A Review on Polycyclic Aromatic Hydrocarbons: Their Transport, Fate and Biodegradation in the Environment

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) are an important group of organic pollutants. They are ubiquitous environmental pollutants generated primarily during the incomplete combustion of organic materials (e.g. coal, oil, petrol, and wood). Emissions from anthropogenic activities are highly responsible for their generation. The demand for processed petroleum products and agricultural produce has exposed our environment to PAH contamination. Globally, the combustion of fossil fuels and wildfires are major sources of PAHs, while road traffic and specific industries frequently dominate urban emissions. The majority of Earth’s hydrocarbon resource is only partially degraded. Bioremediation processes by microorganisms including bacteria, fungi and algae and also phytoremediation are the promising methods in cleaning up PAHs from the environment. These clean-up methods are not only environmental friendly but also present a novel approach in reducing the PAHs ability of causing risk to humans and to the ecosystem. Finally, summarise this review, as the sources of PAHs, their fate of transport, bioaccumulation, toxicity and the bacterial biodegradation aspects of both low molecular weight and high molecular weight PAHs.

Keywords

PAHs, Transport, Biodegradation, Bioaccumulation, Transport.

Introduction

Poly aromatic hydrocarbons (PAHs) are one class of toxic environmental pollutants comprised of fused aromatic rings and have accumulated in the environment due to a variety of anthropogenic activities (Mueller et al., 1991). PAHs are group of chemicals with more than 10,000 compounds that consist of two or more fused benzene rings in different arrangements (Blumer, 1976; Mahyar Sakari, 2012). They are present in high concentrations in coal tar, crude oil and are formed during pyrolysis of organic compounds in various natural and technogenic processes (Srogi, 2007; Baboshin and Golovleva, 2012). Combustion of organic material is the major source of PAHs. Anthropogenic sources of PAHs particularly include fuel combustion, pyrolytic processes and spillage of petroleum products, waste incinerators and domestic heaters (Freeman and Cattell, 1990). Creosote and anthracene oil which are commonly used pesticides for...
wood treatment also serve as a source of PAHs (Bumpus, 1989). Their conversion products are also toxic with mutagenic and carcinogenic properties to living organisms. Contamination with oil, coal tar, and the products of their refinement present the greatest problem to the environment (Mueller et al., 1989). Once exposed to the environment, most of the pollutant do not degrade quickly and may thus reside in the environment for prolonged period of time (Leahy and Colwell, 1990). The persistence of PAH in the environment is mainly due to their low water solubility (Freeman and Cattell, 1990). Their solubility decreases and hydrophobicity increases with an increase in the number of fused benzene rings (Chaillan et al., 2004). Sixteen PAHs in the list of priority environmental pollutants by the United States Environmental Protection Agency (USEPA) (Anatoly, 2008) include: [1] Naphthalene (C_{10}H_{8}), [2] Phenanthrene (C_{14}H_{10}), [3] Anthracene (C_{14}H_{10}), [4] Fluoranthenne (C_{16}H_{10}), [5] Pyrene (C_{16}H_{10}), [6] Chrysene (C_{18}H_{12}), [7] Benzo (a)anthracene (C_{18}H_{12}), [8] Benzo (b)fluoranthene (C_{20}H_{12}), [9] Benzo (k)fluoranthene (C_{20}H_{12}), [10] Benzo (e)pyrene (C_{20}H_{12}), [11] Benzo (a)pyrene (C_{20}H_{12}), [12] Perylene (C_{20}H_{12}), [13] Benzo (ghi)perylene (C_{22}H_{12}), [14] Dibenz(o)anthracene (C_{22}H_{14}), [15] Indeno (cd)pyrene (C_{22}H_{12}), [16] Coronene (C_{24}H_{12}).

Sources of PAHs

PAHs formation can either be natural or anthropogenic. The following three types: pyrogenic, petrogenic, and biological are the major PAH sources to the environment. Pyrolysis is a thermochemical decomposition of organic material at elevated temperatures (350°C to >1200°C) in the absence of oxygen (or any halogen). The destructive distillation of coal into coke and coal tar, or the thermal cracking of petroleum residuals into lighter hydrocarbons are Pyrolytic processes that occur voluntarily. The incomplete combustion of motor fuels in cars and trucks, wood in forest fires and fireplaces, and the fuel oils in heating systems are some of the involuntary pyrolytic processes that lead to the formation of PAHs. PAHs formed during pyrolysis are pyrogenic PAHs, generally found in greater concentrations in urban areas and in locations close to major sources of PAHs (Tolosa et al., 1996). PAHs formed during crude oil maturation and similar processes are termed petrogenic. These PAHs also found in petroleum products. Such petrogenic PAHs are common due to the widespread transportation, storage, and use of crude oil and crude oil products. Some of the major sources of petrogenic PAHs include marine and freshwater oil spills, underground and above ground storage tank leaks, and the accumulation of vast numbers of small releases of gasoline, motor oil, and related substances associated with transportation. (Tolosa et al., 1996; Hussein et al., 2016)

PAHs produced biologically are not well known. For instance, they can be synthesized by certain plants and bacteria or formed during the degradation of vegetative matter. Examples of natural sources of PAHs formation include: forest and brush fires, volcanoes, bacterial and algal synthesis, petroleum seeps, erosion of sedimentary rocks containing petroleum hydrocarbons, and decomposition of vegetative litter fall (Hussein et al., 2016). Emissions from coal combustion for power generation are predominant in emerging countries. Air pollution by PAHs in industrialized nations mainly comes from small combustion units in households. Sediments from urban waterways have an “urban background” signature from a variety of nonpoint sources of PAHs which may confound the interpretation of point sources of PAHs to these waterways.
Common point sources of PAHs include direct or indirect discharges from petroleum terminals, shipyards, aluminium smelting, manufactured gas production plants, tar distillation plants, rail yards, loading/unloading facilities, and spilled or seeped petroleum or coal or oil derived tars and their associated distillation products (Judy Crane, 2010).

Transport of PAHs in atmosphere

The behaviour of PAHs in the atmosphere depends on complex physico-chemical reactions, interactions with other pollutants, photochemical transformations, and dry and wet deposition. Depending on the atmospheric conditions such as ambient temperature, relative humidity, etc., and the nature i.e., origin and properties of the aerosol, and the properties of the individual PAH, in ambient air exist in vapour phase or adsorb into airborne particulate matter (Lima et al., 2005; Ki-Hyun Kim et al., 2013). They are unavoidable by-products of any kind of combustion, in particular incomplete combustion processes. Therefore, these substances are ubiquitous in the polluted atmospheric environment in the ng m$^{-3}$ concentration range. The gaseous state is predominant for the lighter relative molecular mass PAHs, while the substances with more than 4 rings are preferentially associated with the aerosol particles (Gerhard Lammel et al., 2015). The long range transport of organic pollutants in the environment remains a source of interest due to their capacity for long-range atmospheric transport, persistence, bioaccumulation, and toxicity (Kieron, 2008). However, the occurrence of PAHs in the atmospheric environment has been studied for decades, the photochemistry, deposition and, consequently, the long-range transport potential are not well understood. The reason is gas-particle partitioning (GPP) in the aerosol, its sensitivity to temperature and particulate phase composition, and reactivity’s sensitivities towards particulate phase composition. Furthermore, upon deposition to surfaces most PAHs subject to re-volatilisation (Gerhard Lammel et al., 2015). PAHs can enter the environment and reach consumers in multiple ways. Large quantities of PAHs are being released into the environment by leakages or accidents in the extraction, transport, or refinery of petroleum (Published by German Federal Environment Agency, 2012). PAHs enter the environment mostly from man-made and natural sources and thud release into air. Increases in vehicular traffic cause increases in the PAHs load in the form of exhaust emissions. PAHs enter surface waters through storm water runoff, discharges from industries and also through wastewater treatment plants (Jejal Reddy Bathi, 2008). The majority of compounds, and especially those with three or four rings, are semi-volatile and such compounds partition between the vapour and particle phases in the atmosphere. These compounds can deposit to surface water and soils where they have a long lifetime but subsequently re-evaporate to the atmosphere (Ian Keyte et al., 2013). Oil spills is one of the common route for entry of PAHs into the environment. Entire ecosystems breakdown if large quantities of oil are spilled into rivers or lakes, and such breakdown is due to the PAHs contained in such spillages. Accidental oil spills into the seas also destroy ecosystems in vast areas. In 2010, BP’s oil rig Deep water Horizon exploded in the Gulf of Mexico and caused an oil slick. An estimated 600,000 tons of oil were spilled into the sea (Crone and Tolstoy, 2010). This is equivalent to about 1,200 to 45,000 tons of highly toxic PAHs that will harm the Gulf of Mexico for decades (Published by German Federal Environment Agency, 2012). In the costal zones, sewage, runoff from roads, the smelter industry and oil spills are the major routes for PAHs to enter into water while offshore PAHs chiefly enter the water through oil seeps, oil spills, and
produced water discharge from offshore oil installations. In aquatic environments, PAHs generally accumulate in sediments due to their strong hydrophobicity and their resistance to biodegradation (Zaisheng Yan et al., 2015). Depending on how strong the bond is, organic compounds adsorbed on soil particles may migrate down to deeper soil layers and then to ground or surface waters, or they may remain in the surface layer of the soil and, in some cases, end up in plants. These compounds also bioaccumulate in fish and other aquatic organisms (Mira and Petar, 2004).

In coal mining, PAHs mainly enter the environment through dusts. The material in stock piles and tailings also contains PAHs and pollute soils, waters and the groundwater. PAHs can also enter the environment through exhaust gases and waste water through coal processing in coking plants. The soil and groundwater on the premises of former gas works and coking plants are often highly contaminated with PAHs, especially around tar pits and the areas where coal and waste were stored (Published by German Federal Environment Agency, 2012). PAHs enter the ambient air through dusts to which they are bound and also through abrasion from rubber products such as car tyres. Not only the exhaust gases but also abrasion from rubber products can be relevant PAH sources (Zaisheng Yan et al., 2015). Agricultural soil is polluted by PAHs via particles from the air. Also, the low-molecular-weight fraction of these compounds is more often found in agricultural soils than in urban ones because of greater evaporability and easier transport by air (Mira and Petar, 2004).

**PAH toxicity**

PAHs and their derivatives are ubiquitous in the environment such as air, water, soil, sediments and living organisms (Blumer 1976). As ubiquitous pollutants, PAHs are of serious concern because some of them have been identified as carcinogenic and mutagenic compounds e.g. the high molecular weight ones (Law et al., 1997; Wanga and Tam, 2011). Some of the PAHs have been determined to be carcinogenic by several regulatory agencies like (US Environmental Protection Agency (EPA), US Department of Health and Human Services (DHHS) and the International Agency for Research on Cancer (IARC) (Jejal Reddy Bathi, 2008). According to the report given by Nesta Bortey-Sam et al., carcinogenicity of PAHs is higher and Bap contributing 70% of the total PAHs toxicity in the city centres of Ghana and Kumasi (Nesta Bortey-Sam et al., 2014).

In animals, there is adequate evidence for carcinogenicity caused by benzo \([k]\) fluoranthene, and in humans it is classified as a group 2B carcinogen by the International Agency for Research on Cancer (Allyn Maeda et al., 2014). Benzo (a) pyrene (BaP) is the potent and most studied carcinogenic PAH. In cultured mammalian cells it can induce DNA binding, sister chromatid exchange, chromosomal aberrations, point mutation and transformations (Hollstein et al., 1979; De Serres and Ashby, 1981). However, there are no epidemiological studies regarding the carcinogenity of BaP in humans (Albert and Ravendra, 2000). But, the acute and chronic effects of PAHs on human health have been reported. Short-term exposure to PAHs also has been shown to cause impaired lung function in asthmatics and thrombotic effects in people affected by coronary heart disease (Ki-Hyun Kim et al., 2013; ACGIH, 2005). In contrast, occupational exposures to high levels of pollutant mixtures containing PAHs are known to result in symptoms such as eye irritation, nausea, vomiting, diarrhea, etc. Ki-Hyun Kim and few others mentioned that mixtures of PAHs are also known to cause skin irritation and inflammation (Unwin
et al., 2006). Long-term exposure to low levels of some PAHs has been identified as the cause of cancer in laboratory animals (Ki-Hyun et al., 2013; Diggs et al., 2012).

**Bioaccumulation of PAH**

PAHs have a high potential for biomagnification through trophic transfers due to their lipophilic nature (Clements, 1997; Lu et al., 1999; Twiss et al., 2000; Robert and Shigeaki, 2006). PAHs have low solubility in water, but they stick to solid particles of clay, and they readily make bonds in fat tissues of organisms (Stankovic et al., 2011). They tend to attach to particles in the air, water, and sediment and also accumulate in the lipids (i.e., fat) of benthic organisms that are unable to metabolize them. Although fish are able to metabolize PAHs, lessening their accumulation in tissues, PAHs can cause other detrimental effects in fish such as mouth tumours (Judy, 2010). The total PAH concentrations detected per dry weight in a number of marine organisms is as follows, fish 14-315 ng/g (Albert and Ravendra, 2000; Humason and Gadbois, 1982), molluscs 54-2803 ng/g (Dunn and Stich, 1976; Knutzen and Sortland, 1982), crustaceans 52-1600 ng/g (Humason and Gadbois, 1982), algae 284-4665 ng/g (Knutzen and Sortland, 1982).

Little is known about the action of PAHs on higher plants which are often used as bioindicators. Robert Aina et al., exposed Clover plants to soil artificially contaminated with three concentrations of BaP (5, 10 and 20 lg g⁻¹) or Naphthalene (25, 50 and 100 lg g⁻¹), and studied the changes in the DNA content and sequence of roots and shoots. Results showed that BaP and Naphthalene were both genotoxic for white clover, inducing significant changes in root and shoot DNA sequence (Robertta et al., 2005). A greenhouse pot experiment conducted by Sardar Khana et al., to assess the concentrations of PAH using Lactuca sativa L. as a representative vegetable grown in wastewater-contaminated soils showed the concentrations of total PAHs ranged from 1.5 to 3.4 mg kg⁻¹ in the contaminated soils, while 1.2 mg kg⁻¹ in the reference soil (Sardar Khana et al., 2008). Investigation on PAH contamination by Azza Zohair et al., in organically farmed crop varieties showed the accumulation of PAHs ranged from 8.42 ± 0.93 to 40.1 ± 4.9 µg/kg. Desiree potato and Nairobi carrot varieties were more susceptible to PAH contamination (Azza Zohair et al., 2006). Spartina alterniflora plants grown in pots of PAH contaminated sediment, and in native sediment at a marsh contaminated with up to 900 µg/g total PAHs, were measured for PAH contamination. The roots and leaves of the plants were separated, cleaned, and analysed for PAHs. PAH compounds were detected at up to 43 µg/g dry weight in the root tissue and at up to 0.2 µg/g in the leaves of plants grown in pots of contaminated soil, concentrations less than 0.004 µg/g were detected in the leaves of plants grown in native site (Alison et al., 2006). Determination of HMW PAHs by Gas chromatographic-mass spectroscopic analysis of the samples of roots and soils from rooting zone of wild blackberry from the locations near “The Copper Mining and Smelting Complex Bor” (Serbia) resulted in the highest accumulation of Benzo(a)pyrene (Slađana Alagić Vesna et al., 2014).

**Biodegradation of PAH**

Biodegradation is the promising technology for the treatment of contaminated sites, which may refer to complete mineralization of organic contaminants into carbon dioxide, water, inorganic compounds, and cell protein or transformation of complex organic contaminants to other simpler organic compounds by biological agents like microorganisms.
Table 1 Recent information on biodegradation of different low and high molecular weight PAHs by different bacterial strains (incomplete)

| Organism | Sample source | Growth substrate | Initial substrate concentration | Incubation time | % removal | Reference |
|----------|---------------|------------------|---------------------------------|-----------------|-----------|-----------|
| Bacterial consortium 1. *Ochrobactrum sp.*, 2. *Enterobacter cloacae* 3. *Stenotrophomonas maltophilia* | Marine water sample | 1. Phenanthrene | 5,10,20 ppm 100 ppm | >95%  79% | Arulazhagan and Vasudevan (2009) |
|          |               | 2. Fluorene      | 50 ppm 100 ppm                | 89% 78%         |           |           |
|          |               | 3. Pyrene        | 50 ppm 100 ppm                | 74% 52%         |           |           |
|          |               | 4. Benzo(e)pyrene| 2 ppm 5 ppm                   | 51% 21%         |           |           |
| *Rhizobium tropici* 899 | Pure strain | 1. Benzo(a)pyrene | 40 µg mL\(^{-1}\) 60 µg mL\(^{-1}\) | 24h 120h | 25% 48% | Gonzalez-Parades et al., (2013) |
| Bacterial consortium | Oil port seawater | 1. Pyrene | 100 mg/L | 6 days | 100% 50% | Palanisami Thavamani, Mallavarapu Megharaj (2012) |
|          |               | 2. Benzo(a)pyrene in the presence of Cd | 10 mg/L | 60 days |           |           |
| *Ochrobactrum sp.* BAP5 | Marine sediment | 1. Benzo(a)pyrene | 50 mg/L | 30 days | 20%      | Yirui, HE Tengteng et al., (2009) |
| **Microbial consortia** | **Sample type** | **Compounds** | **Initial Concentration** | **Biodegradation Time** | **Degradation Efficiency** | **References** |
|-------------------------|-----------------|---------------|---------------------------|-------------------------|---------------------------|---------------|
| *Ochrobactrum sp. VA1*  | Marine water sample | 1. Anthracene, 2. Phenanthrene, 3. Naphthalene, 4. Fluorene, 5. Pyrene, 6. Benzo(k)fluoranthe ne, 7. Benzo(e)pyrene | 25 ppm 100 ppm 3 ppm 3 ppm 3 ppm 25 ppm 100 ppm 1 ppm 1 ppm | 4-5 days 4-5 days 4-5 days 4 days 4 days 3 days 2 days | 82% 56% 90% 97% 71% 32% 57% 50% | Arulazhagan, N. Vasudevan (2011). |
| Microbial consortium    | Oil port seawater | 1. benzo (a) pyrene | 10 ppm | 14 days | 44.7% | Luo et al., (2009) |
| *Mycobacterium strain A1-PYR,* *Sphingomonas sp. PheB4* | | Phenanthrene Fluoranthene Pyrene | 10mg/L 10mg/L 10mg/L | 3 days 7 days 7 days | 100% 72.1% 50% | Yin Zhong et al., (2011) |
| *Stenotrophomonas maltophilia strainVUN10,003* | Deep sea sediment | Pyrene Fluoranthene Benz[a]anthracene, Benzo[a]pyrene, Dibenz[a,h]anthracene Coronene | 243mg/L 45mg/L 26mg/L 11mg/L 11mg/L 11mg/L | 5-42 days | 98% 45% 26% 22% 22% 55% | Juhasz et al., (2000) |
| *Bacterial consortium* | Deep sea sediment | Spent engine oil | 1%(v/v) | 10 days | 52% | A. Ganesh Kumar et al., (2014) |
| *Bacterial consortium* | Petroleum contaminated soil | Phenanthrene | 100-1000mg/kg | 20 weeks | >85% | Arbabi et al., (2009) |
Many indigenous microorganisms in water and soil are capable of degrading hydrocarbon contaminants (Humason and Gadbois, 1982). Bacterial genera, namely, Gordonia, Brevibacterium, Aeromicrobium, Dietzia, Burkholderia, and Mycobacterium isolated from petroleum contaminated soil proved to be the potential organisms for hydrocarbon degradation. Bacteria isolated from brackish water are capable of hydrocarbon degradation and these include members of the genera Achromobacter, Acinetobacter, Alcaligenes, Arthrobacter, Bacillus, Brevibacterium, Corynebacterium, Enterobacter, Escherichia, Flavobacterium, Norcadia, Pseudomonas, Staphylococcus, and Vibrio (Foght and Westlake, 1987; Atlas and Bartha, 1992; Dunn and Stich, 1976; Nilanjana and Preethy, 2011). Bacillus subtilis, Pseudomonas aeruginosa and Torulopsis bombicola capable of improving bioremediation by solubilising PAHs into the aqueous medium and enhance their bioavailability for degradation (Cottin and Merlin, 2007). Bacterial genera namely Brevibacterium, Aeromicrobium, Gordonia, Dietzia, Burkholderia, and mycobacterium isolated from petroleum contaminated soils were proved to be potential for hydrocarbon degradation (Chaillan et al., 2004). Arthrobacter sulphurous, Acidovorax delafieldii (Samanta et al., 1999), Brevibacterium sp. (Samanta et al., 1999), Burkholderia sp. (Seo et al., 2006), Burkholderia cocovenenans (Wong et al., 2002), Janibacter sp. (Yamazoe et al., 2004), Mycobacterium sp. (Heitkamp et al., 1998; Schneider et al., 1996; Miller et al., 2004; Vila et al., 2001; Cheung and Kinkle, 2001), Pseudomonas paucumobilis (Weissenfels, 1990), (Weissenfels et al., 1990) Pseudomonas fluorescens (Balashova et al., 1999), Pseudomonas aeruginosa (Romero et al., 1998), Staphylococcus sp. (Romero et al., 1998; Kazunga and Aitken, 2000), Sphingomonas sp. (Rentz et al., 2008) are capable of degrading various PAHs.

In conclusion, PAHs are of considerable concern due to their well-recognised toxicity and especially due to the carcinogenic hazard which they present and today biosphere suffers increasing anthropogenic influence. In one way or the other PAHs are the major environmental pollutants and require a greater removal from the environment. Microbial degradation is the major and ultimate natural mechanism by which petroleum hydrocarbons can be removed from the environment, bioaccumulation of PAHs in plants indicate that these plants can be applied in phytoremediation process. Hence both microbes and plants can be used as potential tools for removing PAHs from the environment. Therefore it can be concluded that increasing attention has to be paid to the environmental behaviour of PAHs on a global scale due to their long-range transport potentials. Due to the health risk posed by their exposure there is a need for their control through air quality management. Refinery industries are required to monitor and regulate their discharges. There is a greater need for the considerable efforts to be applied in the field of research to degrade and monitor potentially hazardous substances to control, predict and to avoid negative effects of PAHs pollution. The ultimate goal of any bioremediation process is still to be achieved better.

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