis*, have the ability to degrade low molecular weight PAHs. Mandal and Das (2018) reported that *Hanseniaspora opuntiae* NS02 and *Debaryomyces hansenii* NS03 can be used to remove BghiP.

4.2. Physical/chemical treatment methods

4.2.1. Membrane

A membrane is a material that makes a thin barrier capable of selectively resisting the movement of diverse constituents of a fluid, thereby enabling separation of the constituents. Different membrane filtration systems such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis are employed in water and wastewater treatment (Mojiri et al., 2013). The removal of organic contaminants from potable water by membrane processes is strongly related to the type of membrane selected. When choosing an appropriate membrane it is important to consider the molecular weight cut-off (MWCO), which is stated in Daltons and indicates the molecular weight of a hypothetical non-charged solute that is 90% rejected by the membrane (Plakas and Karabelas, 2012). Membranes have the disadvantage of requiring pretreatment and energy consumption (Zazouli and Kalankesh, 2017).

Smol et al. (2016) removed 59% to 72% of PAHs by reverse osmosis, while Smol and Włodarczyk-Makuła (2012) studied the removal of \sum 16PAHs from industrial wastewater using an ultrafiltration process and achieved a removal efficiency of 66.6% to 85.0%. Wang et al. (2015) reported 95% NAP removal by using nanofiltration in acidic solution. Gong et al. (2017) removed 50% of ACE, 91% of ANT, 88.3% of FL and 85.9% of PHE from wastewater by low-pressure reverse osmosis.

4.2.2. Adsorption

Balati et al. (2015) reported that adsorption is one of the simplest, most effective, quickest, and broadly applicable methods among different types of remediation technologies. Adsorption can be used for the remediation of various pollutants including organic compounds and heavy metals. Different adsorbents including activated carbon (Dowaidar et al., 2007), bentonite (Karaca et al., 2016), biochar (Guo et al., 2018), chitosan (Crisafully et al., 2008), graphene (Li et al., 2018), nano-tubes (Paszkiewicz et al., 2018), and zeolite (Vidal et al., 2011) have been used to eliminate PAHs. Smol and Wlodarczyk-Makula (2017) reported that the recycling of sorbents and subsequent treatment of PAHs are difficult, which may present a risk of secondary contamination. However, adsorption is relatively simple, convenient, and easy to design when compared to other methods of PAHs abatement, and adsorption systems may be operated with very little technical know-how. Yakout and Daifullah (2013) used different adsorbents including bone charcoal, activated rice husk, peat moss, activated carbon, and pyrolysis residue to remove PAHs such as naphthalene, pyrene and phenanthrene (Table 7).

Hedayati (2018) investigated the removal of PAHs (ANT, FL, FLU, PHE and PYR) via clinoptilolite and modified forms of clinoptilolite, such as didodecyldimethylammonium bromide (DDAB), cetylpridinium

Table 6

Some plants reported to be capable of removing PAHs from water sources.

PAHs	Source	Plant	In presence of bacteria or substrate	Removal	References
$\sum 16$ PAHs	Contaminated water by diesel (Synthetic wastewater)	Lepironia articulate	Sands and Gravels	79.6% to 96.9%	Al-Sbani et al. (2016)
$\sum 16$ PAHs	Wastewater	Phragmites australis and Arudo donax	Gravel and Soil	68.2% to 79.2	Fountoulaksi et al. (2009)
PHE	Stormwater	Dianella revoluta	Activated carbon, zeolite, bentionite and sand	53.6% to 92.2%	Lamichhane (2017)
$\sum 16$ PAHs	Landfill Leachate	Reed and Cattail (Up-flow system)	Zeolite, Gravel and Soil	0.0% to 58%	H. He et al. (2017) and J. He et al. (2017)
$\sum 16$ PAHs	Wastewater treatment plant	Phragmites australis	Sand and Gravel	63%	Cui et al. (2015)
PHE	Sediment	Vallisneria spiralis	-	53.3% to	Liu et al. (2014)
PYR				59.6%	
				50% to	
				53.6%	
PHE	Sediment	Potamogeton crispus L.	-	18.3% to	Meng and Chi (2015)
PYR				34.1%	
				14.1% to	
				27.8%	

chloride (CPC), and tetramethylammonium chloride (TMA) and hexadecyltrimethylammonium bromide (HDTMA-Br). The results revealed that clinoptilolite and TMA removed around 66% of PAHs while CPC, DDAB, and HDTMA-Br removed >93% of PAHs. During 24 h of contact time, 95.6% of ACE, 100% of NAP and 99.89% of PHE were eliminated by the soybean stalk–based carbon (Kong et al., 2011). BAP was completely removed from landfill leachate by an activated carbon filter column (Kalmykova et al., 2014), while approximately 88% of \sum 12PAHs were eliminated using modified diethylamine/bentonite (Karaca et al., 2016) and 99.9% of BaP and 98.5% of PYR were removed by iron oxide nanoparticles (Hassan et al., 2018).

4.2.3. Advanced oxidation processes

Advanced oxidation processes (AOPs) using combinations of oxidants, catalysts and ultraviolet irradiation to generate hydroxyl radicals (OH•) in solutions have attracted interest for the degradation of hazardous organic compounds or biorefractory in wastewater (Badawy et al., 2006). Organic contaminants are oxidized by free radicals and mineralized to water, mineral salts and carbon dioxide. Several AOPs

Table 7

Capacity for adsorption of PAHs from water sources of some common adsorbents.

(e.g., Fenton's reagent, ozonation, electrochemical oxidation, and UV) that have been applied for the oxidation of a diversity of contaminants are known to transform the parent compounds into more innocuous and biodegradable intermediate products (Vagi and Petsas, 2017). AOPs have some disadvantages such as energy consumption and high maintenance costs. For example, the disadvantages of the photo-Fenton process contain the need for low pH values and for removal of the iron catalyst after the reaction has terminated (Machulek et al., 2012).

Approximately 95% of fenthion was removed with UV-TiO₂ in a study conducted by Petsas et al. (2013). Włodarczyk-Makuła (2011) reported a high efficiency of the removal of hydrocarbons by UV-rays, based on the number of rings. The removal efficiency reached up to 94% for naphthalene. Ates and Argun (2018) investigated PAHs removal by Fenton and ozone oxidation and found that the removal efficiencies ranged between 6% and 40%. Lin et al. (2014) found that ozone has been efficiently applied in an advanced oxidation process (AOP) for treatment of various organic pollutants due to its high oxidation and disinfection potential. Some pathway mechanisms for PAHs oxidation

Compounds	Adsorbent	Adsorption isotherm	Adsorption capacity (mg/g)	References
ACE	NH2-SBA-15 organic-inorganic nanohybrid material	Pseudo-Second-Order	1.41	Balati et al. (2015)
NAP			1.92	
PHE			0.76	
ANT	Modified clinoptilolite, cetylpridinium chloride	Pseudo-Second-Order	4.91	Hedayati (2018)
FL			9.36	
FLU			9.83	
PHE			9.72	
PYR			9.93	
ANT	Modified clinoptilolite, didodecyldimethylammonium bromide (DDAB),	Pseudo-Second-Order	4.96	Hedayati (2018)
FL			9.62	
FLU			9.94	
PHE			9.88	
PYR			9.97	
ACE	Pine wood	Pseudo-Second-Order	1.372	Xi and Chen (2014)
NAP			2.653	
PHE			2.212	
PYR	Dias Usels Diasker	Desuda Casand Ordan	0.364	Gue et el (2018)
PHE	Rice Husk Biochar Biocher	Pseudo-Second-Order	16.2	Guo et al. (2018)
NAD	Biochar	Langmuir	41.0	N Listal (2017) and L Listal
INAP	Zeonite	Eroundlich	1.022	N. LI EL al. (2017) allu L. LI EL al.
		Proudo Socond Order	0.256	(2017)
ACE	Activated Carbon	Not specified	0.230	Kong et al. (2011)
NAD	Activated Caliboli	Not specified	1 295	Kong et al. (2011)
PHF			0.038	
ACE	Modified Organosilica	Pseudo-second-order	1.01	Vidal et al. (2011)
FI	wounce organostica	i seduo-second order	0.91	
FIII			0.72	
NAP			1 54	
PYR			1 36	
ACE	Commercial granular activated carbon	Langmuir	$6.15(10^{-3})$	Radwan et al. (2018)
BaP	Iron oxide nanoparticles	Pseudo-second-order	0.96	Hassan et al. (2018)
PYR			0.99	
ACE	Activated carbon	Langmuir-Freundlich	575	Haro et al. (2011)
FLU			532	
NAP			481	
NAP	Graphene	Langmuir	5.98	Das et al. (2016)
PAHs (not specified)	PK-PSAC (Pyrolysis-assisted potassium hydroxide induced palm shell activated carbon)	(not specified)	131.7	Kumar et al. (2019)
BaP	Fe ₃ O ₄ @polyaniline nanoparticle	Langmuir	10.6 to 12.3	J. Zhao et al. (2019) and Q. Zhao
FLU			12.9 to 16.1	et al. (2019)
PYR			14.2 to 15.8	
ACE	RBP (Rice bran powder)	Freundlich	2.36 ± 0.04	Lu et al. (2018)
	BP (Bamboo powder)		0.87 ± 0.02	
	RBB (Rice bran biochar)		3.79 ± 0.05	
	BB (Bamboo biochar)		3.05 ± 0.06	
NAP	TiO ₂ /NiO (visible phase)	Langmuir	322.1	Sharma and Lee (2015)
NAP	Graphene nanoplatelet/MIL-101 (Cr) nanocomposite	Langmuir (Linear isotherm)	740.7	Bayazit et al. (2017)

with different AOPs are presented below. The general mechanism for PAHs removal by ozone is suggested as Eqs. (1) to (5) (Miller and Olejnik, 2004):

Ozone in aqueous solution reacts with hydroxyl anion giving hydroperoxide anion:

$$O_3 + OH^- \rightarrow HO_2^- + O_2$$
 (1)

$$HO_2 - + H^+ \rightarrow H_2O_2 \tag{2}$$

$$O_3 + HO_2 \rightarrow HO_2 + O_3^{-}$$
(3)

When PAHs are introduced in this system, they undergo degradation through the direct reaction:

$$PAH + O_3 \rightarrow products$$
 (4)

or through the radical reaction (indirect reaction):

$$PAH + OH \rightarrow products$$
 (5)

Ledakowicz et al. (2001) reported a general pathway mechanism for PAHs removal by O_3/UV (Eqs. (6) to (11)). Ozone in the aqueous solution in presence of UV radiation supplied oxygen atom which reacts with water to hydrogen peroxide.

$$O_3 + h\nu \rightarrow O_2 + O \tag{6}$$

 $0 + H_2 O \rightarrow H_2 O_2$

Hydrogen peroxide photolyses to hydroxyl radicals:

$$H_2O_2' + hv \rightarrow 2OH' \tag{7}$$

or dissociates to hydroperoxide anion:

$$H_2 O_2 \leftrightarrow H O_2^{-} + H^+ \tag{8}$$

Reactions (7) and (8) are of minor significance since UV absorption coefficient of hydrogen peroxide is small and the rate constant of H_2O_2 dissociation is low. The ionic form of hydrogen peroxide rises also in the reaction of ozone with hydroxyl anion:

$$O_3 + OH^- \rightarrow HO_2^- + O_2 \tag{9}$$

Hydroperoxide anion, in turn, reacts with ozone donating hydroperoxide radical and ozonide radical anion:

$$O_3 + HO_2^- \to HO2^+ + O3^-$$
 (10)

When PAHs are introduced in this system, they may undergo oxidation in three ways:

$$PAH + \begin{pmatrix} O_3 \\ \frac{h\nu}{OH} \end{pmatrix} \rightarrow Products$$
(11)

Manan et al. (2019) removed 76.4% to 91.0% of PAHs by photo-Fenton oxidation process. The general pathway of PAHs oxidation with the photo-Fenton can be discussed as Eqs. (12) to (19) (Manan et al., 2019).

PAHs degradation with the photo-Fenton reaction consists of the absorption of light by, leading their excitation.

$$PAHs + hv \rightarrow PAH*$$
(12)

The excited PAH may therefore return to the ground state and dissipate its energy

$$PAH * \rightarrow PAH \tag{13}$$

or be transformed into a radical cation (PAH+) and a solvated electron (eaq)

$$PAH \rightarrow PAH^{+} + e_{aq} \tag{14}$$

Meanwhile, O_2 from the water may react with the e_{aq} causing to O_2^- or 1O_2 formation.

$$e_{aq} + O_2 \rightarrow {}^1O_2 \tag{16}$$

$$PAH/PAH^{+} + O_2^{-} \rightarrow Intermediates$$
(17)

$$PAH/PAH^{+} + {}^{1}O_{2} \rightarrow Intermediates$$
(18)

Intermediates
$$\rightarrow$$
 CO₂ + H₂O + Inorganic ions (19)

4.2.4. Coagulation

Coagulation is applied for the abatement of colloidal suspensions and to decrease the content of organic compounds, such as PAHs, in aqueous solutions. Coagulation attended by chemical precipitation is frequently applied in high-effectiveness technologies for water and wastewater treatment (Smol and Wlodarczyk-Makula, 2017). In previous studies, several coagulants including inorganic salts (alum, aluminum chloride, ferric chloride and ferric sulphate), polymeric coagulants (polyaluminium chloride, polyferric chloride, polyferric sulphate), organic polyelectrolytes (polydiallyldimethyl ammonium chloride, anionic polyacrylamides) and composite inorganic–organic coagulants have been applied (Matilainena et al., 2010). The principal disadvantages of application of coagulation solution to wastewater treatment are the problems associated with the highly putrescible sludge formed, and the high operating costs of chemical addition (IWA, 2016).

Kim et al. (2002) reported that the abatement efficiencies of pyrene, fluoranthene, anthracene and phenanthrene were about 75%, 57%, 40% and 30%, respectively, during PAHs removal by coagulation-precipitation.

4.2.5. Combined treatment methods

Integrated physical/chemical-biological methods, such as using a powerful oxidant and adsorption or membrane methods have indicated promising results for efficient solubilization and degradation and complete elimination of many high-molecular-weight PAHs (Yerushalmi et al., 2006). PAHs removal (50% to 100) from wastewater by membrane bioreactors has been reported (González et al., 2012) and low molecular weight PAHs were shown to be more easily removed than high molecular weight PAHs. Additionally, 94.1% to 100% PAHs (ACE, ACY, ANT, FL, FLU, NAP and PHE) removal from wastewater was reported using integrated electrocoagulation and low-pressure reverse osmosis (Gong et al., 2017).

5. Conclusions

In the past few decades, organic pollutants such as PAHs have been found to be widespread in aquatic environments. Therefore, monitoring these kinds of pollutants and removing them with different techniques has attracted a good deal of attention. In this study, we reviewed several research papers to investigate the occurrence of PAHs in water sources and methods for their removal. The key conclusions of this study are as follows:

- 1. The minimum and maximum reported concentration of PAHs were 0.03 ng/L (seawater; southeastern Japan Sea, Japan) and 8,310,000 ng/L (domestic wastewater in South Africa), respectively.
- PAHs and their substituents might be found in all water sources. Substituted-PAHs such as nitrated or oxygenated derivatives may be formed by reactions between PAHs and atmospheric oxidants such as O₃, NOx, and OH.
- 3. PAHs have great impacts on microorganisms, humans, animals, and plants.
- 4. Among PAHs measurement methods, GC/MS and HPLC have been widely applied in the literature.
- 5. Biological methods such as bioreactors, phytoremediation and bioremediation, physical/chemical methods such as membrane, coagulations, advanced oxidation process and adsorption, and combined treatment methods have been used to treat PAHs.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2019.133971.

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