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Aromatic Hydrocarbon Compounds as Environmental Pollutants and Factors Affecting the Degradation Process By Some Microorganisms

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Abstract
Petroleum hydrocarbons (PAHs) are one of the most important and complex natural and heterogeneous organic compounds which consist of hydrocarbons, small percentages of sulfur, ***,*** *oxygen, some trace elements, and some compounds that contain a heterogeneous nitrogen atom, in addition to many other minerals such as copper, nickel, iron, and vanadium. . This review reported the different types of PAHs which have been isolated and characterized. Degradation of these materials with different methods was also discussed. PAHs have been divided into three main sources which are pyrogenic, petrogenic, and biogenic in the environment. These Polycyclic aromatic hydrocarbons materials are consisting of two or more benzene rings and classified into two groups, materials with low molecular weights and other with high molecular weights compounds. The first are characterized by higher solubility and higher volatility and are easier to degrade by microorganisms while Polycyclic aromatic hydrocarbons with high molecular weight are difficult to be removed or degraded by different methods and need new protocols to be applied*

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I. Introduction

Hydrocarbons are among the largest environmental pollutants in the world derived from crude oil. Where numerous studies and reports related to the study of its toxicity confirm the important need to remove them from the terrestrial and water environments (Ławniczak et al., 2020). Since it is one of the common pollutants in the environment, there are many concerns regarding the polluting compounds associated with oil (Suliman et al., 2021).

Petroleum hydrocarbon compounds are used as fuel widely in all parts of the world, and due to the increasing energy demand, it results in many environmental problems, such as pollution resulting from these compounds as a result of exploration, transportation, maintenance, production, storage, accidental accidents of oil tankers and also as a result of many natural processes, which lead to all these factors lead to major environmental problems that affect marine environments. Approximately 5.74 million tonnes of oil have been lost due to transportation accidents. There are also other sources of these pollutants, such as oils leaked from engines, car workshops, diesel, jet fuel, and others (Spierings and Mulder, 2017). When an oil spill occurs in the water environment, the oil is less dense than the water, and it forms a layer on the surface of the water according to the amount of oil spilled, as it happens that the components of the oil volatilize after the spill process through the process of evaporation (Ahmed and Fakhruddin, 2018; Srivastava et al., 2019).

II. Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are considered to be organic pollutants that are characterized by their chemical stability and resistance to biodegradation processes. They are also widely distributed in the environment and are considered hydrophobic compounds. They are formed as a result of the incomplete combustion of oil, coal, wood, gas, garbage. They are toxic nonpolar organic compounds consisting of more than 100 individual moieties, containing carbon and hydrogen atoms, and consisting of many compact aromatic rings (Rehmann et al., 2008). PAHs molecules contain two important compounds, carbon, and hydrogen. They had two to many rings, where the number of compounds consisting of 2-5 rings is about 20 compounds and compounds that contain 6-8 rings are about 500 compounds. These compounds are usually formed by condensation of small organic compounds, as these compounds decompose at high temperatures. Polycyclic aromatic hydrocarbons with larger rings are formed at lower levels than PAHs with fewer rings, as this is done by the addition of many rings due to the process of kinetic constraints and the presence of many isomers in these compounds (Motarjemi et al., 2013).Crude oil contains many PAHs compounds, which constitute 0.2 to 7% of the crude oil. These compounds consist of many sources in the environment, including fossil fuels, waste materials, and the combustion of organic materials. They are produced by thermogenesis processes, both in refined and crude oil (Coppock and Dziwenka, 2019).

2.1 Physical and Chemical Properties of PAHs

Polycyclic aromatic hydrocarbons are several organic compounds consisting of two or more benzene rings, usually colorless solid or pale yellow or white (Abdel-Shafy and Mansour, 2016) They are usually arranged in a linear, cluster, or angular configurations. A linear skeletal structure is unstable compared to cluster, or angular configurations and this, in turn, affects many parameters such as sorption and solubility. Photochemical and chemical reactions, vapor pressure, high boiling and melting points, and ionization processes were reported. Based on the variety of structures, PAHs have been divided into non-alternative classes and alternative classes. Non-alternant PAHs contain a benzene ring as well as four or five carbon rings such as fluoranthene and fluorene, while alternate PAHs consist only of molten benzenoid rings such as pyrene, phenanthrene, anthracene (Harvey, 1991; Chauhan et al., 2021).

They are also classified according to their number of aromatic rings (Table 1) into PAHs high molecular weight and PAHs low molecular weight. Polycyclic aromatic hydrocarbons contain two or three aromatic rings such as phenanthrene, naphthalene, anthracene, and fluorene, while PAHs contain more than four rings such as chrysene, fluoranthene, pyrene, benzo(β)fluoranthene, and benzo[α]pyrene (Dai et al., 2021).

Polycyclic aromatic hydrocarbons are characterized by their low solubility in water and increased susceptibility to the formation of fats, and therefore these compounds are more rebellious and aggressive in the environment, and most of them have a boiling point greater than 100°C and a melting point greater than room temperature. Polycyclic aromatic hydrocarbons character depends on the type of cyclic structure and the number of benzene rings in the compound, as the stability of these compounds increases with the increase in the number of rings and thus negatively affects the rates of degradation of these compounds in the environment by microorganisms (Lima et al., 2005). PAHs with LMW compounds are characterized by having higher solubility and higher volatility and are easier to degrade and remove by microorganisms. Polycyclic aromatic hydrocarbons with HMW is absorbed by sediments and soils due to their increased hydrophobicity and are therefore less available to microorganisms in the environment (Froehner et al., 2018).

2.2 Source and Distribution of PAHs

Polycyclic aromatic hydrocarbons are ubiquitous in the environment in air, sediment, water, and soil (Agbozu et al., 2019; Kungwani et al., 2022).Polycyclic aromatic hydrocarbons occur naturally in fossil fuels (Gao et al., 2018, Al-Shiti, 2017; Dhanya and Kalia, 2020). The sources of PAHs have been divided into three main sources which are pyrogenic, petrogenic, and biogenic in the environment (Lin et al., 2020).

2.2.1 Pyrogenic Sources of PAHs

Pyrogenic PAHs are formed by incomplete combustion of organic matter and by pyrolysis. These compounds are formed by exposure of organic matter to high temperatures in conditions of low or no oxygen. Many unintended processes occur for the distillation of coal into coke or coal tar, in addition to the bio-cracking of the residues of hydrocarbon compounds and their conversion into light compounds, all of which are produced by the pyrolysis of these compounds. In addition, many other processes occurred, which represent the incomplete combustion of wood, the incomplete combustion of fuel in engines, and the incomplete combustion of fuel used in heating (Stout et al., 2015). These processes can occur at temperatures from 350°C to more than 1200°C, and PAHs can form at lower geological temperatures of 100–150°C and pressures over millions of years (Abdel-Shafy and Mansour, 2016, Marris et al., 2020).

Table 1. Physicochemical properties some PAHs pollutants

No	PAHs	CF	Rings	MW	BP	MP	PD	SW
1	**Naphthalene**	$C_{10}H_8$	2	128.17	218	80	Gas	31
2	**Acenaphthene**	$C_{12}H_{10}$	3	154.21	279	95	Gas	3.8
3	**Acenaphthylene**	$C_{12}H_8$	3	152.19	207	92	Gas	16.1
4	**Anthracene**	$C_{14}H_{10}$	3	178.23	342	216	Particle-gas	0.045
5	**Phenanthrene**	$C_{14}H_{10}$	3	178.23	340	100	Particle-gas	1.1
6	**Fluorene**	$C_{13}H_{10}$	3	166.22	295	116	Gas	1.9
7	**Fluoranthene**	$C_{16}H_{10}$	4	202.26	384	110	Particle-gas	0.26
8	**Benz[*a*]anthracene**	$C_{18}H_{12}$	4	228.29	435	160	Particle gas	0.011

* CF**, Chemical formula;** MW- **Molecular weight;** BP- **Boiling point (°C);** MP- **Melting point (°C);** PD- **Phase distribution;** SW- **Solubility in water (mg/L).**

2.2.2 Petrogenic Sources of PAHs

Polycyclic aromatic hydrocarbons are made from carbon compounds that are formed by thermal effects deep in the earth. These compounds are found in effusive rocks, rift zones, mineral oils, and hydrothermal systems. Petrogenic PAHs enter the environment and soil through oils, erosion of hard rock, and creosote-treated railways. In addition, there are other sources of these compounds, such as small releases caused by transportation of gasoline and other materials, diesel fuel, petroleum spills in different environments, lubricating oils, and leaks resulting from storage facilities (Sakari, 2012; Hussain et al., 2018). Petrogenic PAHs consist mainly of PAHs (LMW) compared to pyrogenic PAHs which consist mainly of different sources of PAHs (HMW), (Marris et al., 2020).

2.2.3 Biogenic Sources of PAHs

Polycyclic aromatic hydrocarbons are produced as a result of the biotransformation process by some types of microorganisms, macrophytes, and algae or by degradation of the plant material that formed these compounds. Biogenic sources of PAHs are formed either from natural sources or anthropogenic sources (Abbas et al., 2018).

There are many sources of biogenic PAHs such as microbes, plants, wastewater treatment plants, lignin compounds, tobacco, microbial lipids, aromatic amino acids, grilled and smoked food, incinerators, burning wood, emission from burning of motor fuels, wax on the surface of leaves, cigarette and industries (Abdel-Shafy and Mansour, 2016; Geng et al., 2019).Tropical wood contains PAHs such as perylene, naphthalenes, and phenanthrene. Many researchers believe that organisms accumulate PAHs and others believe that they are made by bacteria, algae, and plants, and therefore the possibility of their accumulation by living organisms is the most important (Smith et al., 2001, Belis et al., 2011, Mandal et al., 2018).

2.3 Fate of PAHs in the Environment

Polycyclic aromatic hydrocarbons can enter various environments through many industrial activities such as gas-processing activities (Zhang and Tao, 2009, Sakari, 2012, Abdel-Shafy and Mansour, 2016, Hussain et al., 2018). There are three primary modes of exposure to PAHs: inhalation, ingestion, and direct contact. One of the most important of these factors is the period of exposure to these compounds. Many symptoms may accompany short exposure to PAHs such as eye and skin irritation, diarrhea, vomiting, headache, nausea, dizziness, and confusion, and these symptoms occur when exposure to these compounds at high concentrations. Polycyclic aromatic hydrocarbons are widely present in aquatic environments, in marine mammals, fish, benthic invertebrates, sediments, seabirds, water, and other aquatic organisms (McNutt et al., 2012, Pittman, 2016, Alonso et al., 2019, Kungwani et al., 2022).

2.4 PAHs removal techniques

Treatment processes work to reduce pollutants in different environments through the processes of degradation and transformation in water, air, and soil until the limits of safe levels for these pollutants are reached. The pollutants caused by PAHs are considered as pollution sources of concern all over the world due to their harmful and toxic effects on the environment. Therefore, to restore environments polluted by these compounds, many strategies have been used to reduce the impact of these pollutants and remove them from the environment. There are many techniques used in the treatment of hydrocarbons, and these methods are Physical/chemical methods, and biological methods are used. In this review the biological methods was discussed.

2.4.1 Bioremediation

Bioremediation is the process of converting pollutants of interest, either completely or partially into its basic components by microbial such as fungi, bacteria, algae, protozoa, and nematode. This process can also be called biorestoration or reclamation. Microorganisms play an important role in converting pollutant compounds (PAHs) into cellular biomass, water, and carbon dioxide. Bacteria, fungi, and algae are considered microorganisms that are successful in the process of degrading PAHs in the environment. Several researchers have found that there are more than 100 genera and more than 200 species. Mandal and Das (2018) reported the removal of PAHs using yeast consortium. Mansouri et al. (2017) reported the ability of some bacteria such as *Bacillus subtilis, Mycobacterium* sp. and *Alcaligenes denitrificans* to degrade PAHs (LMW).

2.4.2 Degradation of PAHs by bacteria

Many physical and chemical processes degrade PAHs, but microbial biodegradation is one of the most promising processes for removing PAHs. Biodegradation is an important environmentally friendly process for removing pollutants from the environment. Many microbes degrade PAHs pollutants by using these compounds as an energy source and decomposing these compounds into CO_2 and water, thus transforming these pollutants into less toxic or non-toxic compounds. The biodegradation process of PAHs depends mainly on the ability of bacteria to degrade these compounds and on the bioavailability of these organisms, which depends on their availability or decrease through degrees of alkylation and number of rings. Bacteria that degrade PAHs can be obtained from environments contaminated with these compounds, where there is a large diversity of microorganisms, usually bacteria, that bioremediate these compounds. Many types of bacteria metabolize PAHs that have been isolated from various sources such as *Pseudomonas* sp., *Alcaligenes denitrificans*, *Rhodococcus* sp, and *Mycobacterium* sp.

Several studies have revealed the presence of many types of bacteria that degrade PAHs. Samanta et al. (2001) in their study on Nap degradation which revealed the presence of many types of bacteria such as *Vibrio* sp., *Alcaligenes denitrificans*, *Pseudomonas putida*, *P. fluorescens*, *P. cepacia*, *P. vesicularis*, *P. testosteroni*, *Corynebacterium venale*, *Rhodococcus* sp., *Moraxella* sp., *Bacillus cereus*, *Cyclotrophicus* sp., *Streptomyces* sp., *Mycobacterium* sp., and *Bacillus* sp. Amran (2020) in his study which was conducted on the degradation of PAHs by halophilic bacterial consortium isolated from the Red Sea coast of Jeddah, Saudi Arabia, where several bacterial strains such as *Propionispira*, *Microbacterium*, *Ochrobactrum*, *Martelella*, *Azospira*, *Gordonia*, *Flavobacterium*, *Fusobacteria*, *Bacillus*, *Mycobacterium*, *Rhodococcus*, *Pseudomonas*, and *Aeromonas* were detected. Bacteria isolated from a polluted environment are considered to have a higher ability to cause biodegradation and are more efficient than bacteria isolated from a non-contaminated environment, due to the adaptation of bacteria in polluted areas. It was reported that marine bacteria have high efficiency and degradation rates for petroleum compounds, reaching 100%. Table 2 showed some of the major bacterial genera that are involved in the biodegradation of PAHs and petroleum hydrocarbons.

2.5 The Role of Enzymes in the Degradation of Petroleum Hydrocarbons

They can be defined as biological catalytic compounds that facilitate the conversion of substrates into compounds or products by providing appropriate conditions that reduce the energy required to activate the reaction. Microorganisms utilize different enzymes to degrade petroleum, producing degradation by means of enzymatic metabolism. There is also the possibility that petroleum compounds are degraded by genes in the plasmid or chromosomal DNA. Oxidoreductase is produced and secreted by various types of bacteria, fungi, and higher plants, and oxidative coupling detoxifies compounds. Oxidoreductase releases chloride ions, CO_2, and methanol by oxidizing the compound by transferring electrons from the reducing agents to the oxidant. When pollutants are broken down by oxidoreductases, heat or energy is generated and used by microorganisms for metabolism activities (Khare and Prakash, 2017). There are several types of enzymes grouped. There are two types of biodegradations of pollutants using microbes: aerobic degradation and anaerobic degradation.

2.6 Aerobic Degradation

The hallmark of aerobic degradation is the interference of oxygen into the degradation pathway (Pieper, 2005; Rojo, 2010). Most reactions to aliphatic and aromatic hydrocarbons are carried out by monooxygenases, which add oxygen (one) to the hydrocarbons. Under aerobic conditions, Dioxygenase works by adding one atom to two of O_2 to PAHs, whereby O_2 acts on the primary enzymatic activation of these compounds and also as a terminal electron acceptor. When oxygen is incorporated into the rings of PAHs, reactions are then catalyzed by mono- or dioxygenases (Gibson, 1978). Under oxidative conditions a biochemical strategy is used to activate PAHs by introducing a hydroxyl group (dihydroxylation by dioxygenases) to the rings of aromatic compounds.

2.7 Anaerobic Degradation

Nitrogen, sulfate, and iron are reduced in hydrocarbons degraded by bacteria. There are two classes of anaerobic bacteria: strict and facultative.Anaerobes bacteria are almost entirely responsible for reducing sulfate. Anaerobes can degrade hydrocarbons either with or without oxygen; however, facultative anaerobes are oxygen-independent.In anaerobic degradation nitrogen and sulfur are used by bacteria to complete the degradation.Bacteria convert aromatic compounds into fatty acids, thus the main carbon chain is degrading through the free radical mechanism (Boll et al., 2002).

Table 2. Some major bacterial genera involved in the biodegradation of PAHs and petroleum hydrocarbons.

Genus	Reference	Genus	Reference
Propionispira	Amran (2020)	*Alcanivorax*	Zadjelovic et al. (2020)
Marinobacter	Chernikova et al. (2020)	*Oleispira*	Yakimov et al. (2007)
Alcaligens	Durán et al. (2019)	*Ralstonia*	Purnomo et al. (2019)
Corynebacterium	Zhang et al. (2016)	*Martelella*	Amran (2020)
Microbacterium	Wang et al. (2009)	*Streptomyces*	Baoune et al. (2019)
Advenella	Changmei et al. (2022)	*Acidovorax*	Liao et al. (2021)
Desulfovibrio	Qian et al. (2021)	*Gordonia*	Amran (2020)
Stenotrophomonas	Changmei et al. (2022)	*Thallassolituus*	Mahjoubi et al. (2018)

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