

Review

# An Overview and Evaluation of Highly Porous Adsorbent Materials for Polycyclic Aromatic Hydrocarbons and Phenols Removal from Wastewater

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Abstract: Polycyclic aromatic hydrocarbons (PAHs) and phenolic compounds had been widely recognized as priority organic pollutants in wastewater with toxic effects on both plants and animals. Thus, the remediation of these pollutants has been an active area of research in the field of environmental science and engineering. This review highlighted the advantage of adsorption technology in the removal of PAHs and phenols in wastewater. The literature presented on the applications of various porous carbon materials such as biochar, activated carbon (AC), carbon nanotubes (CNTs), and graphene as potential adsorbents for these pollutants has been critically reviewed and analyzed. Under similar conditions, the use of porous polymers such as Chitosan and molecularly imprinted polymers (MIPs) have been well presented. The high adsorption capacities of advanced porous materials such as mesoporous silica and metal-organic frameworks have been considered and evaluated. The preference of these materials, higher adsorption efficiencies, mechanism of adsorptions, and possible challenges have been discussed. Recommendations have been proposed for commercialization, pilot, and industrial-scale applications of the studied adsorbents towards persistent organic pollutants (POPs) removal from wastewater.

Keywords: adsorption; phenols; polycyclic aromatic hydrocarbons; wastewater

# 1. Introduction

Organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), phenols, and their derivatives, are one of the major pollutants that are frequently detected in wastewater. They are usually originated from both natural and anthropogenic sources, such as bush-burning, volcanic eruptions, petroleum exploration and refining, mining etc. [1,2]. They are usually run-off into the water bodies where



they reside in the ground and surface water, thus resulting into environmental problem for decades. They are highly lipophilic, semi volatile, and accumulative in water and can be transported into various water compartments due to their non-biodegradable and persistent nature [3,4]. They are categorized as persistent organic pollutants (POP's) in the water. When present in the living cells, they are capable of undergoing bioaccumulating and biomagnifying through food webs into different trophic levels of organisms, and subsequently into humans [5].

PAHs are organic compounds consisting of two or more fused benzene ring molecules. They mostly result from petroleum refining, fuel combustion, coal mining and processing, coke production, oil shale pyrolysis, chemicals production, and other industrial processes. Various research conducted in different parts of the world have shown considerable increase in the PAHs concentrations in the receiving water bodies. Thus, PAHs could be ubiquitously transported into the rivers [6,7], fresh and terrestrial water [8], municipal wastewater treatment plants [9], and consequently exposed to the human being and aquatic organism. They are classified based on the number of rings they contained into low molecular weight (LMW-PAHs) and higher molecular weight (HMW-PAHs). The former comprised those PAHs with two to three benzene rings, while the latter are made of four to six rings. In comparisons, the latter are more toxic and persistent to the water due to their low solubility [10,11]. The United States Protection Agency (USEPA) has listed 16 PAHs as potential toxic and carcinogenic and thus they have priority concerned. The structures, molecular weight, and log *p* values of the PAHs are highlighted in Table 1.

Compound	Formula	Abbreviation	Structure	Molecular Weight (g/mol)	Partition Coefficient
Naphthalene	C <sub>10</sub> H <sub>8</sub>	NAP		128.17	3.37
Acenaphthylene	C <sub>12</sub> H <sub>8</sub>	ACE		152.1	4.0
Acenaphthene	C1H10	ACE		154.21	3.92
Fluorene	C <sub>13</sub> H <sub>10</sub>	FLU		166.22	4.18
Phenanthrene	C <sub>14</sub> H <sub>10</sub>	PHE		178.23	4.57
Anthracene	C <sub>14</sub> H <sub>10</sub>	ANT		178.23	4.54

Table 1. Chemical structures and log *p* values of polycyclic aromatic hydrocarbons (PAHs).

Compound	Formula	Abbreviation	Structure	Molecular Weight (g/mol)	Partition Coefficient
Fluoranthene	C <sub>16</sub> H <sub>10</sub>	FL		202.25	4.58
Pyrene	C <sub>16</sub> H <sub>10</sub>	PYR		202.25	4.58
Benz[a]anthracene	C <sub>18</sub> H <sub>12</sub>	B[a]A		228.3	6.14
Chrysene	C <sub>18</sub> H <sub>12</sub>	CRY	$\sim$ $\sim$ $\sim$	228.29	5.30
Benzo[b]fluoranthene	C <sub>20</sub> H <sub>12</sub>	BbF		252.3	5.74
Benz[k]fluoranthene	C <sub>20</sub> H <sub>12</sub>	BkF		252.3	5.74
Benz[a]pyrene	$C_{20}H_{12}$	BaP		252.3	6.74
Benzo[ghi]perylene	$C_{22}H_{12}$	BhP	• • •	276.3	6.52
Dibenz[a,h]anthracene	C <sub>22</sub> H <sub>14</sub>	DahA		278.4	6.20
Indeno[1,2,3-c,d]pyrene	C <sub>22</sub> H <sub>12</sub>	IP		276.3	6.20

Table 1. Cont.

Phenols are a class of organic compounds having hydroxyl group (–OH) attached to the carbon atom that is part of the aromatic ring. They have found numerous applications as intermediates for industrial production of petrochemicals, dyes, herbicides, pesticides, pharmaceuticals and cosmetics, etc. [12]. Apart from that, they are also used in household products such as flavorings, cleaners and mouthwash. Thus, they are usually discharged in the wastewater and effluents from those industries [13,14]. Even though small quantity of phenols are produced from the decomposition of plants and animals, but anthropogenic sources has been a major contributor of phenols in the environment wastewater effluents in high concentration up to even several thousands of mg/L [15,16]. Considering their adverse

effect to living organisms in the environment, they are also classified as priority pollutants [17,18]. The frequent discharge of phenolics without treatment leads to environmental toxicities to the extent that they are posing serious threats to human health. Some of their toxic effects includes; skin and eyes irritations, respiratory complications, weight loss, diarrhea, vertigo, salivation, and dark coloration of urine etc. [19,20]. Table 2 presents some of the most frequently reported phenols and their dissociation constants (PKa).

Compound	Formula	Structure	Molecular Weight (g/mol)	Partition Coefficient
Phenol	C <sub>6</sub> H <sub>6</sub> O	OH	94.11	10.0
Cresol	C7H8O	CH <sub>3</sub>	108.14	10.28
Resorcinol	C <sub>6</sub> H <sub>6</sub> O <sub>2</sub>	HOOH	110.1	19.15
Hydroquinone	C <sub>6</sub> H <sub>6</sub> O <sub>2</sub>	но-Он	110.11	9.9
Pyrogallol	C <sub>6</sub> H <sub>6</sub> O <sub>3</sub>	но ОН	126.11	9.03
Naphthol	C <sub>6</sub> H <sub>8</sub> O	OH	144.17	9.51
Salicylic acid	C <sub>7</sub> H <sub>6</sub> O <sub>3</sub>	ОНОН	178.23	2.97

<b>Table 2.</b> Chemical structures and $\log p$ values of polycyclic aromatic hydrocarbons studied
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Compound	Formula	Structure	Molecular Weight (g/mol)	Partition Coefficient
Picric acid	C <sub>6</sub> H <sub>3</sub> N <sub>3</sub> O <sub>7</sub>	OH O <sub>2</sub> N NO <sub>2</sub> NO <sub>2</sub>	229.1	0.38
Bisphenol A	$C_{15}H_{16}O_2$	но он	228.29	10.29
Chlorophenol	C <sub>6</sub> H <sub>5</sub> ClO	OH	128.6	9.12
Nitrophenol	C <sub>6</sub> H <sub>5</sub> NO <sub>3</sub>	OH NO <sub>2</sub>	139.10	7.15

Table 2. Cont.

In recognition of the mobilities and high toxicities of these pollutants in environmental waters, a prompt response from various environmental monitoring and protection is needed to reduce their concentrations, or to ideally completely eliminate them. Thus, considerable attention has been paid to develop suitable techniques for their effective remediation [21,22]. Over the last three decades, extensive studies have been conducted using different technologies including coagulation and flocculation, phytoremediation, reverse and forward osmosis, chemical oxidation, photocatalytic degradation and adsorption [23–25]. Table 3 highlights the pros and cons of some of the remediation techniques employed. Generally, most of these techniques failed to address the persistent problem, partly because the pollutants are highly lipophilic. In a conventional wastewater treatment plant (WWTPs), they are resistant to bioremediation and photolysis is insignificant for their degradations [26]. Additionally, the excessive chemical remediation might cause secondary pollution because of the unreasonable proportions of the reagents used and formation of oxygenated species that are potentially hazardous [27,28]. Photocatalytic degradation has been limited by the semi conducting property of the materials and the utilization of light energy. Additionally, most of the techniques are non-feasible and non-economical.

Method	Pros	Cons
Coagulation	<ul> <li>The colloidal particles are easily settled down by the coagulants added to the wastewater</li> </ul>	<ul> <li>It is time-consuming</li> <li>Incomplete removal of the organic pollutants are always the case</li> <li>It resulted in secondary pollution inform of sludge</li> </ul>
Flocculation	<ul> <li>Availability of various flocculants that have been commercialized.</li> <li>Some of the flocculants have shown a high affinity to the organic pollutants.</li> </ul>	<ul> <li>It is highly dependent on the physicochemical parameters of the pollutants; such as pH and ionic strength.</li> <li>The flocculants often resulted in secondary pollution.</li> </ul>
Bioremediation; including the use of microbes such as bacteria, fungi, and algae.	<ul> <li>It was employed for the treatments of PAHs at the contamination site and wastewater treatment plants</li> <li>Availability and low-cost of various microbes.</li> <li>It is a simple process</li> </ul>	<ul> <li>The method requires nutrients for the microbes to flourish.</li> <li>Temperature optimization for the condition has been a challenge.</li> <li>A long time is required to degrade organic pollutants.</li> </ul>
Reverse Osmosis	<ul> <li>No chemicals are added to the pollutants; hence no secondary pollutants are generated.</li> <li>It is available for large scale application using sophisticated reverse osmosis membranes systems</li> </ul>	<ul> <li>It is costly, especially for small and medium-scale applications.</li> <li>It is a time-consuming process.</li> </ul>
Chemical oxidations	<ul> <li>The organic pollutants such as PAHs and phenols are easily oxidized to carbon dioxide and water.</li> <li>No sludge is generated in the process; thus, it is environmentally friendly.</li> </ul>	<ul> <li>The high cost of operation and maintenance</li> <li>Since it involved the use of hydrogen peroxide, it may result in harmful effects to humans if not considerably removed.</li> </ul>

Table 3. Pros and Cons of Wastewater treatments process used for PAHs and phenols remediation.

# Table 3. Cont.

Method	Pros	Cons
Solvent extraction and ion exchange process	<ul> <li>It is a rapid process with high efficiency for the removal of PAHs and phenols in wastewater.</li> <li>The cost of operation is cheap</li> <li>The extractants are mostly reusable, thus, it is economical.</li> </ul>	<ul> <li>Some solvents are toxic; thus, cross-contamination may occur.</li> <li>The use of volatile solvents may result in fire emission.</li> <li>Not suitable at the low concentration of the pollutants.</li> </ul>
Photocatalytic degradation	<ul> <li>Complete mineralization of the pollutants to carbon dioxide and water</li> <li>The photocatalysts are upon reusable from the economic point of view.</li> <li>It is an environmentally friendly technique.</li> <li>It is mostly operated under room temperature.</li> </ul>	<ul> <li>A sufficient light source of energy is required to excite the photocatalyst.</li> <li>Some of the photocatalysts are expensive, thus not economical for industrial-scale applications.</li> <li>The photocatalytic reactor is expensive for both purchasing and maintenance.</li> </ul>
Adsorption process	<ul> <li>It is simple to design and operate.</li> <li>Availability of various forms of locally synthetic adsorbents materials.</li> <li>Low-cost adsorbents and commercial adsorbents are available.</li> <li>Complete adsorption of the pollutants can be achieved within a short time.</li> <li>It is an environmentally friendly technique.</li> <li>Reusability of the adsorbents.</li> </ul>	<ul> <li>Adsorbent regeneration is often difficult and costly.</li> <li>A high temperature is needed to dissolve high molecular weight pollutants.</li> <li>Spent adsorbent may be considered hazardous but are usually incinerated.</li> </ul>

However, adsorption has been recognized as the major technique for the effective removal due to the high affinity the various adsorbents for the organic pollutants. Additionally, the process has been recognized for its simplicity, low-cost, availability of various adsorbents materials, non-environmental toxicity, ease of design, etc. Figure 1 presents the literature survey conducted via web of science and for publications reported and patents for various remediation for PAHs and phenols. The prominence of adsorption is clearly seen among the techniques. It is thus justifiable that this review was conducted for the removal of PAHs and phenols using adsorption techniques. Critical analysis was conducted using various advanced porous adsorbents and the adsorptive performance of the materials have been highlighted. To some extent, the mechanism for the adsorptions have been discussed.



**Figure 1.** Trends of (**a**) publications and (**b**) patents on various treatment techniques for PAHs and phenols remediation: Data obtained from the web of science using keywords: Adsorptions, Advanced oxidation, Coagulation, Flocculation, PAHs, Phenols, Phytoremediation, Photocatalytic degradation, Reverse osmosis.

#### 1.1. Sources of PAHs and Phenols

Industrial activities driving by energy and power demands have been the major source of PAHs and phenolic compounds in the environment. In particular, the refining of crude oil is usually associated with the production of various by-products such as petrol, diesel, coal tar, etc. [29]. Crude and refined oil leakage have also been widely recognized as one of the major sources of PAHs and phenols in the water [30]. It has been estimated, around 5–30 million tons of crude oil spilt into the ocean every year [31], as a result of either accident or leakage oil tankers. One of the most recent ones is the oil spill which occurred in Ambarnaya river and Mauritius coast in May and July 2020, respectively. In both cases, the spills were reported to occupy large area and consequently washed into the oceans. Petroleum and industrial sludges have also been a major deposit of PAHs and phenols [32,33].

Apart from petroleum and petrochemicals, coal processing to generate heat and power, as well as coke production, has also been identified as another way through which PAHs and phenolic compounds get into the water [34]. The generation of electricity from coal has been widely practiced, taking about one third of the total electricity generation [35]. According to the Environmental Protection Agency, coal fire plants contributed to about a third of the total global water pollution due to the various toxic

chemicals such as PAHs, phenolic compound, and heavy metals discharged into the rivers, streams, and other water bodies [36]. Thus, the concentrations of these pollutants in the coal wastewater is high when discharged into the atmosphere and are said to be contributing to the persistent problem of global warming [37].

PAHs and phenols have been identified as one of the major contaminants in wastewater, ans usually exist in complex. The textile and dyeing industry have been a major contributor to PAHs. According to the China environmental statistics yearbook of 2012, discharge of wastewater from dyeing industries was about 2.37 billion. Accordingly, Ning et al., (2014), reported that the total number of 16 different PAHs with concentration range of 1463–16,714 ng/g has been detected in textile dyeing sludge from Guangdong [38]. Natural and anthropogenic sources of phenols have been discussed by Anku et al., (2016), highlighting the contribution of dissolved organic matter in increasing the phenols concentrations in the environment [39]. Floodwater is another source of which PAHs and phenolic compounds get into wastewater. The catastrophe of flooding has been a common phenomenon in different places of the world, particularly in the tropical areas, where rainfall is very high over a long period of the year [40]. For example, Chukwujindu et al. (2017) reported on the prevalent occurrences of flood along the River Niger and River Benue in Nigeria, as a result of excessive rainfall during the wet season [41]. Even though no established finding between the magnitude and frequency of a flood event has been found (especially large floods), the resulting environmental changes has been reported. However, Ciesielczuk et al. (2014) described that PAHs and phenols are usually adsorbed on the humic acids in the floodwaters and thus migrated to the water [42].

#### 1.2. PAHs and Phenols in Wastewater Treatment Plants (WWTPs)

WWTPs has been the secondary source of PAHs and phenols. WWTPs serve as the major reservoir of wastewater collected from a wide range of domestic and industrial sources. Thus, various pollutants from these sources have been found to concentrate along with the solid particles in the wastewater [25]. According to the J. Liu et al. (2011), PAHs were detected in domestic WWTP in Xian, China for a period of one year, originated from domestic usage of petrochemical substances such as oil and grease, cosmetics, etc. Therefore, WWTPs is acting as the secondary source of PAHs from the different sources [43].

### 1.3. Toxicities of PAHs and Phenols to Humans

Much attention has been given to the study of toxicities of PAHs and phenols to humans. The source of human exposure has been mostly linked to consumption of contaminated water, air inhalation or from infected foods [44,45]. The exposure is more pronounced to communities living at the site of petroleum exploration and refining or near the coastal areas where discharged wastewater is released into rivers, oceans or seas [46]. Industrial water has higher risk of contaminations than the municipal WWTPs as in the former, the wastewater is directly coming from the source such as coke production plants which contains mixture of complex compounds such as PAHs, phenols and nitrogen containing aromatics [47].

The level of bioaccumulation of PAHs in humans is determined by the toxicities which increased with molecular weight. Long term exposure usually resulted in acute toxicities, associated with carcinogenicity and mutagenicity [48]. High exposure increased the risk of cancer of various organs such as lung, breast, prostate, kidney, bladder, stomach, and skin [26]. They are also reported to suppress the immune of the body, thus they are labeled as endocrine disruptors [49]. Chukwujindu et al. (2017), studied the effect of exposure to PAHs due to overflooding of PAH contaminated water with the associated health risk. They employed the technique of incremental lifetime cancer risk (ILCR) to determine the extent of the PAHs exposure among various age groups in the study area. The finding shows that on the average, the total ILCR values obtained in 2014 were 443 and 308 chances in a million among the children and adults, respectively, and that the number of the risk of cancer was 6450 and 4480 chances for children and adults, respectively, about a 15-fold decrease in the average total

ILCR values. Thus, they estimated high risk of exposure among adults than children, attributed to the prolong duration of exposure for the adults [41].

On the other hand, phenols are potential human carcinogens. They are found to exert toxic effects even at lower concentrations. At higher concentrations, phenols has the tendency to coagulate with the proteins in the body, which may result in cellular and cytoplasmic permeability, hence, could cause damage of the sensitive cells, cardiovascular, and central nervous system [50]. Prolonged ingestion of phenols may result into mouth sore, dark urine, and even diarrhea [51].

#### 1.4. Environmental Regulations on PAHs and Phenols

In an effort to eliminate the effect of PAH and other toxic chemicals into the environmental waters, the United Nation has set the regulation to stop the establishment of coal power plants around the globe to tackle the climate change and water pollution challenges [47,52]. The United States Protection Agency (USEPA) has categorized 16 PAHs as priority pollutants of carcinogenic and mutagenic effect in water [25]. To limit the PAHs and other toxic contaminants from the environmental water, E.U has regulations to forbid sludge disposal. The Ministry of Environmental Protection of China has set the level of toxic PAHs and B[a]PYR in wastewater treatment plant as 50  $\mu$ g/L and 0.03  $\mu$ g/L [2,9] respectively.

Phenol has been detected in various water samples ranging from hazardous waste sites, surface water, rainwater, ground water, sediments, industrial and urban runoff, as well as the drinking water. The level of toxicity of phenol in human and aquatic animals has been identified as 9–25 mg/L [53]. Considering the toxicity and environmental impact of phenolic compounds in water, the USEPA has included them under the listed of priority pollutants with limited environmental discharge of less than 1 mg/L in the treated effluents and set their maximum content in potable water as 0.5 mg/L [54]. Similarly, the World Health Organization (WHO) has regulated the concentration of phenol in potable drinking water at 0.001 mg/L [55].

#### 2. PAHs and Phenols Remediation in Wastewater

Considering the negative impact of PAHs and phenols with their derivatives in wastewater, various forms of remediation have been proposed. Some of these techniques reported includes conventional methods such as coagulation, flocculation, activated sludge, and bioremediation [42,56]. Other techniques such as membrane technology, photocatalytic degradation, advanced oxidation, and electrochemical catalysis have also been reviewed [57,58]. Some of these techniques have shown considerable promise [26], while some are facing challenges of incomplete removal or degradation of the pollutants, generations of other toxic contaminants [24,59], complexity of the method, as well as the cost of application and maintenance. Hence, the quest for more alternative techniques has seemed endless.

Adsorption techniques using various adsorbent materials from natural and synthetic origins have been well studied due to their prospects to effectively removed the pollutants from water. These technique are simple, economical, and practicable [60]. The ease of operation and non-generation of secondary pollutants are other advantages enjoyed by the method.

## 2.1. PAHs and Phenols Removal by Adsorption

In the adsorption process, pollutants in the solution are trapped on the surface of suspended particles, known as the adsorbent materials. Adsorption technology is motivated by the availability of a broad spectrum of low-cost adsorbents obtained from abundant naturally occurring and waste substances such as mineral deposits, agricultural waste products, particulate organic matters, and solid industrial wastes [61]. Thus, it provides an alternative for transforming waste materials into useful material for environmental sustainability.

Apart from the naturally occurring materials, synthesized particles with higher surface area and pore volumes such as organic and inorganic polymers, porous carbon materials, graphene, silica-based

materials, have been successfully utilized as adsorbents for the PAHs and phenols removal from the wastewater [62,63]. Adsorption process is usually carried out in a batch reactor on a small scale, non-continuous process such as in research laboratories [64], or using a more advanced technique of fixed bed reactors or column for a pilot-scale industrial and water treatment plants application [65]. Villegas et al. (2016) [53] discussed on various methods for phenols removal in wastewater, however, emphasis was laid mainly on activated carbon (AC), those other highly porous adsorbents such as silica and metal-organic frameworks (MOFs) have not been considered.

In this article, we aimed at reviewing and discussing past and present scenario of using various adsorbent materials for the removal of PAHs and phenols from water. The review also provides scientific analysis and propose future research directions on the use of promising adsorbent materials for PAHs and phenols adsorptive removals in the water.

#### 2.2. Adsorption of PAHs and Phenols onto Porous Carbon Materials

The use of porous carbon materials as adsorbents for organic pollutants removal from aquatic environment was first mentioned by Walters and Luthy (1984) [66]. They argued that porous carbon adsorbents are superior to soils, sediments, and suspended organic matters in terms of removal of PAHs. This claim has catalyzed discoveries of various porous carbon materials for wastewater remediation. The use of porous carbon materials such as activated carbon (AC) prepared from various agricultural waste materials [67,68], biochar [69,70], carbon nanotubes (CNTs) [71,72], and other derived carbon materials for the adsorption of PAHs have been studied.

# 2.2.1. Biochar

Biochar is a carbonaceous solid obtained from waste materials such as agricultural wastes, sewage sludge (SSL), and petroleum sludge (PS). For example, sewage sludge is generated as a by-product of sewage treatment. It is composed of many organic and inorganic substances as well as hazardous biological materials [73,74]. Thus, these solid wastes are harmful to the environment and requires proper disposal. Interestingly, pyrolysis is used as an alternative thermochemical technique to convert the waste sludge into useful biochar for effective industrial and environmental applications. It has been identified as one of the major forms of porous derived carbon. Godlewsky et al. (2019) investigated the effect of atmosphere nitrogen (N<sub>2</sub>) and carbon dioxide (CO<sub>2</sub>) for the production of sludge derived-biochar under pyrolysis temperatures of 500–700 °C. The use of CO<sub>2</sub> atmosphere was able to generate biochar with improved properties such as higher BET surface area, porosity as compared to those obtained under N<sub>2</sub> atmosphere [56].

Biochar that are derived from biomass-derived is by far the most important means to produce the porous carbon materials. It has been estimated that, the world production of dry biomass is about 220 billion tons annually [75], thus pyrolysis treatment to energy and biochar is interesting option [76,77]. Agricultural waste materials such as rice husk [78], wheat straw [79], and palm kernel seeds [80] have been reportedly used for PAHs and phenol adsorption in water.

The review by Bedia et al. (2018) focused mainly on the use of biomass derived biochar for emerging contaminants adsorptions in water streams [81]. In comparison to sludge derived-biochar, biomass derived-biochar has been found to possess higher specific surface area and pore volumes which was attributed to the higher carbon content in the latter [77]. Thus, high removal of PAHs (pyrene and benzo[a]pyrene) has been reported by Qiao et al. (2018) using biomass derived biochar generated from *Enteromorpha prolifera* at 200 °C. The Langmuir monolayer maximum adsorption capacities were estimated to be 187.27  $\mu$ g/g and 80.00  $\mu$ g/g, for the pyrene and benzo[a]pyrene, respectively [82]. Recently, Arshad et al. (2019) reported on response surface method (RSM) optimizations of phenol adsorption onto oil–palm bunch derived biochar [80]. Lee et al. (2019) reported on the effective removal of phenol using biochar generated from food waste Factors that affect the adsorption of PAHs and phenols onto biochar include pore size and volume of the adsorbent as well as the concentration of the pollutants [20]. The review by Lamichhane et al., 2016 [25] also highlighted some advantages

of biochar in the removal of PAHs. Different biochars for the adsorptions of PAHs and phenols in water streams are summarized in Table 4.

Adsorbent	Pollutant	Concentration (mg/L)	Q <sub>e</sub> (mg/g)	Equilibrium Time	Ref
RHB Magnetic modified RHB	PHE	5	17 42	1 h	[78]
Biochar graphene/biochar composite	PHE	1	2 3	3 days	[83]
Rice-350-M	PHE Ba A	1	43 19	24 h	[69]
Elephant grass biochar	BaP	0.1	19 18	250 min	[84]
Poplar Catkins biochar	PHE	30	8 384	1120 min	[85]
Sewage sludge char	PHE PYR	0.8	-	24 h	[56]
Wood biochar Wheat straw biochar	PYR PHE FLU PYR	9.1 10.0 11	- - -	48 h	[79]
Chitin biochar	phenol 2-nitrphenol	500	184 206	12 h	
<i>Hizikia fusiformis</i> biochar	phenol	50	30	1440 min	[86]
Black spruce biochar White birch biochar	phenol	200	233 150	18 h 4 h	[87]
Chinese herb biochar	phenol	50	-	360 min	[88]
scots pine bark biochar spruce bark biochar	phenol	500	149 84	24 h	[89]
Bamboo biochar Oak wood biochar	phenol	10	17 13	24 h	[90]
Food waste biochar	phenol	10	3	30 min	[20]
Japanese red pine char Yellow poplar char	Phenol Phenol	200	6 7	30 min	[61]
Pine fruit shell biochar	Phenol	50	27	24 h	[91]

Table 4. Adsorptions of PAHs and phenols using biochars.

## 2.2.2. Activated Carbon (AC)

AC are highly porous carbon materials obtained from agricultural wastes, with high carbon contents such as cellulose, hemicellulose, lignin, lipids, proteins, simple sugars, and starch [67,92]. The production of AC involved thermal decomposition of these materials under limited supply of air, followed by the activation to generate highly porous carbon material. Thus, precursors for AC are usually low-cost agricultural waste materials such as husk [93], shells [94], wood [95], sawdust [96], rice husk [66], etc. The use of other materials such as bones, cartilages, coke etc. have also been employed [97]. It has been widely employed for a variety of industrial applications such as gas separations and purifications, liquid and gas storage, super capacitors, electrodes, catalysis, and removal of toxic substances in contaminated wastewater etc. [98]. They are usually characterized by a large surface area and large porosity than most of the conventional adsorbents ever reported. However, the properties are largely dependent on the precursor material, pyrolysis temperature, and the activation conditions. In some cases, the AC produced might contain heteroatoms and mineral matter (ash content) depending on the nature of the raw material used as precursor. A literature has shown that over 26,000 articles have been published by Elsevier on the use of AC for the adsorption of pollutants in aqueous medium. Danish and Ahmad 2018, has reviewed on the production of AC from wood biomass as adsorbent materials. Pore size, pore diameter distribution, of AC as the major contributing factor for the adsorption of the pollutants [92].

Different forms of AC such as AC powdered activated carbons (PAC) and granular activated carbons (GAC) [99,100] from different biomass has been reportedly used for organic pollutants removal from water (Figure 2). Additionally, the use of modified AC to regulate its pore structure and chemical properties for the effective removal has been studied [101]. Wu et al. (2020) recently reported on the preparation of walnut shell AC (WAC) with a surface area of 438.5 m<sup>2</sup>/g via microwave-assisted KOH activation process at 900 W for the competitive adsorption of NAP and PHE in aqueous medium. Equilibrium adsorption of 40 min, with the adsorption capacities of 39.58 and 63.37 mg/g for the NAP and PHE respectively were obtained [98]. More porous AC were produced via modified coal-base AC (MCAC) and iron-modified coal-base AC (Fe-MCAC) with BET surface area of 1062 and 1079.67 m<sup>2</sup>/g respectively for PAHs adsorption [102]. Similarly, optimizations studies for the phenols adsorption was studied using AC obtained from *Terminalia chebula* (TCAC). Equilibrium time of 24 h with the monolayer adsorption capacity of 36.77 mg/g were observed. Table 5 presented more literature on the application of AC as adsorbent for PAHs and phenols removal.



**Figure 2.** Kinetic studies and adsorption capacities for the removal of NAP and PHE using walnut shell activated carbon (WAC) under (**a**) single and (**b**) binary systems [103]. Copyright: Royal Society of Chemistry (2020).

Adsorbent	Pollutant	Concentration (mg/g)	Qe (mg/g)	Equilibrium Time	Ref
	ANT		143	4 h, 5 min	[(0]
Commercial PAC	PYR	-	142	4 h, 10 min	[00]
	PHE	120	46		
Commercial AC	FLA	80	36	24 h	[104]
	BaA	12	9.7		
	NAP	8	14		
Rice husk AC	PHE		17	24 h	[66]
	PYR		18		
Flamboyant pod back AC			4.7		
Milk bush kernel AC	ACE	50	4	2 h	[105]
Rice husk AC			5.6		
	NAP		15		
Rice husk AC	PHE	8	17.5	24 h	[106]
	PYR		18		
Pice buck AC	PHE	10	-	60 min	[107]
Rice husk AC	FLU	10	-	30 min	
Rice husk AC	ACE	50	46	3 h	[93]
<i>Vitis vinifera</i> leaf AC (Zncl <sub>2</sub> ) <i>Vitis vinifera</i> leaf AC (H <sub>3</sub> PO <sub>4</sub> )	PHE	1	94 89	180 min	[108]

Table 5. Adsorptions of PAHs and phenols using different forms activated carbon (AC).

Adsorbent	Pollutant	Concentration (mg/g)	Qe (mg/g)	Equilibrium Time	Ref
Walnut shell AC	NAP PHE	10 10	49.6 63.8	40 min	[103]
flamboyant pod AC milk bush kernel shell AC	NAP	50	294.2 63.3	2 h	[94]
Coal-based AC	NAP PHE PYR	200	78.2 111.4 117.2	20 min	[98]
Fe-modified Coal-based AC	NAP PHE PYR	30	167.8 190.4 20.3	40 min	[101]
Terminalia chebula AC	phenol	100	26.2	24 h	[109]
Commercial AC	phenol	1000	132.3	360 min	[110]
Commercial AC	phenol				[]
Miswak root AC	phenol	100	142.2	120 min	[111]
Coal-based GAC	r		169.9		[]
Coal-based PAC	phenol	300	176.6	180 min	[112]
Coconut shell PAC	1	000	213	100 11111	
Cherry stone AC	phenol	25	80	24 h	[113]
Agave Utahensis AC	I		1.7		[]
Euphorbia Resinifira AC	Phenol	14	1.9	3h	[114]
Banana peel AC	Thener	11	2.0	on	[]
Oily sludge AC	phenol	100	434	30 min	[115]
Tea waste AC	phenol	100	132.5	4 h	[67]
Babul sawdust AC	phenol	50	15.3	60 min	[96]
Olive stone AC	phenol	800	44.269	4 h	[116]
Palm oil bunch AC	phenol	40	55.3	2 h	[117]
Wood fiberboard AC	phenol	250	208	120 min	[95]
Avocado kernel AC	phenol	0.7	3.3	100 min	[118]
Date pit AC	Phenol	50	46	1 h	[119]
Sugar cane bagasse AC	Phenol	100	27	30 min	[120]
Lantana camara AC	Phenol	150	112.5	7 h	[121]
Magnetic AC	phenol	80	107.5	15 min	[122]
	Phenol.		4.9		[]
Granular activated carbon (GAC)	2-chlorophenol	50	4.3 4.5	60 min	[123]
Red nine AC	i enoropricitor		454 5		
Yellow poplar AC	Phenol	200	625	30 min	[61]
Commercial AC	1 1101101	200	500	50 11111	[01]

Table 5. Cont.

Generally, AC produced from agricultural wastes displayed better removal capacity and shorter equilibrium time for the adsorption of PAHs and phenols in the aqueous medium as compared to biochar. This is perhaps associated with the higher porosities of the AC. However, in terms of sources, biochar can be generated from different varieties and abundant agricultural waste feedstocks [70]. Additionally, from economic point of view, biochar has cheaper production cost than the AC. Unlike in the latter, the former consumes less energy and activation is necessary in most cases [79]. However, both materials are considered superior to raw biomass in their affinity towards PAHs and phenols [124]. In both cases, the adsorption occurs via hydrogen bonding and  $\pi$ - $\pi$  electron-donor-acceptor interactions between the active sites on the surface of the AC with the benzene rings of the organic pollutants and the functional groups on the phenol as seen in the Figure 3. For both AC and biochar, better removal capacities are achieved by surface modifications of the porous carbon materials. Additionally, the surface modifications could result in more stable adsorbents [98].



**Figure 3.** Mechanism for the adsorption of phenols onto activated carbons via hydrogen bonding and  $\pi$ - $\pi$  interactions [125]. Copyright: Korean Society of Chemical Engineers (2020).

### 2.3. Other Porous Carbon Materials as Adsorbents for PAHs and Phenols

Apart from biochar and AC, other porous carbon nanomaterials such as graphene, carbon nanotubes (CNTs), nanohorns, nanofibers, fullerenes, soot, molecular carbon sieves, and other carbonaceous-derived materials [126] have been studied. They are chemically heterogenous and high carbon content materials that are usually obtained from either partial or complete combustion of plant materials [127]. Many of the carbon-based nanomaterials have been investigated as adsorption material for water remediation. The most widely employed are graphene and CNTs. They are recognized with extra-ordinary physical and chemical properties that are superior to other carbon-based materials. They possess good chemical and thermal stability with much higher surface area than AC and biochar [62,128]. Their improved surface properties allow them to interact with organic pollutants via covalent and non-covalent bond formation such as hydrogen bonding, electrostatic forces,  $\pi$ - $\pi$  stacking, van der Waals forces, and hydrophobic interactions [14].

#### 2.3.1. Graphene, Graphene Oxides, and Reduced Graphene Oxides

Graphene is a form of nanomaterial having a honeycomb-like structure of sp<sup>2</sup> hybridized carbon atoms with single atom of graphite layer. The allotrope of graphene is called graphite, having a planar structure [129]. It exhibits outstanding physical and chemical properties, such as good electrical and thermal conductivity, high strength, high specific surface area and pore volumes, flexibility, and negligible thickness [130,131]. Apart from that, graphene chemical mobility, which allow for its modification to form other functionalized advanced carbon materials. Graphene oxides (GOs) and reduced graphene oxide (rGO) are among the most advanced graphene-based engineering materials (Figure 4) [132,133] They are regarded as the fascinating forms of graphene materials and thus have versatile applications in various fields such as biomedicines, sensors, metrology, electronic devices, as well as environmental pollutant's remediation in environmental waters [134].



**Figure 4.** Chrystal structures of (**a**) graphene, (**b**) graphene oxide, and (**c**) reduced graphene oxide [133]. Copyright: Elsevier (2020).

In adsorption studies, their unique chemical properties and high specific surface area has been emphasized. Sun et al. (2013), has reported on the batch adsorptions of NAP, ANT, NAP, and PYR onto graphite, GOs and RGOs. The performance of the materials in decreasing order were rGOs > GOs > graphite, with the former having adsorption capacities of 15.92, 247.17, and 369.06 mg/g for NAP, ANT, and PYR, respectively [135]. Recently, application of graphene wool for the removal of PHE and PYR from aqueous environment has been reported by Adedapo et al. (2019). Effective removal of 99.9 and 99.1% were achieved for PHE and PYR respectively, within 24 h of contact between the PAHs and the adsorbent. The adsorbent material can be repeatedly used with little or no decreased in percentage removals over 8 successive cycles [136].

In the case of phenols removal, graphene and functionalized graphene materials have been reportedly used as adsorbents. Zhou et al. (2017) reported on the fabrication of graphene composite Fe<sub>3</sub>O<sub>4</sub>@PANI-GO for the batch removal of bisphenol A, t-octyl phenol and  $\alpha$ -naphthol in water. The complete removal of the pollutants within shortest time (5 min) for bisphenol A, t-octyl phenol respectively, whereas the  $\alpha$ -naphthol experienced slightly longer time, (300 min) to equilibrium. The promising application of the material was demonstrated by its effective reusability (10 successive cycles) without decreased in the removal capacity of the pollutants [137]. The report of Wang et al., (2018) have shown that the adsorption of phenols pollutants; 2-phenylphenol (PPE), bisphenol A (BPA), 4-isopropylphenol (IPE), 4-methylphenol (ME), and phenol (PE) were more effective onto the rGO (Figure 5) as compared to the GO, attributed to the decreased in electron density in the latter, hence the strong  $\pi$ - $\pi$  interactions between the phenols and the adsorbent [138]. Tian et al. (2019), reported on the rapid adsorption of bisphenol A (3.4 s), onto hybrid material of graphene oxide with amino functionalized polypropylene (PP-g-DMAEMA/GO). The adsorbent possessed dual channel structure which allows bisphenol A to easily access the surface of the GO through the nanochannels of the propylene molecule [139].

In most cases, graphene was shown to have better performance in terms of adsorption capacity as compared to GO and rGO. In comparison to rGO, GO has more oxygen containing functional groups and lesser  $\pi$  electron density [138]. It has been established that the hydrophobic effects of the organic contaminants were the reasons for their strong absorption onto the graphene which occur though  $\pi$ - $\pi$ interactions [63,140]. In the case of GO, the dense carbonyl groups at the edge of the carbon layers were responsible for the strong adsorption of hydrophobic organic pollutants onto the surface of the material through electrostatic, hydrogen bonding, and anion- $\pi$  interactions. More details of the adsorptions of PAHs and phenols onto the graphene, GO, rGO, and their derivatives have been highlighted in Table 6.



**Figure 5.** Adsorption capacity (**a**) graphene and (**b**) graphene oxide for phenols removal. Reproduced from ref [138].

**Table 6.** Adsorptions of PAHs and phenols onto graphene, GO and reduced graphene oxide (rGO) and their composites.

Adsorbent	Pollutant	Concentration (mg/L)	Q <sub>e</sub> (mg/g)	Equilibrium Time	Ref
Cranhana waal	PHE	0.8	5	241	[10(]
Graphene wool	PYR	5	200	24 h	[126]
	NAP	30	16		
rGO	ANT	0.05	247	7 days	[125]
	PYR	0.1	369		
GO/FeO·Fe <sub>2</sub> O <sub>3</sub>	NAP	0.1	6	48 h	[64]
Graphene GO	NAP	4.6	1 66	4 h	[63]
Graphene GO	NAP	-	9 8	10 min	[141]
Sulfonated graphene	NAP	0.39	2	5 days	[142]
rGO@Sand	NAP ACE	0.8	4 6	120 min	[143]
Magnetic graphene nanosheet	PHE	0.5	0.02	1 h	[72]
Magnetic GO			14		
Magnetic chemically GO	PHE	1	30	24 h	<b>[60]</b>
Magnetic rG			26		
0	PHE	4.2	1.2		
TiO <sub>2</sub> -Graphene	FLAN	4.5	0.9	180 min	[59]
	BaP	2.5	0.5		
GO			21		
rG	NAP	20	52	24 h	[144]
Reduced graphene			145		
	Bisphenol A	5	14	5 min	
Fe <sub>3</sub> O <sub>4</sub> @PANI-GO	t-octyl phenol	5	31	5 min	[137]
	α-naphthol	10	23	300 min	
	2-phenylphenol	0.470	12	60 min	
	bisphenol A	0.350	1.56		
rGO	4-isopropylphenol	0.587	1.117		[138]
	4-methylphenol	0.550	0.841		
	phenol	0.850	0.483		
CTAB-modified graphite	bisphenol A	300	125	10 min	[129]
Amino-functionalized GO	Bisphenol A	1	2.3	3.4 s	[139]

Adsorbent	Pollutant	Concentration (mg/L)	Qe (mg/g)	Equilibrium Time	Ref
rGO GO	phenol	50	16 23		
GO	bisphenol A 4-nonylphenol tetrabromobisphenol A 2,4,6-trichlorophenol	20	21 10 11 18	1400 min	[14]
Graphene	4-chloro-2-nitrophenol	10	25	60 min	[145]
ĜO rGO	Phenol	50	23 16.32	100 min	[146]
MGO-IL	phenol	30	741	12 min	[147]
Graphene	Bisphenol A	10	87	5 h	[62]
Sulfonated graphene	1-naphthol	0.7	2.41	5 days	[142]
Graphene	phenol	50	28	400 min	[148]
GO sheet GO sponge	2,4,6-trichlorophenol	5	10 21	30 min 12 min	[149]
ĜO	Bisphenol A	-	49	30 min	[150]
GO	tetrabromobisphenol A	1	41	120 min	[151]
GO	p-nitrophenol	200	138	24 h	[51]
GO			21		
Chemically GO	1-naphthol	50	52	24 h	[144]
rG GO Ultrasonic GO	2 chlorophonol	50	145 32 134	50 min	[152]
Conventional GO Ultrasonic GO	2-chiorophenor	50	50 209	50 mm	[132]

Table 6. Cont.

## 2.3.2. Carbon Nanotubes (CNTs)

CNTs are sheets of pure form of carbon atoms covalently bonded to one another in hexagonal arrays which are rolled into a hollow cylindrical shape, with the outer diameter ranging between 1–100 nm, while the size up to several tens of micrometers [153]. Since its discovery in 1992, CNT have been identified with unique physical and chemical properties such as high strength and specific gravity, good conductivity and thermal stability, high porosity [154,155], making them potential materials have for various applications such as in optical appliances, energy conversion, electrochemical sensors, catalyst and adsorbent, micro analytical devices, biomedical devices, and drug delivery [156,157]. The application of CNTs in environmental remediation have also been investigated [158,159]. CNTs have been employed as potential adsorbents for organic and inorganic pollutants removal from wastewater due to its superior properties and more tailored surface chemistry, presenting higher surface porosities and strong chemical and physical interactions with the organic pollutants than the AC and corresponding carbon-derived adsorbents [160,161]. The surface morphology of the CNTs and the functional groups present in the organic pollutant has been contributing factor for their higher affinity for the pollutants, thus, rapid removal with higher adsorption capacities [162].

The potential of CNTs for environmental remediation of organic pollutants has been well studied. Wu et al. (2016) explored on the affinity of various aromatic compounds onto CNTs. About 22 different organic pollutants including phenols, substituted benzenes, and PAHs, have been considered and the findings confirmed positive correlations between the adsorption capacities of the CNTs based on their mesoporous surfaces to the properties of the pollutants [163]. A review on the prospects and applications of carbon nanostructured materials in water treatment was presented by Selvaraj et al. (2020) [164]. Apur et al. (2015) discussed the adsorption of organic pollutants including PAHs and phenols onto the CNTs [165]. Another report was presented by Ahmad et al. (2019) on the removal

of organic pollutants in wastewater by CNTs [166]. The preparations, properties, and modifications of the CNTs have been discussed. The adsorptions of PAHs, pharmaceutical and personal care products, surfactants and pesticides has been explored and the nature of the interactions between the pollutants and the CNTs has been highlighted. However, in most of the review articles written detailed of the optimized adsorption conditions of phenols and PAHs onto the CNTs were not mentioned.

Earlier work on the removal of PAHs from wastewater was conducted by Yang et al. (2006) using commercially obtained single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). The adsorptions were found to be pH independent due to the neutral nature of the PAHs [167]. The adsorption modeling of PAHs onto CNTs has been studied by Kah et al. (2011). Various concentrations of the PAHs were analyzed, achieving higher adsorption capacities at both lower and higher concentrations, attributed to the higher surface area of the CNTs in the range of  $200-500 \text{ m}^2/\text{g}$  [168]. Zhang et al. (2019) reported the use of magnetic CNTs for the removal of PHE from water. The magnetization of the CNTs has been shown to increase the removal efficiency as well as ease of the regeneration and reusability of the adsorbents [72]. The non-linearity of the PAHs and their non-ionic and hydrophobic nature, enable them to interact with CNTs through  $\pi$ - $\pi$  bonding [169]. Hybrid simulation analysis was employed by Yali et al. (2019) to predict the adsorption coefficient of PAHs onto MWCNT using quantitative structure-property relationship (QSPR) using genetic algorithm-multiple linear regression (GA-MLR). The data was subjected to training and testing. The explanation given was that the electrostatic and steric parameters of PAHs were the major factors responsible for the higher adsorption capacities for the MWCNT. Hence the molecular docking simulation was accurate and reliable to predict the sorption efficiency [170]. Wu et al. (2020), studied the sorption mechanisms for PAHs onto CNTs through density functional theory (DFT) and molecular dynamics simulation. The sorption energies for the individual PAHs (NAP and PHE, -33.48 and -42 kcal/mol, respectively) were calculated from the compass force field. The simulation studies were able to predict that CNTs and PAHs interacted through  $\pi$ - $\pi$  stacking and the Van der Waals interactions [171].

For the adsorption phenol onto CNTs, researchers have identified multiple interactions via  $\pi$ - $\pi$ , hydrogen bonds, and electrostatic interactions. Kragulj et al. (2015) reported on the removal of chlorinated phenols (2,4-dichlorophenols, 2,4,6-trichlorophenols, and 2,3,4,5-tetrachlorophenols) onto originally synthesized MWCNT via catalytic chemical vapor deposition (CCVD) and acid functionalization (FMWCNTs). The FMWCNTs has outstanding performance compared to the pristine form as shown by the increased in the BET surface area functionalization (61.3  $m^2/g$  and 600  $m^2/g$ for MWCNT and FMWCNT, respectively). Except for 2,4,6-trichlorophenol, which was slightly lower than 2,4-dichlorophenol, the adsorption was found to be positively correlated with molecular weight of the pollutants [172]. The mechanism for phenol adsorption onto SWCNTs and MWCNTs using quantum mechanics molecular modeling was investigated by Rezakazemi et al. (2018) [136]. The strong affinity of CNTs onto the phenols has been emphasized by the formation of  $\pi$ - $\pi$  interactions between the delocalized  $\pi$  electrons in the surface of the sp<sup>2</sup> hybridized carbon structure of the CNTs. However, modified CNTs such as oxidized CNT (CNTO) can form hydrogen bonding with the phenol, thus better affinity than the pristine CNT (Figure 6). Other factors affecting the adsorption of phenols onto the CNTs include pH, ionic strength, as well as the dispersion of the CNTs in the solution. Table 7 discussed the adsorptions of phenols and PAHs onto various CNTs.



**Figure 6.** Schematic diagram for the mechanism of phenol adsorption onto carbon nanotubes (CNT). Reproduced from ref [146].

**Table 7.** Adsorptions of PAHs and phenols onto CNTs and their composites.

Adsorbent	Pollutant	Concentration (mg/L)	Qe (mg/g)	Equilibrium Time	Ref
MWCNT	NAP	200	69.2	5 days	[173]
	NAP		10.9	5	
Commercial MWCNT	PHE	-	10.6	5 days	[167]
	PYR		10.6	2	
Magnetic SWCNT Magnetic MWCNT	PHE	500	24.82 19.37	30 min	[72]
MWCNT	phenol	0.147	0.13	3 days	[174]
	3-chlorophenol	1.25	0.38	Juays	
CNT-polymer bead	p-chlorophenol	100	24	1200 min	[71]
CNT-polyethylene glycol	phenol	20	27	40 min	[54]
CNT-polymer composite	phenol	30	262	50 min	[162]
SWCNT	nhonol	50	51	54 min	[127]
MWCNT	prienoi	50	65	36 min	[127]
CNT CNT-Fe <sub>2</sub> O <sub>3</sub>	phenol	2	1.1 2.8	2 h	[175]
Magnetic-N <sub>2</sub> H <sub>4</sub> -SH/MWCNT	phenol	40	39	700 min	[176]
Chitosan hydrogel-CNT	phenol	200	404	120 min	[177]
	phenol		1.1		
CNT	4-chlorophenol	2	1.3	120 main	[24]
CNT-Al <sub>2</sub> O <sub>3</sub>	phenol	2	2.8	120 11111	[34]
	4-chlorophenol		2.8		
CNT	phonol	50	62	24 h	[12]
CNT-deep eutectic solvent	prietioi	50	120	2411	
Chitosan-MWCNT	phenol	150	88	24 h	[17]
	phenol		24	50 min	
	2-chlorophenol		86	50 min	
CNT	4-chlorophenol	150	52	30 min	[178]
CNT-activated	phenol	150	64	50 min	[170]
	2-chlorophenol		240	50 min	
	4-chlorophenol		105	30 min	
MWCNT	hisphonol A	0.37	111	3 dave	[170]
Nitrogen doped-MWCNT	Displicitor A	0.57	440	Juays	
MWCNT	phenol	25	32	300 min	[180]
SWCNT			20		
SWCNTO	phonol	50	21	100 min	[146]
MWCNT	priction	50	18	100 11111	
MCNTO			16		
CNT-Mg-Al hydrovide	phenol	50	13		[181]
Cryr-wig-minyaroxide	4-chlorophenol	50	14		[101]
MWCNT	2,4,6-trichlorophenol	120	2.3	48 h	[182]

Thus, the efficiency of the carbon-based materials as adsorbents for PAHs and phenols removal has been well established. AC to a larger extent has been the most widely reported adsorbent. It has been prepared from the various agricultural waste materials under different conditions. Additionally, the commercial AC has been available in the markets. Thus, their removal efficiency towards the PAHs and phenols is superior to that of the corresponding biochar derived from similar agricultural waste. Generally, CNTs have been shown to offer better removal efficiency than their corresponding graphene adsorbents. This is attributed to their remarkable high surface area. The strong affinity of the materials to the pollutants is made possible by the presence of hydrogen and  $\pi$ - $\pi$  bonding interactions as illustrated in Figure 5.

#### 2.4. Other Porous Materials for PAHs and Phenols Adsorption

Porous polymers (e.g., polymer beads, molecular imprinted polymers (MIPs)) [27], mesoporous chitosan [183], and advanced porous materials, such as mesoporous silica (examples; MCM-41, MCM-48 and SBA-15) [184], metal-organic frameworks (MOFs) [185], and other corresponding composites materials synthesized or are usually synthesized from other known materials. They have been known to possess physical and chemical properties superior to that of precursor components and that of the conventional porous materials. They have ultra-high porosity and surface area, high crystallinity, uniform surface morphology, high chemical and thermal stabilities, and other promising features. Thus, they have found numerous applications in various fields such as catalysis, gas storage, sensors, actuators, drug delivery, microextraction, and wastewater remediation [186,187].

The applications of synthesized polymers and advanced porous materials as adsorbents for PAHs and phenol removals have been investigated by various researchers. In most cases, the efficiency of these materials surpasses that of carbon-based materials such as AC, due to their larger pore size and volumes providing more adsorption sites for the pollutants [188,189]. Additionally, some of these materials have displayed good selectively for the PAHs and other phenolic compounds with the formation of hydrogen bonding and  $\pi$ - $\pi$  interactions.

# 2.4.1. Chitosan

The applications of chitosan, a polymer material obtained from the shell of seafoods and natural substances for adsorption of pollutants from wastewater have received considerable attention. The advantages of the material are the low-cost and large content of hydroxyl functional groups on the surface of the materials [190]. The price of commercial chitosan is much lower compared to that of the AC and zeolites and it can be easily synthesized with much less energy consumption [191]. The adsorptions of PAHs and phenols onto the chitosan are predominated by hydrogen bonding, hydrophobic, and  $\pi$ - $\pi$  interactions [192,193].

Bibi et al. (2015) used membranes synthesized from chitosan and poly vinyl pyrrolidone (CW) and carbon nanotube (CNT). The membranes with a surface area of 246 and 253 m<sup>2</sup>/g were used for the adsorption of NAP from aqueous solution. The membranes were applied for the adsorption of NAP from aqueous solution with the removal efficiency of 93 and 97% were achieved within 120 and 150 min, respectively [194]. Crisafully et al. (2008) reported on the comparisons of various natural and commercial adsorbents materials including chitosan for the removal of PAHs [4]. Filho et al. (2018) reported on the synthesis of pectin/chitosan and pectin/chitosan/cyclodextrin polymers for the adsorption of 3 PAHs, PYR, B[b]F, and B[a]P. The adsorption of PAHs and phenols onto chitosan is summarized in Table 8. Efficient removal of the pollutants was achieved with good reusability of the adsorbents [195].

Alves et al. (2019) reported on the modification of chitosan with carbon nanotube composite with enhanced BET surface area of 1130 m<sup>2</sup>/g for the adsorption of phenol. Equilibrium was reached within 120 min with adsorption capacity of 404.2 mg/g [177]. A similar finding was reported Bahmani et al. (2019) using chitosan grafted with ZIF-8 for the phenol removal, achieving adsorption capacity of 152.3 mg/g within 30 min [196]. Guo et al. (2019) comprised the of adsorption of phenol using

chitosan and MWCNT modified chitosan. The composite material achieved higher adsorption capacity of (86.96 mg/g) than the pristine chitosan with (61.69 mg/g) [17]. Karamipour et al. (2020) reported the adsorption of phenol using cellulose acetate/chitosan (CA/Chitosan) composite. The adsorption capacity achieved was 97.43 mg/g within 9 h. A modification of the CA/Chitosan with iron (III) oxide, (Fe<sub>2</sub>O<sub>3</sub>-CA/Chitosan) improved the physical stability of the adsorbent, achieving good reusability with the adsorption capacity to 163 mg/g [197].

### 2.4.2. Molecularly Imprinted Polymers (MIPs)

MIPs are obtained from the interactions of organic and or inorganic monomers of polymerizable capabilities which are assembled around a specific template known as the 'imprint', resulting into crosslinking of the monomers to form a solid framework [198]. They are well known for their high porosity resulting from the removal of the template molecules, forming large cavities allows for the easy access of the molecules to the recognition sites [27,199]. The MIPs of inorganic molecularly imprinted materials are more flexible in comparison to those obtained from the organic monomers due to the more availability of the inorganic monomers to be selected from, hence the rigidity of the solid network as well as the adaptable porosity [200].

The adsorptive properties of the MIP towards pollutants removals in wastewater has been emphasized on their large porosity, providing more adsorption sites for the pollutants [201], as highlighted in Table 6. They also offer high selectivity to the pollutants, mainly via hydrogen bonding, ion-pair interactions, hydrophobic interactions, and Van der Waals forces [28]. The adsorptions of PAHs onto the functionalized silica aerogels has been recently reported by Saad et al., (2020). The adsorbent material has proven to be effective in the removal of NAP, ANT, and PYR, with the fluorescence measurements showing effective adsorption and selectivity of the material in the highest order of NAP, ANT, and PYR, respectively [200]. Wei et al. (2015) reported on the selective recognition of PAHs by MIPs. In comparison to the hydrophilic MIP (H-MIP) and co-monomer glycidyl methacrylate (G-MIP), the conventional MIP has higher binding capacity to the PAHs, attributed to the hydrophobic nature of the material [202].

Bhatnagar and Anastopoulos (2017) reported the adsorption of bisphenol A and other phenolic compounds in water [190]. Bayramoglu et al. (2016) reported on the synthesis and selectivity of MIP and non-imprinted polymer (NIP) for the adsorption of bisphenol A, 4-aminophenol, p-toluidine, and 2-napthol in the binary and multiple component system. The adsorption capacity of bisphenol A for the MIP and NIP were 76.7 and 59.9 mg/g, respectively, indicating the selectivity and removal efficiency of the latter [203]. Adsorptive recognition of phenolic compounds bisphenol A, phenol, 4-nitrophenol, 2-amino-4-chlorophenol and 2-napthol has been reported by Lyu et al. (2020), using MIP and ionic liquid mediated MIP (IL-MIP). Higher selective of the bisphenol A has been achieved by the IL-MIP as compared to the conventional MIP, attributed to the complementary of the imprinted cavity of the IL-MIP to the bisphenol structure [198].

Adsorbent	Pollutant	Concentration (mg/L)	Qe (mg/g)	Equilibrium Time	Ref
	Chi	itosan			
	PYR		0.1		
Pec-β-CD/Chitosan	B[b]F	2	0.2	1200 min	[195]
	B[a]P		0.2		
Chitosan	phenol				
Chitosan/CNT	phenol	400	404	120 min	[177]
	phenol	150	34		
Chitosan-cyclodextrin	p-chlorophenol	157	180	-	[193]
	p-nitrophenol	150	21		

Table 8. Adsorption of PAHs and phenols using chitosan and molecularly imprinted polymers (MIPs).

Adsorbent	Pollutant	Concentration (mg/L)	Qe (mg/g)	Equilibrium Time	Ref		
chitosan-g-PNVCL/ZIF-8	Phenol	20	152	30 min	[196]		
CA/Chitosan Fe <sub>2</sub> O <sub>3</sub> -CA/Chitosan	phenol	100	97 163	5 h	[197]		
Chitosan MWCNT-Chitosan	phenol	150	62 87	24 h	[16]		
Fe-EDA/β-CD/Chitosan	phenol p-nitrophenol p-cresol	50	243 274 298	120 min	[204]		
Magnetic chitosan	phenol	15	52	50 min	[19]		
Resin-chitosan	phenol 4-chlorophenol	50	189 99	200 min 350 min	[205]		
EDTA/Chitosan/TiO <sub>2</sub>	phenol	50	209	240 min	[206]		
Chitosan-Carbon composite	phenol	10	34	1 h	[ ]		
Magnetic graphene-Chitosan	2-napthol	40	169	60 min	[207]		
0 1	phenol		60				
	2-chlorophenol		71				
β-cyclodextrin-chitosan	4-chlorophenol	120	96	3 h	[22]		
	2,4-dichlorophenol		315				
	2,4,6-trichlorophenol		376				
Magnetic-Chitosan	bisphenol A	100	55	120 min	[208]		
Glutaraldehyde-Chitosan	phenol	50	21	60 min	[209]		
	phenol o-chlorophenol	100	71 51	240 min	[210]		
	M	IPs					
	BaP		75.9				
MIP	PYR	1	7	3 h	[211]		
	CHR		7				
MIP	PHE	0.6	16	90 min	[212]		
MIP	PYR	1	11	90 min	[213]		
MIP	bisphenol A	150	77	120 min	[203]		
vermiculite MP magnetic vermiculite MP	bisphenol A	1000	217 274	90 min	[214]		
0	phenol		86				
Surface-MIP	p-nitrophenol p-tert-butylphenol	0.75	8 7	4 h	[50]		
MIP/PES/SiO <sub>2</sub>	phenol p-nitrophenol	4000	47 37	8 h	[215]		
AMPS-Am-MIP	Phenol	50	97	180 min	[18]		
MIP II -MIP	Bisphenol A	100	87 116	20 min	[198]		
Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> @PNP-SMIP	4-nitophenol	210	134	60 min	[216]		
MIP SMIP	bisphenol-A	20	24 23	240 min	[217]		

Table 8. Cont.

However, it should be stressed that pristine chitosan, upon long term usage as adsorbents, swells. This has drastically limited its application as potential adsorbent material in wastewater remediation. However, surface modification of the chitosan with other ideal functional materials helps to improve the performance of the material and improve its long-term usage. Some reported chitosan composites used for PAHs and phenols adsorption have been highlighted in Table 6.

# 2.4.3. Mesoporous Silica

With the advancement in nanotechnology and the attempted reproduction of silica nanoparticles in 1990 in Japan [218], researchers all over the world continue to explore the potential of the materials

in various areas. Mesoporous nano silica particles such as Mobil Crystalline Materials (such as MCM-41 and MCM-48) and Santa Barbara Amorphous (such as SBA-15 and SBA-16) have been synthesized from rice husks, characterized and commercialized for various applications such as gas storage, drug delivery, oil–water separations, catalysis, and pollutant remediation [186].

Mesoporous silica has well-defined pore structures (hexagonal mesopores), narrow pore size distributions (4–12 nm), as well as high BET and Langmuir surface-area. Table 6 shows mesoporous silica that have been used for the PAHs and phenols adsorption. The adsorption capacity and equilibrium time have been stated.

Nasreen et al. (2018) synthesized highly porous mesoporous silica SBA-15 and MSU-H with BET surface area of 521 and 580 units, respectively. Both SBA-15 and MSU-H were found to be effective in the removal of NAP and PHE with the complete removal achieved within 20 min [218]. Costa et al. (2020) have reported the adsorptions of PAHs from aqueous solution using mesoporous silica MCM-41-NH<sub>2</sub> that have been hydrothermally synthesized, possessing high retention rate of the pollutants with efficiency in the range of 93–98% [219]. Comparison for the removal efficiency of MCM-48 and SBA-15 for the adsorption of NAP was investigated by Balati et al. (2014) with the latter having better efficiency, attributed to its higher larger pore diameter [220]. Yuan et al. (2018) investigated the mechanism for the PAHs adsorption onto the mesoporous silica. They have identified  $\pi$ - $\pi$  as the predominant interactions between the pollutants and the adsorbents [221].

The applications of mesoporous silica materials for phenols adsorptions has been investigated. Nasreen et al. (2018) reported that SBA-15 and MSU-H were able to adsorb 4-aminophenol and p-nitrophenol from the aqueous medium. The removal efficiency achieved was above 90% within 20 min. It was suggested that the initial properties of the silica has been retained even after a series of regeneration and reusability, attributed to the high silicon content in the materials [218]. Highly porous mesoporous silica SBA-10 and PA-10 with a BET surface area of 925 and 353 m<sup>2</sup>/g were synthesized by Yangui et al. (2017) for the adsorption of phenolic compounds. Adsorption efficiency of 75% and 67% for phenol removal were reported within 2 h when the SBA-15 and PA-10 were modified with amine functional groups [222].

### 2.4.4. Metal-Organic Frameworks (MOFs)

The application of MOFs in wastewater remediation have recently gained more recognitions due to the great potential of these materials. They are made from the coordination interaction of the metal ion with organic moieties [223]. The metal node is the central building block, while organic moiety is the ligand and together forming a framework of higher tunability [224]. They are known to possess ultra-high porosity (over 90% by volume is empty) with BET surface area higher than that of AC and mesoporous silica (up to 9000 m<sup>2</sup>/g) [225]. MOFs have higher crystallinity, thermal and moisture stability, and they can be easily regenerated from the aqueous medium through simple decantation or magnetic separation [226]. The topology of the framework also made it possible to modify the MOFs with other functional groups and materials for versatile and particular applications.

The use of MOFs for PAHs adsorptions recently been published by our groups [227]. The application of Zr-based MOFs; UiO-66(Zr) and NH<sub>2</sub>-UiO-66(Zr) have been reported for ANT and CRY adsorptions from the aqueous medium. The BET surface area of the MOFs was 1420 and 985 m<sup>2</sup>/g for the UiO-66(Zr) and NH<sub>2</sub>-UiO-66(Zr), respectively. Higher removal efficiency has been achieved within short equilibrium time (25–30 min) with percentage removals of 98.6 and 97.9% for the ANT and CRY, respectively, using UiO-66(Zr) MOF. Interestingly, all the reported MOFs have displayed good reusability for the PAHs adsorption. We have also reported on the molecular docking interaction of the Fe-based MOFs, MIL-88(Fe) and NH<sub>2</sub>-MIL-88(Fe) with CRY. Figure 7 has illustrated the binding interaction of the MOFs with the CRY molecules with the MIL-88(Fe) having the most stable complex structure with the higher binding energy ( $\Delta G_{bind}$ ) of –3.88 kcal/mol and lower inhibition constant (*Ki*) of 1.58 mM. Thus, the CRY molecules preferred to reside in the internal pores of the MOF. However, for the NH<sub>2</sub>-MIL-88(Fe), the binding energy is slightly lower, and the inhibition

constant is higher, -3.80 kcal/mol and 1.65 mM respectively, resulting into lower binding stability of the complex. In the NH<sub>2</sub>-MIL-88(Fe), the CRY molecules preferred to adsorb on the outer pores of the MOFs. The high selectivity of the MIL-88(Fe) has been emphasized by the larger BET surface area and pore size of 1242 m<sup>2</sup>/g and 12.5 nm respectively. The BET surface area and the pore size of the NH<sub>2</sub>-MIL-88(Fe) was 941 m<sup>2</sup>/g and 8.8 nm respectively [228]. Table 9 highlights the various MOFs used for PAHs and phenols adsorptions in wastewater.



**Figure 7.** The molecular docking structure of (**a**) MIL-88(Fe) and (**b**) NH<sub>2</sub>-MIL-88(Fe) with the CRY molecule, for the binding interactions [229]. Copyright: Elsevier (2020). Metal (Fe), carbon (C), oxygen (O), and nitrogen (N) are colored as yellow, green, red, and blue, respectively. The CRY is indicated in purple.

Adsorptions of phenolic compounds onto MOFs have also been studied. Most of the reported MOFs are promising adsorbents for the removal of the pollutants studied. The famous HKUST-1 has shown good removal efficiency for the p-nitrophenol, with complete removal achieved within 40 min (equilibrium adsorption capacity of 400 mg/g). The high adsorption capacity of the MOF for the p-nitrophenol was attributed to the high affinity of the MOF to the NO<sub>2</sub> side of the pollutant, resulting into hydrogen bonding interactions [229]. The high selectivity and removal efficiency of the crystalline and water stable Zr-based MOF, NH<sub>2</sub>-UiO-66(Zr) for the adsorptions of 2,4,6-trinitrophenol, 2,4,6-tri-nitrotoluene, and 2,4-dinitrotoluene in water was also investigated. Higher equilibrium adsorption capacities was achieved with the hydrogen bonding formation between the MOF and the pollutants [230]. Luo et al. (2019) reported on the enhanced removal of bisphenol A using Al-based composite Al-MOF/SA-CS. The adsorption capacity was 136 mg/g. The adsorption mechanism has been explained by the formation of hydrogen bonding,  $\pi$ - $\pi$  stacking and cation- $\pi$  interactions [231].

Adsorbent	Pollutant	Concentration (mg/L)	Qe (mg/g)	Equilibrium Time	Ref
	Mesop	orous Silica			
MCM-41	NAP	15	61	20 min	[184]
	NAP		1.5		
D 11	ACE		0.9		
Periodic mesoporous	FLU	10	0.9	24 h	
organosnica	FLUO		0.7		
	PYR		1.3		
Fe-SBA-15	PYR	0.1	0.034	20 min	[188]
	NAP		1.9		
NH <sub>2</sub> -SBA-15	ACE	1	1.4	24 h	[232]
	PHE		0.8		_

Table 9. Ads	orption of PAHs and	phenols onto mesor	porous silica and	metal-orga	anic frameworks (	MOFs).
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Adsorbent	Pollutant	Concentration (mg/L)	Q <sub>e</sub> (mg/g)	Equilibrium Time	Ref
	NAP		17.83		
	B[b]FLUO	<b>2</b> 00 /T	20	041	[000]
МСМ-41-РАВА	B[k]FLUO	200 µg/L	19	24 h	233
	BIDIPYR		19		
MCM-48	-[-]		07		
SRA 15	NAP	7	0.4	24 h	[220]
3DA-13	4 -1-1		1.41		
NUL CDA 1E	4-chlorophenol	250	141	<i>c</i> 1	[00.4]
NH <sub>2</sub> -SBA-15	4-bromophenol	250	115	6 h	234
	4-iodophenol		72		
MCM-41-	phenol	50	12	24 h	[222
WICIVI-41-	o-chlorophenol	50	30	2411	[222]
MCM-48-Chitosan	Phenol	500	149	24 h	[183]
	4-aminophenol		10	•••	
SBA-15	4-nitrophenol	-	8	20 min	235
Rice husk silica	phenol	15	4	60 min	[236]
MCM-41	phenol	60	1	00 11111	[200
1010101-41	phenoi	00			
	MC	)Fs			
	ANT		24	30 min	
UiO-66(Zr)	CRY		24	25 min	
$NH_2-UiO-66(Zr)$	ANT	4	22	30 min	227
	CRV		10	25 min	
MII $99(E_0)$	CKI		19	23 mm	
NIL-00(Fe)	CRY	4	24	25 min	[228]
$NH_2$ -MIL-88(Fe)			22		
MIL-88(Fe)			24		
Mixed-MIL-88(Fe)	ANT	4	23	30 min	237
NH <sub>2</sub> -MIL-88(Fe)			21		
MIL-88(Fe)	D\/B	4	24	40	[000]
NH <sub>2</sub> -MIL-88(Fe)	PIK	4	23	40 mm	[238]
MIL-53(Al)			329	90 min	
MIL-53(AI)-F127	bisphenol A	250	473	30 min	239
MIL-68(Al)/PVDF	p-nitrophenol	10	126	720 min	[240]
HKUST-1( $C_{11}$ )	p-nitrophenol	200	400	30 min	[220]
C = C = C = C = C = C = C = C = C = C =	p-introprietor	200	522	30 mm	[241]
$SIO_2 \otimes IVIIL-00(AI)$	annine	3000	552	40 S	[241
[Zn(AIA)(BPD)]	2.4-dichlorophenol	60	-	90 min	[242]
MOF-VII	, , , , , , , , , , , , , , , , , , , ,		-		
[Zn(TDC) MOF	2, 4-dichloropheno	60	-	180 min	243
MIL-68(Al)	phenol	1000	118	120 min	[244]
CNT@MIL-68(Al)	phenol	1000	257	120 11111	[ <del>244</del>
	2,4,6-trinitrophenol		23		
	Styphnic acid		24		
NH2-UiO-66(Zr)	2.4-dinitrophenol	100	30	36 h	[230]
	2 4 6-tri- nitrotoluene		0.5		1
	2,1,0 the introtoluono		2		
	dichlorophonovaractic		2		
MIL-53(Al)	acid	150	556	1 h	[245]
MIL-68(Al)	aciu		271	17 h	_
$MII = 68(\Delta I)/CO$	p–nitrophenol	300	332	17 h	[51]
MH_MII 88(E_)	216 trinitronhanal	25	16/	10 min	[246]
MOE 100(Cm)	$2, \pm, 0$ -unnutophenol	33	10 <del>4</del> =0	40 IIIII 200 main	[240]
MOF-199(Cu)	rnenol	500	58	500 min	[247]
	<i>p</i> -nitro phenol		68	30 min	
AI-MOF/SA-CS	bisphenol A	50	137	18 h	231
Cu-BDC MOF			60		
Cu-BDC@GrO	bisphenol A	200	182	40 min	[248]
Cu-BDC@CNT	_		164		

# Table 9. Cont.

#### 2.5. Pilot and Industrial Scale Adsorption of PAHs and Phenol in Wastewater

Pilot and industrial wastewater treatment plants have been the major repository of PAHs and phenols. In most cases, these pollutants are washed from the soil by heavy rain or directly emitted from petroleum refineries, petrochemical industries, and coal mining and processing areas. Thus, they are deposited into the municipal wastewater and wastewater treatment plants. Various studies have shown the presence of these pollutants at much higher concentrations than the maximum tolerable limit in the water. Thus, the target of wastewater remediation technologies has been the wastewater for both industrial and municipal treatment plants. Application of adsorption technologies at pilot and industrial wastewater has been well explored [250].

Activated carbon has been the adsorbent most widely used for the post-treatment of non-biodegradable pollutants such as PAHs and phenols in the wastewater treatment plants due to its large surface area and uniformity of surface microspores [251]. With the availability of various low-cost and highly porous adsorbents materials, some researchers have endeavored to explore the adsorptive removals of the PAHs and phenols at both pilot and industrial scale treatment plants. Kalmykova et al. (2014) have explored the performance of sand, granular activated carbon (GAC), and peat moss filters for PAHs and phenol adsorption at pilot-plant. The performance of each filter has been assessed with the GAC achieving complete removal of phenols, while the peat filter was able to remove 50–80% of the phenols. For PAHs removal, the percentage achieved by GAC and peat were 50% and 63%, respectively [252]. The application of resin adsorbent, Amberlite XAD16, was reported by Frascari et al. (2016) for the removal of phenols from olive mill wastewater using continuous flow column at pilot-scale. The Amberlite XAD16 was well characterized with BET surface area of  $800 \text{ m}^2/\text{g}$  and average particle size of 0.63 mm. The adsorption capacity of the material was evaluated by the Langmuir model at 40 mg/g with over 70% removal efficiency achieved by the material [253]. El-Nass et al. (2016) have reported on the pilot-scale removal of phenol from refinery wastewater. About 250 kg of GAC with a particle size of 1.5 mm was used as the bed in the packed column of 1.8 m and 0.6 m length and internal diameter respectively. Over 90% removal of the phenols was achieved by the adsorbent, indicating the efficiency of GAC in removal of organic pollutants from the refinery wastewater [251].

## 3. Conclusions

Ubiquitous transport, distributions, and the fate of PAHs and phenols in wastewater has been a major environmental challenge for decades. These pollutants have been well identified as highly toxic to both fauna and flora and pose carcinogenic and mutagenic effects to humans. Hence, their environmental abatement, particularly from industrial, municipal, and wastewater treatment plants (WWTPs), has been highly desired. Various wastewater remediation technologies have been proposed. This article presented comprehensive review on the use of adsorption technology for PAHs and phenols remediation from water. The application of various porous adsorbents materials has been studied.

Porous carbon materials biochar and AC have discovered various applications in environmental remediation. Those materials have been successfully prepared from various agricultural wastes and been widely used as adsorbents for PAHs and phenols removal. In comparison to biochar, AC has shown more promising and potential removal capacity, attributed to the higher porosity of the AC. Apart from the biochar and AC, other derived porous carbons such as the graphene and CNTs have been widely employed. Their potentially higher removal efficiencies have been commended. This is attributed to their remarkable high surface area and strong affinity to the pollutants through the formation of hydrogen and  $\pi$ - $\pi$  bonding interactions, as we have illustrated.

Under similar conditions, the use of synthetic polymers, chitosan and MIPs, have been presented. Despite the selectivity of the polymers, the swelling effect of the chitosan limits their applications. However, modified chitosan has been shown to offer better removal capacity than the pristine form. To the larger extent, the potentials of advanced porous materials; mesoporous silica (such as MCM-41,

MCM-48, and SBA-15), and MOFs had been discussed. The superior adsorption capacity of the silica and MOFs adsorbents had been in all way connected to their ultra-porosities and high stability of the materials. Their strong affinity to the PAHs and phenol has been highlighted.

Thus, application of various porous adsorbents has been presented. Findings from previous works from literature have been compiled and analyzed. This review thus highlighted the potential of adsorption technique for the removal of PAHs and phenols from environmental wastewater.

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