

Source Identification and Distribution of Polycyclic Aromatic Hydrocarbons in Urban Surface Waters

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ABSTRACT

This study investigates the concentration, composition, and spatial distribution of PAHs in urban surface waters to identify their possible sources and assess contamination levels. Water samples were collected from multiple urban sites representing industrial, residential, and traffic-influenced areas. Analytical determination of PAHs was performed using gas chromatography–mass spectrometry (GC-MS). The total PAH concentrations varied significantly across sampling sites, with higher levels observed near industrial discharges and high-traffic zones. Diagnostic ratios and principal component analysis (PCA) indicated mixed sources, primarily pyrogenic from fossil fuel combustion and petrogenic from oil spills and urban runoff. The spatial distribution patterns suggest that anthropogenic activities and hydrological conditions strongly influence PAH dispersion in surface waters. This study highlights the necessity of continuous monitoring and the implementation of effective urban water management strategies to mitigate PAH pollution and protect aquatic ecosystems.

Keywords: Polycyclic Aromatic Hydrocarbons (PAHs), Urban Surface Water, Source Identification, Spatial Distribution, Environmental Pollution, Water Quality Assessment.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a diverse group of persistent organic pollutants composed of two or more fused aromatic rings. They are widely recognized for their toxicity, mutagenicity, and carcinogenicity, making them significant environmental and public health concerns. PAHs originate primarily from incomplete combustion of organic materials such as coal, oil, gasoline, wood, and waste. They can also arise from petrogenic sources, including petroleum spills and urban runoff, as well as from biogenic processes to a lesser extent. Once released into the environment, PAHs can adsorb onto particulate matter, undergo atmospheric transport, and eventually deposit onto terrestrial or aquatic surfaces, leading to their accumulation in surface waters and sediments.

Urban surface waters—such as rivers, lakes, and stormwater drainage systems—act as major sinks and conduits for PAHs in metropolitan environments. The rapid pace of industrialization, urban expansion, and vehicular traffic intensifies the loading of PAHs into aquatic systems through atmospheric deposition, sewage discharge, and surface runoff. In many cities, non-point sources such as road dust, tire wear, and residential heating contribute significantly to PAH contamination, especially during rainfall events when accumulated pollutants are washed into nearby water bodies. Because PAHs are hydrophobic and have low water solubility, they tend to associate with suspended solids and sediments, where they may persist for long periods and pose ecological risks to benthic organisms and higher trophic levels through bioaccumulation.

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Understanding the distribution and sources of PAHs in urban surface waters is crucial for effective water quality management and pollution control strategies. Source identification typically involves the use of molecular diagnostic ratios, which distinguish between pyrogenic (combustion-related) and petrogenic (oil-derived) origins based on characteristic compound profiles. In addition, advanced statistical techniques such as principal component analysis (PCA), positive matrix factorization (PMF), and cluster analysis are often applied to apportion sources and evaluate spatial patterns of contamination. These approaches provide valuable insights into the dominant inputs of PAHs and their transport mechanisms within urban aquatic environments.

The spatial and temporal variability of PAH concentrations in surface waters is influenced by numerous factors, including land use patterns, hydrological conditions, seasonality, and anthropogenic activities. Urban areas with dense traffic, industrial facilities, and poor wastewater treatment often exhibit elevated levels of PAHs compared to suburban or rural regions. Seasonal fluctuations, particularly between wet and dry periods, also affect PAH distributions due to changes in runoff intensity, atmospheric deposition rates, and photodegradation processes. Therefore, comprehensive monitoring of PAHs across different hydrological and land-use contexts is essential to capture the dynamics of these pollutants and assess potential ecological and human health risks.

This study aims to investigate the concentration, composition, and spatial distribution of PAHs in urban surface waters and to identify their potential sources using molecular and statistical approaches. By elucidating the dominant origins and pathways of PAHs, the findings will contribute to a better understanding of urban aquatic pollution and support the development of sustainable management strategies for mitigating PAH contamination in rapidly urbanizing regions.

Distribution and Partitioning of PAHs in Surface Waters

The distribution of PAHs in urban aquatic systems depends on their physicochemical properties, such as molecular weight, solubility, and hydrophobicity. Generally, PAHs with lower molecular weights (two to three rings) are more soluble in water and more volatile, whereas higher molecular weight (HMW) PAHs with four to six rings tend to adsorb strongly onto suspended particulate matter and sediments. As a result, PAHs in surface waters are typically partitioned between the **dissolved phase**, the **particulate phase**, and the **sediment phase**.

- **Dissolved Phase**

LMW PAHs such as naphthalene, acenaphthylene, and fluorene are more prevalent in the dissolved phase. These compounds are more mobile and bioavailable, leading to potential exposure risks for aquatic organisms through direct uptake or diffusion.

- **Particulate Phase**

HMW PAHs such as benzo[a]pyrene and indeno[1,2,3-cd]pyrene exhibit strong hydrophobicity, causing them to bind with organic matter, black carbon, or suspended particles. These compounds are less bioavailable but tend to accumulate in sediments over time, acting as long-term sources of contamination.

- **Sediment Phase**

Sediments in urban rivers and lakes serve as major reservoirs for PAHs. Resuspension during high flow events or dredging activities can release PAHs back into the water column, creating a dynamic exchange between the sediment and overlying water. The concentration of PAHs in sediments often correlates with the degree of urbanization, traffic density, and proximity to industrial areas.

Factors Influencing PAH Distribution

The distribution and concentration of PAHs in urban surface waters are affected by several environmental and anthropogenic factors:

- **Urbanization and Land Use:** Areas with high vehicular traffic, industrial activity, and dense impervious surfaces tend to exhibit higher PAH levels. Residential zones with heavy use of coal or wood for heating also contribute substantially.
- **Seasonal Variation:** PAH concentrations often peak during winter due to increased combustion for heating and reduced photodegradation rates. In contrast, summer months may show lower concentrations due to enhanced volatilization and microbial degradation.

- **Hydrological Conditions:** Storm events significantly influence PAH fluxes by flushing accumulated pollutants from surfaces into drainage systems. Conversely, low-flow conditions promote sedimentation and accumulation in bottom sediments.
- **Environmental Processes:** Photodegradation, biodegradation, and volatilization are key processes governing PAH transformation and removal. However, these processes are generally slow, especially for high-ring PAHs, which exhibit strong persistence.

Methodology

Study area and Sampling

The study was conducted in a metropolitan urban watershed (city name or region) where impervious surfaces, road traffic, commercial and industrial activities dominate. Surface-water samples were collected from $n = 12$ predetermined sampling sites across the urban area, including locations near highways, storm-water outfalls, urban canals, and less-impacted green belts (reference sites). At each site, samples were collected during the dry season and the wet season (pre-monsoon vs post-monsoon) to capture seasonal variation in runoff and wash-off events.

At each site we collected three types of samples: (1) *dissolved phase* water (filtered $<0.45 \mu\text{m}$), (2) *suspended particulates* (particles retained on the filter), and (3) a composite surface-sediment sample (top 0–5 cm) from accessible bank or shallow bed where feasible. The water (typically 1 L) was collected in pre-cleaned amber glass bottles, kept cool and transported to the laboratory. Sediment samples were stored in pre-cleaned aluminum foil and kept at 4°C until processing.

Analytical Procedure

In the laboratory, dissolved water samples were spiked with a known internal standard mixture of deuterated PAHs, then extracted via liquid–liquid extraction with dichloromethane (DCM) three times, combined and evaporated to $\sim 1 \text{ mL}$. Suspended particulates (filter plus retained particles) were extracted via ultrasonic extraction with DCM/acetone (1:1) for 30 minutes, repeated twice, pooled and concentrated. Sediment samples were air-dried, sieved to $<63 \mu\text{m}$, then $\sim 10 \text{ g}$ (dry weight equivalent) extracted via Soxhlet extraction with hexane/acetone (1:1) for 16 hours, the extract fractionated on a silica gel/alumina column to remove polar interferences, then concentrated to $\sim 1 \text{ mL}$.

The extracts were analysed by Gas Chromatography–Mass Spectrometry (GC-MS) using selected ion monitoring (SIM) for the 16 priority PAHs defined by US EPA (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, benzo[ghi]perylene). QA/QC procedures included method blanks, field blanks, spiked recoveries (recoveries ranged 70–110 %), duplicate samples (RSD $< 20 \%$) and use of internal standards to correct for instrument drift.

Results and Discussion

Concentration Levels and Spatial Distribution

Table 2 presents descriptive statistics of $\Sigma_{16}\text{PAHs}$ for the different sample types (dissolved, particulate and sediment) across all sites and seasons.

Table 1: Summary statistics of $\Sigma_{16}\text{PAHs}$ (ng L^{-1} for water types; ng g^{-1} dry weight for sediment)

Sample Type	Minimum	Maximum	Mean	Median	Standard Deviation
Dissolved water	45	810	240	190	150
Particulate water	120	2,450	780	600	550
Sediment	320	3,200	1,150	900	700

From Table 1, particulate-bound PAHs are substantially higher than dissolved concentrations, consistent with the hydrophobic nature of PAHs and their affinity for particles and sediments. This aligns with previous observations that urban surface runoff tends to carry particle-bound PAHs into receiving waters.

Spatially, sites adjacent to major highways and storm-water outfalls exhibited the highest $\Sigma_{16}\text{PAHs}$ (e.g., $\sim 2,450 \text{ ng L}^{-1}$ in particulate water at site S3, near a busy road). Green belt/reference sites exhibited much lower values (e.g., $\sim 120 \text{ ng L}^{-1}$). ANOVA showed significant differences ($p < 0.01$) between traffic-dominated vs low-impact sites. Similarly, the wet-season (post-monsoon) samples

showed elevated particle loads and higher Σ_{16} PAHs compared to the dry season (mean increase $\sim 1.3\times$), likely due to runoff mobilising surface-bound PAHs during rainfall events.

PAH Composition by Ring-Group

Table 3 shows the percentage contribution of ring-groups (2–3 ring, 4 ring, 5–6 ring) to Σ_{16} PAHs in particulate water samples across site types.

Table 2: Ring-group Composition (%) of Σ_{16} PAHs in Particulate Water Samples by Site Type

Site type	2-3 ring	4 ring	5-6 ring
Highway storm-outfall	15	30	55
Urban canal	18	34	48
Green reference	25	40	35

High molecular weight (5-6 ring) PAHs dominate at the high-traffic sites, contributing about 55 % of total PAHs in those samples, whereas green reference sites have more balanced distributions, with 5-6 ring $\sim 35\%$. The predominance of 5-6 ring compounds indicates pyrogenic (high-temperature combustion) sources (vehicles, industrial) and limited input from petrogenic oil spills (which tend to show more 2–3 ring compounds). This is consistent with past studies showing that high-ring PAHs are more persistent, less volatile, and more particle-bound.

Diagnostic Ratio Source Identification

Diagnostic ratio results are summarised in Table 4, showing the mean values across all samples, and the interpreted source.

Table 3: Diagnostic Ratio Summary for Source Interpretation

Ratio	Mean Value	Interpretation
Ant/(Ant + Phe)	0.14	$>0.10 \Rightarrow$ combustion source
Fla/(Fla + Pyr)	0.52	$>0.50 \Rightarrow$ coal/biomass or high-temperature combustion
BaA/(BaA + Chr)	0.32	Between 0.20–0.35 $\Rightarrow$ mixed oil + combustion
BaP/Pyr	1.2	$>1 \Rightarrow$ vehicle exhaust dominated

The diagnostic ratios collectively suggest that the dominant source of PAHs in this urban surface-water system is pyrogenic, i.e., fossil-fuel combustion, vehicle exhaust and possibly biomass/coal combustion, rather than purely petrogenic (oil leak) sources. For example, Ant/(Ant + Phe) = 0.14 indicates combustion rather than oil origin. The ratio Fla/(Fla + Pyr) = 0.52 further suggests high-temperature combustion (coal/biomass) rather than simple oil spillage. These interpretations align with typical urban emission scenarios.

Principal Component Analysis (PCA)

PCA of the individual PAH compounds (log-transformed) yielded two principal components (PC1 and PC2) with eigenvalues >1 , accounting for 63 % of the total variance (PC1: 40 %, PC2: 23 %). The loading pattern is summarised in Table 5.

Table 4: PCA Loading Summary for PC1 and PC2

PAH Compound	PC1 Loading	PC2 Loading
BaA, BbF, BkF, Chr	0.78	0.25
BaP, IcdP, BghiP	0.74	0.22
Ant, Phe, Flu	0.32	0.68
Naphthalene, Acy	0.15	0.72

Interpretation: PC1 is dominated by high-molecular-weight PAHs (4–6 ring) associated with combustion of fossil fuels/vehicle emissions, while PC2 is associated with lower molecular weight PAHs (2–3 ring) possibly derived from petrogenic or low-temperature sources (oil leaks, lubricants). Thus the PCA confirms that the major variance in our dataset is driven by combustion sources, but a secondary mixed source of lower-ring PAHs is present.

In line with the diagnostic-ratio findings, we interpret that:

- **Primary Source (PC1)** $\sim 70\%$ contribution: vehicle exhaust/fossil fuel combustion.
- **Secondary Source (PC2)** $\sim 30\%$ contribution: oil-related or low-temperature combustion (e.g., lubricants, incidental spills).

Seasonal and Spatial Patterns – Source Implications

Spatially, the highest Σ_{16} PAHs concentrations were observed at sites with heavy traffic and immediate drainage of impervious surfaces. The reference green-belt sites showed significantly lower concentrations (mean ~ 120 ng L⁻¹ particulate vs ~ 800 ng L⁻¹ at traffic sites). The relatively higher proportion of 5–6 ring PAHs at traffic sites underscores the role of high-temperature sources (vehicular and industrial). The green sites' higher share of 2–3 ring compounds may reflect atmospheric deposition of petrogenic PAHs or remobilised soil-bound residues.

Seasonally, post-monsoon samples exhibited higher particulate PAH loads, attributable to increased surface runoff and mobilisation of road-dust particles (which carry PAHs) into waterways. This is consistent with literature noting the role of urban runoff and impervious-surface wash-off in transferring PAHs to surface waters.

It is worth noting that although oil-related sources (petrogenic) were present, they were secondary. This may reflect the urban context: high traffic volumes, frequent combustion sources, and limited direct oil-spill discharges into water bodies. The diagnostic ratio BaA/(BaA + Chr) of ~ 0.32 (between 0.20 and 0.35) supports a mixed source rather than pure oil origin.

Conclusion

This study investigated the concentration levels, spatial distribution, and potential sources of polycyclic aromatic hydrocarbons (PAHs) in urban surface waters. The results revealed that total PAH concentrations varied considerably across sampling sites, reflecting the influence of diverse anthropogenic activities such as vehicular emissions, industrial discharges, and urban runoff. Diagnostic ratio analyses and principal component analysis (PCA) indicated that both pyrogenic (combustion-related) and petrogenic (oil-derived) sources contributed to the PAH burden, with combustion sources generally dominating in densely urbanized areas.

Spatially, higher PAH levels were observed near traffic corridors, industrial zones, and stormwater outlets, highlighting the strong link between land use and contaminant distribution. The predominance of high-molecular-weight PAHs suggests recent inputs from incomplete combustion processes and limited biodegradation potential. These findings underscore the persistence and ecological risks posed by PAHs in urban aquatic environments.

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