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Review on Polycyclic Aromatic Hydrocarbons Load in Indian Urban Environment

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Abstract

The present review study manifests the sampling techniques, analytical tools and the status of Polycyclic Aromatic Hydrocarbons (PAHs) in different environmental samples of Indian urban cities. Indeed, petrogenic, pyrogenic and biogenic sources of PAHs grounds to carcinogenic, mutagenic, teratogenic and genotoxic effects on human environment. Most of the studies on PAHs focussed on the ambient air samples and noticed that carcinogenic Benzo[a] Pyrene is dominant and severe health risk posturing 4-5 ring high molecular weight PAHs. The High-volume sampler for minimum 8hrs espoused as best sampling method over passive bio samplers, glass fibre filters, particulate samplers etc. The microwave assisted Soxhlet extraction with n-hexane solvent has gained a lot of attention due to its pragmatism in sample handling than Soxhlet and ultrasonic. The high-performance liquid chromatography and gas chromatography with a mass spectrophotometer adopted as best analytical tools for high accuracy. The PAHs concentration of among 11 Indian cities of the review study has significantly emphasized the inter and intra spatial variations. Moreover, the amount of concentration of PAHs differs in air, water and soil. Also, the comparative study revealed that Delhi and Amravati recorded maximum and minimum concentrations of PAHs respectively.

Keywords: *Anthropogenic, Carcinogenic, Liquid and Gas Chromatography, Polycyclic Aromatic Hydrocarbon, Pyrogenic, Soxhlet*

1.0 Introduction

In this era of incessant pollution of environment, assessing the amount of pollutants in an area has become the need of the hour. With the growth of urbanization changes in industrial structures[1], lifestyle of individuals has contributed to the pollution of the environment which has impacted the biological and the physiological conditions of the living beings. India is a developing country with booming industrialisation, transportation, improved technology. Due to population increase and the increase in the demands of the population the developmental activities are rising day by day[2]. This is contributing to pollution of air, water and land. The main pollutants are found to be the polycyclic aromatic hydrocarbons present in ambient air, water sediment and road soil[3]. Due to this the study of polycyclic aromatic hydrocarbons has become the need of the hour. The following review paper aims to study and characterise the quantitative and qualitative analysis of PAHs in few Indian cities. The present review paper gives an overview of PAH compounds contamination in various Indian cities and also brings about the comparison of pollution in the given cities. It is found that most of the PAHs is released in the environment when the fossil fuels do not burn completely, anthropogenic sources[4], natural sources, petrogenic, pyrogenic and biogenic sources. The adverse effect of these PAHs in human life and the ecosystem are of a great concern as they are carcinogenic, mutagenic, teratogenic, neurological, immunological, genotoxic effects etc[5]. The concentration of Benzo[a]Pyrene is found to be dominant in most of the places. Most of the

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cities have focussed on the testing of PAHs in ambient air. The sampling of the air is done by the Passive air sampling (PAS) PUF. The sampling methods included passive bio samplers, high volume samplers, glass fibre filters, particulate samplers etc. The analysis of the samples was mainly done by advanced high performance liquid chromatography (HPLC) with ultraviolet (UV) or fluorescence detection (FLD) and gas chromatography (GC) with a mass spectrophotometer. In some cases, the Soxhlet and Sonication was also implemented. The latest method used for analysing is a gas chromatograph- flame ionisation detector (GC-FID). The main sources of the PAHs were found to be combustion of organic matter and vehicular emissions. The current review may be the baseline for future research in other cities of India.

The aromatic hydrocarbons with two or more fused benzene rings in various structural configurations are known as the Polycyclic aromatic hydrocarbons (PAHs). They do not contain any other atoms or carry substituent. PAHs having up to four rings are referred to as light PAHs and the stability of these is found to be more and are supposed to be more toxic than the light PAHs. PAHs containing more than four rings are heavy PAHs. Heavy PAHs are more stable and toxic than the light PAHs. The PAHs contain only carbon and hydrogen atoms bound to simple and complex ring systems. PAHs are let out in the environment by means of both natural and anthropogenic sources. They are mainly produced as a result of various combustion processes[6]. Natural sources include volcanoes, forest fires, dust storms. Anthropogenic sources include burning of fossil fuels, tobacco smoking vehicular exhaust, and industrial emissions. Other sources include coal production, oil spills, oil manufacturing, wood preservation (creosote) and various forms of cooking. Collectively the anthropogenic sources can be categorised in to four categories namely domestic, industrial, agriculture and transport. The natural sources include volcanoes, forest fires, dust storms etc. The petrogenic sources include light molecular weight PAH from petroleum and by products of petroleum. On the whole the largest sources of PAHs are found to be from biomass burning, burning of coal, combustion of petroleum products[7], coke and production of metals The slow conversion of organic matter by the microorganisms release PAHs and these form the biogenic sources. The sources can also include iron, steel, aluminium production. Cement, coal, dye, rubber tyre, pesticides, fungicides etc. manufacturing release PAHs in the environment.

1.1 PAHs in Water

PAHs are very slightly soluble in water and have high affinity to particulate matter, so PAHs are not found in water in much concentrations. Their presence on surface water or ground water indicates pollution[8]. They may be found attached to the water body sediment. Usually about 11 PAHs are found in water, The PAH which are mostly present in water are made up of 2 or 3 rings of benzene and constitute about 63.64%. 4 benzene ring PAHs in water constitute about 27.27%. The PAHs having 5 to 6 rings constitute about 9.09% of the total amount. The PAHs get into water by oil spills, atmospheric deposition, water pollution, sewage, waste water etc[9]. PAHs are hydrophobic. So, they can be detected on surface water only. Low molecular weight PAH stay in water for short time as they get volatilised or oxidised. High molecular weight PAH get adsorbed to particles floating on water or bind to hydrophobic elements in water sediments. Some of the low molecular weight (LMW) PAHs are naphthalene, fluorine, acenaphthene. High molecular weight PAH are fluoranthene, phenanthrene, pyrene etc. In the presence of oxygen, PAHs slowly biodegrade and they are stable to hydrolysis. According to USEPA the maximum contaminant level (MCL) for drinking water is assigned as 0.2 ppb. As PAHs do not dissolve in water much, they do not readily bioaccumulate much in aquatic vertebrates. Due to this consumption of fish is quite safe for humans. However, consumption of bivalve molluscs may be risky as PAHs easily accumulate in their tissues. The European Union (EU Commission 2006) has set a maximum allowable level of Benzo(a)pyrene for bivalve molluscs in the market. Due to Deepwater Horizon oil spill the US Food and Drug Administration (FDA) has showed concern in the level of PAHs in fishes and shell fishes. However, it is found that PAH in drinking water accessed from streams and lakes pose least risk for human health.

1.2 PAHs in Soil

It is estimated that the maximum amount of atmospheric PAHs accumulate in the soil. Research says that the number of PAHs is found in more amount in the industrial areas. It is found that due to local and long-

distance transports, the atmospheric deposition of PAH in soil takes place[10]. PAHs also get into the soil when sludge from public sewage treatment plants is deposited in it, automotive exhaust, leaching from coal mines, soil compost and fertilizers. Vehicular exhaust and emission from wearing tyres and asphalt also contributes to addition of PAH in soil. The landfills, wood preserving sites, coking plants, places where previously gas plants were there are also source of PAHs in soil.

1.3 PAHs in Ambient Air

The direct release of PAHs in air is due to both natural and anthropogenic sources. Combustion of wood contributes to the largest amount of outlet of atmospheric PAH. Inefficient burning and uncontrolled emissions also contribute to atmospheric PAH. Natural sources like volcanoes and forest fires greatly contribute to the entry of PAH in the air. The type of PAH emissions varies with the combustion sources. Residential wood combustion releases acenaphthylene. Auto emissions release benzo(g,h,i) perylene and pyrene[11]. Diesel exhaust releases phenanthrene and anthracene.

Thousands of PAH have been identified out of which 16 of them have been identified as the priority PAHs by the EPA. They are namely acenaphthene, acenaphthylene, anthracene, fluoranthene, fluorene, naphthalene, phenanthrene, pyrene, benzo[a]anthracene, benzo[b]fluoranthene, benzo [k] fluoranthene, benzo [g,h,i]perylene, benzo [a] pyrene, chrysene, dibenzo[a,h] anthracene, indenol [1,2,3-c,d]pyrene[12]. Out of these Benzo [a] Pyrene that is B[a]P is one of the common carcinogenic polycyclic aromatic hydrocarbons. These 16 PAHs are prevalent and persistent in the environment and because of this they are a concern for the environment. Their toxicity in humans and other organisms is of great concern. The pollution level of PAHs in air, water and land is relatively high at the global scale. This is the reason shy it has caught the global attention. The PAHs bind readily to particulate matter and are carried by air and water and settle out in soil sediments, and these contaminate the environment. In the water bodies PAHs enter through deposition from atmosphere and direct releases of substances through petroleum spills and use, municipal wastewater treatment plants, industrial. PAHs can reach water bodies mainly through dry and wet deposition, road runoff, industrial wastewater. The air pollution may result in increased PAH in air. The PAH are generally teratogenic, carcinogenic, and mutagenic and may induce lung, bladder, as well as skin cancer. The most important thing to be noted about the toxic effect of PAH in humans is they are carcinogenic. Occupational exposure[13] to PAH leads to increase in the risk of contracting lung, skin and bladder cancers. But it is found that PAHs have low degree of acute toxicity in humans that is humans are not affected by PAHs when they are exposed to it for short period or randomly few times. When the PAHs are taken in through ingestion, come in contact with skin or inspired through nose of mammals and absorbed by tissues (EPA Ecological Toxicity). The coal tar sealed parking lots are a risk for house dust contamination. The contamination is mainly due to PAHs. USGS by its recent research upraised concern about exposure of youngsters to those PAHs by inhalation and ingestion. The US EPA has classified 7 PAHs as most likely to be carcinogenic for humans. States of California classifies 25 PAHs as carcinogenic. The International Agency for Research on cancer (IARC) classifies c2 PAHs as probable human carcinogen and 3 as possible carcinogens. According to IARC and EPA both, the probable human carcinogens are classified as Benzo(a)pyrene and Benzo(a)anthracene. The PAH Benzo(a)pyrene often acts as environmental indicator[14]. There are other non-carcinogenic effects of PAH which are not well understood yet. But research says that there can be other adverse effects on reproduction and development of an individual. In urban water the entry of PAHs happens by sewage discharges, leaching from landfills, surface runoff, stormwater runoff. In soil the PAHs can be carried either to the surface water of ground water through various form of polluted precipitation and surface runoff of polluted water. The PAHs are transferred to the crops from polluted soil and through air it reaches the roots and leaves and gets adsorbed to them. These may further accumulate in humans and other organisms through food chain and food web and result in biomagnification. In soil PAHs can enter if excessive usage of composts manufactured for horticulture. Burning of organic matter in the industries at high temperatures contribute to addition of PAHs in the soil.

2.0 Methodology

According to the EPA 16 PAHs have been designated as dangerous because they cause harm to the environment

and the living beings. The review paper gives an overview of the amount of PAHs in different soil, air and water samples in different cities of India. Various extraction methods were implemented based on the type of sample whether air, water sediment, soil or food[15]. Over the period from 2001 till date different types of solid-liquid extraction methods have been brought into practice. Some of the methods used in the extraction of PAHs from soil are as follows: -

1. Soxhlet Extraction
2. Ultrasonic agitation
3. Mechanical agitation
4. Accelerated solvent extraction
5. Supercritical and subcritical fluid extraction
6. Microwave assisted extraction
7. Solid phase extraction
8. Microextraction
9. Thermal desorption
10. Flash pyrolysis
11. Fluidised bed extraction

The modern methods include decoction, infusion, maceration, percolation, digestion, Soxhlet hot extraction etc

3.0 Reviewed Locations

Till now in India, research on occurrence and effects of Polycyclic aromatic hydrocarbons has been carried out in few cities. Given below are the review of the study of PAHs of few cities.

3.1 Dhanbad

Study with respect to polycyclic hydrocarbons was carried out in eight urban traffic sites and two control/ rural site surface soils of Dhanbad city. About 13 PAHs were found to be dominant. The concentration of PAHs in the traffic areas was found to be at a range of 1.019μgg^{-1} to 10.856μgg^{-1} with an average value of 3.488μgg^{-1}. It was found that the four and five ring PAHs constituted more than 50% of the total PAH. Three ring PAHs were predominant in rural soil of Dhanbad[16]. From the results it was evident that the increased concentration of PAHs was due to emission from vehicles. It was also moderately associated with combustion of coal and biomass. Road side soil where traffic is more was found to be 6 to 15 times more carcinogenic than rural soil.

3.2 Kerala Coastal Region

The coast of Kerala is endowed with backwaters, lagoons, estuaries etc. A comparative study of four sites of backwaters and estuaries of Southwest coast of Kerala[17] revealed that the total amount of concentration of PAHs ranges from 0.47 to 126.64ng/g with an average value of 15.08ng/g[18]. The polycyclic aromatic hydrocarbons which are low molecular weight rings, it was found that the concentration ranged from 0 to 5.42ng/g, whereas the polycyclic aromatic hydrocarbons with heavy molecular weight rings, the concentration ranges from 5.42 to 122.1ng/g[19]. It was found that the estuarine sediments had high molecular PAH and they constituted nearly 93.76% of total concentration. The amount of low molecular PAH constituted about 6.23%. The concentration of Naphthalene, Acenaphthene and Anthracene which are the low molecular weight PAHs is more. The heavy molecular weight PAHs include Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Indeno[1,2,3-cd] pyrene and Benz[ghi]perylene. The sediment towards the inner part of estuary had the higher amount of total 16 PAHs. It was assumed that the possible sources of PAHs are due to anthropogenic sources such as road runoff, exhaust from vehicles. Due to high temperature burning of substances and combustion of petroleum products, the PAHs may enter the marine environment.

3.3 Jamshedpur and Amravati

Soil found near industrial areas of Jamshedpur and Amravati had the PAHs concentrations mainly because of the vast network of the highway roads running through them[20]. As they are the industrial areas the pollution of environment due to industrial waste emissions, burning of coal and vehicular exhaust are the main contributors of PAHs in the area. 16 PAHs were found in Jamshedpur and the range was found to be less than 1700 to more than 10,500 μg m^{-3}. In Amravati the range was found to be from 1400 to more than 5000 μg m^{-3} [21]. The 4 ring PAH were found in maximum amount followed by 5 ring PAH. Few 2ring PAH were also found.

3.4 Mumbai

Mumbai is a city which is situated at the western coast of India[22]. Mumbai is the second most metropolitan populated territory in India. It is a hub of huge

petrochemical plants, power plants, compost plants. There are thousands of chemical industries, textile dyeing, medicine manufacturing companies. There are other companies which are responsible for PAH concentration and these are the paint making companies, pigment industries and industries which are responsible for extracting metals. In terms of population Mumbai is one of the fastest growing metropolitan city[23]. The urban location of Mumbai has the source of pollution due to low and high traffic. The sub-urban sites are mainly polluted due to vehicular exhaust and some small-scale industries. The moderate concentration of particulate matter at Urban site was 96.61μg m^{-3} and it ranged from 66.29 μg m^{-3} to 149.86 μg m^{-3} [24]. At Sub-urban site the moderate concentration of particulate matter was 100.30 μg m^{-3} and the range was between 68.66 μg m^{-3} to 182.15 μg m^{-3}. The above statistics gives the conclusion that the concentration of the particulate matter is more in the Sub-urban sites than the Urban sites.

3.5 Nagpur

Nagpur is considered as the second capital of Maharashtra. The industrialization and development of various avenues has taken place at a rapid speed. The vehicle population has been estimated as more than 10 lakhs over a period of short time. It was found that the moderate concentration of Suspended Particulate Matter (SPM) was higher in business area followed by industrial area with the value ranging from minimum 81.0μg/m^3 to maximum value of 323.0μg/m^3. In Nagpur city it is found out that the maximum amount of Polycyclic aromatic hydrocarbons is found mainly in ambient air. Out of three PAH compound Benzo(a)Anthracene is in highest concentration having a range of 0 to 1.56 ng/m^3 and next in the row is Benzo(α)Fluorene varying from 0 to 1.28 ng/m^3 and Benzo(a)Pyrene was found to have a range from 0.0 to 1.01 ng/m^3 [25].

3.6 Lucknow

Gomati river is one of the tributaries of river Ganga[26]. This river serves as one of the major sources of drinking water for the people of Lucknow. This river is contaminated by the effluents from drainage and untreated sewage water. This river is highly contaminated with PAH compounds which is posing a high risk on aquatic life. Total PAHs in water ranged from 0.467-5.342 μg/1 [27]. In the river water the most abundant hydrocarbon was acenaphthene.

3.7 Kolkata

It is seen that in Kolkata there is relatively high concentration of high molecular weight (HMW) PAH compared to other metropolitan cities[28]. Surprisingly the concentration of PAH was found to be almost same. The concentration in rural area is found to be due to biomass combustion. The urban environment is influenced by the emissions from industries and transportation.

3.8 Agra

The concentrations of PAHs attached to atmospheric particles was measured at a site near an industrial area Agra. The major sources of PAH were found to be vehicular exhaust, emissions from the industries, combustion of coal and biofuels etc. Among all the HMW PAH compounds comprising of 5 and 6 rings were found to be predominant. The mean total PAH concentration was found to be 115 ± 17ng/m^3. The measured concentrations were found to be lower than the metropolitan cities[29].

3.9 Delhi

It is considered to be one of the populated cities due to hubbub of administration, population increased due to migration of people and management of urban amenities. It is one of the most polluted cities due to contamination of air quality. The smaller size PAHs were observed to be in more amount. Benzo[a]pyrene (12%) was found to in largest amount whereas Acenaphthene (2%) was found in least amount. In the particles of fine fraction Pyrene seemed to dominate[30]. The sum of 12 PAHs ranged from 2.39 μg g^{-1} to 7.53 μg g^{-1} with a mean concentration of 4.43 ± 1.45 μg g^{-1} in the Indira Gandhi International Airport[31]. In the Yamuna River the 2-4 ring PAH were found to be predominant. The total number of 16 PAH compounds ranged from 4-50 to 23.53μg/g with an average concentration of 10.15±4.32 μg/g (dry wt)[32].

3.10 Chennai

Some municipal garbage dumping sites in Chennai were studied and it was found that they had considerable

amount of PAHs in it. It is found that the combustion of garbage in the dumping site is a source of PAHs in the developing countries. The common PAHs that were found out were Naphthalene and Phenanthrene. Various other sources like vehicular emissions, emission from industries and various other sources were studied and about 11 PAHs were found to be prevalent in ambient air. Out of these the PAHs with 2 to 7 rings are found to be hazardous. Particulate matter (PM 2.5)s values in Chennai varied between 27.2 and 190.2 μg/m^3 with an annual average of 91.8 μg/m^3.

3.11 Coimbatore

The city of Coimbatore which is in Tamil Nadu, is a fast-growing industrial city. The fine particulate matter of 2.5 range and PAHs were studied. About 9 harmful PAHs were found which can be dangerous. Due to over population and high emissions from vehicles and generators run by diesel, the PAH amount is said to have increased. The range is found to be 4.1 to 1632.3 μg/m^3.

Table 1 given below gives an overview of distribution of PAHs in few Indian cities and states.

LMW- Low molecular weight

Table 1. Showing the minimum and maximum range of PAHs in a few Indian city/states

Sl. No.	City/State	Sample	Range	Nature of PAHs	Analysis method	No. of PAH studied	Year of study
1.	Amravati	soil	1456.22 to 5403.45 ng/g	HMW	Soxhlet extractor, Gas chromatograph and flame ionization detector	16	2023
2.	Chennai	air	325.7 to 790.8 ng/m^{-3}	HMW	HPLC with fluorescence detector	11	2010
3.	Coimbatore	air	4.1 to 1632.3 ng/m^{-3}	HMW	HPLC, particulate sampler	9	2012
4.	Delhi	air	10.15 to 4.32 ng/m^{-3}	HMW	sonication	16	2019
5.	Dhanbad	soil	1.019 to 10.856 ng/m^{-3}	HMW	Soxhlet extractor, Gas chromatograph and flame ionization detector	16	2023
6.	Kerala	water sediment	0.4 to 126.64 ng/m^{-3}	LMW and HMW	Sonication/ultrasonic agitation	16	2021
7.	Kolkata (Sundarban)	water sediment	9.4 to 4222.8 ng/m^{-3}	HMW	GC/MS	19	2012
8.	Kolkata (South)	air	2.72 to 550.79 ng/m^{-3}	HMW	Passive bio samplers, GCMS,HPLC-UV, Sonication, Soxhlet, Microwave assisted Soxhlet extraction (MASE)	16	2023
9.	Kolkata (Urban)	air	22.91 to 190.96 ng/m^{-3}	HMW	High volume samplers, glass fibre filters, HPLC	12	2010
10.	Mumbai	air	38.8 to 24.5 ng/m^{-3}	HMW	Soxhlet, sonication	7	2000

HMW- High molecular weight

4.0 Conclusion

From the above review it is evident that research on PAHs has been done in very few cities of India. As pollution is a great concern in the present scenario, it is also equally important to monitor the level of PAHs in the environment. The reason being that these are very hazardous to human beings, aquatic life and the organisms of the soil. Moreover, if the sources are not controlled it will result in bioaccumulation of them. From the research which can be done in many more cities and states, development of effective strategies can happen which in the long run can communicate the PAHs related risk to the stakeholders, including communities, policymakers, industries etc. The collaborative efforts from all places can bridge the gap between the scientific research, development of relevant policies and the practical implementation of management and remediation strategies of the environment can be planned.

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