

Polycyclic Aromatic Hydrocarbon Derivatives in an Oil-Impacted African River System: Distribution, Source Characteristics, Bioaccumulation and Human Health Risks in the White Nile, South Sudan

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Abstract

Polycyclic aromatic hydrocarbon derivatives (PAH derivatives), including nitro-polycyclic aromatic hydrocarbons (NPAHs), oxygenated polycyclic aromatic hydrocarbons (OPAHs), and hydroxy-polycyclic aromatic hydrocarbons (OHPAHs), are emerging contaminants of increasing environmental concern because of their persistence, bioaccumulation potential, and enhanced toxicity relative to their parent PAHs. However, information regarding their occurrence and ecological implications in African aquatic ecosystems remains extremely limited. This study investigated the occurrence, distribution, source characteristics, bioaccumulation behaviour, and potential human health risks of PAH derivatives in sediments and three commercially important fish species (*Clarias gariepinus*, *Lates niloticus*, and *Oreochromis niloticus*) collected from the White Nile near the Melut oil fields in South Sudan.

Sediment and fish samples were extracted and analysed using gas chromatography–tandem mass spectrometry (GC-MS/MS). Nitro-PAHs were the dominant group of contaminants in both sediments and fish tissues, with 1-nitronaphthalene identified as the most abundant individual congener. Low-molecular-weight PAH derivatives accounted for more than 75% of total PAH derivatives across all sample matrices, indicating the predominance of compounds with relatively high environmental mobility and bioavailability. No significant differences in total PAH derivative concentrations were observed among sampling sites, suggesting common regional contamination influences.

Diagnostic ratio analysis, contaminant composition patterns, and multivariate statistical assessment collectively suggested a substantial petroleum-related influence associated with activities in the Melut oil-producing region, although mixed-source contributions from combustion emissions, atmospheric deposition, and environmental transformation processes could not be excluded. Principal component analysis revealed distinct compositional groupings among PAH derivative classes, indicating the contribution of multiple environmental processes to contaminant distribution.

Bioaccumulation analysis demonstrated that PAH derivatives were bioavailable to aquatic organisms and accumulated in fish tissues, with accumulation patterns varying among compounds and species. Low-molecular-weight derivatives generally exhibited greater apparent accumulation than high-molecular-weight derivatives, whereas relationships between contaminant concentrations and fish lipid content were weak or non-significant. The estimated carcinogenic potency associated with selected nitrated PAHs in fish was below commonly applied screening thresholds, suggesting low carcinogenic risk under the assumptions of the present assessment. However, toxicological uncertainty remains for many oxygenated and hydroxylated derivatives.

This study provides one of the first comprehensive datasets on PAH derivatives in the aquatic environment of South Sudan and contributes important baseline information for environmental monitoring, ecological risk assessment, and management of oil-impacted freshwater ecosystems in sub-Saharan Africa.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are among the most widespread organic contaminants in aquatic environments due to their persistence, bioaccumulation potential, and adverse effects on both ecological and human health (Bandowe and Meusel 2017). These compounds are primarily released through incomplete combustion of organic materials, petroleum extraction and processing, industrial activities, urban runoff, and accidental oil spills (Tomaz et al. 2018). While extensive research has focused on parent PAHs, increasing attention is being directed toward their transformation products, collectively known as polycyclic aromatic hydrocarbon derivatives (PAH derivatives), which include nitro-polycyclic aromatic hydrocarbons (NPAHs), oxygenated polycyclic aromatic hydrocarbons (OPAHs), and hydroxy-polycyclic aromatic hydrocarbons (OHPAHs).

PAH derivatives are formed through both primary and secondary processes (Albinet et al., 2008). Primary sources include direct emissions from fossil fuel combustion, petroleum extraction activities, industrial processes, and vehicular emissions. Secondary formation occurs through atmospheric photochemical reactions, biological transformation, oxidation, nitration, and other environmental degradation processes involving parent PAHs. Consequently, PAH derivatives are frequently detected in air, water, sediments, soils, and biota, often occurring alongside their parent compounds (Bandowe and Meusel 2017; Rykowska and Andrzejewski 2022). PAH derivatives are increasingly recognized as environmentally relevant contaminants because of their widespread occurrence and diverse toxicological properties (Lundstedt et al. 2014).

The environmental significance of PAH derivatives extends beyond their occurrence. Several studies have demonstrated that many PAH derivatives exhibit enhanced toxicity, mutagenicity, and carcinogenicity compared with their parent PAH (Bekki et al. 2013). OPAHs can generate reactive oxygen species that induce oxidative stress and cellular damage (Clergé et al. 2019; Li et al. 2019). NPAHs have been identified among the most potent direct-acting mutagens in environmental matrices (Albinet et al. 2008; Bandowe and Meusel 2017). Unlike many parent PAHs that require metabolic activation before exerting toxic effects, several PAH derivatives possess intrinsic biological activity, making them contaminants of growing environmental concern.

Once released into aquatic environments, PAH derivatives partition between water, suspended particles, sediments, and living organisms according to their physicochemical properties. Sediments frequently serve as long-term reservoirs for these contaminants because of their affinity for organic matter and particulate materials (Burgess et al. 2013; Umeh et al. 2020). Under changing environmental conditions, sediment-bound PAH derivatives may be remobilized into the water column, increasing their bioavailability to aquatic organisms. Fish are particularly important indicators of aquatic contamination because they can accumulate contaminants through dietary uptake, direct absorption from water, and sediment interactions. Bioaccumulation of PAH derivatives represent fish species represents a potential pathway for human exposure through food consumption (Musa et al. 2014; Yu et al. 2019).

Despite increasing global concern regarding PAH derivatives, available information remains highly uneven across geographical regions. Most studies have been conducted in Europe, North America, and Asia, while information from sub-Saharan Africa remains scarce (Abayi et al. 2021; Bandowe & Nkansah 2016). This knowledge gap is particularly significant because many African aquatic ecosystems are increasingly exposed to petroleum exploration, oil production activities, urbanization, industrial development, and agricultural expansion. The lack of environmental monitoring data limits accurate assessment of ecological risks and human exposure associated with these emerging contaminants.

The White Nile represents one of the most important freshwater systems in Africa, supporting fisheries, transportation, agriculture, domestic water supply, and regional biodiversity. The river traverses areas experiencing extensive petroleum exploration and production activities, particularly around the Melut Basin in South Sudan. The Melut oil fields constitute one of the largest oil-producing regions in the country and have raised concerns regarding the release of petroleum-related contaminants into surrounding aquatic ecosystems. While studies have reported the occurrence of petroleum hydrocarbons and other pollutants in oil-producing environments, information concerning PAH derivatives in the White Nile ecosystem is virtually nonexistent (Abayi et al. 2021).

Furthermore, fish species such as *Clarias gariepinus*, *Lates niloticus*, and *Oreochromis niloticus* are economically and nutritionally important to local communities that depend heavily on fish consumption. Understanding the occurrence, distribution, and bioaccumulation of PAH derivatives in these species is therefore essential for evaluating both ecological impacts and potential human health risks associated with petroleum-related contamination (Musa et al. 2014; Ololade et al. 2021).

The present study investigated the occurrence, distribution, source characteristics, bioaccumulation behaviour, and potential human health implications of PAH derivatives in sediments and fish collected from the White Nile near the Melut oil fields in South Sudan. Specifically, the study aimed to: (i) determine the occurrence and composition profiles of NPAHs, OPAHs, and OHPAHs in sediments and fish tissues; (ii) evaluate potential contamination sources using diagnostic ratios and principal component analysis; (iii) assess relationships between contaminant concentrations and sediment organic carbon or fish lipid content; (iv) examine bioaccumulation patterns using biota–sediment accumulation factors; and (v) evaluate potential human health risks associated with consumption of contaminated fish. To the best of our knowledge, this study represents one of the first comprehensive investigations of PAH derivatives in the aquatic environment of South Sudan and among the few studies conducted in sub-Saharan Africa. The findings provide valuable baseline information for environmental monitoring, ecological risk assessment, and sustainable management of oil-impacted freshwater ecosystems within the Nile Basin.

2. Materials and Methods

2.1 Study Area

The study was conducted along the White Nile in the vicinity of the Melut oil fields located in Upper Nile State, South Sudan (Fig. 1). The Melut Basin constitutes one of the major petroleum-producing regions in South Sudan and is characterized by intensive oil exploration, extraction, transportation, and associated anthropogenic activities. These activities have raised concerns regarding the release of petroleum-derived contaminants into adjacent aquatic ecosystems.

Three sampling sites designated as Site A, Site B, and Site C were selected along the White Nile based on their proximity to oil production infrastructure and potential contamination sources. Site A was located nearest to the oil production facilities and was expected to experience the greatest influence from petroleum-related activities, while Sites B and C represented downstream locations. Sediment and fish samples were collected to evaluate the occurrence, distribution, and bioaccumulation of PAH derivatives within the aquatic ecosystem.

2.2 Sample Collection

Sampling was conducted during the study period at the three selected locations. Surface sediment samples (0–10 cm depth) were collected using a stainless-steel grab sampler. At each site, multiple sediment subsamples were collected and composited to obtain representative samples. The sediment samples were transferred into pre-cleaned amber glass containers, transported on ice, and stored at – 20°C until analysis.

Three commonly consumed fish species, namely *Clarias gariepinus*, *Lates niloticus*, and *Oreochromis niloticus*, were collected from local fishermen operating within the sampling areas. Fish samples were transported on ice to the laboratory, where muscle tissues were excised, homogenized, wrapped in aluminum foil, and stored at – 20°C pending extraction and analysis.

2.3 Chemicals and Reagents

All analytical standards, solvents, and reagents used in this study were of analytical or chromatographic grade. Standards of nitro-polycyclic aromatic hydrocarbons (NPAHs) oxygenated polycyclic aromatic hydrocarbons (OPAHs), and hydroxy-polycyclic aromatic hydrocarbons (OHPAHs) were obtained from certified commercial suppliers. Dichloromethane (DCM), n-hexane, acetone, methanol, and other solvents used during extraction and clean-up procedures were of GC-grade purity.

Hydroxy-PAH standards and extracts were derivatized using BSTFA/TMCS (99:1) prior to instrumental analysis to improve volatility and chromatographic performance.

2.4 Extraction and Clean-Up of Sediment Samples

Sediment samples were freeze-dried, homogenized, and sieved before extraction. Approximately 5 g of sediment was extracted using ultrasonic-assisted extraction with an appropriate solvent mixture. The extracts were concentrated and subjected to clean-up procedures to remove matrix interferences. Following clean-up, the extracts were reduced to a suitable volume under a gentle stream of nitrogen and transferred into GC vials for instrumental analysis (Oluseyi et al. 2011; Peng et al. 2012).

2.5 Extraction and Clean-Up of Fish Samples

Fish muscle tissues were homogenized and extracted using accelerated solvent extraction (ASE). Extraction conditions were optimized to ensure efficient recovery of PAH derivatives. The resulting extracts were concentrated and purified using clean-up procedures designed to remove lipids and co-extracted matrix components. Clean extracts were concentrated under nitrogen and transferred to GC vials prior to analysis (Gbeddy et al. 2020).

For OHPAHs, derivatization was performed using BSTFA/TMCS to convert hydroxyl-containing compounds into their corresponding trimethylsilyl derivatives, thereby improving chromatographic separation and detection sensitivity (Cochran et al. 2012).

2.6 GC-MS/MS Analysis

Quantitative analysis of PAH derivatives was performed using gas chromatography coupled with tandem mass spectrometry (GC-MS/MS). Chromatographic separation was achieved using a capillary column suitable for semi-volatile organic compounds. The mass spectrometer was operated in multiple reaction monitoring (MRM) mode to maximize selectivity and sensitivity.

Compound identification and quantification procedures were consistent with previously reported analytical approaches for oxygenated and nitrated PAHs (Albinet et al. 2006; Cochran et al. 2012).

Target analytes included fifteen nitro-PAHs, five oxy-PAHs, and three hydroxy-PAHs. Compound identification was based on retention times, quantifier ions, qualifier ions, and corresponding collision energies obtained from authentic standards. Quantification was performed using external calibration curves generated from multi-point standard solutions covering the expected concentration ranges in environmental samples (Cochran et al. 2012; Mueller et al. 2019).

2.7 Quality Assurance and Quality Control

Quality assurance and quality control procedures were implemented throughout sampling, extraction, and instrumental analysis. Procedural blanks, solvent blanks, duplicate samples, and spiked samples were analyzed to assess contamination, analytical precision, and extraction efficiency.

Method performance was evaluated using recovery studies, limits of detection (LOD), and limits of quantification (LOQ). Recovery values were determined by spiking representative sediment and fish samples with known concentrations of target analytes prior to extraction. Only analytical batches meeting established quality control criteria were accepted.

Instrument calibration was verified regularly using calibration standards and continuing calibration checks to ensure analytical stability throughout the study.

2.8 Determination of Total Organic Carbon and Lipid Content

Total organic carbon (TOC) content of sediment samples was determined using standard titrimetric analytical procedures following removal of inorganic carbon fractions. Fish lipid content was determined gravimetrically following solvent extraction and was expressed as a percentage of wet tissue weight.

TOC and lipid content measurements were subsequently used to normalize contaminant concentrations during evaluation of sediment–biota relationships and calculation of biota-sediment accumulation factors.

2.9 Source Apportionment of PAH Derivatives

Potential sources of PAH derivatives were investigated using diagnostic ratios and principal component analysis (PCA). Diagnostic ratios based on low-molecular-weight and high-molecular-weight PAH derivatives were calculated to provide preliminary information regarding potential petrogenic or pyrogenic origins.

Principal component analysis was conducted using standardized concentration data to identify compositional patterns among PAH derivatives and to explore potential source groupings. Components with eigenvalues greater than unity were retained for interpretation, and loading patterns were examined to identify compounds contributing most strongly to each principal component.

2.10 Biota-Sediment Accumulation Factors

Bioaccumulation of PAH derivatives in fish relative to sediments was assessed using biota-sediment accumulation factors (BSAFs). BSAFs were calculated using lipid-normalized contaminant concentrations in fish tissues and organic carbon-normalized concentrations in sediments according to established environmental risk assessment approaches (Burgess et al. 2013; Y. Zhang et al. 2011).

The calculated BSAF values were used to evaluate the accumulation behaviour of individual PAH derivatives and to examine the influence of molecular characteristics on bioaccumulation potential.

Biota–sediment accumulation factors (BSAFs) were calculated according to:

$$\text{BSAF} = \frac{C_{\text{fish}}/f_{\text{lipid}}}{C_{\text{sediment}}/f_{\text{OC}}}$$

where C_{fish} is the concentration of a PAH derivative in fish tissue, f_{lipid} is the lipid fraction of fish tissue, C_{sediment} is the concentration of a PAH derivative in sediment, and f_{OC} is the fraction of organic carbon in sediment.

2.11 Statistical Analysis

Descriptive statistics including means, standard deviations, minimum values, maximum values, and detection frequencies were calculated for all target analytes. Data were evaluated for normality and homogeneity of variance prior to statistical analysis. Differences among sampling sites and fish species

were assessed using one-way analysis of variance (ANOVA) followed by appropriate post hoc comparisons where applicable.

Pearson correlation analysis was used to investigate relationships between PAH derivative concentrations and sediment total organic carbon (TOC) or fish lipid content. Principal component analysis (PCA) was performed to evaluate contaminant patterns and potential source groupings. Statistical significance was accepted at $p < 0.05$.

Non-detect values were replaced with one-half of the corresponding limit of detection (LOD/2) prior to statistical analysis to reduce bias associated with left-censored environmental data.

3. Results and Discussion

3.1 Occurrence and Distribution of PAH Derivatives in Sediments

PAH derivatives were detected in sediment samples collected from all sampling sites along the White Nile, indicating widespread contamination throughout the study area (Table S1; Fig. 2). Nitro-polycyclic aromatic hydrocarbons (NPAHs) were the dominant class of contaminants at all locations, followed by oxygenated PAHs (OPAHs) and hydroxy-PAHs (OHPAHs). Mean concentrations of Σ NPAHs were approximately 314.4, 307.9, and 181.7 ng g⁻¹ dw at Sites A, B, and C, respectively. Corresponding concentrations of Σ OPAHs were 58.2, 38.4, and 39.1 ng g⁻¹ dw, while Σ OHPAHs averaged 53.1, 47.0, and 17.7 ng g⁻¹ dw at Sites A, B, and C, respectively. These results demonstrate that nitrated derivatives contributed the largest proportion of total PAH derivative contamination throughout the study area.

Among individual analytes, 1-nitronaphthalene (1-NNAP) was the most abundant compound detected in sediments, followed by 2-nitronaphthalene (2-NNAP) and 9-nitroanthracene (9-NANT). The predominance of these low-molecular-weight derivatives is consistent with their relatively greater environmental mobility and persistence compared with higher molecular weight compounds. Figure 2 further illustrates that NPAHs accounted for the majority of total PAH derivatives at all sampling locations, emphasizing the importance of nitrated transformation products in the contaminant profile of the White Nile sediments.

Although Site A generally exhibited the highest concentrations of most contaminant groups, followed by Site B and Site C, statistical analysis revealed no significant differences among sampling sites ($p > 0.05$). The absence of significant spatial variation suggests that contamination may be influenced by regional processes operating throughout the river system rather than by localized point sources alone. Petroleum exploration and production activities within the Melut Basin likely contribute substantially to the observed contamination; however, atmospheric deposition, riverine transport, surface runoff, and environmental transformation of parent PAHs may also influence contaminant distribution.

The predominance of NPAHs is environmentally significant because many nitrated derivatives possess enhanced mutagenic and carcinogenic properties relative to their parent PAHs. Several NPAHs have been identified as potent direct-acting mutagens capable of inducing biological effects without prior metabolic activation (Albinet et al. 2008; Kanae et al. 2013). Consequently, environmental assessments based exclusively on parent PAHs may underestimate ecological and human-health risks in petroleum-impacted aquatic environments. The results of the present study therefore highlight the importance of including PAH derivatives in routine environmental monitoring programs, particularly in regions undergoing intensive petroleum development.

The sediment contamination patterns observed in this study are generally consistent with reports from other oil-impacted aquatic systems (Bandowe and Nkansah 2016; Wei and Han et al. 2015), where nitrated and oxygenated PAH derivatives have been detected in association with petroleum activities and secondary environmental transformation processes. However, available information from sub-Saharan Africa remains limited, emphasizing the importance of the present study as a baseline assessment for future monitoring and risk-evaluation efforts within the Nile Basin.

3.2 Occurrence and Distribution of PAH Derivatives in Fish

PAH derivatives were detected in all three fish species investigated, namely *Clarias gariepinus*, *Lates niloticus*, and *Oreochromis niloticus*, confirming their bioavailability and transfer into aquatic food webs (Tables S2, S3, and S4; Fig. 3). Similar to sediment samples, nitro-polycyclic aromatic hydrocarbons (NPAHs) constituted the dominant class of contaminants in fish tissues, followed by OPAHs and OHPAHs. The widespread occurrence of PAH derivatives across all species indicates continuous environmental exposure and uptake from contaminated aquatic habitats.

Among individual analytes, 1-nitronaphthalene (1-NNAP) and 2-nitronaphthalene (2-NNAP) were generally among the most abundant compounds detected in fish tissues, reflecting the dominance of low-molecular-weight PAH derivatives observed in sediment samples. Detection frequencies varied among compounds and species, suggesting differences in contaminant uptake, metabolism, and elimination. Several compounds, including 6-OHPHE, were consistently detected at relatively high frequencies, indicating widespread exposure throughout the study area.

Differences in contaminant profiles among fish species likely reflect variations in feeding ecology, habitat utilization, trophic position, and physiological characteristics (Ololade et al. 2021; Yu et al. 2019). *Lates niloticus*, a predatory species occupying a relatively high trophic position, exhibited elevated concentrations of several PAH derivatives, whereas *Clarias gariepinus* and *Oreochromis niloticus* displayed distinct accumulation patterns for other compounds. These observations suggest that species-specific ecological and biological factors influence contaminant uptake and retention.

Low-molecular-weight PAH derivatives accounted for most total PAH derivative concentrations in fish tissues, consistent with the patterns observed in sediments. The similarity between sediment and fish contaminant profiles supports the hypothesis that sediments represent an important environmental

reservoir and source of exposure for aquatic organisms. Sediment-associated contaminants may become available to fish through direct contact, ingestion of contaminated food items, and resuspension processes that increase contaminant availability within the aquatic environment.

The occurrence of PAH derivatives in edible fish species is environmentally and public-health relevant because these species constitute important protein sources for local communities along the White Nile. Detection of nitrated, oxygenated, and hydroxylated PAH derivatives in fish tissues demonstrates the potential for transfer of petroleum-related contaminants from environmental compartments to humans through dietary exposure. Although the concentrations observed in the present study were generally low compared with those reported for heavily industrialized aquatic systems, continued monitoring remains warranted because of the enhanced toxicological properties associated with many PAH derivatives and the increasing petroleum development activities occurring within the Melut Basin region.

3.3 Source Apportionment of PAH Derivatives

3.3.1 Diagnostic Ratios

Diagnostic ratios based on low-molecular-weight (LMW) and high-molecular-weight (HMW) PAH derivatives were used as a preliminary tool for source identification (Tables S5, S21, and S22). Mean LMW/HMW ratios ranged from 5.99 at Site C to 8.91 at Site A, with an overall mean ratio of 7.29. Importantly, all sediment samples exhibited LMW/HMW ratios greater than unity, indicating a consistent predominance of low-molecular-weight PAH derivatives throughout the study area.

The dominance of low-molecular-weight derivatives is commonly associated with petroleum-related contamination (Alegre et al. 2017; Han et al. 2021; Wang et al. 2021), because crude oil and petroleum products generally contain larger proportions of lower molecular weight compounds than combustion-derived sources. However, diagnostic ratios should be interpreted cautiously because environmental transport, degradation, transformation, volatilization, and differential partitioning may modify contaminant signatures after release into the environment.

Despite these limitations, the consistent predominance of LMW derivatives across all sampling sites suggests a substantial petroleum-related influence within the study area. This interpretation is consistent with the extensive oil exploration and production activities occurring within the Melut Basin and agrees with the contaminant composition profiles observed in sediment samples.

3.3.2 Principal Component Analysis

Principal component analysis (PCA) was performed to further evaluate relationships among PAH derivatives and to explore potential contamination patterns (Tables S16, S17, and S18; Figs. 3 and 4). The first principal component (PC1) explained 37.13% of the total variance, while the second principal component (PC2) explained 10.89%. Together, the first two components accounted for 48.03% of the total variance in the dataset.

The loading structure indicated that several nitrated PAHs, including 1-NNAP, 2-NNAP, and 9-NANT, contributed strongly to PC1, reflecting the dominant role of nitrated derivatives in sediment contamination. In contrast, oxygenated and hydroxylated derivatives, including FN, CZ, ACr, Qui, 2-OHBP, 1-OHPYR, and 6-OHPHE, contributed more strongly to PC2. The separation of these contaminant groups suggests that multiple environmental processes influenced their occurrence and distribution.

The PCA results indicate that contamination patterns cannot be attributed to a single source (Hoang et al. 2021; Jang et al. 2013). Instead, the observed distribution of PAH derivatives likely reflects a combination of petroleum-related inputs, atmospheric deposition, combustion-derived emissions, and environmental transformation of parent PAHs. Petroleum activities associated with the Melut oil fields are likely a major contributor to the contamination profile; however, secondary formation processes involving oxidation, hydroxylation, and nitration reactions may also contribute substantially to the occurrence of OPAHs and OHPAHs.

When interpreted together, the diagnostic ratios, contaminant composition profiles, and PCA results provide consistent evidence for a substantial petroleum-related influence within the study area while also indicating the contribution of mixed environmental sources and transformation processes. This interpretation is consistent with the complex environmental conditions associated with oil-producing aquatic systems.

3.4 Relationship Between PAH Derivatives and Sediment Organic Carbon or Fish Lipid Content

Pearson correlation analysis was conducted to investigate relationships between PAH derivative concentrations and sediment total organic carbon (TOC) or fish lipid content (Tables S6, S19, and S20). Overall, most PAH derivatives exhibited weak or non-significant correlations with sediment TOC, indicating that organic carbon was not the primary factor controlling contaminant distribution throughout the study area. Only three compounds, namely 1-NNAP, 3-NFA, and 2,7-DNF, exhibited statistically significant positive correlations with TOC, all at Site B. These results suggest that while organic carbon may influence the retention of specific compounds under certain environmental conditions, its overall influence on PAH derivative distribution was limited.

The lack of strong TOC dependence indicates that multiple environmental processes likely contributed to contaminant occurrence and distribution. Potential controlling factors include direct petroleum-related inputs, atmospheric deposition, hydrodynamic transport, sediment resuspension, environmental transformation reactions, and compound-specific physicochemical properties. The importance of these processes is further supported by the source-apportionment results, which indicated mixed environmental influences on contaminant profiles.

Similarly, no statistically significant relationships were observed between fish lipid content and PAH derivative concentrations following correction of fish–lipid matching and recalculation of the dataset. The absence of significant lipid-associated accumulation suggests that contaminant uptake and

retention cannot be explained solely by equilibrium partitioning into lipid-rich tissues. Instead, bioaccumulation patterns are likely influenced by a combination of ecological and physiological factors, including feeding behaviour, trophic position, habitat utilization, metabolic transformation capacity, growth rates, and exposure history.

These findings highlight the complexity of PAH derivative behaviour in natural aquatic ecosystems and suggest that simple partitioning models may not adequately describe their environmental fate and biological accumulation. Similar observations have been reported in other studies where contaminant accumulation was influenced by multiple interacting environmental and biological factors rather than by lipid content alone (Umeh et al. 2020; H. Zhang and Kelly 2018). Consequently, assessment of PAH derivative bioaccumulation should incorporate both physicochemical characteristics and species-specific ecological processes to better understand contaminant dynamics in aquatic food webs.

3.5 Bioaccumulation of PAH Derivatives in Fish

Biota–sediment accumulation factors (BSAFs) were used to evaluate the accumulation behaviour of PAH derivatives in fish relative to sediment concentrations (Table S12, S13, S14, and S15). BSAF values varied considerably among compounds, sampling sites, and fish species, indicating that bioaccumulation was influenced by multiple environmental and biological factors.

In general, low-molecular-weight PAH derivatives exhibited greater apparent accumulation than high-molecular-weight derivatives. This pattern is consistent with the relatively higher mobility, bioavailability, and reduced sediment binding of lower molecular weight compounds. In contrast, high-molecular-weight derivatives tend to associate more strongly with sediment organic matter, reducing their availability for biological uptake.

Species-specific differences in accumulation patterns were also observed. Although accumulation varied among compounds, the results suggest that ecological characteristics such as feeding strategy, habitat preference, trophic position, and metabolic capacity influence contaminant uptake and retention. These factors likely contribute to the differences observed among *Oreochromis niloticus*, *Clarias gariepinus*, and *Lates niloticus*.

Interpretation of BSAFs should be approached cautiously because these values are sensitive to sediment concentrations, organic carbon content, fish lipid content, and treatment of non-detect values. Furthermore, PAH derivatives may undergo biotransformation and metabolic elimination within fish tissues, meaning that field-derived BSAFs represent indicators of apparent accumulation rather than strict equilibrium partitioning between sediments and biota (Burgess et al. 2013; Y. Zhang et al. 2011).

Despite these limitations, the results clearly demonstrate that PAH derivatives are bioavailable within the White Nile ecosystem and can be transferred from contaminated environmental compartments into aquatic organisms. The observed accumulation patterns provide evidence that fish are exposed to these contaminants under natural environmental conditions and support the use of fish species as indicators of petroleum-related contamination within the study area.

3.6 Influence of Physicochemical Properties on Bioaccumulation (Log Kow Relationships)

The relationship between biota–sediment accumulation factors (BSAFs) and octanol–water partition coefficients (Log Kow) was examined to evaluate the influence of hydrophobicity on the bioaccumulation behaviour of PAH derivatives. No consistent relationship was observed among compounds, fish species, or sampling sites. Compounds with relatively high Log Kow values did not necessarily exhibit higher BSAFs, and several lower molecular weight derivatives displayed greater apparent accumulation than more hydrophobic compounds.

The absence of a uniform BSAF–Log Kow relationship indicates that hydrophobicity alone was not sufficient to explain the observed accumulation patterns. Although Log Kow is widely used as an indicator of bioaccumulation potential (Wang et al. 2021; H. Zhang and Kelly 2018), accumulation under field conditions is influenced by numerous interacting environmental and biological factors. These include aqueous solubility, sediment sorption, contaminant bioavailability, feeding behaviour, trophic transfer, habitat utilization, growth dilution, and metabolic transformation processes.

The interpretation of PAH derivative behaviour is further complicated by the presence of functional groups such as nitro, carbonyl, and hydroxyl moieties, which substantially alter physicochemical properties relative to parent PAHs. These structural modifications influence polarity, environmental mobility, degradation pathways, and biological uptake processes. Consequently, physicochemical properties reported for parent PAHs cannot always be used to predict the environmental behaviour of their derivatives.

The weak relationship between BSAFs and Log Kow observed in the present study is consistent with the lack of significant correlations between contaminant concentrations and fish lipid content. Together, these findings suggest that bioaccumulation of PAH derivatives in the White Nile ecosystem is governed by a combination of contaminant-specific properties and species-specific ecological processes rather than by hydrophobic partitioning alone.

Overall, the results indicate that Log Kow provides useful information regarding the environmental behaviour of PAH derivatives but should not be considered a standalone predictor of bioaccumulation under natural field conditions. Accurate assessment of PAH derivative accumulation therefore requires consideration of both physicochemical characteristics and ecological factors influencing contaminant exposure and uptake.

3.7 Human Health Risk Assessment

The potential human health risks associated with consumption of contaminated fish were evaluated using toxic equivalency factors (TEFs) available for selected nitrated PAHs (Table S7,S8, S9, S10, and S11). The estimated total carcinogenic potency was approximately 1.8×10^{-3} ng g⁻¹ wet weight, with

Oreochromis niloticus contributing the highest proportion of the calculated toxic equivalent concentration (TEQ).

Although several NPAHs were detected at moderate concentrations, compounds possessing relatively high toxic equivalency factors contributed disproportionately to the overall carcinogenic potency. This observation demonstrates that contaminant concentration alone is not sufficient to evaluate potential health risks and highlights the importance of incorporating toxicological potency into environmental risk assessments.

The calculated toxic equivalent concentrations remained below commonly applied screening thresholds, suggesting that consumption of the investigated fish species is unlikely to result in substantial carcinogenic risk from the selected NPAHs under the assumptions used in the present assessment. These findings are encouraging because the studied fish species constitute important dietary protein sources for communities residing along the White Nile.

However, interpretation of the results should be approached cautiously because toxic equivalency factors are currently unavailable for many oxygenated and hydroxylated PAH derivatives detected in the samples (Wei et al. 2015; Yadav et al. 2018). Consequently, the present assessment may underestimate total toxicological risk associated with complex mixtures of PAH derivatives. In addition, long-term exposure, cumulative effects, and potential interactions among contaminants remain insufficiently characterized.

Overall, the results indicate that the estimated carcinogenic potency associated with selected NPAHs is low, but continued environmental monitoring and expanded toxicological investigations are warranted. Future studies should focus on developing toxicological reference values for OPAHs and OHPAHs and on evaluating combined exposure effects in populations that rely heavily on fish consumption from the White Nile ecosystem.

4. Conclusions

This study provides one of the first comprehensive assessments of polycyclic aromatic hydrocarbon derivatives (PAH derivatives) in sediments and fish from the White Nile near the Melut oil-producing region of South Sudan. Nitro-PAHs (NPAHs), oxygenated PAHs (OPAHs), and hydroxy-PAHs (OHPAHs) were detected in all environmental matrices investigated, demonstrating the widespread occurrence of these emerging contaminants within the aquatic ecosystem.

NPAHs constituted the dominant class of PAH derivatives in both sediments and fish tissues, with low-molecular-weight compounds accounting for the majority of the total contaminant burden. Diagnostic ratios, contaminant composition profiles, and principal component analysis collectively indicated a substantial petroleum-related influence associated with activities in the Melut Basin, although contributions from atmospheric deposition, combustion sources, and environmental transformation processes were also evident.

The detection of PAH derivatives in edible fish species confirmed their bioavailability and transfer within the aquatic food web. Bioaccumulation patterns varied among compounds and fish species and were influenced by a combination of physicochemical properties, ecological characteristics, and biological processes rather than hydrophobicity alone. The weak relationships observed between contaminant concentrations, fish lipid content, and Log Kow values highlight the complexity of PAH derivative behaviour under natural environmental conditions.

Human health risk assessment indicated that the estimated carcinogenic potency associated with selected nitrated PAHs was low and remained below commonly applied screening thresholds. Nevertheless, uncertainties remain because toxicological information is unavailable for many oxygenated and hydroxylated PAH derivatives detected in the study.

Overall, the findings provide valuable baseline information on the occurrence, distribution, source characteristics, bioaccumulation behaviour, and potential health implications of PAH derivatives in an understudied African freshwater ecosystem. The study contributes to the growing body of knowledge on emerging petroleum-related contaminants and highlights the importance of continued monitoring and risk assessment in oil-impacted aquatic environments within the Nile Basin and sub-Saharan Africa.

5. Recommendations

Based on the findings of this study, several recommendations are proposed to improve understanding and management of PAH derivatives in the White Nile ecosystem and similar oil-impacted freshwater environments.

Long-term environmental monitoring programs should be established to evaluate temporal and spatial trends of PAH derivatives in sediments, water, and aquatic organisms. Future monitoring efforts should include both parent PAHs and their transformation products because PAH derivatives may exhibit enhanced toxicity despite occurring at lower concentrations.

Environmental regulators, petroleum operators, and other stakeholders should strengthen environmental surveillance and pollution-management practices within the Melut Basin. Routine monitoring of petroleum-related contaminants and implementation of effective pollution-control measures are necessary to minimize contaminant release and protect the ecological integrity of the White Nile.

Further research is required to improve understanding of the environmental fate, transport, transformation pathways, bioavailability, and toxicological effects of PAH derivatives in tropical freshwater ecosystems. Particular attention should be directed toward oxygenated and hydroxylated PAH derivatives, for which toxicological reference values remain limited or unavailable.

Future investigations should evaluate seasonal variability, trophic transfer, biomagnification potential, and species-specific accumulation mechanisms of PAH derivatives within aquatic food webs. Such

studies would provide a more comprehensive understanding of contaminant dynamics and ecological risks in oil-impacted environments.

Finally, the absence of region-specific environmental quality guidelines for PAH derivatives highlights the need for the development of environmental standards and risk-assessment frameworks tailored to African freshwater ecosystems. Such frameworks would support evidence-based environmental management, regulatory decision-making, and protection of public health in regions experiencing increasing petroleum exploration and production activities.

Declarations

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Author contribution statement

Deo Ssentongo: Conceptualization, Methodology, Formal analysis, Investigation; Methodology, Writing - original draft.

Henry Matovu: Conceptualization, Methodology, Writing, Review & Editing.

Jane Namukobe: Writing, Review & Editing.

Florence Nantaba: Conceptualization, Writing, Review & Editing, Supervision.

Edward Mubiru: Writing, Review & Editing.

Ivan Špánik: Writing, Review & Editing.

Silver Odongo: Writing, Review & Editing.

Patrick Ssebugere: Conceptualization, Writing, Review & Editing, Supervision, Funding acquisition.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☒ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Ethical approval

This study did not involve human participants. Fish samples were obtained from local fishermen, and environmental samples (sediments) were collected in accordance with applicable institutional and national guidelines for environmental research. Therefore, formal ethical approval for human subjects was not required.

Consent to participate

Not applicable. This study did not involve human participants.

Consent to publish

Not applicable. This manuscript does not contain any individual person's data, images, or identifiable information requiring consent for publication.

Data Availability

The datasets generated and analyzed during the current study will be made publicly available in an appropriate public repository upon publication of this article. Repository details and the associated DOI will be provided upon publication.

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Figures

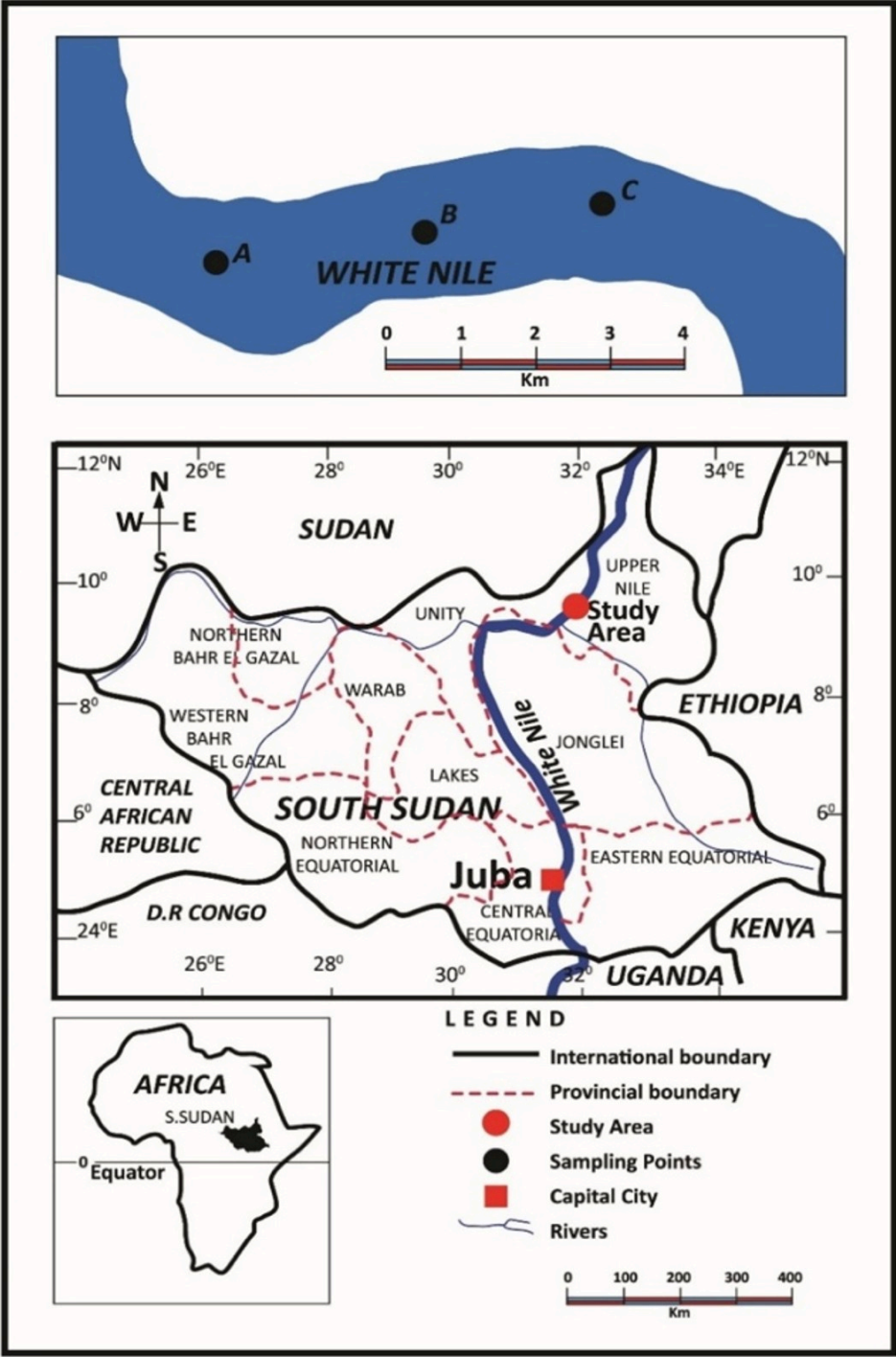


Figure 1

Map showing the study area

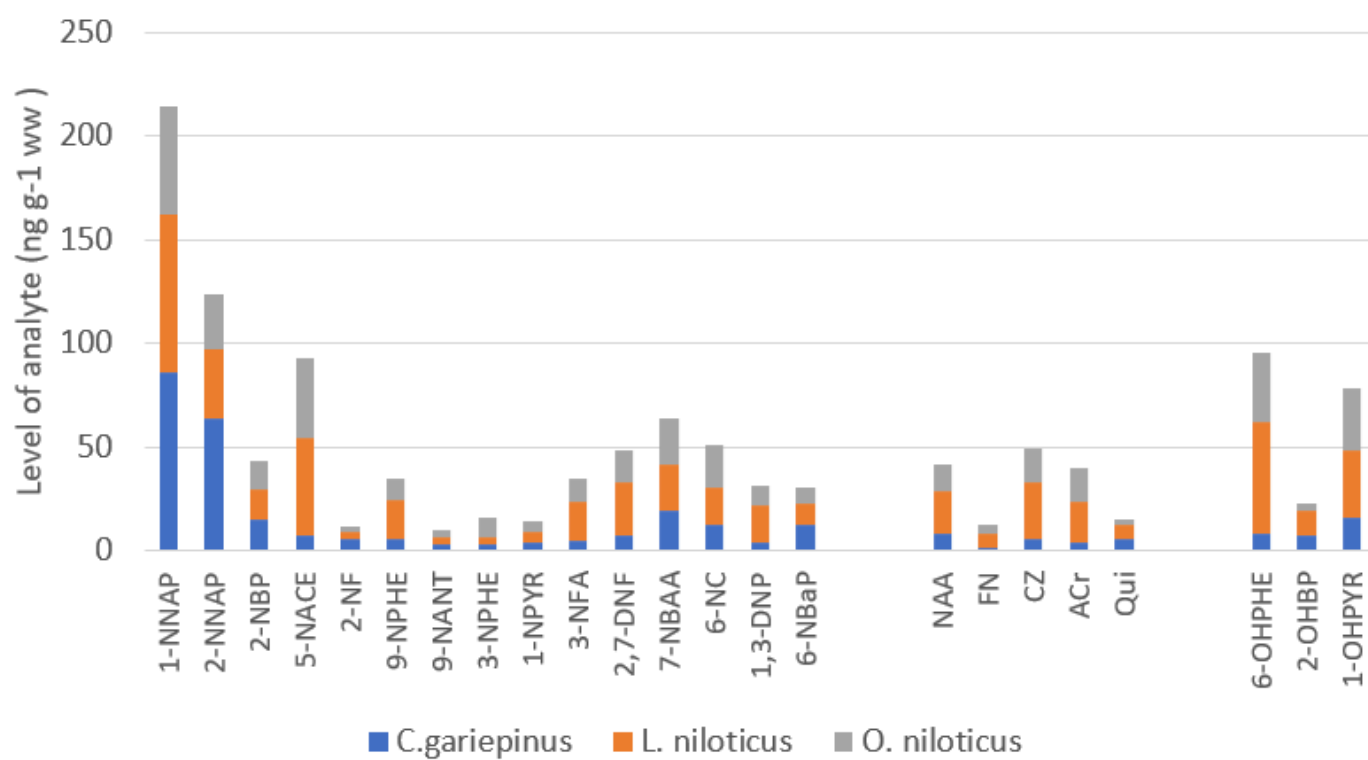


Figure 2

Levels of NPAHs, OPAHs and OHPAHs in three fish species from the White Nile (all sites A, B, and C).

2D and 3D PCA loading plot for Sediments

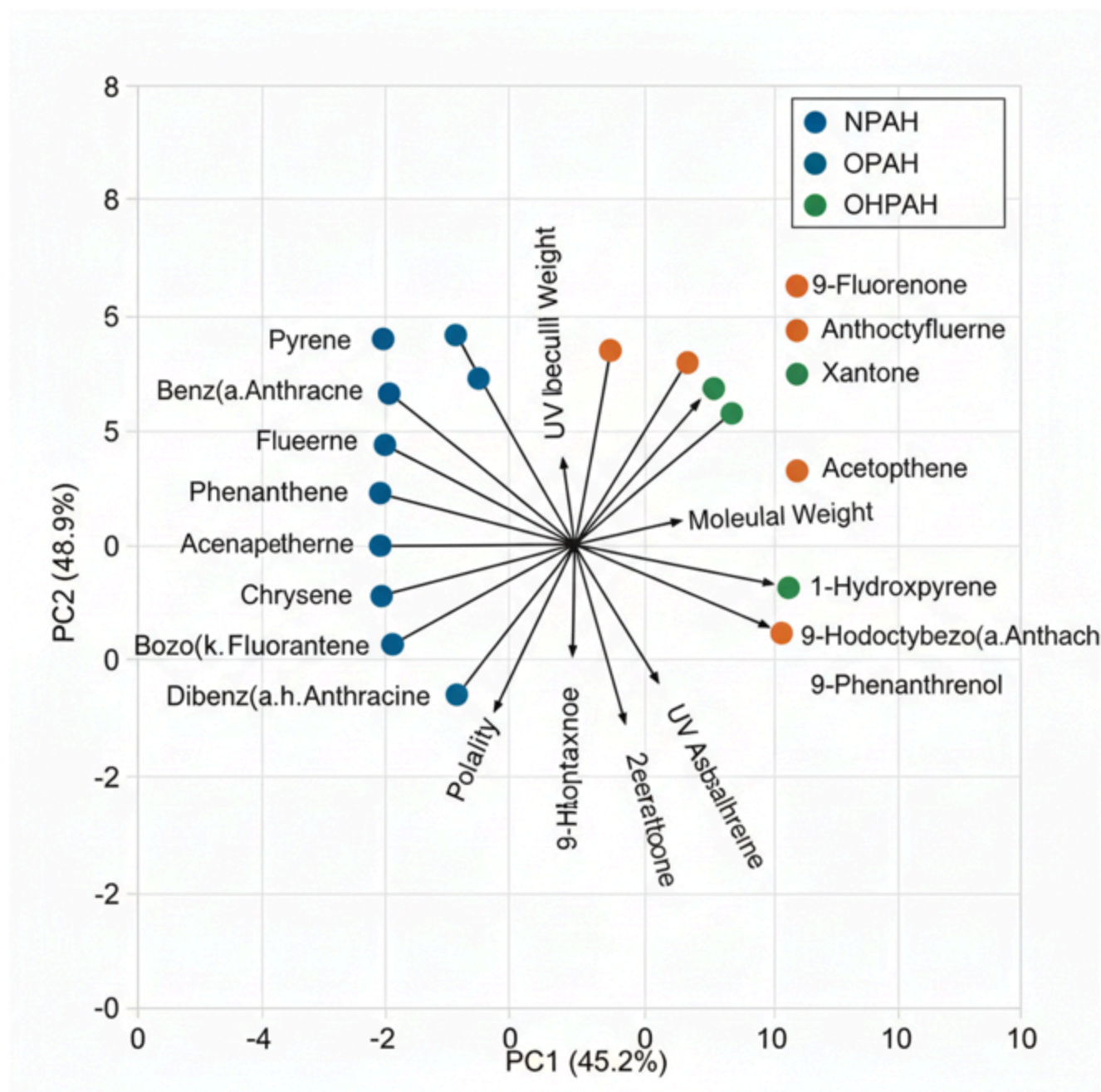


Figure 3

Principal Component Analysis (PCA) loading plots for OPAH, NPAH, and OHPAH analytes. 2D loading plot (PC1 vs. PC2) illustrating the primary variance. Vectors represent the relative contribution and correlation of each analyte; small angles between vectors suggest high positive correlation.

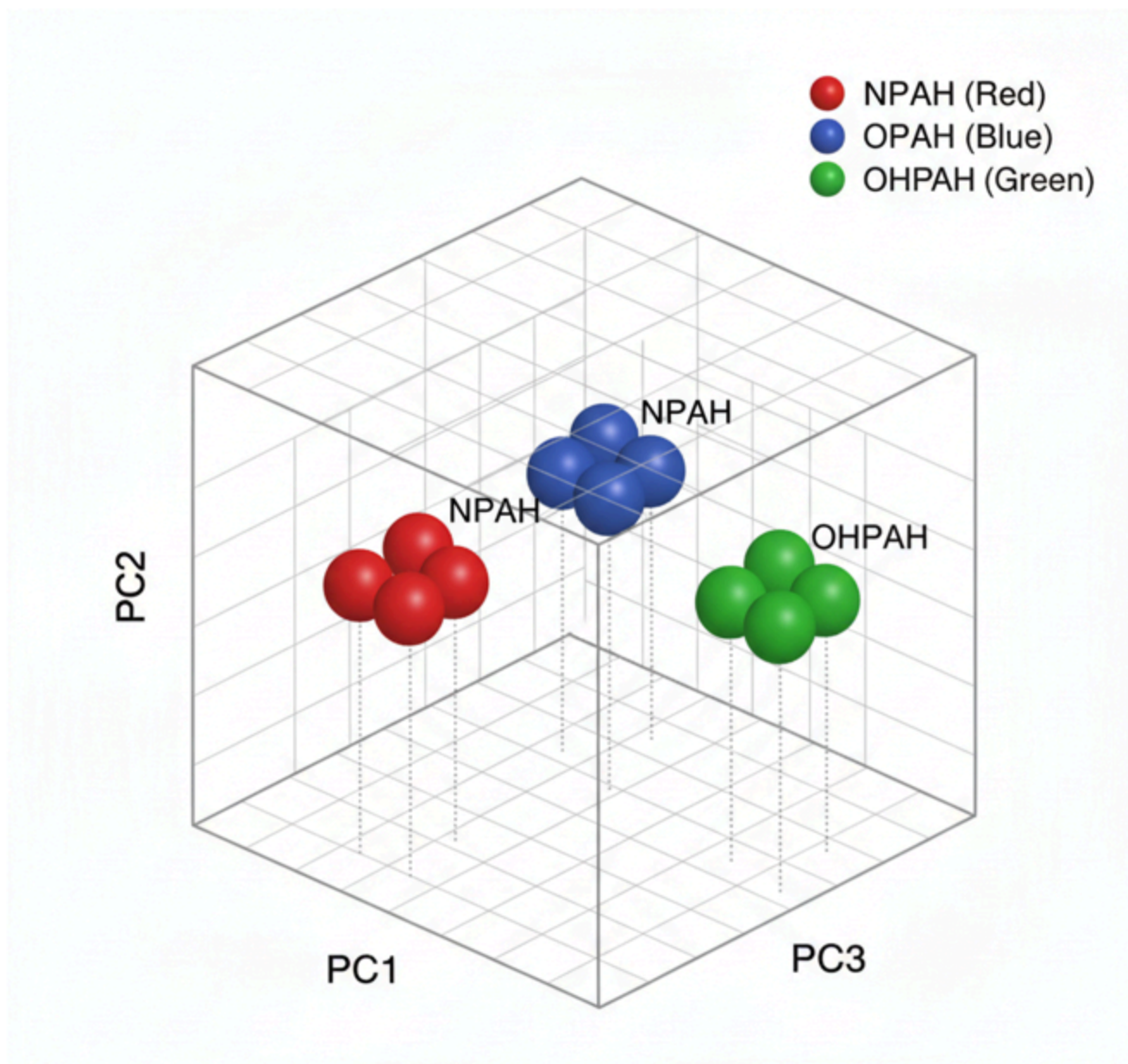


Figure 4

Principal Component Analysis (PCA) loading plots for OPAH, NPAH, and OHPAH analytes. 3D loading plot (PC1, PC2, and PC3) providing a more comprehensive view of the chemical distribution. Color-coding highlights analyte groups, while the sphere size and vector length indicate the strength of the loading on each principal component.

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