



## REVIEW

### A Review on Deposition, Distribution of Polycyclic Aromatic Hydrocarbons in Different Environmental Matrix and Study its Toxicity and Carcinogenic Effect

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The main objectives of review work are to establish an association of polycyclic aromatic hydrocarbons (PAHs) signatures in bulk deposition in soil, street dust, river water and sediment. Polycyclic aromatic hydrocarbons are a unique class of organic pollutants containing 2 or more fused aromatic rings, which are very lethal and potent carcinogens. They are extensively studied compounds and their occurrence has been reported from various places over the world which indicates their ubiquitous nature. USEPA has already listed 16 PAHs as most priority ones to be analyzed in various environmental matrices. From the literature survey, it might be concluded that meteorological parameters played a significant role in atmospheric deposition of PAHs with temperature dependent scavenging for low molecular weight polycyclic aromatic hydrocarbons. The exposure risk for PAHs in all the environmental matrices was found to be the maximum during dry season. Finally, atmospheric bulk deposition was revealed as a significant contributor to other environmental matrices of the environment.

**Keywords:** PAH, Environmental matrix, Soil, Street dust, River, Sediment, Organic pollutant, Carcinogens, USEPA.

## INTRODUCTION

In the group of the non-halogenated organic compounds polycyclic aromatic hydrocarbons (PAHs) are among the most notorious semi volatile organic pollutants and are considered as hazardous air pollutants (HAP) along with benzene, phenols, aldehydes, *etc.* [1]. They are the class of compounds with two or more fused benzenoid rings made up of only carbon and hydrogen [2]. Based on the molecular structure PAHs are commonly classified into two categories, namely low molecular weight (LMW) PAHs with four or fewer aromatic rings and PAHs with five or more rings as high molecular weight (HMW). The United States Environmental Protection Agency (USEPA) has notified 16PAHs as the most priority ones which has been studied, monitored and analyzed in different environmental matrices [3].

Polycyclic aromatic hydrocarbons are formed in incomplete combustion processes of any organic matter including fossil fuels and vegetation [4]. Polycyclic aromatic hydrocarbons formation gets enhanced in thermochemical processes operating under rich fuel conditions [5]. The hydrogen abstraction  $C_2H_2$  addition (HACA) mechanism is now popularly considered as the major reaction route leading to the formation of PAHs in combustion flames [6,7]. In the combustion environment

the simple gaseous PAH molecules, such as naphthalene, phenanthrene, pyrene, *etc.*, can undergo a sequential build-up process and eventually lead to the formation of larger PAHs, fullerenes, bowl-shaped nanostructures and solid-phase species including carbonaceous dust, graphene particles and soot [6].

Depending on the volatility and molecular weight PAHs can adsorb on soot surface and can remain in the gas phase as well [5]. Such particles then appear in different environmental matrices such as ambient air, soil, street dust, water, sediment, *etc.* and can be inhaled by human being or consumed with food, leading to major health problems, such as tumors, birth defects and a variety of pulmonary diseases [8].

**Toxicity and carcinogenicity of PAHs:** Environmental PAHs have harmful effects on different types of organisms of ecosystem [9]. Polycyclic aromatic hydrocarbons attracts considerable attention among researchers with the continuous rise in death toll caused by cancer, which account for about 13 % of all human deaths worldwide in 2007 [10]. Carcinogenic and mutagenic properties of few PAHs members have already been well established by researchers [11,12] in experimental animals and humans. It is revealed from studies in experimental animals that PAHs may trigger various health effects, such as toxicity in cell, immunotoxicity, genetic toxicity, carcinogenicity, reproductive toxicity, *etc.* [1]. Organizations including

United states agency for toxicity for toxic substance & disease registry (ATSDR), National occupation safety & health (OSHA), USEPA, International agency for research on cancer (IARC) and Department of health & human services (DHHS) classifies PAHs as carcinogenic materials. As a reference of PAH toxicity the benzo(a)pyrene is more toxic and called as 'gold standard' due to its carcinogenic nature [13]. Carcinogenicity of benzo(a)pyrene through inhalation was tested only in hamsters [14,15]. Risk analysis inferred that about 9 per 100000 individual spending a lifetime in ambient air exposed to the average level of benzo(a)pyrene of 1 ng/m<sup>3</sup> may die from respiratory tract cancer [15,16]. Low molecular weight polycyclic aromatic hydrocarbons (LMWPAHs) are generally associated with relatively lower toxicity than high molecular weight polycyclic aromatic hydrocarbons (HMWPAHs). Car exhausts (petrol, diesel), domestic coal-stove emissions and tobacco smoke releasing mainly 4 to 7 ring PAHs are found to exhibit nearly all the carcinogenic potential [17]. Elevated levels of DNA adduct, mutations, reproductive effects, cancers of respiratory channel and urinary bladder are found to be associated with exposure to PAHs [18,19]. Pankow *et al.* [20] observed that the cancer causing risk of inhaled PAHs depends upon whether it enters into lung in the gas form or particulate; in case of particulate PAHs carcinogenicity prevails for a longer period of time.

**Distribution in the environment (air, soil, street dust, water and sediment):** Aerial movement is one of the major pathways for environmental distribution and trans boundary deposition of PAHs [21]. Remote areas far from emission sources also get exposed to PAHs as they undergo long range transport [14,22]. Eventually, PAHs settle down in soils, street dust and enter into the aquatic environment [23,24]. Soil and street dust acts as a direct sink of atmospheric PAHs near to traffic and other combustion sources. From these environmental compartments rainwater and storm water easily washed away PAHs to nearby aquatic bodies. Due to hydrophobic nature, PAHs in aquatic environments are preferably partitioned and accumulate into the particulate phase of sediment [25]. Polycyclic aromatic hydrocarbons thus, occur in a multicompartamental system in the environment [26,27] and paved the way for multiple routes of exposure to this class of carcinogen.

**Atmospheric deposition of PAHs:** Among earlier studies, Lunde *et al.* [28] performed studies on organic micro pollutants in precipitation in Norway. Snow and rain samples were collected and analyzed for four chemical groups, namely alkanes, esters of phthalic acid, PAH and ethyl ester fatty acid. Both particles adsorbed and water dissolved phase organic components were fractionated for analysis. They reported presence of alkanes and PAHs in precipitation samples and considered fossil fuel to contribute these organic micro pollutants in precipitation.

Van Noort *et al.* [29] determined the temporal variation of PAHs concentration in rain during three precipitation events. The rain samples were collected simultaneously at ground level and at an altitude of 200 m. The concentration of phenanthroline and fluoranthrene was remarkably higher in former than that of later as reported in the study. The presence of phenanthroline was considered to be the result of below-cloud gas-phase scavenging. The range of study PAHs was BDL to 110 ng/L.

To estimate PAHs fluxes [30] within the Lake Michigan basin to coastal waters adjacent to the Chicago urban area and found that dry deposition loading for PAHs were three times higher than that of wet deposition.

Polkowska *et al.* [31] estimated PAHs in atmospheric precipitation, including rain and snow in Poland. Phenanthroline, fluoranthrene and pyrene were the dominant PAHs in precipitation samples. Seasonal attributes were recognized in both rain and snow with maximum PAHs concentrations during winter, which was attributed by them to be primarily from residential heating. Another significant PAHs source was identified as traffic emission by them.

Bulk deposition and gas exchange of PAHs were estimated in the Galveston Bay, Texas by Park *et al.* [32] and reported atmospheric PAHs in the vapour phase and in the rain dissolved phase. They had found phenanthroline and naphthalene to be the dominant individual PAHs in air vapour and rain dissolved phase. Particulate phase of both air and rain samples were found to be dominated by 5 and 6 ring PAHs. Moreover, when the comparison has been made with wet deposition, air to surface water gas exchange was recognized as a principal deposition process for PAHs. Burning and petroleum deposition were identified as major sources of PAHs in the studied area. Levels of PAHs in bulk (wet and dry) precipitation were conducted by Manoli *et al.* [33] in the main plain of central Macedonia in Northern Greece. They observed maximum PAHs concentration during cold months. For deposition flux, the greatest values were revealed during large precipitation events coinciding with high PAHs concentrations. In an urban region of Poland, Gryniewicz *et al.* [34] estimated 16 PAHs in bulk precipitation (dissolved in water and adsorbed on solid particles) in which compounds were recovered using solid phase extraction method and analyzed by GC-MS. In this study naphthalene, phenanthroline + anthracene, fluoranthrene and pyrene were found to be dominant compounds with high concentration values. On the other hand, trace levels of acenaphthylene, acenaphthene, fluorene, benzo(b)fluoranthene + benzo(k)fluoranthene, iden(123-cd)pyrene, benzo(a)-anthracene and benzo(ghi)perylene were also reported. High PAHs concentration in winter samples and the major contribution of coal fed heating to average PAHs load was observed. Vehicular traffic was identified to be a major source of total PAHs in precipitation.

A similar method of PAHs estimation was adopted by Golomb *et al.* [35] while determining wet and dry deposition of PAHs near New England coastal water. High wet and dry deposition was observed during heating season. It was also reported that precipitation during non-convective events brought more wet deposition of PAHs than convective events. They identified diesel and gasoline fuelled vehicle and wood burning as major sources of PAHs.

Seasonal variation of PAHs in atmospheric precipitation in a rural site of Lake Balaton, Hungary was examined [36] and higher level of 3-6 ring PAHs was observed during the winter (3-350 ng/L) than during the summer (1-54 ng/L) with a higher wet deposition rate of 412 µg m<sup>2</sup>/year. The most abundant compounds in snow samples reported by the group were phenanthroline, fluoranthrene and pyrene.

Atmospheric bulk deposition PAHs were also studied by Ollivon *et al.* [37] during 1999-2000 in Paris, France. High abundance of fluoranthrene and pyrene was observed in bulk deposition samples by them. Vivid seasonal variation was observed with the highest concentration during winter. The temperature was found to play an important role in scavenging of LMWPAHs. The heating contributed 41 % of the overall annual PAH load. In another study, Montelay-Massei *et al.* [38] estimated PAHs in bulk deposition during 2001-2002 on a weekly basis at Evereux, West of Paris (France). Similar trends were reported and dominant phenanthroline, fluoranthrene and pyrene were the species contributing 69 % of total PAHs. Bulk deposition of PAHs at remote, rural village and an urban area of Beijing-Tianjin, North China were studied during 2007-2008 [39]. The high deposition flux of phenanthroline (35.3 %) and fluoranthrene (25.7 %) during the fall and spatially, urban areas (8.28  $\mu\text{g}/\text{m}^2/\text{day}$ ) received high deposition flux than that of rural ones (3.91  $\mu\text{g}/\text{m}^2/\text{day}$ ).

Polycyclic aromatic hydrocarbons flux was monitored in Seine estuary, France to identify the major pathways of transport and accumulation. Transport of PAHs from the atmosphere to watershed outlet was monitored for the two rural and urban sampling locations of watersheds within the Seine Estuary. High loading of atmospheric input of PAHs was observed in rural areas relative to urban areas [14].

Yan *et al.* [40] conducted a large-scale survey on the distribution and deposition flux of PAHs in rainwater samples in Shanghai, China and reported annual deposition flux of PAHs was found to be 4148 kg/yr. Naphthalene, phenanthroline, anthracene and fluoranthrene were revealed as the most dominant PAHs in rainwater.

**Polycyclic aromatic hydrocarbons in soil:** Aamot *et al.* [41] investigated levels of nine selected PAHs in surface soils from southern and central Norway. Concentration in southern Norway was observed to be higher than that of central Norway in general; which was approximately ten times higher in the case of 4 and 5-ring PAHs. Atmospheric transport was found as a major contributor source of PAHs in soil after comparing with air quality data in south Norway. Naphthalene concentration in the soil was found to be very high compared to many other studies. A similar study was carried out in seven locations across the Seine river basin, France was estimated by Montelay-Massei *et al.* [38]. They observed distinct spatial variation with concentration industrial and urban areas to be in order of magnitude higher than those in the remote areas. High percentage contributions of carcinogenic PAHs (approximately 40 %) were reported. Spatial distribution and sources of PAHs in soil of Valasske Mezirici were investigated by Plachá *et al.* [42]. Polycyclic aromatic hydrocarbons concentration in urban, agriculture and forest sampled soil was investigated and observed that the maximum density was found to be at high altitude of forest soil. The maximum PAH level was found to be higher than that of the forest soil in Western and Northern Europe. Meteorological condition with frequent calm condition associated with inversion state was considered as a major contributing factor for such high PAH concentration at the high altitude forest. The most significant source of PAHs in soil was identified due to Industrial processes transport and local heating activities.

Wang *et al.* [39] monitored and reported air-soil gas exchange flux in Beijing-Tianjin region, North China with annual median PAHs flux of 42.2  $\text{ng}/\text{m}^2/\text{day}$  from soil to air. Acenaphthylene and acenaphthene were observed as major contributor in total exchange flux. The air-soil gas exchange fluxes of PAHs were higher at the urban sites than those at the remote and rural sites. Seasonal variation also found to affect the mechanism with more gaseous PAHs volatilized from soil to air in summer with rise of temperature and increased rainfall. However, more gaseous PAHs were recognized to be deposited from the atmosphere to soil during winter because of higher PAH emissions and lower temperature.

**Polycyclic aromatic hydrocarbons in street dust:** Takada *et al.* [43] reported molecular distributions of PAHs in street dust samples in Tokyo. The level of total PAHs concentration was found to be a few  $\mu\text{g}/\text{g}$  of dust. Individually, 3 and 4 ring PAHs such as phenanthroline, fluoranthrene and pyrene were recognized as the dominant ones in the street dust. Pengchai *et al.* [44] also studied PAHs profile in urban road dust in Tokyo. Seven categories of PAHs sources were recognized, including diesel and gasoline vehicle exhaust, vehicle exhaust, tire, asphalt or bitumen, asphalt pavement, petroleum products and the burning emitted products except for those in vehicle engines.

Netto *et al.* [45] monitored and reported the seasonal variation in street dust in the center of Niteroi, having tropical climate in Brazil, at the boundaries of Guanabara Bay. The maximum PAH concentrations were found in the month of July when the temperature was minimum. Individually, the maximum geometric mean was detected in the case of pyrene, fluoranthrene and naphthalene. Carcinogenic PAHs contribution was found to vary from 29 to 45 %. Polycyclic aromatic hydrocarbons showed significant correlations with daily mean and minimum temperature, as well as the overall temperature during the sampling period. This indicates that meteorological parameter mainly temperature plays an important role in the PAHs distribution in street dust.

Liu *et al.* [46] performed a study of PAHs in road dust samples collected from central Shanghai during winter and summer. Seasonal variation was reported with higher level of total PAHs during winter than that of summer. High molecular weight polycyclic aromatic hydrocarbons were found to be dominated in all areas. Mix source of traffic and coal combustion was identified for road dust PAHs. Pyrogenic and petrogenic source contribution was 71.0 and 11.4 %, respectively, during winter, while 64.9 and 14.1 %, respectively during the summer.

Samimi *et al.* [47] investigated PAHs contamination level in the form of settling street dust, in Tehran, Iran. Very high PAHs contamination was observed in the vicinity of the highway. Naphthalene and phen were found as most common individual PAH in all the samples. The presence of these compounds was an indication of emission from incomplete combustion of vehicular emission sources and consequently adsorption to the atmosphere.

**Polycyclic aromatic hydrocarbons in river water and sediment:** Smith *et al.* [48] measured PAHs in waters from three rivers which flow through the largest cities in southeastern

Australia. Smaller ring compounds were found to be more abundant with a higher concentration than the larger ring ones in this study.

Skrbic *et al.* [49] estimated PAHs bank sediment of Danube covering the entire city and found that total PAHs content in bank sediments were higher. The overall percentage contribution of benzo(a)pyrene was found to be 6-14 % of the total PAHs. The total carcinogenic potency for most of the sites was observed to be on the higher side than recommended levels.

Spatial distribution of PAHs in surface water, solid particulate matter and Daliao river watershed composed of the Hun, Daliao and Taizi river sediment were investigated by Guo *et al.* [50]. Composition of PAHs was found to be dominated by 4-6 ring PAHs. Low molecular weight polycyclic aromatic hydrocarbon concentration were very high in water. In industrial area high pollution load in water and sediment indicated input from industrial wastewater.

Zhao *et al.* [51] has assessed PAHs river sediments (Yangtze river Delta) in Xincheng, China. The total loads of PAHs in upstream contaminated water current were influenced by factors including particle size, land use and the hydrological conditions in the stream network as recommended by many researchers.

Major inputs and sources of PAHs into surface water, stream and precipitation along the Sacramento river, California was investigated by Kim and Young [52]. Concentration of total PAHs in the precipitation was found to be lower than surface water, but according to the observation the dissolved PAHs was having almost similar content in both the matrix. Individually naphthalene, phenanthroline, pyrene and benzo-(ghi)perylene were identified as most dominant compounds in both surface water and rain. In dissolved phase more than 4 ring PAH compounds were rarely occurred. During the wet season significant input of PAHs in surface water was observed through indirect deposition (*i.e.*, wash-down of atmospheric aerosol particles previously deposited in the soil) in California's Mediterranean climate. During winter time although there was a remarkable increase in aqueous PAHs level no apparent seasonal change was noticed in dissolved PAHs concentrations. Marker PAH ratios suggested pyrogenic origin of rainwater PAHs whereas complex and variable origin of PAHs was recognized in the surface water study.

Monitoring and investigated of PAHs water sediment sampled from the bank of Aojiang river and its estuary is done by Li *et al.* [53]. The water and sediment concentration of PAHs were found to be relatively high while considering the population size of the area. Petrogenic sources were identified in sediment using diagnostic ratio. The composition profile of PAHs in sediment was found to of dominance of 4-6 ring PAHs.

Chemical analysis and bio toxicity was evaluated by Fu *et al.* [54] to find its occurrence, origin, environmental and biological risk and ecotoxicity in sediments of Huaihe river. Diagnostic ratio revealed mainly pyrogenic origin of PAHs in the river sediment. Some of the individual PAHs were found to pose occasional biological risk, however, no sample was identified for frequent biological impairment. However, some of the sites were observed with high carcinogenic potential value.

Levels, dispersion patterns, seasonal variation and sources of PAHs in Hun river of Liaoning Province, China were conducted by Zhang *et al.* [55]. The total PAHs concentrations were prominently elevated periodically in the order of level period > flood period > dry period. Dominance of LMWPAHs was noticed particularly for 2-3-ring PAHs. Predominance of LMWPAHs with high concentration during flood period was considered as a relatively recent local source of PAHs. Selected PAHs ratio was used for source identification which indicated petrogenic origin of PAHs during dry season, while the mixed pyrogenic and petrogenic source during the wet season. After comparing the contamination level of PAH among different types of area it was prescribed that atmospheric deposition was the most important mechanism of PAHs entry into the water system.

### Conclusion

The PAHs in different environmental matrices were studied by various researchers in the past. However, literature survey for the interrelationship of PAHs among all the environmental compartments is rather limited. Atmosphere being a principal pathway for dispersion of PAHs in the environment, its contribution to other environmental matrices need to be estimated. Composition of PAHs was found to be dominated by 4-6 ring PAHs in the entire three mediums. The high occurrence of LMWPAHs in water and solid particulate matter was recommended for recent local source and atmospheric deposition. High pollution load in water and sediment near the industrial area were suggested by industrial wastewater. It was also observed that PAHs levels are relatively higher in water and solid particulate matter and lower in case of sediments. In our literature survey, an attempt has been made to study the contribution of bulk deposition PAHs into various other environmental compartments including soil, street dust, river water and sediment.

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