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Comprehensive Review of Polycyclic Aromatic Hydrocarbons in Water Sources, their Effects and Treatments

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 environment and humans, 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 ∑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 physical/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.

Keywords: Polycyclic aromatic hydrocarbons; Treatment; Water; Wastewater

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 Z 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 large amounts in industrial activities, after which they are discharged in to various water compartments, where they can persist, causing severe health and environmental problems (Zambianchi et al., 2017). Recent studies have reported 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 C 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).

Physical/chemical and biological ways have been used for the elimination of organic contaminants from water and wastewater (He J. 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 methods 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 contaminants produced by anthropogenic activities associated with industrialization and urbanization, as well as through natural activities (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 condensed benzene rings bonded in linear, angular of cluster arrangements. Most PAHs are colorless, white or pale-yellow solids (Pogorzelec and Piekarska, 2018). PAHs (Figure A.1 in supplementary file) in the environment are primarily of pyrogenic, petrogenic, and biologic origin (Hąc-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).

Table 1: Most Reported Polycyclic Aromatic Hydrocarbons (PAHs)

2.2. 16 PAHs

The USEPA has categorized 16 of the PAHs (Table 1) as priority-contaminants based on their toxicity, possible for human exposure, 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, 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 Y 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 swallowed by animals or taken up by plants (MDH, 2015).

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 NO_3 radicals, Cl atoms and O_3.

Anthracene (ANT), which is mainly generated during the incomplete combustion of organic materials, is a common organic contaminant in water sources 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 (Bihaic 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 A 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 pollutants 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 WK 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 T 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, diesel-fueled and gasoline-fueled engines, roofing tar, coke ovens, kerosene-fueled stoves and oil flames (Ding Z 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 AP 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.

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, 2015).

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 W et al., 2018). Wang J 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 µg/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 (1073–1273 K), especially in gasification or pyrolysis procedures (Wang Q et a., 2018). Pyrene is a chief component of the PAH compounds broadly present in the environment (Makelane et al., 2019). Zhou W et al. (2018) stated that pyrene is widely distributed in municipal sewage sludge from different countries.

2.3. 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 atmospheric oxidants such as NO_x, O_3, and OH to procedure nitrated or oxygenated derivatives. Substituted PAHs may be added in aquatic ecosystems 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 chemical characteristics and biological reactivity 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 significant 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, groundwater, and both marine and freshwater environments. 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) (Fillippo et al., 2015). As urban areas grow, forests and undeveloped land are being replaced by impervious types of surface cover such as commercial parking lots, residential driveways, and roads, which increases surface runoff during rain and snow events and works as a vital transport pathway for substituted PAHs (SPAHs) in addition to entry of PAHs to urban streams (Witter and Nguyen, 2019).

Table 2: Most Reported NPAHs, OPAHs and SPAHs

2.4. Presence of PAHs in water and wastewater

PAHs have been detected in sediments, water sources, wastewater and crustaceans as mixtures and typically cooccur 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 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 creosote-impregnated 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.

Table 3: Most reported PAHs in water sources

*This table has been drawn based on Table A.1 in the supplementary file.

2.4. Effects of PAHs on Humans and 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 (Ajiboye et al., 2011). 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-Shafya and Mansourb (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 Z 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 Plants

The uptake of PAHs by plants is vital when considering their transfer from soils into the food chain (Zhang S et al., 2017). PAHs are toxic to plants, influencing the growth and development of plants growing in areas of PAH contamination (Somtrakoona and Chouychai, 2013). Organic pollutants including PAHs have been shown to accumulate in vegetation, and they may cause important phytotoxicity. Indeed, PAHs toxicity has been observed in multiple plant species and the documented impacts contained inhibition of germination, growth, and photosynthesis (Pašková et al., 2009). Spinedi et al. (2019) reported that anthracene in vascular plants can be solubilized in the thylakoid membranes and may cause conformational changes, resulting in transformation in electron flux, as well as reductions in the pool of co-enzymes and biomass. Henner et al. (1999) reported that 5 µM fluoranthene reduced cowpea root elongation. They also showed that the percentage of corn seeds that germinated decreased with increasing concentrations of phenanthrene up to 80 mg/kg. Plant germination and growth are intensely inhibited by the presence of volatile, water-soluble low molecular-weight PAHs (Henner et al., 1999). Additionally, Chouychai et al. (2007) reported that the root lengths of four plants (corn, groundnut, cowpea, and mungbean) were affected by increasing concentrations of PAHs; specifically, their root lengths normally diminished progressively with increasing pyrene or phenanthrene concentrations up to 100 mg/kg and 200 mg/kg, respectively.

2.4.3. PAHs Impacts on Fishes and 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. (2010) 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. Diggs et al. (2011) may cause several kinds of cancers, such as breast tumors, in animals.

2.4.4. PAHs Impacts on Human

Rengarajan et al. (2015) reported that PAHs may decrease immune function, kidney, cataracts and liver damage (e.g., jaundice), asthma-like symptoms, breathing problems and lung function abnormalities, and that repeated contact with skin can induce skin inflammation and redness. Naphthalene, a specific PAH, may cause the breakdown of red blood cells if inhaled or ingested in large amounts. With exposure to PAHs, the harmful impacts that can occur largely depend on the way in which the individual is exposed. Moorthy et al. (2015) reported that PAHs have caused lung cancer, while Abdel-Shafy et al. (2016) reported that anthracene and benzo(a)pyrene are skin sensitizers that can cause allergic reactions in human skin. Additionally, Balcıogˇlu (2016) reported that occupational exposure to high levels of contaminant mixtures comprising PAH resulted in symptoms such as eye irritation, vomiting, convulsion and nausea; however, it was not known which of the mixture components were responsible for these effects. Health impacts from long-term exposure to PAH can include kidney problems, liver damage, jaundice and cataracts. Naphthalene can cause the breakdown of red blood cells if inhaled or ingested in large amounts. Ewa et al. (2017) found that adduct formation is the outcome of covalent binding between reactive electrophilic substances and nucleophilic sites in DNA and proteins. The capability of a chemical to bind to DNA, either directly or after metabolic activation, is obtained as indication of mutagenic and carcinogenic potential, and the genotoxicity of PAHs is well-established. PAHs also cause reduced fertility. For example, there is a significant association between reduced fertility and smoking among female smokers (Ramesh et al., 2012). Shiue (2016) reported that urinary polyaromatic hydrocarbons, such as 4-hydroxyphenanthrene, were associated with celiac disease, and that urinary 2-hydroxyfluorene, 3-hydroxyfluorene, 1-hydroxyphenanthrene, 1-hydroxypyrene, and 2-hydroxynapthalene were meaningfully associated with kidney stones. Parida et al. (2019) stated that BaP can cause behavioral alterations, neuronal damage, teratogenicity, tumor formation, immunosuppression and cognitive and memory dysfunction. Rachna et al. (2018) reported that chrysene has negative impacts on distorted chromosomal DNA and liver tumors in mice, even at levels as low as 0.002 mg/kg. The PAHs in human biological samples are listed in the appendix (Table A.3) and some important effects of PAHs on human health are listed in Figure A.2.

4. 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 (Adeniji 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.

Table 4: Methods used to measure PAHs in aqueous solutions

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 monitoring 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.

5. Removal of PAHs from Aqueous Solutions

During the last few decades, several researchers have focused on efficient 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 physical/chemical and biological treatment methods for PAHs removal from aqueous solutions are listed in Table 5.

Table 5: PAHs removal by physical, chemical and biological methods

5.1. Biological Treatment Methods

5.1.1. Bioreactor

Municipal wastewater treatment plants (MWTPs) accept domestic 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 biodegradation might occur both aerobically and anaerobically. Among 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 pollutants could be more easily biodegraded/biotrans formed during biological treatment.

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

5.1.2. 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 bioindicators and bioaccumulators because of their extensive surface distribution and specific responses. Tree leaves are so effective at trapping PAHs and thus play an unique role in diminishing the level of respirable fine particulates that cause serious human diseases. Li N 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 that assimilation of PAHs from matrices into plants 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). Lower molecular PAHs are main in plants (Tao et al., 2006) in comparing with high molecular PAHs. Reynoso-Cuevas (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 J 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.

Table 6: Some plants reported to be capable of removing PAHs from water sources

5.2. Physical/Chemical Treatment Methods

5.2.1. 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, nanofiltration, ultrafiltration, 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 pre-treatment 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 ∑16PAHs from industrial wastewater using an ultrafiltration process and achieved a removal efficiency of 66.6% to 85.0%. Wang XD et al. (2015) reported 95% NAP removal by using nanofiltration in acidic solution. Gong C 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.

5.2.2. 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 W et al., 2018), chitosan (Crisafully et al., 2008), graphene (Li B 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,.

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 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 more than 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 ∑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).

Table 7: Capacity for adsorption of PAHs from water sources of some common adsorbents

5.2.3. 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 (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 (Vagı 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 Jr. et al. 2012).

Approximately 95% of fenthion was removed with UV-TiO_2 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 C 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.

5.2.4. 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 accompanied by chemical precipitation is frequently applied in high-effectiveness technologies for water and wastewater treatment (Smol and Włodarczyk-Makuła, 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 (Matilainen 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 Y 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.

5.2.5. 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 C et al., 2017).

6. 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.

2. 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 O_3, NOx, and OH.

3. PAHs have great impacts on microorganisms, humans and animals.

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, and physical/chemical methods such as membrane, coagulations, advanced oxidation process and adsorption have been used to treat PAHs.

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