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Treatment of Water Contaminated with Polycyclic Aromatic Hydrocarbons (PAHs): A Review of Various Techniques, Constraints, and Field Procedures

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Abstract: Clean water is vital in the creation of energy and sustenance of life. However, the pollution of water and the absence of potable water are global problems resulting from agricultural and industrial activities. We have witnessed significant growth in the pollution of water by organic compounds like PAH. Experts have made an effort to establish favorable techniques for the treatment of PAH polluted water. These techniques are either thermal, biological, physical or chemical. Bioremediation, chemical oxidation, solid-phase extraction, coagulation, photocatalytic degradation and adsorption using graphenes, mesoporous silica and agricultural wastes are techniques that are already in use in the field treatment of PAHs while electrokinetic remediation and nanoremediation are still in their developmental stage. Several reviews on the treatment of sediments and soils contaminated with PAHs have been published, but only a few reviews center mainly on the removal of PAHs in water. Therefore, this review aims to provide information on the techniques used in the treatment of water contaminated with PAHs. Techniques that are already in use and those that are in their developmental stage were reviewed. The successes of these methods, limitations, constraints and field procedures were analyzed and this will help to inform decision making.

Keywords: PAH, water pollution, water treatment, agricultural wastes, bioremediation, photocatylitic degradation

Abstrak: *Air bersih merupakan kebutuhan utama dalam pembuatan energi dan kehidupan berkelanjutan. Namun, polusi air telah menurunkan ketersediaan air minum dan masalah global karena adanya aktivitas pertanian dan industri. Kita telah menyaksikan peningkatan polusi air oleh senyawa organik seperti PAH. Para ahli telah mengembangkan berbagai teknik untuk pengolahan air yang tercemar PAH. Teknik-teknik ini mencakup perlakuan termal, biologis, fisika ataupun kimiawi. Bioremediasi, oksidasi kimiawi, ekstraksi fase padat, koagulasi, degradasi fotokatalitik dan adsorpsi dengan grafen, silika mesopori, dan limbah-limbah pertanian merupakan teknik yang banyak digunakan dalam pengolahan limbah PAH di lapangan sedangkan remediasi elektrokinetika dan nanoremediasi masih dalam tahap pengembangan. Beberapa ulasan mengenai pengolahan sedimen dan tanah yang tercemar PAH sudah bayak diterbitkan, namun masih sedikit ulasan yang membahas mengenai penghilangan PAH dalam air. Oleh karena itu, artikel ini bertujuan untuk memberikan informasi mengenai teknik-teknik yang digunakan untuk mengolah air yang tercemar PAH. Teknik-teknik yang telah digunakan dan masih dalam tahap pengembangan diulas dalam artikel ini. Keberhasilan, kekurangan, keterbatasan dan prosedur lapangan metode-metode ini dianalisis sehingga dapat membantu untuk pengambilan keputusan.*

Kata kunci: *PAH, polusi air, pengolahan air, limbah pertanian, bioremediasi, degradasi fotokatalitik*

INTRODUCTION

Water is life and clean water is vital in the creation of energy and sustenance of life. Potable water is calculated to be about 0.01 % of the entire global water (Ritchie & Roser 2021). The pollution and absence of potable water are global problems resulting from agricultural and industrial activities (Wang *et al*. 2019). In addition to this challenge, water is not evenly distributed around the world and this is evident with some regions mainly in low latitudes facing water shortage and globally, millions of people suffer from this shortage (UNICEF 2012).

One of the major goals of the World Health Organization (WHO) is to provide access to the required supply of clean water to people irrespective of developmental, economic or social status. Also, the Sustainable Development Goals (SDGs) aim to offer solutions to the challenges surrounding water which include water pollution, flooding and drought. Certain successes have been achieved in this aspect; however, there are need to develop cheaper, eco-friendly and long-lasting measures.

When a given water body is polluted, the pollutants migrate from the water animals drink to humans when the animals' meat is consumed, hence introducing the pollutants into the food chain. Polluted water can equally be consumed directly by humans and this can result to infectious diseases like typhoid and cholera. This is referred to as microbial water pollution (Ghafoor *et al*. 1994). If polluted water is frequently consumed, it may affect the kidney and the human heart. Poor circulation of blood, impairment of the nervous system, skin sores and vomiting are some of the health issues that can also be linked to polluted water. Indeed, issues arising from polluted water are claimed to be the largest causes of death for humans all over the world (Ghafoor *et al*. 1994) as over 5 million people die every year from water-related complications (World_Water_Day 2002).

Significant growth in the number of organic compounds being introduced into the worldwide market has been witnessed in recent decades. The majority of them, which include pharmaceuticals, PAHs, surfactants and pesticides are employed in industrial and agricultural practices and eventually released into bodies of water, thus causing serious environmental and health issues (Zambianchi *et al*. 2017). Research has shown the presence of organic pollutants (e.g PAHs) in diverse aquatic environments (Grandclément *et al*. 2017). The presence of organic compounds was reported in Asia (Lin *et al*. 2017), Africa (Edokpayi *et al*. 2017), Europe (Wen *et al*. 2017) and America (Gilliom 2007).

Polycyclic Aromatic Hydrocarbons (PAHs) are hydrocarbons - compounds of organic origin that occur in crude oil, coal and gasoline. Some PAHs (especially the carcinogenic ones) are dangerous organic micropollutants that are pervasive and resistant to degradation. They are generally white, colorless or pale-yellow solids (Duan *et al*. 2015; Ghosal *et al*. 2016). They exhibit distinct physical and chemical properties and possess two or more benzene rings bonded together by C-C bonds (Mohan *et al*. 2006; Okere & Semple 2012). Over 400 varieties of PAHs and their effects have been reported (Pan *et al*. 2006). However, sixteen (16) PAHs molecules, as shown in Table 1, have been identified by the United States Environmental Protection Agency (US-EPA) as pollutants of great concern. The acceptable PAH content in drinking water for benzo(a)pyrene, a toxic PAH molecule, is estimated at 200 ng/L (Mastral & Callén 2000; ATSDR 2009). PAHs are often found in water bodies that are close to gas production, exploitation of crude oil and industries that deal with coal/wood (Sun *et al*. 2009; Zhao *et al*. 2011; Abdel-Shafy & Mansour 2016). The major channel of exposure to these compounds is mainly through polluted water and a typical example is how PAHs in coal and coal waste seep into mine water (Hendryx *et al*. 2008) and subsequently discharge into marine ecosystems when mine water is released (Goodarzi & Mukhopadhyay 2000).

PAHs have certain commercial uses as intermediaries in pharmaceuticals, photographic products, lubricants, agricultural chemicals, thermosetting polymers, among others (Kaminski *et al*. 2008). However, acute exposure to PAHs can lead to irritation of skin and eye, nausea or even vomiting and inflammation, while chronic exposure can result in cancer, organ failure, cell damage, DNA damage, mutation of genes and even death. Polycyclic Aromatic Hydrocarbons are categorized into two; the lower molecular weight (LMW) and the higher molecular weight (HMW). Structurally, LMW comprises of 2 to 3 aromatic rings and is less toxic, while HMW comprises of 4 to 7 aromatic rings and are more difficult to degrade (Kuppusamy *et al*. 2016a). A good number of the treatment of sediments and soils contaminated with PAHs have been published (Wise 2000; Samanta *et al*. 2002; Mohan *et al*. 2006; Gan *et al*. 2009; Kuppusamy *et al*. 2017; Maletić *et al*. 2019), but only a few studies centered mainly on the removal of PAHs in water (Mojiri *et al*. 2013; Adeola & Forbes, 2020). There is a great need for a more comprehensive literature review that will focus on remediation methods for PAH contaminated water. This literature review provides information on remediation technologies for water contaminated with PAHs. The successes of these methods, limitations, constraints, field potential and procedures have been reviewed, and this will help in decision making and undertaking remediation measures.

SOURCES OF PAHs

The major PAH sources are pyrogenic source, petrogenic source and biological source. Pyrogenic PAHs form through pyrolysis when organic materials are burnt under high temperatures with little or no oxygen present. Thermal cracking of petroleum remnants, destructive distillation of coal into coal tar or coke are examples of pyrolytic processes that occur deliberately. Some unintentional processes also occur during the incomplete burning of wood in fireplaces or partial combustion of petroleum fuels. Pyrogenic PAHs are primarily found in urban areas and areas proximal to large sources of PAHs and for pyrogenetic processes to occur, the required

Table 1. 16 PAHs identified by US-EPA

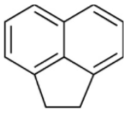
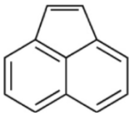
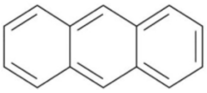
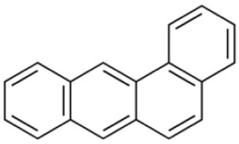
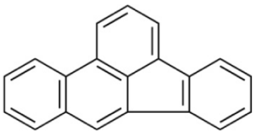
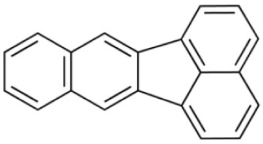
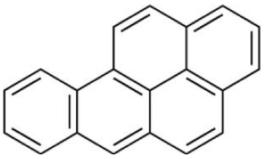
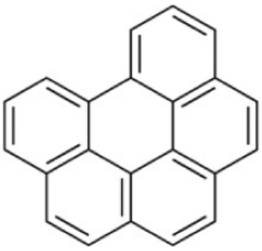
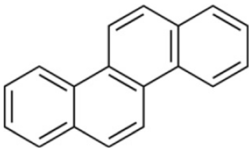
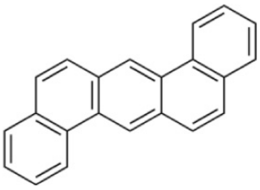
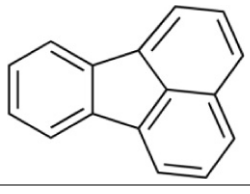
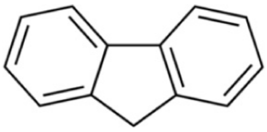
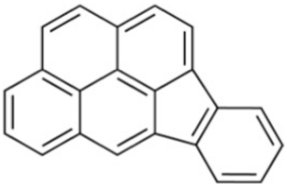
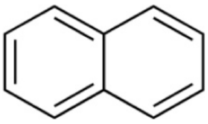
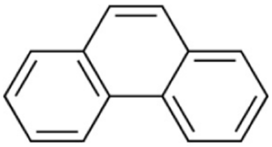
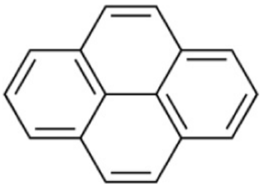
PAHs	Benzene Ring	Chemical Formula	References
Acenaphthene, (ACE)		$C_{12}H_{10}$	Lerda 2011 Zelinkova & Wenzl 2015
Acenaphthylene, (ACY)		$C_{12}H_8$	Zelinkova & Wenzl 2015 PubChem 2005
Anthracene, (ANT)		$C_{14}H_{10}$	Lerda 2011 Lawal 2017
Benz[a]anthracene, (BaA)		$C_{18}H_{12}$	Gen & Hartwig 2012 Yan et al. 2004
Benzo[b]fluoranthene, (BbF)		$C_{20}H_{12}$	WHO 2017
Benzo[k]fluoranthene, (BkF)		$C_{20}H_{12}$	Abdel-Shafy & Mansour 2016
Benzo[a]pyrene, (BaP)		$C_{20}H_{12}$	Lerda 2011
Benzo[ghi]perylene, (BghiP)		$C_{22}H_{12}$	ECHA 2018
Chrysene, (CHY)		$C_{18}H_{12}$	Rachna *et al*. 2018
Dibenz[a,h]anthracene, (DahA)		$C_{22}H_{14}$	Zelinkova & Wenzl 2015
Fluoranthene, (FLU)		$C_{16}H_{10}$	Abdel-Shafy & Mansour 2016

Table 1. 16 PAHs identified by US-EPA (cont.)

PAHs	Benzene Ring	Chemical Formula	References
Fluorene, (FL)		$C_{13}H_{10}$	Abdel-Shafy & Mansour 2016
Indeno[1,2,3-cd]pyrene, (IcdP)		$C_{22}H_{12}$	Lerda 2011
Naphthalene, (NAP)		$C_{10}H_{8}$	Lawal 2017
Phenanthrene, (PHE)		$C_{14}H_{10}$	Lerda 2011
Pyrene, (PYR)		$C_{16}H_{10}$	Lawal 2017

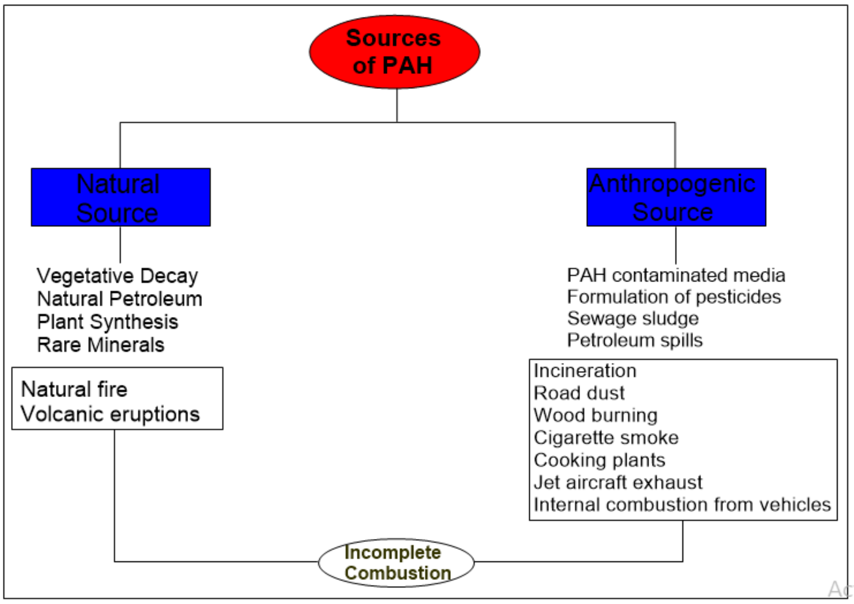


Figure 1. Sources of PAHs (modified from19)

temperature ranges from about 350°C to over 1200°C.

Also, PAHs can equally form under lower temperatures of about 100 – 150°C over millions of years. Therefore, PAHs that form during fossil fuel maturation and other related processes are termed petrogenetic PAHs. Primary sources of petrogenetic PAHs are leaks of underground and overhead storage tanks, freshwater and oceanic oil spills, and the gradual accumulation of gasoline and other derivatives during transport. The high usage of petroleum and its derivatives, widespread transportation and storage make petrogenetic PAHs common.

PAHs can also be generated by some bacteria and plants or forms when vegetative matter degrade. Generally, PAHs form through natural activities such as algal and bacterial synthesis, forest and bush fires, petroleum seeps, volcanoes and erosion of petroleum host or reservoir rocks. It can equally form through anthropogenic activities, which can be large point sources such as industrial processes and incinerators, or small point sources such as emissions from automobiles, jet aircraft exhaust, cigarette smoke, smoke from firewood, etc. Figure 1 illustrates the various modes of PAH formation (Tolosa *et al*. 1996; WHO 2003; Zhang *et al*. 2004; Masih & Taneja 2006).

VARIOUS METHODS FOR PAHs REMOVAL

Bioremediation

Bioremediation is one treatment technology with the highest approval regarding the removal of PAHs in media, including soil, subsurface material and water. It is done by altering the conditions of the environment to stimulate the growth of microorganisms, plants, and enzymatic reactions, which will then degrade the target pollutants. Bioremediation is commonly applied in the treatment of oil spills, leaks of underground pipes, crime scene cleanups and the treatment of soils that are contaminated with acidic mining drainage (Reavill 2007). The method is adjudged to be ecologically friendly, cheap and safe since there is no pollutant transfer to other (Mohan *et al*. 2006; Kuppusamy *et al*. 2016b). Reviews of PAH degradation were made (Juhasz & Naidu 2000; Haritash & Kaushik 2009; Duan *et al*. 2015). Species of bacteria like *Pseudomonas*, *Alcanivorax*, *Microbulbifer*, *Sphingomonas*, *Micrococcus*, *Cellulomonas*, *Dietzia*, *Gordonia marinobacter* and fungal species such as *Aspergillus* sp., *Fusarium oxysporum* and *Trichocladium canadense* have proven to be capable of degrading PAH compounds (Wu *et al*. 2011).

To achieve an optimum result, temperature, pH, oxygen content, nutrient and other relevant parameters have to be considered as they are reported to have considerable influence on a successful bioremediation process. For instance, temperature between 30 and 38°C is suitable for the degradation of PAHs as it enhances the growth of microbes (Abdou 2003; Antizar-Ladislao *et al*. 2008; Moscoso *et al*. 2013). Temperature rise will lead to a fall in oxygen solubility which will, in turn, reduce the dissolved oxygen available for the growth of microbes in the bioreactor. Therefore, a higher temperature of about 40 and 45°C will lead to a significant decrease in PAHs removal (Viñas *et al*. 2005; Alegbeleye *et al*. 2017).

Chemical Oxidation

Chemical oxidation stands out as one of the *in-situ* treatment methods to remediate lower molecular and higher molecular weight PAHs in soils and industrial/municipal landfill leachates. Chemical oxidation, both basic and advanced, has been described for such treatment and over 90% degradation was reported (Li *et al*. 2016). The treatment of chlorinated organic substances affecting groundwater and saturated area using in-situ chemical oxidation method has equally been reported to remediate PAH contaminated soils (ITRC 2005; Huling & Pivetz 2006; Rivas 2006; Siegrist *et al*. 2011; Ranc *et al*. 2016). The method involves the injection of oxidant to degrade pollutants and various oxidants exist and this ranges from the popular ozone and Fenton's reagent to less popular oxidants like as hydrogen peroxide (H_2O_2), potassium permanganate ($KMnO_4$), peroxy-acid, sodium (Na_2þ), or iron (Fe_2þ) activated persulfate (Cheng *et al*. 2016; Kuppusamy *et al*. 2016b). It equally involves the breakdown of the aromatic rings of PAHs using reactive radicals such as •OH, O_3 and SO_4^{2-} SO_4•$^-$ O_3• radicals. Other reactive species such as peroxides and persulfate anion can also be used. The efficiency is dependent on the contact between the contaminants and oxidants, and this can be affected by the short life of some of the oxidants, low availability of contaminants and high oxidant demand by the environmental media.

The *in-situ* degradation of contaminants, successful treatment of a broad range of PAHs, and shorter treatment time are the advantages this method has over other methods. Following the oxidation of non-target components like dissolved/particulate organic substances, the quantity of water to be treated and toxicity, the employment of highly reactive oxidizing agents makes this technique not to be suitable for the treatment of potable water. The use of peroxide as an oxidant requires careful control of pH to be within 2.5 to 4.0 and this is not cost-efficient; hence these oxidants are not economical for treatment of a huge quantity of water (Huang *et al*. 2003). It is much more efficient to integrate chemical oxidation with biodegradation in the treatment of polluted environmental media. The high level of organic contaminants will be lowered by chemical oxidation, allowing microorganisms to work on low concentrations of the remaining organic contaminants leading to a more effective cleanup (Sutton *et al*. 2011; Chen *et al*. 2016; Xu *et al*. 2019a).

Coagulation

Coagulation is seen as a chemical water treatment technique that is done by manipulating electrostatic charges of particles that are suspended in water. The technique is executed by introducing highly charged molecules into the contaminated water to destabilize the charges on colloids, particles or oily materials in suspension. Colloidal suspensions can be abated and organic chemical concentrations like PAHs can be reduced in aqueous solutions using the coagulation technique. The choice of the right coagulant can enhance the overall success of the treatment and research have shown that various coagulants, including organic polyelectrolytes (anionic polyacrylamides, polydiallyldimethyl ammonium chloride), inorganic salts, polymeric coagulants (polyferric chloride, polyaluminium chloride, polyferric sulphate), and composite inorganic-organic coagulants can be used (Matilainen *et al*. 2010). The International Water Association (2016) stated that the high cost of chemical addition and the formation of sludge are the major challenges of applying the coagulation technique to treat wastewater. During PAHs treatment by coagulation precipitation, about 30% of phenanthrene, 40% anthracene, 57% fluroranthene and 75% pyrene were removed (Kim *et al*. 2002).

Adsorption Techniques

This process utilizes adsorbents and contaminants adhere to the adsorbent's surface through electrostatic attraction and other attractions that can be physical or chemical. Certain factors that affect the adsorption of organic compounds include temperature, pH, amount of sorbate, pore size, duration of contact and other physicochemical parameters) (Adeola & Forbes 2019). Among various treatment techniques, adsorption has been reported to be one of the fastest, widely applicable, simplest and most effective methods (Balati *et al*. 2015). Adsorption technique has found use in treating different pollutants, including heavy metals and organic compounds. Polycyclic aromatic hydrocarbons have been treated using various adsorbents such as bentonite (Karaca *et al*. 2016), nano-tubes (Paszkiewicz *et al*. 2018), carbon (Dowaidar *et al*. 2007), graphene (Li *et al*. 2018), chitosan (Crisafully *et al*. 2008) and biochar (Guo *et al*. 2018). The technique poses a risk of secondary contamination (Smol & Włodarczyk-Makuła 2017). Despite that, the adsorption technique is relatively easy to design, simple and convenient when compared to other PAHs treatment techniques. A notable research using this technique is the use of peat moss, bone charcoal, and activated carbon as an adsorbent to degrade pyrene, phenanthrene and naphthalene (Yakout & Daifullah 2013). Another notable research is the removal of phenanthrene, pyrene, fluorine among others through clinoptilolite and its modified forms by (Hedayati 2018). Some materials used in the treatment of PAHs are discussed below.

Biomass as adsorbent sources

Agricultural waste such as sawdust, sugar cane bagasse, coconut shells, rice husks, etc., are seen to be cost-effective because they are readily available and are eco-friendly hence making them a viable option for the treatment of polluted water. PAHs has been reportedly adsorbed in water using agricultural waste like wheat straw (Li *et al*. 2014), palm kernel seeds (Arshad *et al*. 2019) and rice husk (Guo *et al*. 2018). The major components of such agricultural wastes are lignin and cellulose. It can even possess some lignin's polar functional group. These groups are capable of binding aquatic pollutants via various binding mechanisms. According to (Xu et al. 2019a) and (Xu *et al*. 2019b), the conversion of "waste to wealth" encouraged the decision to explore these materials, paving an economical path to eco-friendly material science and waste management. (Qiao *et al*. 2018) published the high removal of benzo[a]pyrene and pyrene (PAHs) using biochar that was obtained from Enteromorpha prolifera at 200°C. Biochar and activated carbon (AC) are all derivatives of agricultural waste. Activated carbon precursors are mostly low-cost agricultural wastes like rice husk (Yakout & Daifullah 2013), wood (Jin & Zhu 2014), sawdust (Ingole & Lataye, 2015), shells (Alade *et al*. 2012), etc. It has been greatly employed in the removal of toxic substances in contaminated wastewater and in other industrial applications (Xiao *et al*. 2015).

Yakout & Daifullah (2013) evaluated pyrene, naphthalene and phenanthrene removal using activated rice husk. The results indicate that a considerable quantity of heat was discharged because the thermodynamic system was exothermic and spontaneous (Yakout & Daifullah, 2013). Similarly, activated carbon gotten from wheat straw was applied in PAH removal and the result indicated that with an increase in the number of aromatic rings, the adsorbent adsorption capacity improved and the adsorbent surface area had the biggest impact on the removal of both HMW and LMW PAHs with the pseudo-second-order model producing the best fit to kinetic experiments (Xu *et al*., 2019a; Xu *et al*., 2019b). Water contaminated with organic compounds was treated using biochar derived from rice straw and the material showed both photodegradation and adsorption activity (Ye *et al*. 2019). However, removal of PAHs using biochar is affected by factors such as concentration of the pollutants, pore size and adsorbent volume (Lee *et al*. 2019). Some other limitations associated with using powdered biomass are complicated regeneration, difficulty in recovering the material after treatment, as well as having a low density. To solve the problem, the biomass was entrapped in a polymeric matrix and there was a great improvement in sorption capacity, the efficiency of biomass, ruggedness, robustness and in recovering biomass after treatment (Aksu 2005). The technique has been regarded as a promising economical alternative in the treatment of aqueous PAH pollution because it was efficient in the remediation of simulated and field water samples (Cabal *et al*. 2009; Zhang *et al*. 2017).

Solid-phase extraction

Solid-phase extraction (SPE) is a remediation technique used for the treatment of LMW and HMW PAHs following the preferred solubility of PAHs in organic solvents. Solid-phase extraction has been widely studied for trace enrichment of PAHs in tap water and surface water and reported by (Van Noort & Wondergem 1985; López García *et al*. 1992; Brouwer *et al*. 1994). However, PAHs have some of its compounds that are difficult to be applied to SPE and this is due to several physical and chemical factors. One of the popular reasons is that the analytes may be lost by adsorption on the walls of the vessels. This is particularly the problem of surface water, whereby filtration is usually needed before sample application, hence leading to an increase in the loss of the analytes. To isolate PAHs from water, suitable nonpolar solvents like dichloromethane, hexane or a mixture of both, rotary evaporator and separatory funnel are some of the basic apparatuses required (Gong *et al*. 2006). The method usually involves solid-phase extraction with various stages of adsorbent. Others are membrane-assisted extraction and liquid-phase micro-extractions (Martinez *et al*. 2004; Egli *et al*. 2015; Hussain *et al*. 2018; Munyeza *et al*. 2018).

Derivatives of graphitic carbon nitride were used as sorbents for the removal of PAHs and the result ranges from 83.3 % to 103.0 % (Nian *et al*. 2019; Feng *et al*. 2020). The environmental impact and cost of solvents make this technique uneconomical to be applied for water resource recovery facilities. Another limitation is that SPE and other extraction methods are limited to the analytical determination and monitoring of water contaminated with PAHs because the technique is not robust and lacks the capacity to treat large volumes of water (Adeola & Forbes 2020).

Nanoremediation

Nanoremediation is an emerging innovative remediation technique that uses nanomaterials to remediate aqueous systems. Some of the reactive nanomaterials that are being used for the removal of contaminants are bimetallic nanoparticles like Pt-Ag, nanodots like TiO_2, metal oxides like MgO, nanoclusters like V^{+4}, and carbon nanotubes and nanocomposites (Rajan 2011; Khin *et al*. 2012; Gehrke *et al*. 2015; Guerra *et al*. 2018). The technique helps to reduce cleanup time, reduce the concentration of contaminants to almost zero and to reduce the cost of treating large-scale contaminated media (Karn *et al*. 2009; Corsi *et al*. 2018). Nanoremediation technique provides a sustainable solution to the remediation of contaminated sites and is relatively cheap compared to other traditional methods like pump treat method, chemical oxidation, thermal decomposition and solvent co-flushing (Yeung 2010; Caliman *et al*. 2011; Patil *et al*. 2016; Khan *et al*. 2019). The technique has received great attention in terms of development and research in the last ten years and it was discovered that it has great potential in the treatment of wastewater and protection of the environment (Kuppusamy *et al*. 2015; Huang *et al*. 2016). It has been used in the removal of contaminants like hydrocarbons or organic compounds, chlorinated compounds, inorganic ions and pesticides in water (Rajan 2011; Teow & Mohammad 2019; Liu *et al*. 2019a; Liu *et al*. 2019b; Zhang & Liu 2020). Successful in-situ treatment of water, wastewater and groundwater using nanoremediation is dependent on using nanocatalysts, nanoadsorbents and nanocomposite membrane systems. The good permeability ability of nanomaterials and its excellent adsorption properties permit the rapid treatment of wastewater.

Membrane-based technology

A membrane can be defined as a selective barrier that can stop the movement of various constituents of fluid and therefore enables the successful separation of the constituents. Water and wastewater treatment employ the use of some membrane processes, which include nanofiltration, ultrafiltration, microfiltration, reverse osmosis and nanostructured membranes (Mojiri *et al*. 2013). In literature, the remediation of PAH polluted water has been reported using ultrafiltration (Dudziak *et al*. 2003), microfiltration, nanofiltration, reverse osmosis and ultrafiltration (Smol *et al*. 2014). When choosing a perfect membrane, bear in mind the Molecular Weight Cut-Off (MWCO) expressed in Daltons, which reflects the molecular weight of a speculative uncharged solute that the membrane rejects 90 %. This is due to the fact that the choice of membrane determines the effectiveness of removing organic pollutants from water (Plakas & Karabelas 2012). Reverse osmosis used for the remediation of landfill leachates and water shows improved efficiencies with HMW PAHs due to the fact that the procedure is managed by diffusion and sieving techniques (Smol *et al*. 2016). Also, Smol *et al*. (2016) recorded 59% to 72% of PAHs removal using reverse osmosis, while (Smol *et al*. 2012) were successful in the removal of 66.6% to 85.0% of Σ16PAHs from industrial wastewater by utilizing the ultrafiltration process. 50% ACE, 85.9 % PHE, 88.3% FL and 91% ANT were removed from water using reverse osmosis at low pressure by (Gong *et al*. 2017). Membrane-based technologies are equally successful in the treatment of produced water; however, the technique has some limitations, such as high energy consumption and membrane fouling (Chang *et al*. 2019). Although back-washing solves the problem of membrane fouling, it subsequently results in the decline of the functionality of the membrane.

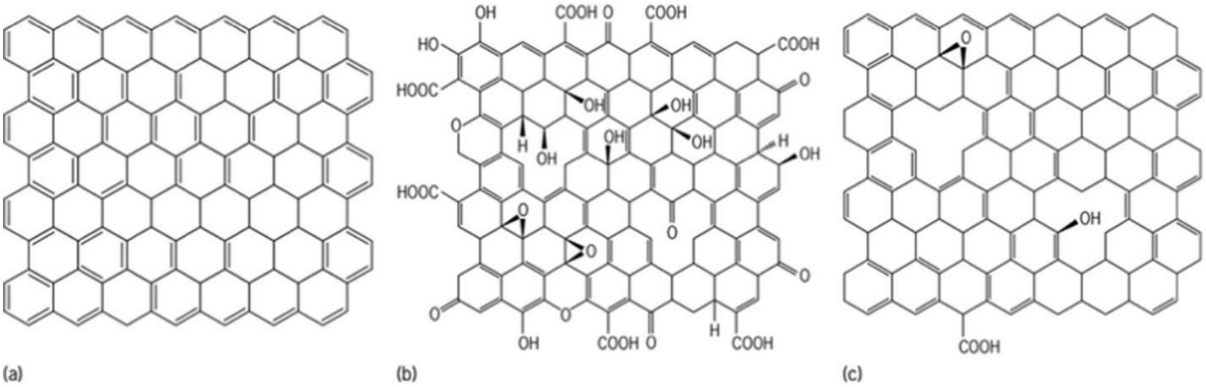


Figure 2. The crystal structure of (a) graphene, (b) GO and (c) rGO (Bi *et al*. 2012).

Graphene, graphene oxides, and reduced graphene oxides

Graphene has numerous remarkable traits, but its hydrophobic trait is probably most useful in the treatment of water. Naturally, water does not pass through graphene, but this change when small openings are created in the graphene. Studies have been made using graphene sheets that are perforated with miniature holes as a technique of water filtration since they are capable of allowing passage of water and hindering substances and contaminants (Chandler 2014). Graphenes are of various types like pristine graphene, reduced graphene oxide (rGO), graphine oxide (GO) and shown in Figure 2, and functionalized graphene. Aside from that, graphene-based polymer composites also exist and are currently being investigated for remediation purposes (Bi *et al*. 2012).

Recently, graphene was greatly studied because of its unique physical, chemical and mechanical features. It is appropriate to be used as an adsorbent in the handling of wastewater due to its enhanced active sites, very high surface area, good chemical stability and large delocalized π-electron systems (Bi *et al*. 2012; Amin *et al*. 2014; Chowdhury & Balasubramanian 2014; Gopalakrishnan *et al*. 2015). It has been effectively applied as a sorbent in the treatment of water, especially in organic and inorganic (Zhao *et al*. 2011; Wang *et al*. 2014; Ersan *et al*. 2017; Ali *et al*. 2019). Different forms of graphene in water and PAHs have a mechanism of interaction described by adsorption and partitioning and it follows the second-order reaction kinetics. The performance of graphenes is controlled by dosage and amount of PAH, which is controlled by reaction temperature and the solution's ionic strength (Lamichhane *et al*. 2016).

Choice of adsorbent for water remediation is influenced by factors like nontoxicity, material efficiency and availability, robustness, flexibility, among others. Due to wool-associated flexibility and reusability, coupled with high PAH removal efficiency of greater than 98%, graphene wool is preferable to other graphenes in water treatment. Because of its high porosity and ratio of mass to volume, graphene wool is an ideal packing material for filters or membrane separations (Adeola & Forbes 2019).

Mesoporous silica

Mesoporous nano-silica particles like santa barbara amorphous (e.g., SBA-15 and SBA-16) and mobil crystalline materials (e.g MCM-41 and MCM-48) were made out of rice husks, commercialized and characterized for several uses like treatment of pollutant, catalysis among others (Jiang *et al*. 2018). The synthesis of β-cyclodextrin-functionalized periodic mesoporous organosilica (PMO) was applied to remove five different PAHs from water was reported by Topuz & Uyar (2017) with the adsorption capacities of 0.3 – 1.65 mg/g. Phenanthrene was remediated using pentynyl β-cyclodextrin and efficiency above 95% was reported by (Choi *et al*. 2017). Table 2 outlines mesoporous silica that has been employed to adsorb PAHs. Highly porous mesoporous silica SBA-15 was synthesized with BET surface area of 521 and MSU-H with 580 units and both completely removed PHE and NAP in 20 minutes (Nasreen *et al.* 2018). Also, the adsorption of PAHs by applying hydrothermally synthesized mesoporous silica MCM-41-NH_2, was a success with a rate ranging from 93-98%, as reported by (Costa *et al*. 2020). The technique of adsorption of PAHs onto mesoporous silica was investigated by Yuan *et al*. (2018). Krupadam *et al*. (2014) discovered that the adsorbents and pollutants mainly interacted with the π-π. Adsorption efficiencies and capacities in aqueous solutions recorded 40 % to 70 % and 0.72 to 1.69 mg/g respectively, when functionalized silica with phenyl groups was used to investigate the degradation of five PAHs (Vidal *et al*. 2011).

Electrokinetic Remediation

The electrokinetic remediation technology (EKRT) is an emerging technology that has proven to be efficient and interesting following its ability to be applied in-situ (Rosestolato *et al*. 2015; Ferrucci *et al*. 2017) and compared to other methods; it is more environmentally friendly (Vocciante *et al*. 2016). The technique operates theoretically, exploiting electrons

Table 2. Adsorption of PAHs onto mesoporous silica 144

Adsorbent	**Pollutant**	**Concentration (mg/L)**	**Q_e (mg/g)**	**Equilibrium Time**	**Ref**
MCM-41	NAP	15	61	20 min	Albayati, & Kalash 2020
Periodic mesoporous Organosilica	NAP	10	1.5	24 h	
	ACE		0.9		
	FLU		0.9		
	FLUO		0.7		
	PYR		1.3		
Fe-SBA-15	PYR	0.1	0.034	20 min	Zhang *et al*. 2008
NH_2-SBA-15	NAP	1	1.9	24 h	Balati *et al*. 2015
	ACE		1.4		
	PHE		0.8		
MCM-41-PABA	NAP	0.2	17.83	24 h	Costa *et al*. 2019
	B[b]FLUO		20		
	B[k]FLUO		19		
	B[b]PYR		19		
MCM-48 SBA-15	NAP	7	0.7	24 h	Balati *et al*. 2014

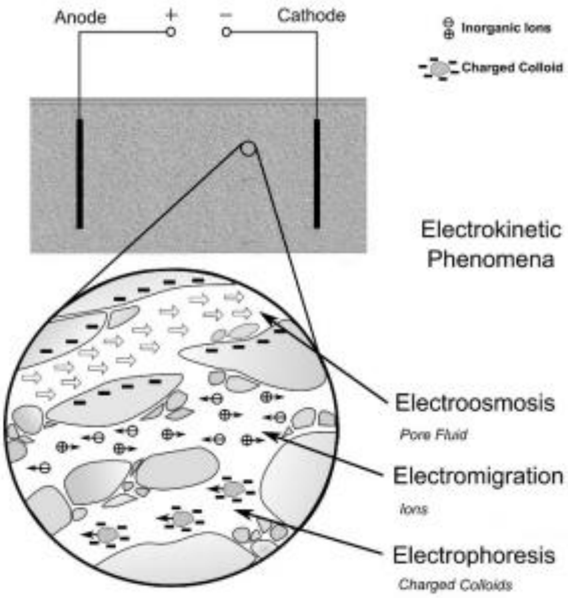


Figure 3. Illustration of the main mechanisms involved in EKRT. Adapted from Gill *et al*. (2014).

as its sole reagent (Millán *et al*. 2020). However, in order to get a good outcome within a short time, the use of complexing agents or electrolytes is usually mandatory. EKRT is an established technique in soiltreatment; however, its application in treating water contaminated with PAHs is yet to be applied on the field. The technique involves producing low electric current over an area of the environmental media that is to be treated, with the aid of electrodes that are suitably positioned in the subsurface (Reuss 1809). The applied electric field has the possibility of reaching subsurface contaminants of great depth, something other techniques are unable to achieve (Schnarr *et al*. 1998; Reddy & Saichek, 2003). The main mechanisms involved in EKRT are graphically illustrated in Figure 3.

This technique has its own challenges as well, arising from the insolubility/hydrophobicity of PAHs and its inability to readily ionize in water. Therefore, if the efficiency of PAH remediation on-field water samples using this technique is to be improved, surfactants which will help minimize tension within molecules of water and pollutants, cyclodextrins and co-solvents should be considered as additives (Saichek & Reddy 2005; Pourfadakari *et al*. 2019).

Photocatalytic Degradation

A photocatalyst is seen as a substance that can accelerate reaction without being involved in the reaction and can be activated by adsorbing a photon (Fox 1988). These substances are invariably semiconductors and in recent years, great attention has been given to semiconducting oxide photocatalysts due to their great function in environmental purification. Organic contaminants in both air and water have been successfully treated using heterogeneous semiconductor photocatalysis. The photocatalytic degradation technique has the merit of degrading organic contaminants rather than transforming them under the physical variables in the environment and this makes the technique a promising one. (Zhang *et al*. 2008) investigated the synergistic role of chemical catalysts and photocatalysis on the removal of PAHs using titanium oxide (TiO_2) catalyst and UV radiation. One of the most effective photocatalysts that have stood out over the years in water treatment is titanium oxide. It is used following its thermal stability, low cost, non-toxic, chemical and biological inert nature (Mandelbaum *et al*. 1999). The acceleration of the photocatalytic degradation of benzo(a)pyrene, pyrene and phenanthrene under UV irradiation followed the pseudo-first-order kinetics approach and this was due to an increase in TiO2 because of its proven catalytic activity (Zertal *et al*. 2004; Garg *et al*. 2019). Composites that contain photocatalytic properties were formed and used to remove PAHs (e.g., Pt/TiO_2-SiO_2) (Luo *et al*. 2015).

The application of TiO_2 photocatalysis on an industrial scale needs some limitations to be worked upon. Some of the factors that serve as a constraint to this treatment technique are the lack of cheap and efficient catalyst for high photon efficiency and the use of a wider range of solar spectra wide bandgap (3.2eV). Table 3 outlines the advantages and disadvantages of some of the techniques.

FIELD PROCEDURE

The field procedure for cleaning up PAH polluted sites involves three phases which are (1) Reconnaissance of site and assessment of risk, (2) Evaluation of the remedial options, and (3) Remediation and monitoring (Ashraf *et al*. 2014).

Phase 1: Reconnaissance of Site and Assessment of Risk

This phase investigates whether the water is contaminated with PAHs or otherwise. It also determines whether the PAHs concentration exceeds that of the maximum allowable PAH concentration in drinking water. The outcome of this assessment establishes whether the site is polluted and whether treatment is required. The source of the contamination is first addressed in order to stop further PAH release. After determining the scope of pollution, remediation goals are defined in a variety of approaches, considering the broad national recommendations and risk assessments unique to the location (including ecological, environmental and human). Risk assessments unique to the location can follow this order: problem formulation, exposure assessment, assessment of toxicity (the various effects of PAHs and the dose that leads to such effects), and the characterization of risk (Kuppusamy *et al*. 2017).

Phase 2: Evaluation of the Remedial Options

This involves the detailed evaluation of the various remediation techniques with the aim of selecting the best-fit bearing the cost, site of pollution and its environment in mind. Before the implementation of a technique in a field scale, a laboratory scale experiment will help determine the efficacy of the method. According to (Adeola & Forbes 2020), some factors that affect the field scale treatment of PAH contaminated water are:

i. Pretreatment and post-treatment cost.
ii. Physicochemical condition of the water and sediment. Such conditions can be pH, temperature, organic matter content, mineral content, redox potential, etc.
iii. Microbial/biotic community (diversity, resistance, population, symbiosis, etc.).
iv. Co-existing contaminants and targets contaminants (volatility, solubility, concentration, toxicity, etc.).
v. Non-technical factors such as infrastructure, government and standard regulations, etc.

Phase 3: Remediation and Monitoring

This is the final phase where the selected remediation technique is implemented, followed by long-term monitoring and maintenance. If the treatment fails, the procedure can be taken again or some adjustments can be made (Duan *et al*. 2015).

CONSTRAINTS

The techniques discussed in this review reflect the interest of researchers in solving the problem of water pollution. Most of these approaches have been discovered for quite a long time and are proven to remove PAHs from contaminated water. However, these remediation techniques are not always successful at field-scale due to various limiting factors. These constraints are broadly grouped into technical and non-technical factors.

Table 3. Advantages and disadvantages of some remediation techniques.

Technique	Advantages	Disadvantages
Bioremediation	-It is used in the removal of PAHs in the wastewater treatment plant and in contaminated sites. -It is a simple process with the highest approval. -Microbes are readily available at lowcost.	-Degradation of organic pollutants takes a long period of time. -Optimizing temperature at about 30-38^{o}C is a great challenge. -Microbes requires nutrients to flourish
Chemical Oxidation	-It is eco-friendly since no sludge is generated. -Organic pollutants are oxidized to water and CO_2 easily.	-May be harmful to humans if not well managed because it involves the use of hydrogen peroxide. -It is not cost-effective.
Adsorption	-Adsorbents can be reused. -It is relatively simple, easy to design and convenient. -It is a fast technique and can completely adsorb pollutants. -Different forms of locally synthetic adsorbent materials are available. -It is an eco-friendly approach.	-The spent adsorbent can be hazardous, but is normally incinerated. -Regeneration of adsorbent is expensive and difficult.
Coagulation	-Coagulants can easily make colloidal particles settle down in the water.	-It can lead to secondary pollution in the form of sludge. -Organic pollutants are not completely removed.
Photocatalytic degradation	-It is an ecofriendly approach. -It can be executed at room temperature, which is an advantage. -It completely mineralizes pollutants to CO_2 and water.	-It is uneconomical for industrial-scale applications because some photocatalysts are quite expensive. -To excite the photocatalysts, an adequate light source of energy is required.

Technical Factors

This can be related to the cost, microbes, soil and water, contaminant and co-contaminant. Theseconstraints are site-specific and the effectiveness and efficiency of a remedial technique are greatly affected by temperature, the concentration of PAHs, availability of oxygen, type of environment, microbial competition, water activity, presence of other contaminants, among others (Christofi & Ivshina, 2002). According to (Tang *et al*. 2005), to ensure success at field-scale remediation and improve remedial efficiency, the various factors have to be properly optimized. There is less bioavailability and greater toxicity if the HMW to LMW PAH ratio is higher and this results to a slower rate of degradation because bioavailability affects remediation. Surfactants can be used to boost the bioavailability of PAHs for microbial breakdown and this will solve the problem of low bioavailability (Adrion *et al*. 2016). Clean-up can be accelerated using surface-active components and organisms (fungi and bacteria) producing biosurfactants. However, not much research has been done in the aspect of using biosurfactants in the field treatment of water contaminated with PAHs and it is probably because biosurfactant production is expensive. Also, the selection of the appropriate remedial technique dictates the extent and rate of remediation. The special features of various techniques can be integrated to produce an outstanding technique with better scopes. However, one limiting factor that must be critically considered in such cases is the cost of remediation. The lack of appropriate maintenance and monitoring coupled with inappropriate remediation objectives also leads to remediation failure. In most cases, a good choice of microbes determines the success of bioremediation (Boopathy 2000; Kuppusamy *et al*. 2016d; Kuppusamy *et al*. 2016e). Some of the reasons that lead to the previous failure of bioremediation are tolerance of co-contaminants like heavy metals and the ability to withstand unstable weather conditions (e.g., fluctuating temperature, pH and salinity), among other reasons (Kuppusamy *et al*. 2016c).

Non-Technical Factors

Some non-technical factors that affect the success of PAH remediation are environmental laws and regulations, time limitation and the risk of residual contaminants that remains after remediation. Government and environmental regulations can hinder the use of certain remedial techniques; for instance, genetically modified organisms (GMOs) are yet to be approved (Pisciotta & Dolceamore Jr, 2016). All these rules equally affect the choice of tools to be used. Another constraint is the decline in research funding and this has made it difficult to meet the need of thorough research. A multidisciplinary approach is required where the expertise of a geologist, hydrogeologist, microbiologist, engineer, soil scientist, among other disciplines, is required to carefully work together to achieve remedial success. Various institutions of learning are therefore encouraged to provide relevant training that will equip scholars with the needed remedial skills (Kuppusamy *et al*. 2017).

FUTURE WORK

In preference perspective, as regards the techniques discussed in this review, the use of biomass and agricultural waste stands out as the most preferred remediation technique. This technique has proven to have a very low operational cost as agricultural waste is readily available. It proffers a good channel of waste management as it makes the conversion of “waste to wealth” possible while removing PAHs in contaminated water (Xu *et al*. 2019a; Xu *et al*. 2019b). Usually, open burning is the common technique in the treatment of agricultural waste (UNEP 2018) and this technique impacts negatively to the environment. Therefore, the application of biomass and agricultural waste in the treatment of PAHs contaminated water is truly a sustainable and eco-friendly approach. Studies on the use of mixed agricultural waste for the treatment of water polluted with PAHs can improve the adsorption outcome. There is a great potential to enhance the removal of PAHs from water using a combination of two or more different biochars. However, there are very few reports on the combination of two types of biochar for treatment. This should be the focus of future research.

CONCLUSIONS

The monitoring and treatment of PAHs using different approaches have recently received a lot of attention. Some of the techniques discussed in this review are in the prototype stage, while some have been implemented in the cleanup of PAH polluted sites and are found to be successful. However, some of these existing techniques have limitations including the production of sludge, high cost, long remedial period and incomplete remediation, among others. Emerging techniques are therefore encouraged to bridge the gaps. The three phases involved in the field procedure for the remediation of PAH contaminated water were reviewed alongside technical and non-technical constraints that hinder successful remediation. However, it is sincerely impractical to treat PAH-contaminated water to its original form. As a result, the implementation of the function-directed risk-based green remediation strategy is recommended as the most suitable technique to treat water polluted with PAHs.

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