Review

Polycyclic Aromatic Hydrocarbons (PAHs) in Freshwater Systems: A Comprehensive Review of Sources, Distribution, and Impacts

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Review

Polycyclic Aromatic Hydrocarbons (PAHs) in Freshwater Systems: A Comprehensive Review of Sources, Distribution, and Impacts

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Abstract: This review paper presents a comprehensive synthesis of current knowledge on the contamination of water systems by polycyclic aromatic hydrocarbons (PAHs). PAHs, organic contaminants characterized by two or more fused aromatic rings, are notably problematic due to their persistence, bioaccumulation, and significant health risks. The review commences with an examination of various sources of PAHs in aquatic environments, encompassing both anthropogenic activities, such as industrial discharges, urban runoff, and oil spills, and natural phenomena including volcanic eruptions and wildfires. Subsequently, the discussion shifts to the distribution patterns of PAHs within different aquatic ecosystems, with an analysis of factors that influence their transport. Considerable focus is placed on the entry mechanisms of PAHs into water systems and their interactions with diverse environmental matrices. Additionally, the ecotoxicological effects of PAHs on aquatic life are scrutinized, with particular emphasis on molecular, individual, and ecosystemic impacts. This includes an exploration of PAH bioaccumulation in aquatic organisms and their potential mutagenic and carcinogenic consequences. The review further addresses the challenges associated with detecting and quantifying PAHs in aquatic settings, reflecting on the advancements in analytical methodologies and the complexities introduced by their varied physicochemical properties. The paper concludes with a summary of existing regulatory frameworks and guidelines governing PAH contamination in water systems, underscoring the imperative for global collaboration in the monitoring and management of PAH pollution.

Keywords: polycyclic aromatic hydrocarbons; water pollution; ecotoxicology; environmental distribution; analytical methods; regulatory frameworks

1. Introduction

1.1. Polycyclic Aromatic Hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are a class of semi-volatile organic pollutants, ubiquitous in the environment. PAHs contain two or more aromatic rings and are produced mainly by incomplete combustion of organic matter in the environment. They are classified as low molecular weight (LMW) with two to three fused aromatic rings, and high molecular weight (HMW) that contain four or more fused aromatic rings [1,2]. LMW PAHs are recognized for causing significant and immediate toxicity in living organisms, whereas HMW PAHs are known for their potential to cause cancer, mutations, genotoxicity, and teratogenicity to living organisms [1,3]. Increase in molecular weight among PAH chemical species, will increase hydrophobicity, lipophilicity, melting, and boiling point [4], while water solubility and vapor pressure decreases [5]. These physicochemical properties affect their distribution, transportation, and fate in the environment.

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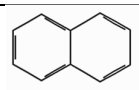
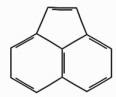
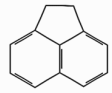
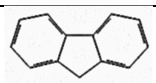
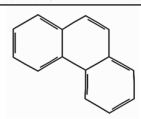
PAH transfer among food web compartments is promoted by their lipophilic nature [6] that promotes bioaccumulation in animal bodies [7], exposing human consumers eventually [7,8]. On the other hand, HMW PAHs are more persistent compounds in the environment making them less biodegradable [9]. These characteristics and environmental behaviors of PAHs are crucial for comprehensive understanding of their impacts on ecosystems and human health.

PAHs originate from pyrogenic, petrogenic, and biogenic processes occurring during natural phenomena or anthropogenic activities. In the environment, these compounds originate primarily from pyrogenic and petrogenic processes [10]. Pyrogenic PAHs results from high temperature combustion, incomplete combustion and/or pyrolysis of organic matter under low or no oxygen conditions, such as the combustion of fossil fuels, wood, wildfires, coal, and volcanic activity [11]. The diagenesis of organic matter occurring over extended periods of time at specific temperature and pressure conditions leads to formation of petrogenic PAHs [12–14]. Common anthropogenic emissions of petrogenic PAHs to the environment, are petroleum-derived products, fossil fuels, oily-lubricant leakage, and crude oil exploration activities from industry, commercial, and transportation development [1,11]. While natural emissions consist of petroleum seepage lakes, marine oil seeps, and crude oil-tar lagoons [14–16]. Lastly, biogenic PAHs are synthetized by microorganisms (bacteria, fungi), plants, or phytoplankton without involvement of diagenesis processes [7,17,18]. Some documented emissions sources of biogenic PAHs are termite nests [19,20], bio-synthetization by magnolia flowers [21], and *Annonaceae* family in Amazon rainforest [22]. Diverse anthropogenic and natural emissions discharge PAHs into the environment through the three main origin processes, and can be tracked by analyzing PAH compositional pattern in sample matrices [23–25].

Globally, a great concern is arising about the presence and persistency of PAHs in aquatic, terrestrial, and atmospheric systems [26]. More than 200 PAHs had been discovered in the environment [10]. However, only 14 to 20 individual PAHs are of regulatory and research interests [11]. Various governmental and non-governmental organizations, such as the United States Environmental Protection Agency (USEPA) and Greenpeace [27], are actively involved in monitoring these pollutants. USEPA established 16 PAHs as priority pollutants for monitoring in the environment [28–30]. These 16 priority PAHs, were selected based on human health risk, exposure, toxicity, and occurrence in environment [31]. Many studies are focused on reporting the concentration and presence of these 16 priority PAHs in the environment. Table 1 presents the summary of the physicochemical properties (e.g., melting point, solubility, etc.), chemical structure, and classification of the 16 USEPA priority PAHs.

Table 1. Physical properties and chemical structure of the 16 USEPA priority PAHs.

No.	Name	CS[(a)]	Rings	Class.[(b)]	MW[(c)]	BP[(d)]	MP[(e)]	S[(f)]	Log K_{ow}[(g)]	VP[(h)]
1	Naphthalene		2	LMW	128.17	209	80	31.0	3.37	$8.89E^{-2}$
2	Acenaphthylene		3	LMW	152.19	290	124	16.1	4.00	$2.90E^{-0.2}$
3	Acenaphthene		3	LMW	154.21	252	108	3.8	3.92	$3.75E^{-3}$
4	Fluorene		3	LMW	166.22	276	119	1.9	4.18	$3.24E^{-3}$
5	Phenanthrene		3	LMW	178.23	326	136	1.1	4.57	$6.80E^{-4}$

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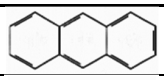
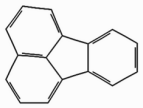
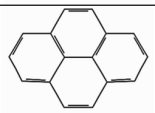
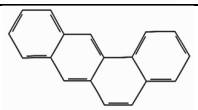
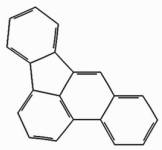
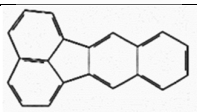
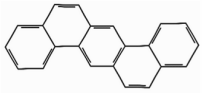
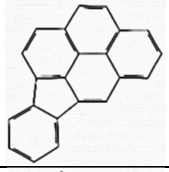
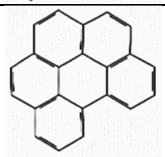
6	Anthracene		3	LMW	178.23	326	136	0.045	4.54	$2.55E^{-5}$
7	Fluoranthene		4	HMW	202.25	369	166	0.26	5.22	$8.13E^{-6}$
8	Pyrene		4	HMW	202.25	369	166	0.132	5.18	$4.25E^{-6}$
9	Benzo[a]anthracene		4	HMW	228.30	400	177	0.011	5.91	$1.54E^{-7}$
10	Chrysene		4	HMW	228.30	400	177	0.0015	5.91	$7.80E^{-9}$
11	Benzo[b]fluoranthene		5	HMW	252.30	461	209	0.0015	5.80	$8.06E^{-8}$
12	Benzo[k]fluoranthene		5	HMW	252.30	430	194	0.0008	6.00	$9.59E^{-11}$
13	Benzo[a]pyrene		5	HMW	252.30	461	209	0.0038	5.91	$4.89E^{-9}$
14	Indeno [1,2,3-cd]pyrene		6	HMW	276.30	498	233	0.062	6.50	$1.40E^{-10}$
15	Dibenz[a,h]anthracene		6	HMW	278.30	487	218	0.0005	6.75	$2.10E^{-11}$
16	Benzo[g,h,i]perylene		6	HMW	276.30	467	218	0.00026	6.50	$1.00E^{-10}$

(a) CS Chemical structure [NCBI 32]; (b) Classification as LMW and HMW; (c) MW Molecular weight [32]; (d) BP Boiling point [33]; (e) MP Melting point [33]; (f) WS Water solubility at 25**°C** [34]**;** (g) Octanol water partitioning [35]; (h) VP Vapor pressure mmHg at 25**°C** [34]**.**

PAHs are ubiquitous in the environment and occur in various environmental compartments, raising concerns of possible ecological and human health impacts. When formed, PAHs reach the atmosphere as particles bound or gas forms [36]. In the atmosphere LMW PAHs remain in vapor phase while HMW PAHs tend to adsorb to particulate matter [37] facilitating there Long-range transport (LRT) [38]. Eventually by wet or dry deposition they enter terrestrial or aquatic environment [11]. In Figure 1, PAH dynamics in the environment involve their mobility upon deposition on soil surfaces [39], often facilitated by runoff or weathering processes, and subsequently be transported into water systems [40]. In water systems PAHs partition, either binding to sediment or present in the dissolved phase, posing ecological and health risks to aquatic organisms and humans [39].

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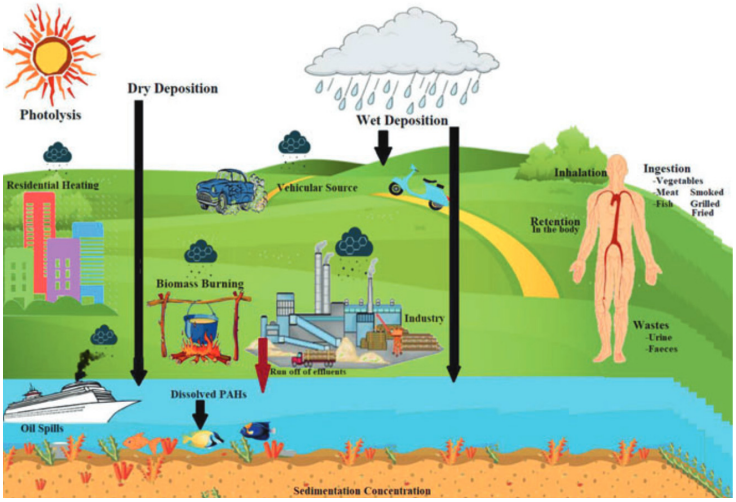

Figure 1. Distribution of PAHs in air, terrestrial, and aquatic environments. Reprinted with permission from ref. [39], Copyright 2019, Springer.

PAHs can enter aquatic systems through various pathways, primarily via wet and dry deposition from the atmosphere, sewage discharges, industrial effluents, and runoff from urban or agricultural areas. In water they can be in dissolved phase, bound to organic matter, or adsorbed onto particulate matter or sediment [41]. Distribution in the water column is influenced by the sediment-water partition coefficient [42]. Sediment-bound PAHs can be resuspended into the water column, increasing exposure risk to aquatic organisms and undergo long-range transport within watersheds before eventually reaching the ocean.

PAHs produces toxic effects on various living organisms that includes humans, animals, and microorganisms. Biochemical persistence derives from the dense π electrons in their ring structure that makes them resistant to nucleophilic attack [5]. Some PAHs are known for their genotoxicity, mutagenicity, carcinogenicity, and teratogenicity, posing risks to the endocrine system [43,44]. HMW PAHs are less volatile, more lipophilic, environmentally persistent, and hydrophobic, raising concerns about their associated risks to humans, the environment, and living organisms [45]. They also bind to dissolved organic matter deposited in sediment thus accumulating in the aquatic environment, increasing toxicity levels in the benthic area of aquatic systems [2,41,46]. Also rain runoff can transfer soil-bound PAHs to water, affecting flora, aquatic life, and food chains, ultimately posing health risks to humans [47]. PAHs are characterized by their diverse toxic effects and persistence in the environment, presenting significant risks to human health and the environment from their toxicity perspective and their potential to bioaccumulation.

1.2. Freshwater Systems

Freshwater systems, comprise 2.5% the world's terrestrial surface area [48,49]. These terrestrial freshwater systems are vital resource for human consumption and ecological integrity [50]. Encompassing diverse water bodies such as lakes, ponds, reservoirs, rivers, streams, groundwater aquifers, estuaries, and wetlands that cover 0.8% of the Earth's surface and are unequally distributed globally [51–53]. Despite their significance, freshwater resources face vulnerability not only by their geographical limitations but also due to human interventions and pollution [52,54]. The broader context of Earth's water distribution reveals that while approximately 71% of the Earth's surface is water-covered, the oceans hold the majority, about 96.5%. Within the limited freshwater supply, the majority is locked in ice and underground compartments, with slightly more than 1.2% constituting surface water, meeting the essential needs of life [48]. Figure 2 illustrates the proportion of water found in different reservoirs, including oceans, glaciers and ice caps, groundwater, lakes, and rivers

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as relative percentage of water stored in each reservoir. Managing and preserving the relatively small portion of freshwater available for sustaining various life and ecosystem's function, is fundamental.

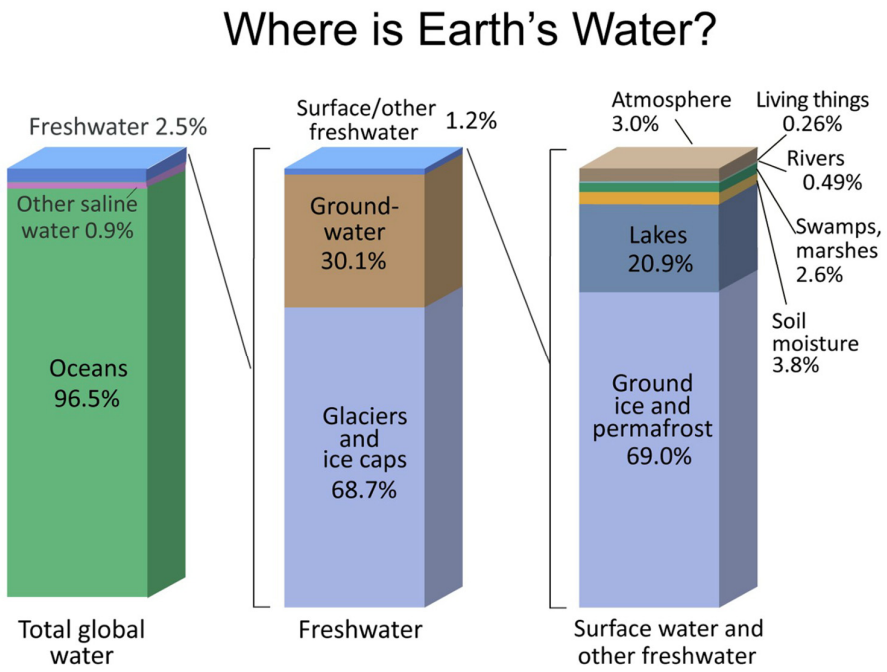

Figure 2. Global water distribution by percentage in different reservoirs. Public domain image courtesy of [55] and data from [48].

Increase in industrialization, agricultural practices, and socio-economic development near freshwater systems, contributes with diverse organic pollutants that impacts environmental quality [56]. Among organic micropollutants, PAHs are one of the most abundant in freshwater systems and present toxic properties even at low doses, leading to bioaccumulation in fishes [57], invertebrates [7], and algae [58]. The prevalence of PAHs in freshwater systems poses a significant environmental concern, with their discharge into aquatic environments occurring through various pathways, including atmospheric deposition, runoff, and wastewater inputs [59]. Their hydrophobic nature promotes the adsorption of PAHs to suspended sediments in diverse aquatic ecosystems composed by wetlands, rivers, lakes, and estuarine environments [60–62]. A comprehensive understanding of the impacts of PAH pollution on freshwater systems globally, is necessary to assess and mitigate its effects in freshwater systems, their biodiversity and human health protection.

PAHs occurrence, distribution, and fate [63] represent an existing global [64] threat to freshwater ecosystems, irrespective of their geographical remoteness [65]. Inputs from both point and non-point sources, continuously introduce these compounds into freshwater systems, resulting in temporal and spatial heterogeneity [28,66]. PAHs, deposited from atmospheric sources, runoff, and water discharge inputs, either adsorb onto suspended sediments or exist in a freely dissolved state within freshwater systems [67]. Despite the growing awareness of atmospheric PAHs, the study of these compounds in terrestrial and freshwater systems is limited [68]. Thus, the widespread distribution, emission sources, and impacts of PAHs in freshwater systems must be thoroughly reviewed and critically examined in recent literature.

There have been numerous and recent literature reviews about PAH dynamics in different environmental topics [1,11,31,63,68–72]. On the other hand, recent publications had examined specialized aspects of PAHs in freshwater systems. [2,31,47,71,73–75]. A comprehensive overview of PAHs in freshwater systems will provide a better understanding of the behavior and dynamics of

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these compounds in natural environments. Thus, is necessary to provide a recent comprehensive literature review that discusses general aspects of PAHs in freshwater systems.

The main objective of this review paper is to synthesize the most recent literature on the sources, distribution, and impacts of PAHs in freshwater systems, with additional focus on regulations and commonly implemented analytical methods for their environmental monitoring and management. A synthesis of this recent information will establish a comprehensive understanding and contribute significantly to the current state of knowledge, thereby informing further research and regulatory direction.

2. Sources of PAHs in Freshwater Environments

PAHs may enter freshwater systems through diverse processes and emission sources. It is possible to identify PAHs origins and emissions through a combination of data analysis models, such as molecular diagnostic ratios, principal component analysis (PCA), positive matrix factorization model, and other methods [76]. These sources can be influenced by geographic features [77], topography [78], hydrological conditions [79], landscape characteristics [80], land use patterns [81], climate variations [82], anthropogenic activities, and natural phenomena [11]. The interaction of these factors can contribute to PAHs discharges on freshwater systems. Additionally, the type of freshwater body with their distinct physical, chemical, and hydrological properties, affect PAHs dynamics in water [11,83,84]. Identifying the sources and understanding the factors influencing PAHs concentration in these systems is crucial for comprehending their ecological impacts and formulating effective mitigation strategies.

2.1. Lakes

The availability of water resources impacts the economic development of a region, involving water supply, recreational opportunities, fisheries, and other ecosystem services that lakes provide. In the case of lakes, surrounded by urban settings, pose unique challenges when diverse emission sources contribute to the presence of PAHs. PAHs sources in lakes, includes those from petroleum, biomass, heavy oils, and natural gas consumption [85]. Studies have reported varying ∑16PAHs of USEPA priority list, with concentrations reaching more than 10,000 ng L^{-1} [86]. LMW congeners and monomeric PAHs dominate in dissolved water phase at high levels, being naphthalene the most abundant, while HMW stay adsorbed into sediments (Figure 3) [87,88]. Comprehensive analyses in Ontario Lakes reveal that local sources, especially non-point atmospheric sources from pyrogenic activities can have a more substantial impact than regional sources [10]. While successful policies for point sources reduction had been implemented in different countries [89,90], non-point sources like atmospheric deposition and surface runoff are major contributors to lake systems [91].

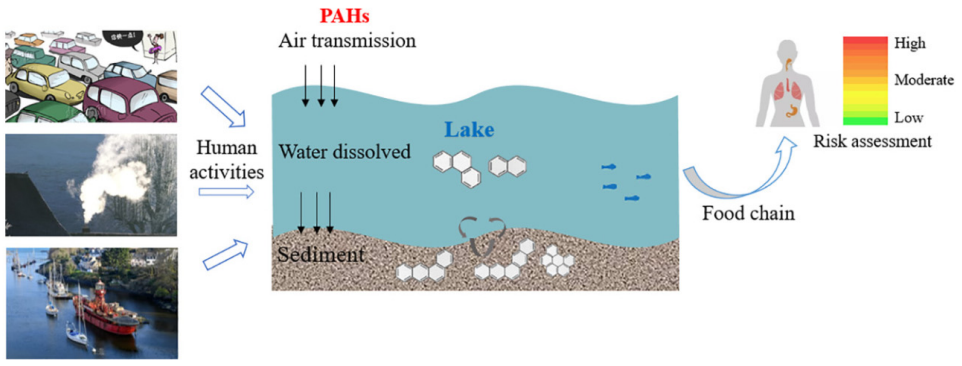

Figure 3. Sources, distribution, and transfer of PAHs through environmental compartments in freshwater lakes until human exposure is achieved. Reprinted with permission from ref. [87], Copyright 2019, Elsevier.

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HMW PAHs with geochemical and environmental significance, dominate recent sediment cores from northern Ontario Lakes and indicated a predominant pyrogenic local sources [92]. While coal-tar sealed pavement contributes to the transfer of PAHs into lakes through various pathways (Figure 4) [93], resulting in higher Σ16PAHs concentrations in Great Lakes Basin watershed compared with other water bodies throughout the world [94]. Urbanization intensifies the presence of PAHs in lakes, associated with the degree of urbanization and increased runoff from impervious surfaces [85,95,96]. Economic development in the Taihu Lake Basin correlates with changes in PAHs sediment composition, highlighting the need for up-to-date ecological risks assessment and protection efforts [97]. In the case of remote lakes less affected by anthropogenic activities, atmospheric deposition from long-range transported PAHs and wildfires can be the main contributors [47,98,99]. On the other hand, studies spanning from 2002 to 2018 emphasize that ubiquitousness of PAHs in lake sediments is caused by anthropogenic combustion, petroleum industries, road traffic, and socioeconomic factors, being China, Egypt, United States of America (USA), and some African lakes as the more polluted [10]. The presence of PAHs in lakes are mainly driven by diverse urban and anthropogenic activities in the surrounding landscape, with wildfires as main contributor in remote lakes.

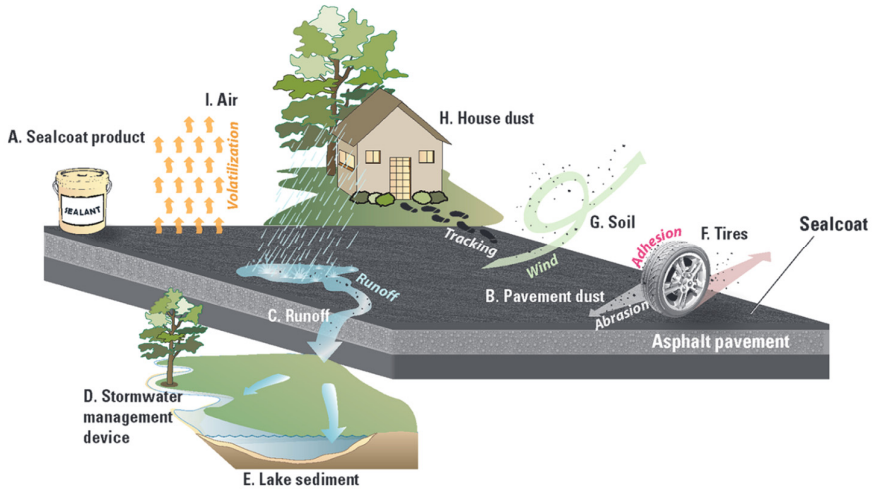

Figure 4. The main pathways of PAHs from coal-tar based pavement, are dried sealcoat product (A), pavement dust (B) transported by surface runoff (C) that discharges dust into stormwater management infrastructure (D), streams, and lakes (E). In addition, dust attaches to vehicle tires (F) that can be transported further to other sites where wind and runoff continue to disperse PAHs to long distances (G). Attached dust in shoes transport PAHs into indoor environments (H). While volatile PAHs are released into the atmosphere by the coal-tar sealed pavement (I). Reprinted with permission from ref. [93], Copyright 2012, ACS.

2.2. Rivers

Rivers play a fundamental role as primary transport pathways for organic pollutants, from upstream to downstream until discharging them in lakes, wetlands, or estuaries [100,101]. Specifically, large rivers with expansive watersheds and upstream tributaries contain PAHs from mixed emission sources [61,102,103]. An example is the Buffalo River Estuary in South Africa, where pollutant levels are influenced primarily by pyrolytic sources, such as effluent discharge from automobile, petrochemical industries, landfill leachate, domestic and industrial wastewater, vehicle emissions on highways, harbor activities, and drains from urban runoff from the city [104]. Oil related activities with weaker combustion contribution are also main non-point sources of PAHs in Nun River, Nigeria [105]. While, anthropogenic expansion promotes deposition of PAHs in surface water, water column layers, and sediments [106], increasing ecological risk in rivers [107]. Euphrates River in Iraq, is the longest river in southwest Asia and presented pollution sources from petroleum

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product consumption influenced by the increase of private car and home electricity generators ownership [108]. Predominance of HMW PAHs in riparian soils from rivers impacted by coal mining areas, indicated origins of pyrogenic sources like vehicular emissions, biomass, and coal combustion [109]. San Joaquin River in Northen California (USA) surface and core sediment analysis estimated that pyrogenic sources increased with time, while biogenic sources were the main contributors in the past [110]. Studies on the River Benue, Nigeria (Africa) emphasize the contribution of diverse anthropogenic activities, with petrogenic and pyrogenic sources of PAHs in equal magnitudes [111]. Specifically, a comprehensive review of PAHs in seven main river basins in China, reported coal and biomass combustion from pyrogenic sources as the main contributors [112]. Further analysis in riverine transport of PAHs in middle-lower Yangtze River, China, determined coal and coke combustions along with vehicle emissions [113]. In this case, sediments acted as secondary emission sources of LMW PAHs and as a sink for HMW PAHs through water-sediment exchange, resuspension, and deposition (Figure 5) [113]. Whereas, 30 river systems in Taiwan reported that PAHs sources in sediments varied across geographic locations with seasons [114]. A assessment of PAHs in USA coastal areas, were various rivers discharge, established that pyrogenic PAHs dominates [115], while estuarine river systems in Mumbai, India resulted in mixed sources [116]. Rivers serve as a conduit for PAHs and large drainage basins presents mixed emissions, been pyrogenic the main contributor due to diverse land use characteristics. There is a need for comprehensive management and analysis that addresses ecological risks along river catchments.

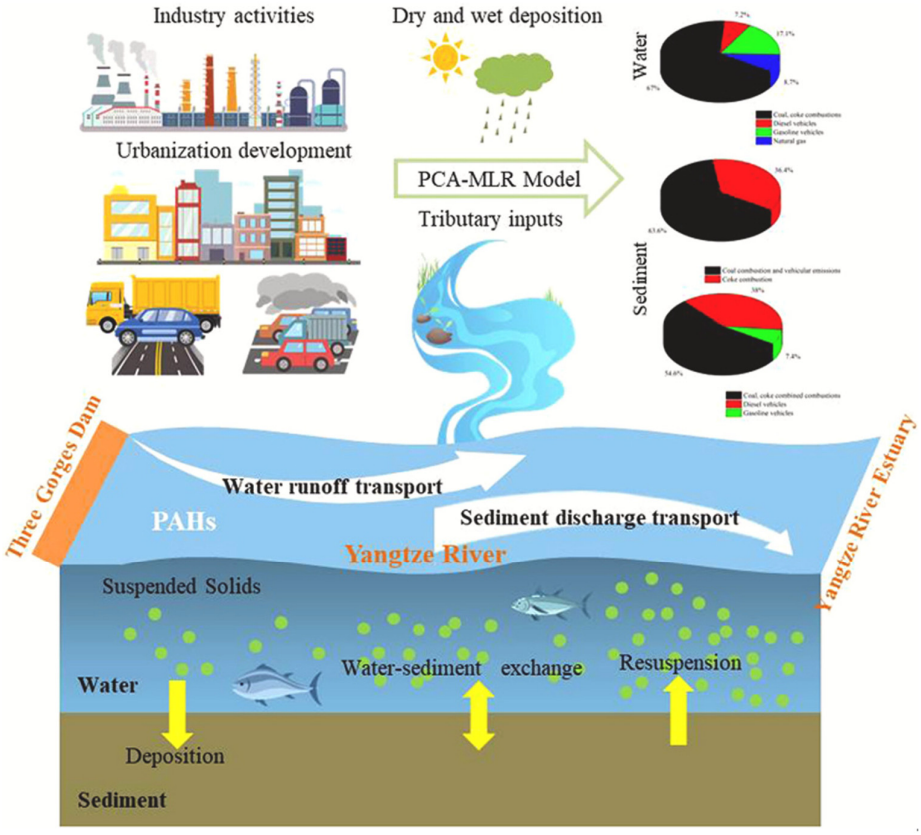

Figure 5. Megacities water runoff from coal and coke combustions sources contributes to transport and increase in catchment retention of PAHs along Yangtze River, China. Reprinted with permission from ref. [113], Copyright 2021, Elsevier.

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2.3. Streams

While most studies have traditionally focused on examining PAHs in large and publicly significant rivers, there is a growing interest in assessing these pollutants in smaller streams to understand primary sources and their relative contribution [94]. Urban streams offer a comprehensive insight into proportion of sources, were in the case of an eco-industrial park in China sources identified were coal/biomass combustion (61%), followed by non-combustion sources (21%), and vehicular emissions (18%), being residential and commercial playing a minimal role [117]. Surface streambed sediment of 10 urban watershed in three regions of USA, suggest that pavement areas drive occurrence of PAHs in urban streams [118]. While bottom sediments in two urban streams of Poland, indicated pyrogenic origins from biomass, coal, and petroleum combustion [119]. Although PAHs can have mixed influence of petrogenic and pyrogenic origins in urban streams, pyrogenic is the dominant source in most of recent reports [120]. In the case of forested streams, post-wildfire runoff contribute substantially with pyrogenic PAHs content through various pathways (Figure 6) [47,121,122]. Although, streams presents mix of petrogenic and pyrogenic origin of PAHs, most of them are emitted in distant parts [123]. PAHs emissions are linked to diverse economic factors [113,124]. Overall, smaller streams highlight the major occurrence of pyrogenic sources of PAHs, with urban streams revealing coal/biomass combustion emissions and forested streams showing significant contributions from post-wildfire runoffs.

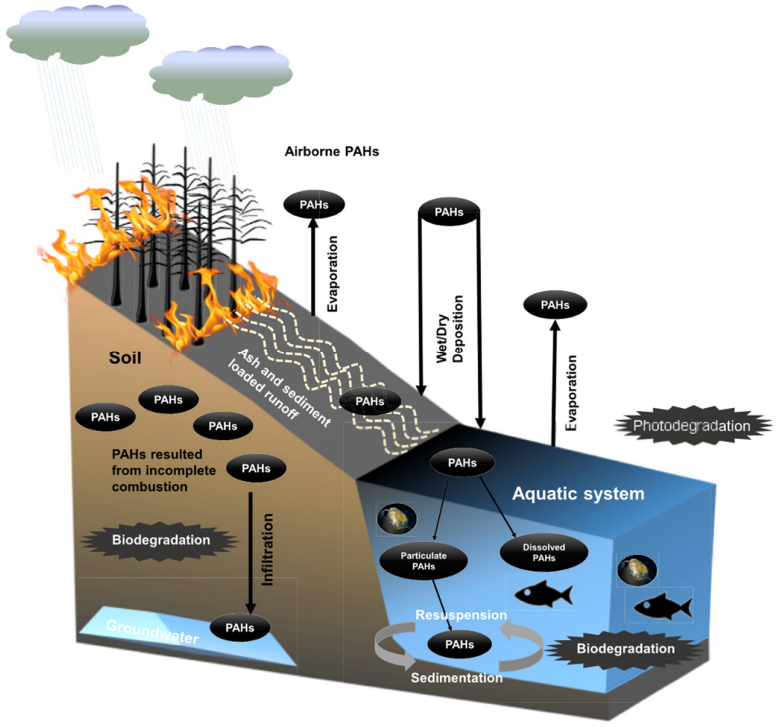

Figure 6. Transfer, distribution, and pathways of wildfire produced PAHs into aquatic systems. Reprinted with permission from ref. [47], Copyright 2021, Elsevier.

2.4. Groundwater

Groundwater serves as the primary freshwater source for domestic, agricultural, and industrial purposes, while being the source of drinking water for one third of the world's population [125]. Reports indicate PAHs are introduced by various pathways, increasing levels in groundwater systems [126]. Upon deposition in soil, they may leach through the soil profile or be transported by surface runoff impacting both surface and groundwater systems [68]. The input of PAHs from surface

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water to groundwater, occurs when a strong correlation and hydrological connectivity exists between them, allowing the former to influence the sources and distribution on the latter [78,127].

In the Yellow River Delta, China, strong correlation of surface water and groundwater indicated that pyrogenic PAHs from petroleum combustion were the primary sources [127]. While, groundwater assessment in the Abou Ali River System in North Lebanon, also indicated pyrogenic inputs from fuel combustion, incineration, and miscellaneous burning [128]. On the other hand, peri-urban forest wildfires significantly impact groundwater quality connected to public supply systems, contributing predominantly pyrogenic PAHs during post-wildfire events [129].

PAHs may migrate vertically from surface soils to groundwater systems [78], but their concentration may decrease with soil depth [130]. In the case of karstic topography, PAHs migrate much faster through conduits, infiltration, sinkholes, and fissures (Figure 7) [131]. While in large abandoned industrial complexes, can introduce mainly pyrogenic origin PAHs, with a minor contribution from petrogenic PAHs to groundwater aquifers through vertical migration [130].

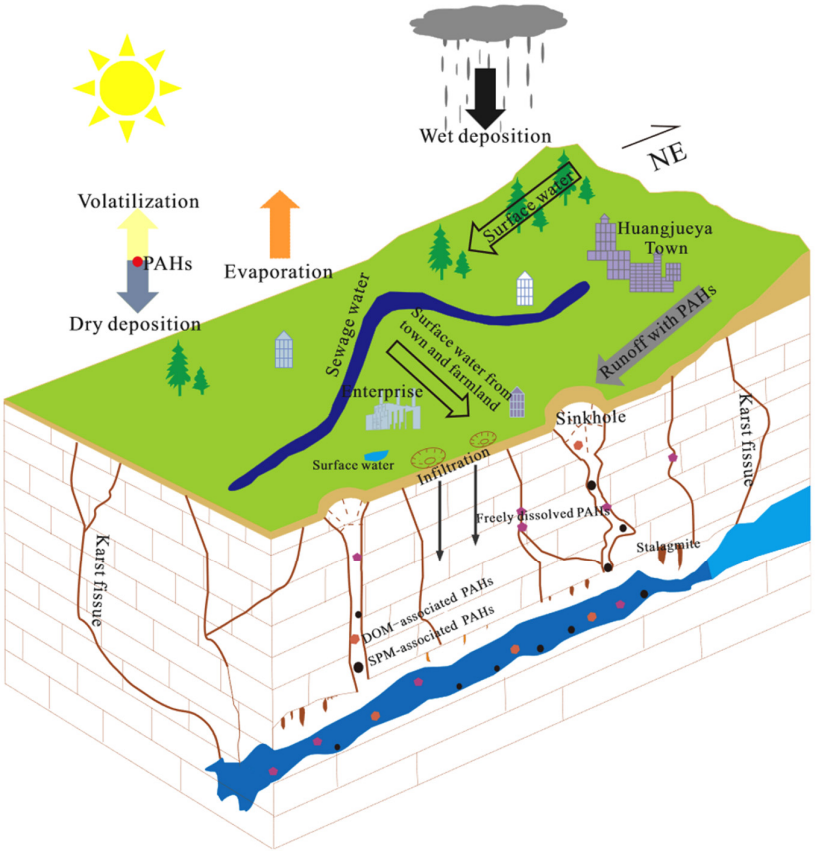

Figure 7. Transport model for PAHs in a karst underground river system. Reprinted with permission from ref. [131], Copyright 2018, Springer.

Despite the slow migration of PAHs over an extended period, emission sources evolved with the economic development of the region. In the case of karstic regions, rapid migration of PAHs to groundwater systems, exhibit a shift in emission sources from coal combustion to vehicular contributions over a 10-year period, while maintaining a pattern of pyrogenic origin [132]. Soil and spring water PAHs were derived mainly by grass, wood, and coal combustion in karstic springs of Southwest, China that are affected by total organic carbon and large water fluxes [78]. Additionally, groundwater samples of major cities in China, indicated pyrogenic combustion as the primary contributor PAHs [133]. Similarly, in the Campania Plain, Italy, the major source of pollution in groundwater was attributed to pyrogenic origins from burning carbon and fuel during illegal

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practices of burning abandoned solid waste piles [134]. While oil-producing areas with petroleum-related activities, compositional patterns of PAHs reflect a balance between major emissions of pyrogenic and petrogenic mixed sources associated with petroleum development activities in areas near to groundwater systems [135]. Although emission sources of PAHs depend on anthropogenic factors in the area where groundwater systems reside, pyrogenic sources present a significant contribution to the input of PAHs into groundwater systems.

2.5. Wetlands

Freshwater wetlands are some of the most biologically diverse, crucial, and productive ecosystems globally, contributing to biogeochemical cycles and functioning as mega-diversity regions [136–138]. Freshwater wetlands constitute about 5 to 8 percent of Earth's land surface [139] and provide essential ecosystem services [140]. They serve as transitional zones between terrestrial and aquatic ecosystems and are recognized for their irreplaceable ecological functions like water and soil conservation, water purification, climate regulation, and biodiversity protection [141,142]. However, industrialization and increasing human disturbances have led to wetland degradation and declining biodiversity, with oil exploitation posing a particularly detrimental and challenging issue [143,144]. Despite their ecological significance, research on PAHs in wetland's environmental media remains limited, although their presence spans in several wetlands throughout the world [138]. The vulnerability of wetlands to PAHs highlights the need for a comprehensive understanding of potential sources to these freshwater systems.

Freshwater wetlands are vulnerable to the presence of PAHs that originate from various pollution sources and impacts wetlands, based on their characteristics. Anzali Wetland is Iran's largest freshwater wetland and pollution is affected by nearby cities, wastewater discharge, tourism, and fishing activities [145,146]. Surface sediment in the western and central part of the lagoon revealed higher levels in coastal areas with a main attribution by oil-related activities dating back to the 1800s [147]. Although, recently a comprehensive study indicated petrogenic origin in water and pyrogenic origin of PAHs in sediments, attributed by the difference in distribution of LMW and HMW PAHs in these environmental matrices (Figure 8) [59]. Another study in Northeastern China wetlands, showed that PAHs originated mainly from coal burning and vehicle emissions, with high concentration in industrial areas [148]. Freshwater wetlands are vulnerable to diverse PAH origins that highlight complex emission sources impacting these ecosystems.

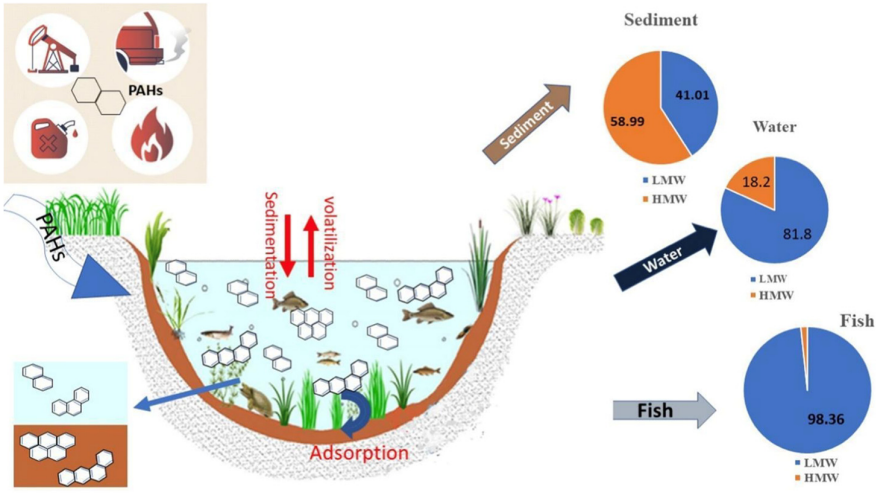

Figure 8. PAHs sources and distribution in environmental matrices of a freshwater wetland. Reprinted with permission from ref. [59], Copyright 2023, Elsevier.

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Constructed and urban wetlands are vital for water treatment, but they face a threat from the continuous discharge of effluents containing PAHs [143,149,150]. PAH assessments in constructed wetlands are focused on the study of pollutant bioremediation [151,152]. A study on a typical constructed wetland specify that PAHs mainly originate from a mix of fossil fuel combustion and petroleum leakage sources, although distribution between sediments and surface water is influenced by molecular weight (Figure 9) [153]. On the other hand, urban wetlands can have diverse sources of PAH pollution, including agricultural practices [154], wastewater discharge [59], and vehicular exhaust emissions [109]. Ashtamudi Wetland, India, was confirmed that petrogenic and pyrogenic origins of PAHs are related to the impact of urban activities. Reports from Shadegan Wetland, Iran revealed anthropogenic pollution sources were dominated by petrogenic origin from oil-related activities, such as oil spills near petrochemical complexes [138,156] and a lesser pyrogenic origin from incomplete combustion, bulrush combustion, vehicular exhaust, and fishing boat emissions [138]. In the case of Hoor-Al-Azim Wetland in Lower Mesopotamia faces challenges from PAHs derived from mixed sources, with a primarily pyrogenic contribution percentage of 62.62% that includes unburned and combusted fossil fuels from fishing boats and vehicle engines, fuel incomplete combustion, and oil leakage [140]. Wetland protection areas like Momoge Wetland, China, highlights the importance of promoting an effective management and conservation strategies for managing multiple petroleum sources of PAHs, generated by oil exploitation [157]. While other protected wetlands considered as Ramsar site importance, are degraded by a complex mix of multiple petrogenic and pyrogenic PAHs sources when they serve as the final receiver of river water [158]. The paradigm of PAH mixed sources freshwater urban wetlands near oil exploitation activity, can be easily affected by extraordinary events, like crude oil spills or discharge [159].

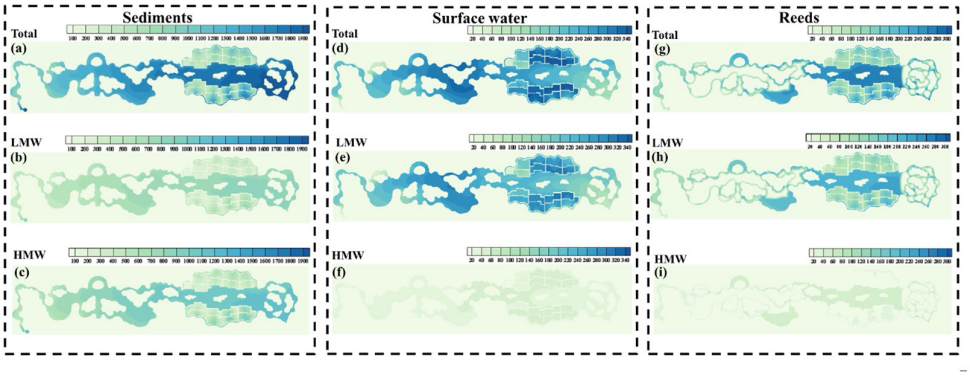

Figure 9. Distribution of Total, LMW, and HMW PAHs in sediments, surface water, and reeds in a constructed wetland that receives water from a wastewater treatment plant. Reprinted with permission from ref. [153], Copyright 2021, Elsevier.

Other types of freshwater wetlands are peatlands, marshes, and bogs. Peatlands can serve as environmental archives of the historical PAH pollution sources [160]. In the case of an ombrotrophic peatland in Great Hinggan Mountain, Northeast China, the main sources of PAHs were petrogenic and were caused by the oil extraction and refining process during the industrial development in the region after 1860 [160]. On the other hand, frequent wildfires in peatlands and bogs contribute to PAH accumulation [161] and eventually becoming the sole source of pyrogenic PAHs [162,163]. Typically oil exploitation activities, contributes mainly to petrogenic PAHs pollution on bogs, while pyrogenic PAHs originates from wildfire that occur sporadically over time [162].

In the case of freshwater marshes cover less than 20% of the world's total wetlands and play a crucial role in supplying essential ecosystem services [164,165]. In the case of Iraq's marshes, there is a concern to focus in petrogenic hydrocarbon pollution when Al-Hammar marshland was recognized with rich petroleum hydrocarbons [166]. Conversion of freshwater marshes to sugarcane cultivation in the Everglades Agricultural Area, Florida, USA, led to increased PAHs in soil layers used for

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sugarcane cultivation [167]. Particularly, illegal burnings associated with agriculture in the Pantanal, Brazil, along with anthropogenic factors, contributed to elevated levels of pyrogenic PAHs in the topsoil during the 2020 wildfires [168]. These diverse freshwater wetlands show distinct patterns for PAH pollution sources, with peatlands acting as historical pollution archives and increasing concerns over petrogenic PAH pollution in marshes and bogs due to the oil exploitation industry.

In summary, freshwater wetlands worldwide, from urban and constructed wetlands, peatlands, marshes, swamps, and bogs, face diverse sources of PAH pollution, including historical industrial activities, urbanization, and oil exploitation. The multifaceted nature of PAH origins is evident. These findings highlight the critical importance of adjusting management and conservation strategies to address the complex and varied sources of PAH pollution in freshwater wetlands globally.

2.6. Glaciers

Recent research reports sources of PAHs in various environments associated with glacial freshwater systems. Attribution ratios found that arctic freshwater environments indicated presence of atmospheric and combustion-derived sources of PAHs [68]. While samples of fresh meltwater from eastern Tibetan Glacier, showed that PAHs in this region are associated to incomplete coal combustion and coking that are deposited from long-range transport and local environmental settings [102]. Strong association of PAHs with soil organic matter in proglacial soils, affects retention in these environments [169]. Melting glaciers act as secondary sources of PAHs, leading to increased concentrations in sediment cores of proglacial lakes and glacial-fed streams, thereby establishing them as the most dominant persistent organic pollutants in the Arctic environment [170]. For instance, near the Kongsfjorden Glacier in Norway, PAH concentrations decrease with distance from the glacier, with local petrogenic combustion processes, long-range atmospheric transport, and historical coal mining identified as the primary sources [171]. Further, dissolved PAHs in the Kongsfjorden Glacier meltwater indicate grass, wood, and coal combustion as predominant sources, which are further influenced by ocean currents and glacier runoff [172].

Antarctica, the most remote continent on Earth, holds the largest reservoir of freshwater. Sources of PAHs had been studied to better understand dynamics in the Antarctic glaciers and their environment. Reports suggests that PAHs in Antarctic waters have a mix of from local consumed fuel combustion and global PAHs that arrived by long-range global transport (Figure 10) [173]. King George Island had PAHs accumulated in terrestrial soils, associated to electricity generators and light-duty gasoline used by scientific research and tourism [174]. Compared with other parts in the world, King George Island has significantly low terrestrial pollution of PAHs [175]. The reduction of fossil fuel consumption is recommended to mitigate PAH emissions in Arctic and Antarctic glacial freshwater environments.

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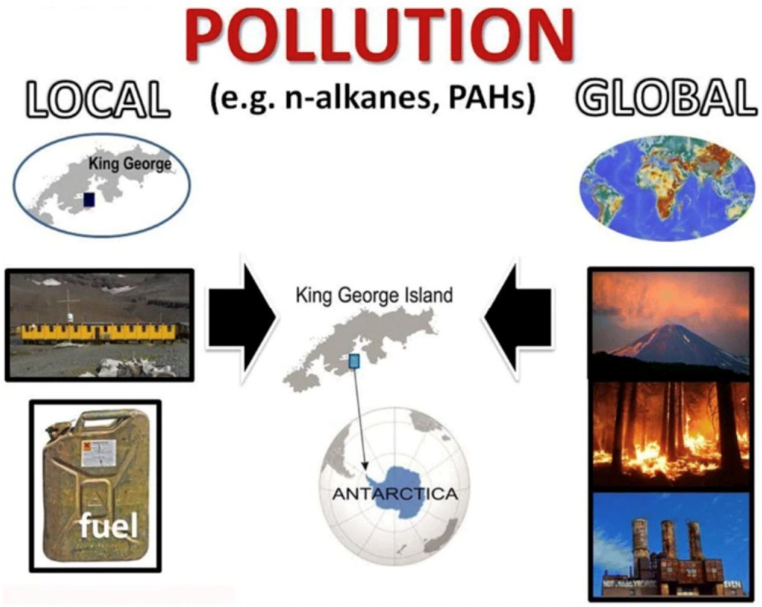

Figure 10. PAH origins in the western shore of Admiralty Bay (King George Island, Maritime Antarctica). Reprinted with permission from ref. [173], Copyright 2019, Springer.

Overall, the presence of PAHs in freshwater systems like lakes, rivers, streams, groundwater, wetlands, and glaciers are influenced by diverse emissions such as petroleum sources, biomass combustion, and urban activities (see Table 2). Lakes, particularly those in urban settings, face challenges from both point and non-point sources of PAHs, with atmospheric deposition and surface runoff playing significant roles. Rivers serve as primary pathways for PAH transport, reflecting mixed emission sources, while wetlands, groundwater, and glacial freshwater systems exhibit unique patterns of PAH contamination, emphasizing the need for comprehensive management, monitoring, research, and policy strategies.

Table 2. Origin and Sources of PAHs in Various Types of Freshwater Systems.

Freshwater system	Name/Location	Environmental matrix	Origin of PAHs	PAH source	Citation
lake	Tangxun and Dong Lake, Wuhan, China	water	mixed	Petroleum biomass heavy oils natural gas	[85]
lake	Soltaire and Fairbank, Central Ontario, Canada	sediment	pyrogenic	particulate deposition grass wood gasoline coal	[92]
lake	Shangai, China	sediment	pyrogenic	vehicle emission coal biomass natural gas combustion	[96]
lake	Lake Baikal, Siberia	aerosol	pyrogenic	wildfires	[98]
lake	Lake Baikal, Russia	water aerosol	pyrogenic	wildfires	[99]

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river	Jiulong River watershed and estuary, southeast China	water	pyrogenic	fossil fuel combustion petroleum combustion biomass combustion	[100]
river	Rur River catchment, North Rhine-West-phalia, Germany	water sediment	petrogenic	wastewater	[101]
river	Liujiang River Basin, China	sediment	pyrogenic	coal wood weed petroleum gasoline kerosene	[103]
river	Buffalo River Estuary, South Africa	water sediment	pyrogenic	crude oil highway runoff biomass combustion domestic wastes stormwater runoff	[104]
river	Nun River, Bayelsa State, Nigeria	water	petrogenic	oil-related activities	[105]
river	Ekulu, Enugu metropolis, Nigeria	water	mixed	agricultural waste burning vehicular emissions power generators	[176]
river	Euphrates River system, Iraq	water sediment	pyrogenic	petroleum combustion vehicular emissions fossil fuel burning illegal waste disposal wastewater discharge	[108]
river	Huaihe River, China	soil	pyrogenic	vehicular emissions biomass combustion coal combustion	[109]
river	River Benue, Nigeria	sediment	mixed	coal combustion crude oil wood burning vehicular emissions	[111]
river	Yangtze River, China	water sediment	pyrogenic	coal burning coke plants biomass vehicular emissions	[113]
river	River systems, Taiwan	sediment fish	pyrogenic	energy consumption petroleum grass and wood burning	[114]
river	Ulhas River, India	sediment	pyrogenic	coal wood burning coke plants coal	[116]
river	Pitimbu River, Natal, Brazil	sediment	pyrogenic	gasoline and diesel biomass coal petroleum	[120]

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river	Eastern Tibetean Plateau, China	water	pyrogenic	refined oil products coal charcoal coking discharge	[102]
river	Great Lakes tributaries, USA and Canada	sediment	petrogenic	coal-tar-sealed pavement	[94]
stream	Suzhou Industrial Park, China	sediment	pyrogenic	biomass coal petroleum	[117]
stream	Oliwski and Strzyza, Gdansk, Poland	sediment	pyrogenic	biomass coal petroleum	[119]
stream	North-central Portugal	water	pyrogenic	biomass wildfires	[121]
stream	Monastir Bay, Tunisia	sediment	mixed	road runoff domestic wastewater industrial wastewater workshops waste oil industrial activity	[123]
groundwater	Yellow River Estuary, China	water	pyrogenic	fossil fuels coal natural gas vehicular emissions	[127]
groundwater	Chongqing, Southwest, China	water soil	pyrogenic	grass wood coal	[78]
groundwater	Abou Ali River-North Lebanon	water	pyrogenic	fuel incineration miscellaneous burning	[128]
groundwater	Peri-Urban Forest Area, Braga Region, NW Portugal	water	pyrogenic	grass wood coal	[129]
groundwater	Banja Luka, Republic of Srpska, Bosnia and Herzegovina	soil	mixed	traffic emissions coal combustion biomass combustion	[130]
groundwater	Nanchuan catchment, Chongqing, Southwest, China	soil water	pyrogenic	coal combustion vehicular emissions	[132]
groundwater	Lanzhou, Shijiazhuang, and Golmud and Du'an County, China	water	mixed	oil pipeline leakage coal combustion wood and straw burning	[133]
groundwater	Campania Plain, South Italy	water	pyrogenic	carbon and fuel	[134]
groundwater	Eleme, Nigeria	water soil	mixed	petroleum extraction	[135]
wetland	Shadegan wetland, Iran	sediment water	mixed	bulrush burning biomass combustion heavy gasoline oils lubricating oils	[138]

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wetland	Anzali Wetland, Caspian Sea, Iran	sediment	petrogenic	used motor oils fuel oil light refined oil	[147]
wetland	Anzali Wetland, Caspian Sea, Iran	water sediment fish	mixed	crude oil combustion diesel fuel biomass coal and oil leakage	[59]
wetland	Coastal Industrial Zone, Tianjin, China	sediment water reed	mixed	fossil fuels combustion petroleum leakage	[153]
wetland	Ashtamudi Wetland, south-west coast, India	sediment	mixed	biomass burning fuel combustion oil leakage	[155]
wetland	Hoor Al-Azim Wetland, Iran	sediment	mixed	fossil fuel combustion oil leakage	[140]
wetland	Momoge Wetland, China	soils	mixed	petroleum combustion coal combustion petroleum spills	[157]
wetland	Miankaleh International Wetland, Iran	water sediment	mixed	fossil fuel sewage discharge port activity	[158]
wetland	Baghjan, upper Assam, India	water sediment fish	petrogenic	oil spill	[159]
peatland	Songkhla Lake Basin Swamp Forest, Thailand	air	pyrogenic	biomass and peat burning	[161]
bog	Southern Taiga of Western Siberia	water peat	pyrogenic	biomass and peat burning wildfires	[162]
marsh	Pantanal Biome, Brazil	soil	pyrogenic	biomass burning wildfires	[168]
glacier	Tibetan Plateau, China	meltwater	pyrogenic	coal incomplete combustion biomass combustion coking discharge	[102]
glacier	Kongsfjorden, Svalbard, Norway	water	pyrogenic	diesel fuel leakage heat and power generation dust from coal mining long-range transport	[171]
glacier	Kongsfjorden, Arctic	water	pyrogenic	grass wood coal	[172]
glacier	Admiralty Bay, King George Island, Maritime Antarctica	water	petrogenic	fuel consumption local human activity	[173]
glacier	King George Island, Antarctica	soil	petrogenic	electricity generators light-duty gasoline	[174]
glacier	Eastern Tibetean Plateau, China	water	pyrogenic	fuel consumption coal charcoal coking discharge	[102]

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3. Distribution of PAHs in Water Systems

3.1. Transport Mechanisms of PAHs in Water Bodies

The distribution and transport of PAHs in freshwater systems are dynamic processes influenced by various environmental factors. Several studies have shed light on these mechanisms, providing valuable insights into how PAHs move and persist in water bodies. Chen et al. [177] investigated the distribution of PAHs in karst spring systems in Central China. In karst regions, characterized by their unique geological structure of extensive fractures and conduits, groundwater is highly susceptible to pollution. This study focused on the infiltration of PAHs, primarily from human activities, into karst groundwater systems in Western Hubei. Through the analysis of various environmental media across ten karst springs, the research highlighted the predominance of low-molecular-weight PAHs and identified their primary sources as high-temperature combustion processes, including coal, biomass, and vehicular emissions. The findings underscore the swift transportation of PAHs within these systems, emphasizing the vulnerability of karst water resources to contamination. The study emphasized the rapid transport of PAHs from the recharge area soil to the discharge area of spring water and sediments, showcasing the dynamic movement of PAHs in karst environments [177]. In another study, Nikanorov et al. [178] documented the concentrations of polycyclic aromatic hydrocarbons and benzo[a]pyrene in water and sediment near the Baikal Pulp and Paper Mill, Lake Baikal, and the Selenga River delta. This research uncovered significant variations in benzo[a]pyrene levels, especially in sediments, indicating considerable pollution around the mill [178]. Conversely, concentrations in other areas generally did not surpass natural background levels. In a historical context, the study by Maier et al. [179] delved into the effects of coal-tar pipe linings on PAHs in drinking water, linking PAH presence to disinfectants like chlorine. Laboratory findings suggested that coal-tar surfaces encourage biofilm development, which may mitigate PAH release. They identified key factors promoting PAH presence, including disinfection practices, water stagnation, and anaerobic conditions, with mechanical disturbances significantly elevating PAH concentrations through biofilm disruption or the dislodgement of PAH-contaminated particles. Studies have shown that PAHs are transported from superficial water to sediment, supporting the idea that both compartments play a crucial role in the fate of PAHs in water bodies. The presence of individual PAHs in both water and sediment, along with their petrogenic or pyrolytic nature, further underscores the complexity of their transport mechanisms [180]. It has been observed that low molecular weight PAHs (2-3 rings) migrate faster in soil profiles than high molecular weight PAHs and are primarily transported in dissolved matter, indicating the significance of molecular weight in their environmental mobility [181]. In another research, Nganje et al. [182] investigated PAH distribution in contaminated soils and found that PAH concentrations decreased with depth from the spill site. The study found PAHs of both petrogenic and pyrogenic origins, and proposed the main mechanism of PAH transport in the area, emphasizing the significance of soil properties in PAH mobility [182]. Li et al. [183] study the spatial distribution of PAHs in Taihu Lake. They found that rivers flowing into Zhushan and Meiliang Bays were major contributors to PAH and sediment in the lake, with concentrations up to 5000 ng/g near river outlets, demonstrating the importance of riverine transport in PAH distribution [183]. This study also explored the origins of perylene in the lake, identifying both anthropogenic and biogenic sources. Near river outlets, perylene's presence was largely anthropogenic, correlated with specific PAHs. Conversely, in areas further from the outlets, perylene was primarily biogenic, indicating that human activities might suppress its natural formation [183]. Gregg et al. [184] revealed that PAHs in the Columbia River estuary are present in suspended particulate material and are concentrated in the estuarine turbidity maximum (ETM) due to a particle-selective hydrodynamic trapping process. This ETM, a sedimentary feature at the river-sea interface, showed PAH concentrations, including environmentally concerning ones like fluoranthene, chrysene, and benzo[a]pyrene, surpassing EPA's threshold levels, indicating higher pollution levels than many heavily industrialized areas [184]. This study highlighted the role of hydrodynamics in PAH transport. Gold-Bouchot et al. [185] investigated PAH distribution in the Perdido Fold Belt area of the Gulf of Mexico, a region influenced significantly by freshwater inputs. Their findings indicated the highest PAH concentrations during a survey conducted in September

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2017, which were attributed primarily to riverine contributions. This underscores the seasonal and spatial variability of PAH levels in freshwater influenced marine environments. The initial survey pointed to pyrogenic (combustion-derived) hydrocarbons, while later surveys highlighted petrogenic (oil-derived) sources [185]. The spatial distribution patterns showed higher PAH concentrations in areas strongly affected by riverine discharges and the complex hydrodynamics of the region. These dynamics were crucial in driving the seasonal fluctuations of PAH levels and their dispersal, which were further influenced by eddy circulation and frontal systems [185]. This study emphasizes the interconnectedness of riverine and marine systems in the dispersion and concentration of PAHs, highlighting the need for integrated management strategies to address pollution in such ecologically sensitive areas. In another study on soil and sediment PAHs in Hanfeng Lake, China, Cai et al. [186] found that water dynamics and the physicochemical properties of PAHs influence their distribution in soils and sediments. The study highlighted the dominance of two- and three-ring PAHs, particularly phenanthrene, in the samples. The spatial distribution indicated that PAH transfer and fate were affected by water dynamics and the PAHs physicochemical properties. Analyses revealed over 75% of PAHs originated from coal combustion and vehicle emissions [186]. In a very recent research, the adoption of innovative methods, specifically thin-layer capping with activated biochar, was explored to address the challenge of mitigating the migration of PAHs from sediments to aquatic systems [187]. This approach, while effective in reducing PAH flux, has been identified to potentially facilitate the mobilization of arsenic (As) under oxygen-rich conditions, thereby underscoring the critical need for strategies that carefully consider the dynamics of multiple pollutants. The study, which used sediment samples from Bureå bay in Sweden, demonstrated that activated biochar effectively decreased PAH diffusion but inadvertently increased As mobility [187]. This finding highlights the importance of a balanced assessment in the application of biochar capping techniques, especially in sediments contaminated with various pollutants, to prevent the exacerbation of contaminant dispersion, particularly with arsenic. In a different context, shifting focus from aquatic bodies to snow, the transport of PAHs within snow cover was explored by Khaustov and Redina [188] at the campus of RUDN University, located in a relatively pristine sector of Moscow's southwest. Despite the general cleanliness of the area, it was found that atmospheric pollutants, particularly PAHs, were significantly deposited in this region, primarily due to vehicular emissions from major roadways and other anthropogenic activities. This study revealed the presence of primary pollutants, including fluoranthene, pyrene, phenanthrene, and benzo[b]fluoranthene, distinct from those identified in prior reports, thereby challenging conventional perceptions of PAH distribution in different mediums such as soil and snow [188]. This discrepancy highlighted the nuances in PAH migration and accumulation, potentially tied to phase-structural shifts within the "snow-water" system during freeze-thaw cycles, and underscored the distinct nature of pyrogenic pollution [188].

In summary, the transport mechanisms of PAHs in water bodies are influenced by a myriad of factors, including their molecular weight, interaction with sediments and organic matter, and environmental processes such as runoff and erosion. The interplay of these factors determines the mobility, distribution, and ultimately the environmental impact of PAHs in aquatic systems. Understanding these mechanisms is crucial for developing effective strategies for mitigating the presence of PAHs and other similar pollutants in water bodies, thereby protecting aquatic ecosystems and human health. Further research in this area will continue to uncover the complexities of PAH transport and provide insights into more effective pollution control and remediation strategies.

3.2. Factors Affecting the Distribution

The distribution of PAHs in freshwater systems is a topic of considerable environmental significance, given the persistence, bioaccumulative potential, and carcinogenic properties of these compounds. PAHs enter freshwater ecosystems through various pathways, including atmospheric deposition, industrial discharges, urban runoff, and natural seeps, and their distribution within these systems is influenced by a complex interplay of physical, chemical, biological, and ecological factors.

The physicochemical properties of PAH molecules themselves are primary determinants of their environmental behavior. Their hydrophobic nature leads to low solubility in water and a strong

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affinity for organic matter, significantly influencing their distribution between water, sediments, and biota in aquatic environments [189]. The molecular weight of PAH compounds is a critical factor; low molecular weight PAHs (with two or three aromatic rings) tend to be more soluble in water and more volatile, whereas PAHs (with four or more rings) are more likely to adsorb to particulate matter and accumulate in sediments [190]. In a study conducted in China several years ago, *Cui* et al. [191] investigated the distribution and contamination levels of PAHs on atmospheric particulates in Jinan. They pinpointed emission intensity and meteorological conditions as critical determinants influencing the distribution of PAHs, and advocated for a numerical model to predict and mitigate PAH air pollution in the area. Also in China, Ou et al. [192] discovered that the seasonal fluctuations in concentrations of PAHs in freshwater ecosystems were modulated by temperature in the overlying waters and by the presence of organic carbon and soot carbon in surface sediments. Their research assessed the pollution levels, along with the temporal and spatial variations of PAHs in aquatic environments, identifying an elevation in PAH concentrations during the dry season. Among the PAHs analyzed, phenanthrene emerged as the predominant compound. The investigation elucidated that while temperature was a key driver for the seasonal variability of PAH concentrations in water, organic and soot carbon significantly influenced the accumulation of PAHs in sediments [192]. Furthermore, the study underscored the role of hydrodynamic conditions and anthropogenic influences in shaping the distribution and sources of PAHs, thereby indicating potential ecological threats to the Yangtze ecosystem. Notably, the concentrations of certain PAHs surpassed ecological and water quality benchmarks, underscoring substantial environmental issues [192]. In a relatively dated publication, Van Hattum et al. [193] observed that hydrodynamic forces and seasonal fluctuations significantly influenced the distribution of PAHs in freshwater ecosystems. In their study, they explored the bioaccumulation of 13 PAHs in freshwater isopods, assessing their distribution across sediments, particulate matter, and aqueous phases within eight different water systems in The Netherlands. They reported marked disparities in the concentrations of PAHs and the presence of individual PAHs among various environmental compartments and geographic locations [193]. Their analysis identified pronounced differences in the PAH profiles between sediments, predominantly characterized by HMW PAHs, and water, where naphthalene was more prevalent. Isopods were found to mainly bioaccumulate fluoranthene and pyrene. The research highlighted that the bioconcentration factors (BCFs) for PAHs in isopods increased with the compounds' hydrophobicity, although lower than expected BCFs were noted for PAHs with a log K_{ow} greater than 6.1, attributed to reduced bioavailability and the impact of growth dilution on accumulation. The study also found that abiotic partitioning coefficients, such as the organic carbon normalized sediment–water partition coefficient (K_{oc}) and the particulate matter–water distribution coefficient (K_{pm}), escalated with the hydrophobicity of PAHs, especially for those with a log K_{ow} less than 6.1, and correlated with the organic carbon content in sediments. The results underscored the significant capacity of isopods to bioaccumulate PAHs, with their tissue concentrations reflecting the spatial and temporal variations in PAH availability in littoral freshwater habitats [193]. In their recent investigation, Soukarieh et al. [194] assessed the contamination levels of 17 PAHs across marine and continental water bodies in Lebanon, finding a significantly higher level of PAH pollution in Lebanese seawater compared to other Mediterranean regions. The research recorded PAH concentrations in the marine domain ranging from 55.7 to 2683.8 ng/L in water and from 19.09 to 2025.03 ng/g in sediments. In contrast, continental aquatic systems presented elevated levels of contamination, with water concentrations spanning from 465.7 to 1399.9 ng/L and sediment concentrations from 72.6 to 1074.7 ng/g. It was determined that PAHs possessing greater numbers of rings and higher molecular weights predominantly settled in sediments, while those with increased solubility and vapor pressure were more likely to remain in the water phase. The study traced the sources of PAHs in Lebanese sediments primarily to combustion processes, notably from industrial activities adjacent to aquatic environments and dense traffic along the coastline. The research suggested that, although certain sites might occasionally pose risks to benthic life, the overarching ecological threat posed by PAHs in Lebanese sediments was deemed low [194]. Huang et al. [195] conducted a study on parent and alkylated polycyclic aromatic hydrocarbons (PAHs and A-PAHs)

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in sediments from six mangrove wetlands across the Taiwan Strait, China, aiming to pinpoint their origins and evaluate the ecological risks posed. The research uncovered significant variations in total PAH concentrations, with a marked underestimation observed when analysis was restricted to only 16 priority PAHs. It identified coal and petroleum combustion as the main sources of PAHs, with the significance of each source differing among the wetland sites and various factors influencing their distribution. Moreover, the research stressed the critical need for creating ecological risk assessment guidelines for individual A-PAHs, due to the absence of specific criteria for these substances [195]. In a recent investigation, Cai et al. [196] conducted an analysis on the occurrence, sources, and pollution levels of PAHs within the surface soil of the Yellow River Delta (YRD) wetland, China. The results indicated PAH concentrations ranging from 14.1 to 47.1 ng/g, with a mean value of 28.3 ng/g, which implies a relatively low level of contamination. The research attributed the predominant sources of PAHs to oil spills, vehicular emissions, and the combustion of coal and biomass. Moreover, the study revealed that the geographical distribution of PAHs was significantly affected by factors such as vegetation type, soil pH, organic matter content, water content, and soil texture [196]. In another study conducted in the Wang Lake Wetland region of China, Shi et al. [197] investigated how various factors, including dynamic water levels and human activities, impact the distribution of PAHs. Sampling across different locations under varying water flow conditions revealed a significant increase in PAHs associated with suspended particulate matter (SPM) following the onset of the rainy season, while the levels of dissolved PAHs remained constant. The analysis indicated that petrogenic sources were primarily responsible for the PAHs, even though pyrogenic sources were also present. The study further emphasized the challenges in evaluating PAH partitioning, noting discrepancies between theoretical and actual partition coefficients, particularly during periods of high-water flow which could lead to the release of higher molecular weight PAHs from sediments [197]. It was determined that human activities were the predominant source of PAHs during low water flow, while during high-flow conditions, factors such as increased rainfall, temperature rise, and fishery activities had a significant impact on PAH concentrations in the wetland [197]. Reback and Martins [198] conducted the initial evaluation of PAHs within the mangrove sediments of the Paranaguá Estuarine System (PES) in South Brazil, focusing particularly on areas influenced by freshwater inputs. They reported concentrations of 16 EPA-listed priority PAHs peaking at 234.3 ng/g, noting these levels were higher than those previously reported in purely freshwater sediment studies [198]. The observed concentrations were comparable to other estuaries affected by human activities but were less than those found in heavily polluted marine environments. The moderate level of anthropogenic contamination was mainly attributed to local shipping activities and domestic sewage that impact the freshwater sections of the estuary. Through the examination of bulk parameters such as the TOC/TN ratio and δ13C values, the researchers delineated the transitional areas within the estuary from freshwater to brackish environments [198]. They pinpointed the primary sites of PAH accumulation, linking them to the presence of organic matter and fine sediments in freshwater regions. The study further indicated that factors such as hydrodynamics and dilution processes predominantly influenced the spatial distribution of PAHs, with higher concentrations found in the estuary's sheltered freshwater regions and progressively lower concentrations as the waters mixed with the more turbulent saline conditions of the outer estuary [198]. This focus on freshwater influences provides crucial insights into how PAHs disperse and accumulate in estuarine systems that begin as freshwater environments.

The studies previously mentioned underscore the complex array of factors that determine the distribution of PAHs in freshwater ecosystems. Hydrodynamic conditions, temperature variations, the organic content of sediments, human-induced pollution, and seasonal variations are among the critical elements that significantly impact PAH pollution levels and their distribution within water bodies. Each of these factors contributes to the overall pattern of PAH contamination, affecting how these hazardous compounds are dispersed and concentrated in aquatic environments. Comprehending the intricate relationships among these variables is vital for the development of efficient strategies aimed at managing and reducing PAH pollution in freshwater systems. This understanding facilitates the identification of primary sources of PAHs, helps predict their behavior

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under different environmental conditions, and enables the implementation of targeted remediation efforts to safeguard aquatic life and water quality.

3.3. Interaction with Environmental Matrices

The interaction of PAHs with environmental matrices, such as sediments and biota, is a critical area of research due to the widespread presence of these compounds and their potential ecological and health impacts. In this context, Kieta et al. [199] conducted a study on the effects of the 2018 Shovel Lake wildfire on the origins of sediment in downstream water bodies in British Columbia, employing both PAHs and color metrics as indicators. From 2018 to 2021, sediment collections were made from three tributaries impacted by the fire and from two locations along the Nechako River. The analysis included comparisons of sediments from areas affected by the fire and those that were not, as well as sediments from riverbanks and areas adjacent to roads, looking at both PAH concentrations and color characteristics. Findings indicated that sediments from burned areas substantially affected the tributaries, in contrast to the Nechako River, where sediment primarily originated from riverbank erosion. The study found that color indicators were more effective than PAHs in tracing sediment sources, indicating that wildfires have a pronounced effect on sediment contribution in smaller waterways, an effect that becomes less apparent in larger river systems. In a separate study, Stephansen et al. [200] investigated sediment and invertebrate samples from nine stormwater retention ponds (SWRPs) and eleven natural shallow lakes in Denmark for the presence of PAHs. The research revealed that the sediment of both SWRPs and lakes contained comparable amounts of total PAHs, although SWRPs were characterized by a higher concentration of heavier PAH compounds. Similarly, invertebrates from both types of aquatic environments displayed analogous levels of PAHs. Through principal component analysis, it was determined that SWRPs receiving runoff from highways shared similarities with natural lakes situated in areas of forest and farmland, as well as with some ponds in residential zones. The findings indicated that SWRPs, particularly those handling highway runoff, featured sediment PAH levels similar to those found in natural lakes, suggesting no increased risk of PAH bioaccumulation in invertebrate communities within these ponds compared to lakes in the same region. In a significant investigation, Jesús et al. [201] explored the distribution of PAHs in freshwater ecosystems and their effects on benthic fauna. The study highlighted the tendency of PAHs to accumulate in sediments due to their hydrophobic nature, presenting a threat to the benthic species residing in these vital habitats. The research revealed that PAH concentrations were substantially higher in sediment layers than in the water column, with a strong link to the hydrophobic properties of the PAHs. Through the use of Species Sensitivity Distributions for toxicity assessment, the study uncovered a broad spectrum of hazardous concentration levels for 5% of species (HC5) associated with various PAHs, which showed an inverse relationship with their hydrophobicity [201]. Within this framework, Raudonytė-Svirbutavičienė et al. [202] undertook an extensive investigation into the pollution of water bodies by heavy metals and PAHs, examining their spatial distribution, sources, and ecological impacts. Chemical assessments confirmed persistent long-term pollution in select areas along various rivers in Lithuania. Lake Talkša, situated in an urbanized zone and subject to direct human-induced stress, exhibited the most significant concentrations of both heavy metals and PAHs. The research further identified vehicular and industrial activities as the predominant sources of these contaminants. Genotoxic irregularities were found to be more prevalent in riverine environments than in lacustrine settings, with the Nemunas River downstream of Alytus and Kaunas presenting the highest levels of genotoxic risk. Interestingly, despite its heavy contamination, Lake Talkša was associated with only moderate genotoxic risks, suggesting an adaptive response by the biota in stagnant water bodies to sustained pollution exposure [202]. In a study focused on lake littoral zones [203], researchers utilized the native clam *Corbicula fluminea* to investigate the impact of pollution sources on ecological risk by analyzing the bioavailability of six trace metals and fourteen PAHs. They discovered substantial spatial variation in contaminant levels across the lake's littoral zone, both in sediments and in clams, indicating localized ecological risks. The study found a strong correlation between the clams' PAH levels and sediment contamination, suggesting sediment as the primary source of PAHs for benthic

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organisms. Additionally, trace metal concentrations in the clams were linked to stormwater runoff, highlighting its role in littoral zone contamination [203]. The research emphasized the difference in bioconcentration of various contaminants within *C. fluminea*, demonstrating the varying bioavailability of trace metals and PAHs. In a recent study, Skic et al. [204] delved into the sorption dynamics of PAHs within bottom sediments. The investigation revealed that PAHs, prone to sediment accumulation due to their hydrophobic nature, were majorly composed of higher-molecular-weight compounds, constituting 73% of the total PAH concentration. The team uncovered a sorption mechanism driven by steric effects, significantly influenced by negatively dissociating structures such as carboxylic and phenolic groups. Specifically, PAH molecules with 5- and 6-rings showed a tendency for size-exclusion effects, with sequestration primarily occurring in pores smaller than 5 μm. The findings also highlighted a decrease in PAH bioavailability in sediment rich in organic matter, evidenced by lowered PAH bioaccumulation factors correlating with increased total organic carbon (TOC) levels. Additionally, the research noted that higher and medium TOC concentrations in samples were associated with greater mortality and growth inhibition of the ostracod *Heterocypris incongruens*, underscoring the role of organic matter in modulating PAH toxicity [204]. In the same vein, addressing the detrimental impacts of PAHs on aquatic life, Rezan Rasheed [205] undertook an investigation into the seasonal fluctuations of PAH concentrations in the muscle tissue of catfish (*Silurus triostegus*) within the Derbendikhan Reservoir, ranking as the second-largest reservoir in the Iraqi Kurdistan region. This study identified a surge in PAH levels during the spring season, a phenomenon attributed to the use of outdated fishing methods and the seasonal variation in water temperatures. The detected PAH concentrations in the fish muscle ranged from 0.098 to 1.271 μg/g, which surpasses the World Health Organization's (WHO) acceptable limit of 0.001 μg/g, indicating a pronounced bioaccumulation and biomagnification of these compounds. As previously demonstrated, PAHs interact with the aquatic matrix in which they are found, including sediments and biota, leading to significant pollution issues in both the environment and aquatic life. In this context, the study conducted by Mdaini et al. [206] on the Tunis Lagoon, an internationally significant coastal wetland, serves as another example. The research measured PAH concentrations in the bodies and excrements of *Marphysa sanguinea* (an aquatic polychaete worm used as a pollution indicator) and in surface sediments, finding total mean concentrations reaching up to 2398 ng/g dry weight (DW) in sediments, 1007.19 ng/g DW in *M. sanguinea*, and 2602.05 ng/g DW in excrements. The study determined that the origins of these PAHs were predominantly pyrogenic. Principal component analysis revealed a clear distinction between PAHs in polychaetes and those in sediments and excrements, implying that sediments may not be the main source of bioaccumulation in *M. sanguinea*. Moreover, PAHs associated with sediments were found to pose a moderate to high toxicity risk to benthic organisms [206].

These studies collectively highlight the complex interactions of PAHs with different environmental matrices. Factors such as fire events, sediment composition, treatment processes, and ecotoxicological risks play crucial roles in determining the fate and impact of PAHs in various ecosystems. Understanding these interactions is essential for assessing ecological risks and developing effective strategies for managing PAH pollution.

4. Ecotoxicological Impacts of PAHs

4.1. Molecular and Cellular Level Effects

PAHs, recognized as pervasive environmental pollutants, have garnered extensive scrutiny for their interactions with cellular components and the consequent induction of oxidative stress. This interaction unfolds through the generation of reactive oxygen species (ROS), either directly by PAHs or indirectly via their metabolic intermediates [207]. ROS, comprising chemically reactive molecules containing oxygen, can inflict considerable damage on cellular structures [208]. Upon cellular entry, PAHs undergo metabolic activation catalyzed by cytochrome P450 enzymes, leading to the formation of PAH quinones, redox-active metabolites capable of redox cycling [209]. This process significantly contributes to the generation of ROS, including superoxide anions (O_2^-), hydrogen peroxide (H_2O_2), and hydroxyl radicals ($^{\cdot}OH$), overwhelming the cellular antioxidant defense mechanisms, such as

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glutathione (GSH), superoxide dismutase (SOD), and catalase (CAT), and resulting in oxidative stress [210]. The oxidative stress induced by PAHs can cause extensive damage to critical biomolecules. Lipids, particularly polyunsaturated fatty acids in cellular membranes, are susceptible to peroxidation, leading to membrane integrity disruption [211]. Proteins can undergo oxidation of side chains, backbone cleavage, and formation of protein-protein cross-links, impairing their function and turnover [212]. The cellular damage inflicted by oxidative stress can disrupt normal cellular functions and signal transduction pathways, potentially leading to a cascade of events culminating in cell death [210,212]. Cells may undergo apoptosis, a form of programmed cell death characterized by specific morphological and biochemical features, as a response to mild to moderate oxidative damage [213]. Alternatively, severe oxidative damage may result in necrosis, a form of uncontrolled cell death associated with inflammation and further tissue damage [213].

Additionally, PAHs can cause DNA damage and subsequent mutagenesis. This concern stems primarily from the capacity of PAH metabolites to interact with DNA, forming PAH-DNA adducts [214]. These adducts pose a critical challenge to genomic integrity, as they can induce mutations during the process of DNA replication if they are not accurately repaired by the cell's intrinsic repair mechanisms [214]. Historically, the formation of PAH-DNA adducts has been recognized as a pivotal event in the initiation of carcinogenesis. The mutagenic potential of these adducts, characterized by their ability to cause errors in the DNA replication process, has been linked to the onset of uncontrolled cellular proliferation and the development of cancerous tumors [215]. The mechanistic pathway involves the metabolic activation of PAHs by cytochrome P450 enzymes to form reactive metabolites capable of covalently binding to DNA. This binding can disrupt the normal base-pairing and template functions of DNA, leading to mutations that, if propagated, can contribute to carcinogenesis [215]. Among the most ubiquitous PAH compounds, benzo[a]pyrene has been extensively investigated for its potent mutagenic and carcinogenic properties. This PAH compound serves as a model for studying the molecular mechanisms underpinning the genotoxic effects of PAH exposure. Benzo[a]pyrene's metabolites, particularly its diol-epoxide derivatives, have been shown to form stable adducts with the nucleophilic sites in DNA, primarily with guanine bases [216]. These adducts are capable of inducing miscoding during DNA replication, thereby increasing the likelihood of mutation occurrence. Research spanning several decades has elucidated that the mutagenic and carcinogenic potentials of PAHs vary significantly among different compounds [217]. This variability is attributed to differences in their chemical structures and the efficiency with which they are metabolized to form DNA-reactive metabolites. Moreover, the cellular context, including the availability and effectiveness of DNA repair mechanisms, plays a crucial role in modulating the mutagenic outcomes of PAH exposure.

PAHs have also been implicated in endocrine disruption, with the potential to mimic or interfere with hormone activities, leading to significant alterations in hormone signaling pathways [218]. This interference primarily manifested in aquatic organisms, where it precipitated a range of developmental and reproductive abnormalities. Notable among these were skewed sex ratios, diminished fertility rates, and the occurrence of developmental anomalies in offspring [219]. The underlying mechanism of endocrine disruption attributed to PAHs entailed a complex interaction with hormone receptors. By binding to or mimicking the structure of natural hormones, PAHs could competitively inhibit or falsely initiate hormone receptor signaling [220]. This aberrant signaling was capable of inducing profound alterations in the normal physiological processes governed by these hormones. Furthermore, PAHs exerted influence over hormone synthesis and metabolism, either by inducing or suppressing the enzymatic pathways involved in hormone production and degradation. These disruptions were not uniform across all PAH compounds [221]; rather, specific PAHs exhibited distinct affinities and mechanisms of action on various hormone receptors. Research into the endocrine-disruptive effects of PAHs underscored the compound-specific nature of these interactions. Investigations into the molecular dynamics of PAH-hormone receptor binding revealed that certain PAH molecules could preferentially bind to estrogen, androgen, or thyroid hormone receptors, among others, leading to differential effects on hormone-regulated biological functions. Such specificity suggested that the endocrine-disruptive potential of PAHs was mediated by their

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chemical structure, the presence of functional groups, and their metabolic biotransformation products [222]. The implications of these findings were particularly pronounced in studies of aquatic ecosystems, where PAH contamination has been linked to reproductive and developmental perturbations in wildlife. Observations of altered sex ratios reduced reproductive success, and developmental defects in fish and amphibian populations in PAH-contaminated environments provided empirical evidence of the endocrine-disruptive capabilities of PAHs. These effects were not confined to direct exposure scenarios but also extended to subsequent generations, indicating the potential for long-term ecological and evolutionary impacts [223].

4.2. Impact on Aquatic Organisms

The impact of PAHs on aquatic organisms is a subject of considerable environmental concern, given their toxicity, persistence, and bioaccumulative properties. The molecular and cellular disruptions caused by PAHs can have significant implications for aquatic organisms. These impacts range from sub-lethal effects, such as changes in behavior and physiology, to lethal outcomes.

Chronic exposure to PAHs, even at low concentrations, can lead to long-term health effects in aquatic organisms, impacting their survival, growth, and reproduction. Here's an in-depth exploration of the two key aspects of this impact: bioaccumulation, and mutagenic and carcinogenic effects.

4.2.1. Bioaccumulation

PAHs are hydrophobic and lipophilic, meaning they have a tendency to accumulate in the fatty tissues of organisms. This leads to bioaccumulation, where the concentration of PAHs in the organism becomes higher than in the surrounding environment. Net et al. [224] conducted a comprehensive analysis of PAHs in Dakar, Senegal, an area characterized by heavy industrial activity and a significant portion of the country's population. This investigation measured the contamination levels of PAHs in both sediments and various aquatic organisms, revealing a wide range of pollutant concentrations ranging from 2 to 636 μg/kg. Notably, higher concentrations of pollutants were found in sediments compared to aquatic organisms, with certain species showing significantly higher levels of specific pollutants. Mercury levels were also assessed, indicating a potential for biomagnification and bioaccumulation within the aquatic food chain. The primary source of PAH contamination was identified as pyrogenic, and an evaluation suggested minimal adverse biological effects. Furthermore, the quality of edible aquatic species met European Union standards for human consumption regarding PCBs, PAHs, and mercury levels. This research is a clear example of the differential accumulation of PAHs in various environmental matrices and organisms. The bioaccumulation factor (BAF) is an essential metric in these studies. It is a ratio that compares the concentration of a substance in an organism to the concentration in water. High BAF values for PAHs in aquatic organisms, especially in species at the bottom of the food chain, highlight the risk of biomagnification. Bioaccumulation patterns exhibit variation contingent upon the molecular weight of PAHs. Studies conducted across various global regions have consistently demonstrated that PAHs with higher molecular weights are predisposed to bioaccumulate to a greater extent compared to those with lower molecular weights [218]. This phenomenon has been substantiated through research findings that underscore the differential propensity for accumulation, which is significantly influenced by the molecular structure and size of the PAH molecules involved [218]. In another notable example, Lettoof et al. [225] carried out a comprehensive broad-scale analysis of contaminants in Australian wetlands, investigating 17 metals and trace elements, 21 organochlorine pesticides, and 14 PAHs in both sediments and the livers of western tiger snakes (*Notechis scutatus occidentalis*) from four wetlands in Perth, Western Australia. The study confirmed the presence of all targeted metals and trace elements in the examined sediments and snake livers, with some concentrations exceeding established limits [225]. While elevated levels of specific pesticides and hydrocarbons were identified in the sediments of one wetland, these contaminants were absent in the tiger snakes. Contaminant concentrations were higher in sediments and snakes from more

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urbanized wetlands, whereas agricultural runoff was implicated as a source of contamination in less urbanized locations [225].

4.2.2. Mutagenic and Carcinogenic Effects

Research into the carcinogenic effects of PAHs on both humans and mammals has identified eight PAHs as potential carcinogens, with benzo[a]pyrene (BaP) recognized for its significant oncogenic potential [226]. Investigations have connected BaP-related mutations to a majority of lung cancer cases and identified its role in cancers of the cervix, bladder, breast, and prostate [227].

In aquatic life, the study of PAHs has linked environmental pollution to increased cancer rates in species such as English sole and flounder, with juvenile sole showing a notably high binding of BaP to liver DNA [218]. This contrasts with Sprague Dawley rats, which are less susceptible to BaP-induced liver cancer. The research in fish has primarily focused on toxicological impacts, including those on early development and reproduction, with medaka and zebrafish being key models in OECD guidelines [218]. Additionally, the issue of PAHs adhering to microplastics in aquatic environments is highlighted, underscoring the broader concern of plastic pollution [228]. Teleost fish embryos are particularly vulnerable to PAHs during two critical stages of their development [218,229]. First, early on, PAHs can mess up the normal signaling needed for proper body orientation development, leading to embryos that are overly developed on one side and do not survive. The second critical stage is during heart development, where PAH exposure can severely affect the heart's function, endangering the fish's survival from a young age to adulthood [218]. This is because a healthy heart is crucial for delivering oxygen throughout the body and for supporting swimming. The Deepwater Horizon oil spill, which released a lot of PAHs, showed how these chemicals can cause heart problems in embryos by interfering with heart muscle cells and the heart's electrical signals [230]. Besides affecting the heart, PAHs from oil can also harm eye development and function, leading to visible abnormalities in larvae. Studies on Atlantic haddock embryos have shown that even low levels of PAHs can stick to the embryos and cause heart and developmental issues [231]. The Japanese medaka, a fish often used in research, helped scientists understand how certain PAHs (specifically, 3-hydroxybenzo[c]phenanthrene or 3-OHBcP) can be toxic, especially during early development stages [218]. Using a precise method called nanoinjection to study this, researchers found that 3-OHBcP exposure could speed up heart development abnormally and affect genes related to heart and eye health, muscle development, and the body's stress response. This shows that 3-OHBcP and similar PAHs can be harmful to the development of fish embryos [218].

Research has highlighted significant concerns about the effects of PAHs on both fish and invertebrates in aquatic environments. In fish, substances like BaP and 7,12-dimethylbenz[a]anthracene, found in pollutants such as cigarette smoke, have been linked to a reduction in bone strength and mass. Similar findings have been observed in mammals, where exposure to atmospheric PAHs correlates with decreased bone density, particularly in postmenopausal women. This suggests a broader impact of PAHs on bone health across different species. Furthermore, fish bone metabolism is disrupted by PAH exposure, evident in species like Pacific herring and medaka, emphasizing the need for more focused studies on fish bone health [218].

Teleost scales, a type of calcified tissue in fish, have been instrumental in studying the effects of PAHs on bone cells [232]. These scales act as calcium reservoirs, mirroring the function of bones in mammals, and have led to the development of assays that can measure the activity of bone-forming and bone-resorbing cells. These studies have shown that hormones and pollutants can significantly affect the health of fish bones, mirroring the effects seen in mammalian bone studies [232].

The liver, another critical organ affected by PAHs, shows significant bioaccumulation of these harmful substances [233]. In fish living in contaminated habitats, PAH levels have been directly linked to adverse effects on liver cells, including carcinogenic and endocrine-disruptive actions. This has been further evidenced by studies in fish, where exposure to certain PAHs altered liver function and disrupted lipid metabolism [233].

PAH exposure also poses a threat to reproductive health in fish, affecting gene expression in both the liver and gonads and disrupting important metabolic and hormonal pathways [234]. Some

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PAH metabolites, due to their structural similarity to estrogen, can bind to estrogen receptors, suggesting a potential for endocrine disruption beyond their direct toxic effects [218,234].

The issue of plastic pollution, particularly microplastics, has introduced a new vector for PAH toxicity in aquatic environments. Microplastics can carry PAHs, potentially introducing these harmful substances to aquatic organisms. While immediate toxicity varies, the presence of PAH-laden microplastics necessitates further research into their long-term effects on marine life [235].

Finally, toxicity studies on invertebrates have revealed varying sensitivities to PAHs, with some species showing significant adverse effects. Recent research on urchins has begun to uncover the impact of PAHs and their metabolites on embryonic development, indicating that PAHs and their conversion products are harmful not only to fish but also to invertebrates at early life stages [236]. This comprehensive look at the effects of PAHs across different organisms highlights the need for continued and diverse research to fully understand and mitigate the impact of these pollutants on aquatic ecosystems.

4.3. Case Studies of PAH Impact in Specific Water Systems

In exploring the impact of PAHs in specific water systems, several case studies offer valuable insights. A study by Lee et al. analyzed PAH contamination in whales, revealing evidence of maternal transfer of PAHs to the fetus, indicating in-utero exposure to PAH contamination [237]. Liver and muscle tissues from stranded individuals were analyzed. The findings revealed significant PAH concentrations, particularly C3-phenanthrenes/anthracenes, C4-dibenzothiophenes, and C4-phenanthrenes/anthracenes, indicating substantial contamination across the sampled tissues [237]. The analysis also differentiated the sources of PAHs affecting each killer whale group; SRKWs showed signs of petrogenic (oil-derived) contamination, while Bigg's killer whales had pyrogenic (fire-related) burdens. This variation likely stems from differences in their living areas, diet, and metabolic processes [237]. A particularly concerning discovery was the evidence of PAH transfer from mother to fetus, observed through a mother-fetus muscle sample pair. It was found that low molecular weight PAHs such as C3-fluorenes, dibenzothiophene, and naphthalene were efficiently transferred to the fetus, showcasing in-utero exposure to these toxic substances [237].

In another research, Zhang et al. [238] in Houshui and Yangpu Bays found that heavy metal pollution had a more significant effect on benthic foraminiferal assemblages than PAHs, highlighting the complex interactions between different pollutants in aquatic ecosystems. Sampling 30 different locations, they looked into the composition of these microorganism communities, alongside pollution levels. They found that most of the organic material in the area came from algae, and the PAHs present were mainly from oil and its combustion [238]. This study grouped 35 common foraminiferal species into two categories and similarly classified the sampling sites by their location. The investigation revealed that heavy metals had the most significant impact on these communities, followed by TOC and then PAHs. One group of species, which included both agglutinated and some resilient hyaline forms, thrived with higher heavy metal concentrations. Another group, mainly consisting of porcelaneous and some hyaline types, was more positively associated with TOC and PAH levels [238].

In relation to fish production and processing for consumption, Bomfeh et al. [239] conducted a recent study to enhance the fish smoking system, developing a method known as the FAO-Thiaroye Technique (FTT) in Ghana to examine its potential in reducing exposure to harmful chemicals, specifically PAHs, found in traditionally smoked fish. The study compared the traditional smoking methods, such as the Chorkor smoker and metal drum oven, to fish smoked using the FTT. Additionally, fish from local markets were tested for PAH levels using sophisticated analytical techniques. Surveys were conducted to gauge the consumption of smoked fish across different regions of Ghana. The research team utilized a targeted risk assessment approach, focusing on benzo[a]pyrene (BaP), to evaluate the health risks. The results revealed that traditional smoking techniques and market-sourced fish contained high BaP levels, presenting a considerable health hazard. Conversely, fish smoked using the FTT method demonstrated significantly lower risks, indicating it as a safer alternative [239].

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In a study conducted by Semenov et al. [240] on the composition, concentration, and origin of PAHs in the surface waters and bottom sediments of Lake Baikal and its tributaries, the concentrations of PAHs in water and sediments were analyzed using gas chromatography and analytical models for source identification. The findings revealed that PAH concentrations in water ranged from 5 to 200 ng/L and in sediments from 50 to 700 ng/g, with lighter PAHs prevailing in water and heavier ones in sediments due to fractionation in the soil-water system [240]. The main sources of PAHs were identified as biomass and fossil fuel combustion. Discrepancies with the data from previous studies were attributed to differences in suspended sediment concentrations, suggesting a need for uniform PAH measurement methods. Correlation analyses between partitioning coefficients indicated levels of ecosystem contamination, with specific patterns suggesting either sediment or water pollution. The study concluded that the distribution of PAHs is influenced by molecule size and particle distribution, confirming that biomass and fossil fuel combustion are the primary sources [240].

5. Characterization and Quantification of PAHs

5.1. Characterization Techniques

Sample preparation stands as a pivotal element within the analytical framework, its primary aim being the concentration of PAHs whilst concurrently eliminating any matrix interferences that could skew results. This step is instrumental in ensuring that subsequent analytical procedures yield accurate and reliable data, tailoring the method of preparation to align with the intrinsic characteristics of the sample matrix as well as the specific requisites of the analysis that follows. Among the most widely used preparation techniques, the following stand out: i) Solid-Phase Extraction (SPE) [241] emerges as a prevalent technique for the preparation of water samples. This method entails the passage of the sample through a cartridge or disk imbued with a solid adsorbent material. The operational principle of SPE hinges on the adsorbent's capacity to selectively retain PAHs, thereby allowing other constituents of the sample to pass through unimpeded. This selective retention facilitates the effective concentration of PAHs, rendering SPE an invaluable tool in the purification and preparation of samples for detailed analysis; ii) Solid-Phase Microextraction (SPME) [242] represents an innovative, solvent-free extraction methodology. This technique is predicated on the exposure of a fiber, which has been coated with an adsorbent material, to the sample. The adsorbent coating harbors the capacity to absorb PAHs directly from the sample, a process that is markedly efficient for both aqueous and gaseous samples. Given its solvent-free nature, SPME stands out for its environmental friendliness and operational efficiency, offering a sustainable alternative for sample preparation; iii) Soxhlet Extraction [243,244], a method steeped in tradition, is tailored for the extraction of PAHs from solid samples, such as soil and sediment. This technique involves the continuous extraction of the sample using a solvent under reflux conditions. The repeated washing of the sample with solvent ensures thorough extraction of PAHs, making Soxhlet Extraction a robust method for preparing solid samples for comprehensive PAH analysis; iv) Liquid-Liquid Extraction (LLE) is a commonly applied extraction technique; however, it is increasingly influenced by principles of "green" chemistry, which advocate for the reduction of hazardous substances in laboratory settings. A notable application of this principle is demonstrated in the work of Termedashev et al. [245] who utilized ultrasound-assisted dispersive liquid-liquid micro-extraction (DLLME). This approach employs a solvent mixture of chloroform-acetone and acetonitrile for the extraction of PAHs from aquatic environments such as Lake Karasun, the Azov Sea, and the Black Sea. The method reported extraction recoveries ranging from 88% to 103%, with relative standard deviations (RSD) spanning from 3.1% to 7.8%; v) Ionic Liquid-Based Dispersive Liquid-Liquid Microextraction (ILDLLME), developed by Peña et al. [246] This technique not only extracts but also concentrates the analyte in a single step, thus enhancing efficiency. Notably, this method avoids the use of chlorinated solvents, which are often employed but pose significant environmental and health risks. The ILDLLME technique has demonstrated extraction yields between 90.3% and 103.8%, with detection limits ranging from 0.1 ngL^{-1} to 7 ngL^{-1}, and a standard deviation from 1.2% to 5.7% [246]; and vi) Supercritical Fluid Extraction (SFE) [247] utilizes supercritical CO_2 as the extracting solvent,

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heralding a host of advantages in terms of efficiency and reduced environmental impact. The supercritical state of CO_2—characterized by fluid-like densities and gas-like diffusivities—facilitates the efficient penetration and extraction of PAHs from solid matrices. The environmental benignity of CO_2, coupled with the efficiency of the extraction process, positions SFE as a particularly effective method for the preparation of solid samples, aligning with the overarching goal of analytical chemistry to devise more sustainable and efficient analytical methodologies.

The comprehensive suite of analytical methodologies employed for the characterization of PAHs encompasses several advanced techniques that cater to various requirements based on the physicochemical properties of the analytes and the complexities of the environmental matrices involved. The primary methodologies include gas chromatography coupled with mass spectrometry (GC-MS) and high-performance liquid chromatography paired with ultraviolet detection (HPLC-UV). These foundational techniques are further augmented by HPLC interfaced with diode-array and fluorescence detection (HPLC-DAD-FLD) as well as HPLC linked with fluorescence and ultraviolet detection (HPLC-FLD-UV). Although these supplementary methods are utilized less frequently, they offer invaluable spectral data and heightened sensitivity for detecting certain PAH compounds under specific analytical conditions [248–251]. GC-MS is particularly noteworthy for its superior sensitivity, selectivity, and operational efficiency, which positions it as the method of choice for analyzing volatile and semi-volatile organic compounds, including PAHs. This technique effectively utilizes the capabilities of gas chromatography to separate complex mixtures based on the volatility and polarity of the constituent compounds. It is complemented by the robust detection capabilities of mass spectrometry, which provides critical molecular weight information that is essential for the definitive identification of compounds [249,250]. The synergy between these components ensures precise quantification and identification, making GC-MS indispensable in environmental analysis. However, the widespread implementation of GC-MS technology encounters obstacles, primarily due to its significant cost. The investment required for state-of-the-art GC-MS systems is considerable, posing a barrier to their adoption in resource-limited settings. This financial burden affects not only the initial acquisition but also the ongoing maintenance and operational costs. Moreover, these systems require highly trained personnel to perform analyses and interpret sophisticated data, further escalating the overall expense [252]. In contrast, HPLC-UV presents a more economically feasible option for analyzing non-volatile PAHs. This technique employs high-pressure pumps to drive liquid samples through a packed column, where separation of analytes occurs predominantly through hydrophobic interactions. Detection via ultraviolet light provides quantitative data based on the absorbance of specific wavelengths by the analytes. While generally less sensitive and selective than GC-MS, HPLC-UV is invaluable for its broad applicability to a wider array of substances, including those that are non-volatile or thermally labile. The analytical repertoire for PAH detection is further enhanced by HPLC-DAD-FLD and HPLC-FLD-UV technologies, which are particularly adept at identifying specific PAHs with high sensitivity owing to their unique fluorescence properties [253,254]. The diode-array detector (DAD) in HPLC-DAD allows for the simultaneous detection of multiple wavelengths, providing a comprehensive spectral fingerprint of the analytes. Similarly, fluorescence detection employed in HPLC-FLD and HPLC-FLD-UV techniques capitalizes on the native fluorescence of some PAHs or employs derivatization techniques to enhance detectability. These methods are especially beneficial in complex matrices, where increased specificity and sensitivity are crucial for accurate analysis [253,255]. Ultimately, the choice of an analytical method for PAH determination is influenced by several factors, including the nature of the sample, the complexity of the matrix, the specific PAHs of interest, and the available resources within the laboratory setting. Collectively, these methods provide a versatile and effective toolkit for the accurate and efficient detection and quantification of PAHs in a variety of environmental, biological, and industrial samples, facilitating comprehensive assessments of their presence and impact in the ecosystem.

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5.2. Advanced Methodologies

Recent advances in nanotechnology have significantly impacted the field of environmental science, particularly in the development of novel methodologies for the extraction, detection, and degradation of PAHs in environmental matrices, especially water. One notable application is the use of nanomaterials to enhance these processes, offering potential breakthroughs in environmental remediation technologies. For instance, Han et al. [256] demonstrated the use of graphene oxide (GO) aggregates combined with sodium chloride to effectively concentrate and separate PAHs in aqueous solutions. Their method, which involves a sample volume of 40 mL, achieved a detection limit ranging between 10 to 30 ng/L. The method also reported excellent analytical performance with a correlation coefficient (R2) exceeding 0.99 and recovery yields ranging from 80 to 111% [256]. This procedure is particularly noteworthy for its simplicity and environmental sustainability, as it significantly reduces the use of organic solvents, utilizing only 1 mL of hexane for GC-MS analysis, thus aligning with green chemistry principles. Another innovative approach involves the use of silver nanoparticles embedded within a porous graphitic carbon nitride matrix (Ag/pg-CN). Lu et al. [257] reported that this nanocomposite material enhances the detection capabilities for PAHs such as fluorene by leveraging the high surface-enhanced Raman scattering (SERS) sensitivity provided by the silver nanoparticles [257]. This method exemplifies the potential of nano-enhanced substrates to improve the sensitivity and specificity of PAH detection. Further studies have explored the utility of gold nanorods array functionalized with 4-dodecyl benzenediazonium tetrafluoroborate as a SERS substrate for the detection of benzo[a]pyrene (BaP), fluoranthene (FL), and naphthalene (NAP) [258]. The reported detection limits for these PAHs were notably low, ranging from 0.026 mg/L for BaP to 3.94 mg/L for NAP, indicating high sensitivity [258]. This material also demonstrated excellent reproducibility and reusability, which are critical features for practical environmental monitoring applications [257,258]. Additionally, metal-organic frameworks (MOFs) have been extensively tested due to their high chemical stability and porosity, which make them ideal for adsorbing PAHs. One such development involves the synthesis of MOF-199 combined with graphene (MOF-199/Gr) [259], which showed a high absorption capacity for various PAHs, with detection limits ranging from 3 to 10 pg/mL and relative standard deviations (RSD) between 5.0 to 7.4% [260]. Another noteworthy innovation is the use of zinc MOF covalently linked to organic frameworks (Zn-MOF/COF) as SPME applications, achieving detection limits from 0.1 to 1 ng/g and high recovery rates in real soil samples [260]. In an attempt to direct trends towards sustainable practices, spent tea leaves from Camellia sinensis have been repurposed as SPME materials for extracting different PAHs from water samples [261]. This method not only supports waste reuse but also achieves high correlation coefficients (0.9952 to 0.9983) and recovery rates (88.0 to 111.4%) under optimal conditions, illustrating its potential as a sensitive, cost-effective, and environmentally friendly analytical tool [261].

The advancement of sensor technology is crucial for the in situ detection of contaminants such as PAHs in environmental matrices. Notably, Ferreto et al. [262] have identified excitation-emission matrix (EEM) fluorescence spectroscopy combined with parallel factor (PARAFAC) analysis as effective methods for identifying and characterizing PAHs in water. These techniques leverage the unique fluorescence properties of PAHs to facilitate accurate and detailed detection. Rosental et al. [263] furthered this research by synthesizing phosphane-gold (I)-naphthalimide complexes to explore the interactions between these complexes and specific PAHs—namely naphthalene, phenanthrene, anthracene, and pyrene. Their study revealed that these complexes exhibit high affinity for anthracene and pyrene, but not for naphthalene, suggesting weak π-π interactions for the latter. This insight into the molecular interactions assists in refining the sensitivity of sensors to different PAH structures [263]. Aside from these chemical methods, biomarkers and bio-monitors represent alternative approaches for detecting PAHs. However, their effectiveness is often constrained by the choice of organisms used and the quantification capabilities within the specific environmental matrix [252]. To address some of these limitations, immunoassay techniques such as chemiluminescence immunoassays (CLIA), fluoroimmunoassays (FIA), radioimmunoassay (RIA), and enzyme-linked immunosorbent assays (ELISA) have been developed [252]. Commercially available kits like the Rapid Assay® PAH test kit and Envirogard® are employed for PAH detection, but these methods can

 doi:10.20944/preprints202404.1983.v1

be time-consuming and may lose sensitivity in matrices that contain a mixture of PAHs [252]. A more innovative approach involves the use of immunosensors, which integrate immunoassay techniques into microdevices. These compact analytical devices are designed with an antigen-antibody complex on the surface and can detect changes through electrical, mechanical, or biomechanical signals. This method boasts several advantages, including minimal sample and solvent requirements and the development of small-scale instrumental devices [252]. A notable application of this technology is the lab-on-a-chip device, which has been developed to detect benzo[a]pyrene (BaP) in water using an electrochemical immunoassay technique [264]. The detection process occurs within a microfluidic cell equipped with a chip, where the electrochemical reactions are analyzed through cyclic voltammetry [264]. Another sophisticated implementation is the in-line fiber optofluidic sensor, which uses an antiresonant-reflecting optical waveguide for the detection of BaP with a sensitivity of up to 23 pm/pM [264]. All these advancements highlight the dynamic nature of sensor development for environmental monitoring, underscoring the potential of nanotechnology and bioengineering to enhance the detection and quantification of hazardous substances like PAHs in different environments.

Computational chemistry plays a pivotal role in advancing environmental chemistry, particularly through the development of methods for studying and managing contaminants such as PAHs. The application of various computational techniques, including molecular mechanics, molecular dynamics, and hybrid methods, enables researchers to delve deeply into the interactions between PAHs, environmental matrices, and potential remediation materials. One of the significant contributions of computational chemistry is in exploring the transformation pathways of PAHs through environmental reactions, such as atmospheric photooxidation. By simulating these processes, researchers can predict the behavior and fate of these contaminants under various environmental conditions, guiding effective mitigation strategies.

The use of density functional theory (DFT), particularly the B3LYP/6-31G level of theory, has been instrumental in comparing the thermodynamic stability of hydroxylated PAHs (OHPAHs) and analyzing the average local ionization energy (ALIE) [265]. These computational studies have been validated against published experimental data, demonstrating that these theoretical approaches provide accurate predictions of PAH stability and reactivity. Furthermore, computational chemistry has facilitated the development of innovative biosensors for detecting PAHs. For instance, a DNA/Cu_2O-GS nanostructure was evaluated using the DFT methods implemented in the QUANTUM ESPRESSO software alongside the BoltzTrap computing code [266]. The results from this study indicated that the DNA/Cu_2O-GS nanostructure could function effectively as a field-effect transistor for PAH detection. This capability is attributed to the observed changes in electrical conductivity when PAHs interact with the biosensor, highlighting the potential of computational tools in designing sensitive and specific detection systems for environmental contaminants [266]. Another interesting study by Zhao et al. [267] assessed the interaction capabilities of PAHs with different media. To do this, molecular dynamics and electrostatic potential surface simulations were conducted to investigate the adsorption of PAHs in soil, focusing on the roles of quartz, leonardite humic acid (LHA), and water. The results indicated that PAHs bound more strongly to LHA than to quartz, primarily due to the hydroxyl and carboxyl groups in LHA and their π-π interactions. Water, while mobile on quartz, adversely affected PAH adsorption on both surfaces. Notably, when the water layer thickness exceeded 2.0 nm, it significantly reduced the adsorption capabilities of both quartz and LHA [267]. In another study conducted by Zeng et al. [268] the oxidation of anthracene (ANT) and phenanthrene (PHE) initiated by OH radicals was explored using quantum chemistry and chemical kinetics. The process begins with PAH-OH adducts reacting with O_2 and NO_2, forming radical species that can either revert, isomerize to oxygenated PAHs (OPAHs), or react with NO. The study found that ANT-9-OH, PHE-4-OH, and PHE-1-OH have slow reaction rates with O_2, leading to the formation of both OPAHs and significant quantities of nitrated PAHs (NPAHs). Conversely, ANT-1-OH and PHE-10-OH react rapidly with O_2 to form only OPAHs. Field measurements confirmed the formation of NPAHs, supporting these findings, while the presence of 9-NPHE suggested an alternative primary source such as biomass burning, due to its unlikely formation from

 doi:10.20944/preprints202404.1983.v1

OH or NO_3 reactions. These recent examples are just one instance of how various simulation and calculation methods can be applied to characterize the genesis of PAHs and their potential processes of degradation and transformation into other species with different environmental implications. Overall, the integration of computational chemistry into environmental science not only enhances our understanding of chemical interactions and transformations in the environment but also drives the innovation of new technologies and methodologies that can be used for the effective detection and remediation of hazardous substances.

6. Regulatory Frameworks

The evaluation of PAHs in surface waters is critically important due to the significant adverse effects these hazardous substances can have on environmental quality, as well as on human and animal health. PAHs are known for their persistence in the environment and their potential to bioaccumulate in wildlife and humans, posing serious health risks including cancer and reproductive issues. Given their toxicological significance, regulatory frameworks have been established globally to monitor PAHs and their derivatives in different environmental matrices, including surface water. To effectively manage and mitigate the risks associated with PAHs in surface waters, it is crucial to continually refine monitoring standards, analytical methods, and regulatory measures to align with the specific uses or potential uses of each water reservoir or watershed. This requires a dynamic regulatory approach that adapts to emerging scientific knowledge and technological advancements in water quality assessment.

The establishment of the Environmental Protection Agency (EPA) in 1970 under the Code of Federal Regulations, Title 40, marked a significant milestone in the United States' commitment to environmental protection. This was followed by the enactment of the Clean Water Act (CWA) in 1972, codified at 33 U.S.C. §1251 et seq., which aimed to "restore and maintain the chemical, physical, and biological integrity of the Nation's waters" [269]. This legislation set the stage for comprehensive federal water pollution control activities, including setting wastewater standards for industry and water quality standards for all contaminants in surface waters [270]. Furthermore, the Safe Drinking Water Act (SDWA), established under 42 U.S.C. §300f et seq., mandates the EPA to protect the quality of drinking water in the U.S., empowering the agency to set minimum standards to safeguard public health. One of the key actions taken by the EPA under the SDWA was to establish a Priority Chemical list that includes several PAH compounds such as Acenaphthene, Acenaphtylene, Anthracene, Benzo[ghi]perylene, Fluorene, Phenanthrene, and Pyrene due to their toxicity and prevalence in the environment [270].

Internationally, approaches to regulating PAHs in freshwater vary significantly. For instance, the European Union under its Water Framework Directive has established standards for 8 specific PAH compounds, reflecting a targeted approach to managing these pollutants in European waters [271–274]. However, not all countries have consistent or comprehensive regulations regarding PAHs in freshwater. For example, while the European Community does not prioritize the same list of contaminants in drinking water as the USA and Turkey, nations like Japan set their water quality parameters according to guidelines established by the WHO, which has set a guideline value of 0.7 µg/L for benzo[a]pyrene due to its potent carcinogenicity [251,275–277]. Research such as that conducted by Yao et al. [276] which examined PAH concentrations at 42 different surface water sites, underscores the necessity for robust monitoring and regulation of PAHs in freshwater. This study highlighted the variable presence of PAHs across diverse aquatic environments and has been instrumental in advocating for more stringent and localized water quality standards, particularly in regions like China where industrial and urban runoff pose significant risks to water quality [276]. Canada's approach, focusing on the protection of aquatic life, demonstrates another dimension of PAH regulation, emphasizing ecological health alongside human health considerations [277]. This perspective is crucial as it recognizes the ecological impacts of PAH contamination, which can affect biodiversity and the functioning of aquatic ecosystems. The global challenge of regulating PAHs in surface water is complex and demands a coordinated, scientifically informed approach that incorporates international cooperation and local action. Continuous improvement of analytical

 doi:10.20944/preprints202404.1983.v1

methods, alongside adaptive regulatory frameworks, is essential for the effective management of PAH pollution and the protection of public and ecological health. This multifaceted approach ensures that water quality management keeps pace with both technological advancements and expanding scientific understanding of the impacts of PAHs.

7. Conclusion

In conclusion, this comprehensive review has thoroughly examined the sources, distribution, ecotoxicological impacts, detection methods, and regulatory frameworks concerning PAHs in water systems. The review illuminates the multifaceted nature of PAH contamination, delineating both anthropogenic and natural origins. The significant anthropogenic sources, including industrial discharges, urban runoff, and oil spills, juxtapose against natural sources such as volcanic activities and wildfires, underscoring the pervasive nature of PAHs across various environmental contexts. The complexity of PAH distribution in aquatic environments has been elaborated upon, revealing how transport mechanisms and environmental interactions, such as with sediments and biota, are influenced by dynamic factors including hydrodynamics and seasonal variations. This distribution is critical to understanding the persistence and movement of PAHs in freshwater ecosystems, which directly impacts their bioavailability and ecological consequences. The section on ecotoxicological impacts has highlighted the profound effects of PAHs at molecular, organismal, and ecosystem levels. The evidence of bioaccumulation, alongside mutagenic and carcinogenic effects on aquatic life, emphasizes the severe risk PAHs pose to biodiversity and ecological health. Case studies have further exemplified the tangible impacts of these compounds in specific water systems, offering insight into the mechanisms of PAH-induced ecological degradation. The discussion of analytical methods for PAH detection has brought to light both traditional and advanced techniques, acknowledging ongoing challenges due to the diverse physicochemical properties of PAHs. The evolution of these methodologies not only enhances our capability to detect and quantify PAHs but also raises critical considerations for methodological accuracy and environmental monitoring. The regulatory frameworks have provided a perspective on the existing measures aimed at managing PAH contamination. A comparative analysis of international standards has revealed significant disparities, highlighting the need for a unified global strategy to address the complexities of PAH pollution effectively. The identified gaps in current regulatory approaches underscore the urgency of refining legal and policy measures to encompass a broader spectrum of PAH-related environmental and health impacts.

In conclusion, this review emphasizes the necessity for an integrated approach that combines robust scientific research with strict regulatory measures. Enhancing our understanding of PAH dynamics and their harmful effects on freshwater ecosystems will guide more effective environmental management and policymaking. Ultimately, addressing the challenges posed by PAH pollution is essential for protecting our water resources and ensuring a sustainable ecological future, necessitating collaborative efforts from researchers, policymakers, and stakeholders alike.

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List of Acronyms

PAH	Polycyclic aromatic hydrocarbon
LMW-PAH	Low molecular weight polycyclic aromatic hydrocarbon
HMW-PAH	High molecular weight polycyclic aromatic hydrocarbon
USEPA	United States Environmental Protection Agency
LRT	Long-range transport
ETM	Estuarine turbidity maximum
BCF	Bioconcentration factor
K_{ow}	Octanol-water partition coefficient
K_{oc}	Organic carbon normalized sediment–water partition coefficient
K_{pm}	Particulate matter–water distribution coefficient
A-PAH	Alkylated polycyclic aromatic hydrocarbon
YRD	Yellow River Delta
SPM	Suspended particulate matter
PES	Paranaguá Estuarine System
TOC/TN	Total organic carbon/total nitrogen ratio
$\delta^{13}C$	^{13}C isotropic chemical shift
SWRP	Stormwater retention pond
HC5	Hazardous concentration levels for 5% of species
WHO	World Health Organization
DW	Dry weight
ROS	Reactive oxygen species
GSH	Glutathione
CAT	Catalase
SOD	Superoxide dismutase
PCB	Polychlorinated biphenyl
BAF	Bioaccumulation factor
BaP	Benzo[a]pyrene
OECD	Organization for Economic Co-operation and Development
3-OHBcP	3-hydroxybenzo[c]phenanthrene
SRKWs	Southern resident killer whales
FTT	FAO-Thiaroye processing technique
SPE	Solid-Phase Extraction
SPME	Solid-Phase Microextraction
LLE	Liquid-Liquid Extraction
DLLME	Dispersive liquid-liquid micro-extraction
ILDLLME	Ionic Liquid-Based Dispersive Liquid-Liquid Microextraction
SFE	Supercritical Fluid Extraction
GC-MS	Gas chromatography coupled with mass spectrometry
HPLC-UV	High-performance liquid chromatography with ultraviolet detection
HPLC-DAD-FLD	High-performance liquid chromatography with diode-array and fluorescence detection
HPLC-FLD-UV	High-performance liquid chromatography with fluorescence and ultraviolet detection
GO	Graphene oxide
Ag/pg-CN	Silver nanoparticles embedded within a porous graphitic carbon nitride matrix
SERS	Surface-enhanced Raman scattering
FL	Fluoranthene
NAP	Naphthalene
MOF	Metal-organic framework
RSD	Relative standard deviation
EEM	Excitation-emission matrix

 doi:10.20944/preprints202404.1983.v1

PARAFAC	Parallel factor analysis
CLIA	Chemiluminescence immunoassay
FIA	Fluoroimmunoassay
RIA	Radioimmunoassay
ELISA	Enzyme-linked immunosorbent assay
DFT	Density functional theory
OHPAH	Hydroxylated PAH
ALIE	Average local ionization energy
LHA	Leonardite humic acid
PHE	Phenanthrene
OPAH	Oxygenated PAH
NPAH	Nitrated PAH
CWA	Clean Water Act
SDWA	Safe Drinking Water Act

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