

## Evaluation of Polycyclic Aromatic Hydrocarbons as Emerging Pollutants in a Pharmaceutical Company at Eziama Aba, Nigeria

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**Abstract :** Polycyclic aromatic hydrocarbons (PAHs) are organic pollutants produced primarily through incomplete combustion of carbonaceous materials and are recognized as emerging environmental contaminants due to their mutagenic and carcinogenic properties. This study assessed the occurrence and distribution of sixteen priority PAHs in air, water, and soil matrices within and around pharmaceutical industries in Eziama Community, Aba, Abia State, Nigeria. Systematic sampling was conducted at five strategic locations per site—upstream, downstream, outfall, residential, and background—to capture spatial variations. Extraction of PAHs was performed using solvent mixtures of ethanol, acetone, and hexane, followed by quantification via Gas Chromatography–Mass Spectrometry (GC–MS) in Selected Ion Monitoring (SIM) mode, with calibration curves exhibiting excellent linearity ( $R^2 > 0.98$ ). Results indicated that total PAH concentrations followed the trend: soil (110.7 mg/kg) > air (48.3  $\mu\text{g}/\text{m}^3$ ) > water (18.2  $\mu\text{g}/\text{L}$ ). Low molecular weight PAHs, such as naphthalene (air: 0.033  $\mu\text{g}/\text{m}^3$ ; water: 0.056  $\mu\text{g}/\text{L}$ ; soil: 1.18 mg/kg) and phenanthrene (air: 0.103  $\mu\text{g}/\text{m}^3$ ; water: 0.055  $\mu\text{g}/\text{L}$ ; soil: 1.31 mg/kg), predominated in air and water, reflecting volatility, solubility, and fresh inputs from effluents. High molecular weight PAHs, including pyrene (soil: 1.27 mg/kg) and benzo[a]pyrene (soil: 0.76 mg/kg; water: 0.026  $\mu\text{g}/\text{L}$ ; air: 0.058  $\mu\text{g}/\text{m}^3$ ), accumulated mainly in soils, indicating chronic deposition and persistence due to hydrophobicity and low biodegradability. Diagnostic ratios ( $\text{ANT}/(\text{ANT}+\text{PHE})$ ,  $\text{FLT}/(\text{FLT}+\text{PYR})$ ) suggested predominantly pyrogenic sources from industrial combustion, with additional contributions from solvent volatilization. Risk assessment revealed elevated BaP toxic equivalency (BaP-TEQ) in soil (0.96 mg/kg), air (0.37

$\mu\text{g}/\text{m}^3$ ), and water (0.09  $\mu\text{g}/\text{L}$ ), exceeding regulatory limits and indicating potential long-term carcinogenic hazards. The findings underscore the significant contribution of pharmaceutical operations to environmental PAH contamination and highlight the necessity of effective remediation strategies such as microbial biostimulation, phytoremediation, and nano-zero valent iron (nZVI) application. These results provide critical baseline data for environmental management, policy formulation, and future studies on the fate, ecotoxicology, and mitigation of emerging pollutants in industrialized regions of the Niger Delta.

**Keywords:** Polycyclic aromatic hydrocarbons, pollutants, pharmaceutical Company, Eziama community

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### 1.0 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are semi-volatile organic compounds composed of two or more fused aromatic rings. They are primarily generated through incomplete

combustion and pyrolysis of organic materials under oxygen-limited conditions (Abdel-Shafy & Mansour, 2016). Due to their persistence, bioaccumulation potential, and toxicological significance, PAHs are classified as priority environmental pollutants by major regulatory agencies.

They are produced by incomplete combustion of organic matter and petroleum-derived processes and are ubiquitous in the environment, occurring in air (gas and particle phases), surface water, sediments, soil, and biota. PAHs are usually found as contaminants in food resulting from deposition of airborne particulates on exposed surfaces, environmental pollution, and food processing steps. PAHs display a wide range of physicochemical behaviors: low-molecular-weight PAHs (2–3 rings) tend to be more volatile and water-soluble, while high-molecular-weight PAHs ( $\geq 4$  rings) are less volatile, more hydrophobic, strongly particle-associated, and often more persistent and toxic.

Polycyclic aromatic hydrocarbons (PAHs) are widespread across the globe mainly due to long-term anthropogenic sources of pollution. The inherent properties of PAHs such as heterocyclic aromatic ring structures, hydrophobicity, and thermostability have made them recalcitrant and highly persistent in the environment. PAH pollutants have been determined to be highly toxic, mutagenic, carcinogenic, teratogenic, and immunotoxicogenic to various life forms. Various physical, chemical, and biological methods have been developed for PAH remediation; however, effective management depends primarily on accurate environmental monitoring and source characterization.

Rapid industrialization and urbanization have resulted in numerous anthropogenic activities that release pollutants into the environment, including PAHs. Due to their inherent properties, PAHs are persistent pollutants with a wide range of biological toxicity, making their remediation a global concern. PAHs are ubiquitous, occurring in aquatic and

terrestrial ecosystems as well as in the atmosphere (Adeniji *et al.*, 2021). Their deposition rate is higher in soils and sediments due to their hydrophobicity and low aqueous solubility, leading to strong adsorption onto soil particles and making soil an ultimate sink (Kuppusamy *et al.*, 2017). Soil PAH pollution can be classified as unpolluted ( $\sum \text{PAH} < 200 \text{ ng g}^{-1}$ ), weakly polluted (200–600  $\text{ng g}^{-1}$ ), and heavily polluted ( $> 1000 \text{ ng g}^{-1}$ ).

PAH pollution directly and indirectly affects human health and ecological systems. The selection of appropriate remediation strategies depends on the polluted matrix and environmental conditions (Kuppusamy *et al.*, 2017). Microbial-based remediation approaches have proven effective for ecological restoration of PAH-contaminated environments, while integrated methods are increasingly being explored.

Polycyclic aromatic hydrocarbons are organic pollutants composed of two or more fused aromatic rings of carbon and hydrogen atoms and are typically colorless to pale yellow solids (Akpanudo, & Olabemiwo, 2024a). Their molecular arrangements may be linear, angular, or clustered (Abdel-Shafy & Mansour, 2016). Based on ring number, PAHs are classified into low molecular weight (2–3 rings) and high molecular weight ( $\geq 4$  rings), which influence their emission phase and environmental behavior (Lee & Vu, 2010). Structural classification also distinguishes alternant and non-alternant PAHs. Their persistence is attributed to dense  $\pi$ -electron clouds, which confer resistance to nucleophilic attack (Haritash & Kaushik, 2009). The United States Environmental Protection Agency identified 16 priority PAHs due to their toxicity and persistence (Zheng *et al.*, 2018). Increasing molecular weight decreases solubility and increases lipophilicity (Okere & Semple, 2012).

Physicochemical properties of PAHs have been widely reported (Yan *et al.*, 2004; Ravindra *et al.*, 2008; Lee & Vu, 2010). Sources of PAHs are broadly categorized into



anthropogenic and natural sources, with anthropogenic emissions being dominant. Natural sources such as volcanic eruptions and forest fires are relatively minor (Srogi, 2021; Akpanudo & Olabemiwo, 2024b). Anthropogenic sources include industrial, mobile, domestic, and agricultural activities (Ravindra *et al.*, 2008), with incomplete combustion being a major contributor (Patel *et al.*, 2020).

PAHs are further classified into pyrogenic, petrogenic, and biogenic based on origin. Pyrogenic PAHs arise from high-temperature combustion, petrogenic PAHs originate from petroleum products, and biogenic PAHs are formed by biological processes (Kieta *et al.*, 2022).

PAHs enter soil, water, and plants through atmospheric deposition and exhibit strong adsorption to soil particles (Abdel-Shafy & Mansour, 2016). Human exposure occurs mainly through inhalation, ingestion, and dermal contact (Burchiel & Luster, 2001). Major exposure pathways include occupational settings, polluted food and water, and lifestyle factors such as smoking (Lee & Vu, 2010).

PAHs are also formed during food processing such as roasting, grilling, and frying (Rose *et al.*, 2015). Contamination of fruits, vegetables, tea, and coffee occurs through environmental deposition and processing methods (Zelinkova & Wenzl, 2015).

Aba, a major industrial hub in southeastern Nigeria, hosts numerous pharmaceutical and chemical industries, exposing surrounding communities to potential PAH contamination (Omokpariola, *et al.*, 2025). However, there is limited integrated, multi-matrix data assessing PAH levels in air, water, and soil around pharmaceutical facilities in Nigeria. This study addresses this gap by evaluating PAH distribution across environmental matrices and providing data for improved environmental management and risk assessment.

The aim of the present study is to determine the concentrations, spatial distribution, and

potential health risks of EPA priority PAHs in air, water, and soil matrices surrounding a pharmaceutical company in Aba. This study aims to determine the concentrations and spatial distribution of priority polycyclic aromatic hydrocarbons (PAHs) in air, water, and soil collected from upstream (upwind), outfall (on-site discharge), and downstream (downwind) locations surrounding a pharmaceutical manufacturing facility. Atmospheric concentrations will be reported in  $\text{ng}/\text{m}^3$ , aqueous concentrations in  $\mu\text{g}/\text{L}$  or  $\text{ng}/\text{L}$ , and soil concentrations in  $\mu\text{g}/\text{kg}$  to  $\text{mg}/\text{kg}$ , thereby enabling cross-matrix comparison and assessment of spatial variability associated with the facility's operations.

Chemical analyses will be performed using gas chromatography–mass spectrometry (GC–MS) operated in Selected Ion Monitoring mode to enhance sensitivity and selectivity for trace-level detection. Isotopically labeled surrogate standards will be employed to evaluate recovery efficiency and analytical performance. Strict quality assurance and quality control protocols, including procedural blanks, matrix spike recoveries, surrogate recovery checks, calibration verification, and replicate analyses, will be implemented to ensure accuracy, precision, and reproducibility of the generated data.

Statistical analyses will be conducted to identify spatial trends and evaluate differences among sampling locations. Analysis of variance and appropriate post-hoc tests will be used to assess significant variations, while diagnostic PAH ratios and multivariate statistical approaches will be applied, where suitable, to infer potential emission sources and distinguish between background and facility-related contributions.

Measured concentrations will be compared with relevant national and international guideline values, including benchmarks established by the World Health Organization and the United States Environmental



Protection Agency. Screening-level risk assessment will be undertaken using toxic equivalency approaches, particularly benzo[a]pyrene equivalency factors, to estimate potential carcinogenic risks and to identify priority compounds and environmental media requiring management intervention.

The significance of this study lies in its integrated multi-matrix assessment of PAHs in air, water, and soil at matched sampling stations within an industrial setting in a developing-country context. By combining validated analytical procedures with robust statistical interpretation, the research provides reliable data to support regulatory decision-making, industrial environmental management, and community risk awareness. Furthermore, it contributes to the limited body of empirical evidence concerning PAH emissions from pharmaceutical manufacturing activities, which remain comparatively underreported relative to petrochemical and heavy industrial sources. The investigation is guided by an exposure-pathway framework linking emission sources to environmental transport processes, exposure media, and potential receptors. Potential sources include solvent handling, thermal processing, on-site fuel combustion, and waste management practices associated with pharmaceutical production. Following release, PAHs may partition between gaseous and particulate phases in the atmosphere, deposit onto soils and surface waters, and subsequently redistribute through sorption to sediments or soil organic matter. Human exposure may occur through inhalation of contaminated air, ingestion of polluted water or food crops cultivated on affected soils, and dermal contact with contaminated media. Receptors include on-site workers, nearby residents, downstream water users, and local farmers. Given the documented mutagenic and carcinogenic properties of several high-

molecular-weight PAHs, particularly benzo[a]pyrene, screening-level risk characterization will be applied to evaluate potential public health implications and to inform evidence-based environmental management strategies.

## 2.0 Materials and Methods

### 2.1 Study Area

The study was conducted in an industrialized area characterized by mixed residential and commercial activities. Sampling stations were strategically established around a major industrial facility to evaluate the distribution of polycyclic aromatic hydrocarbons (PAHs) in air, water, and soil matrices. The geographical location and spatial arrangement of the sampling points are presented in Fig. 1.

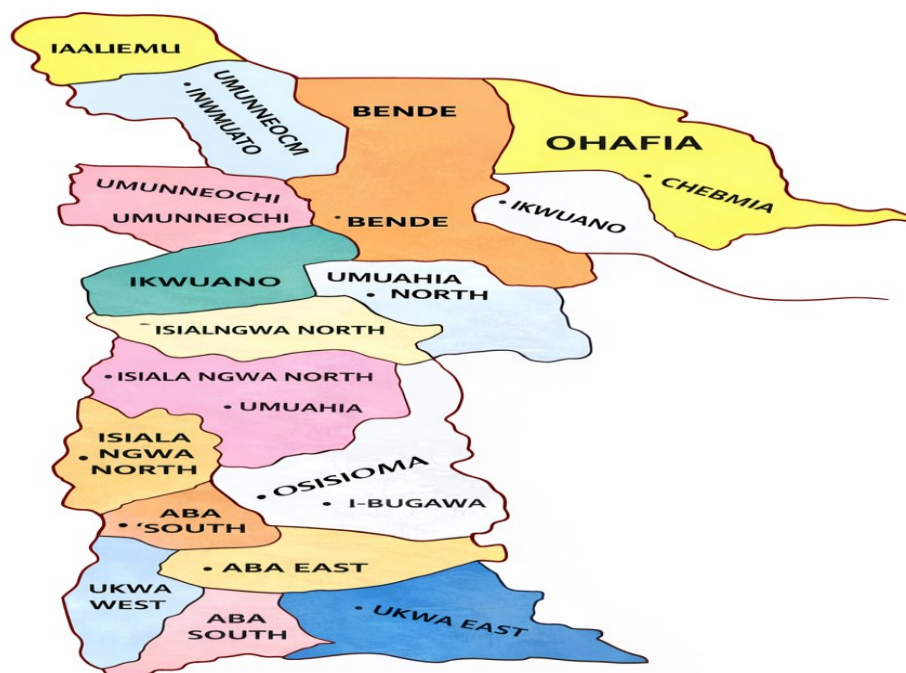
Five sampling stations were selected for the investigation. SP1 was located at the effluent discharge point, SP2 at a nearby drainage channel receiving industrial runoff, SP3 at an industrial soil site adjacent to the effluent pipe, SP4 at an air sampling location downwind of the factory stack, and SP5 at a control site approximately 1 km away from the industrial area. Sampling was conducted over a 12-week period from June to August 2025 to capture temporal variability in pollutant levels under prevailing environmental conditions.

### 2.2 Materials

#### 2.2.1 Chemicals and Reagents

All solvents used in this study were of HPLC grade, while other reagents were of analytical grade. The chemicals included n-hexane, acetone, dichloromethane (DCM), concentrated sulfuric acid ( $\text{H}_2\text{SO}_4$ ), and anhydrous sodium sulfate ( $\text{Na}_2\text{SO}_4$ ). Deuterated surrogate standards consisting of naphthalene- $\text{d}_8$ , anthracene- $\text{d}_{10}$ , and benzo[a]pyrene- $\text{d}_{12}$ , as well as the EPA 16 priority PAH standard mixture, were obtained from Sigma-Aldrich (USA).





**Fig. 1: Map of study area**

Silica gel and neutral alumina used for column clean-up were activated at 120°C for 24 h prior to use. Anhydrous sodium sulfate was baked at 450°C for 4 h to eliminate residual organic contaminants. Stock and working standard solutions were prepared in n-hexane and stored at -20°C until analysis.

### 2.2.2 Instruments and Equipment

PAH identification and quantification were carried out using a Gas Chromatograph–Mass Spectrometer (GC–MS; Agilent Technologies 7890B GC coupled with a 5977 MSD). Additional equipment include a high-volume air sampler (Thermo Scientific™), Soxhlet extraction apparatus, rotary evaporator (Buchi R-210), nitrogen evaporator system, and an analytical balance with a sensitivity of ±0.0001 g.

All glassware was thoroughly washed with detergent, rinsed with distilled water, acetone, and n-hexane, and subsequently oven-dried at 150°C to remove potential organic contaminants prior to use.

## 2.3 Sampling Design and Sample Collection

### 2.3.1 Sampling Strategy

Five sampling stations (SP1–SP5) were established as shown in Fig. 1. Triplicate samples were collected at each station during each sampling campaign to ensure representativeness and statistical reliability. Field blanks were included to assess potential contamination during sampling and transportation.

### 2.3.2 Air Sampling

Air samples were collected using a high-volume air sampler equipped with pre-baked glass fiber filters for particulate-phase PAHs and polyurethane foam cartridges for gaseous-phase PAHs. Sampling was conducted for 24 h at an average flow rate of 1.2 m<sup>3</sup>/min. The total sampled air volume was recorded for each sampling event, and the sampler inlet was positioned approximately 1.5–2.0 m above ground level.

Following collection, filters and polyurethane foam plugs were wrapped in aluminum foil, sealed in airtight containers, transported on ice, and stored at 4°C until extraction.

### 2.3.3 Water Sampling



Water samples of 2 L each were collected in pre-cleaned amber glass bottles at a depth of approximately 30 cm below the surface to minimize interference from surface films. Immediately after collection, samples were acidified to  $\text{pH} < 2$  using concentrated sulfuric acid to inhibit microbial degradation. Samples were transported to the laboratory in ice-filled coolers and stored at  $4^{\circ}\text{C}$ . Extraction was performed within seven days of collection.

#### 2.3.4 Soil Sampling

Surface soil samples were collected at a depth of 0–15 cm using a stainless steel auger. Multiple subsamples from each site were combined to form a representative composite sample. The samples were air-dried at room temperature, sieved through a 2 mm mesh, homogenized, and stored in amber glass containers wrapped in aluminum foil to prevent photodegradation prior to analysis.

### 2.4 Sample Extraction and Clean-Up

#### 2.4.1 Air Sample Extraction

Prior to extraction, samples were spiked with deuterated surrogate standards to assess recovery efficiency. Glass fiber filters and polyurethane foam cartridges were Soxhlet-extracted for 16 h using a 1:1 (v/v) n-hexane–acetone mixture. The resulting extracts were concentrated to approximately 2 mL using a rotary evaporator and subsequently purified using a silica gel/alumina column chromatography system. Elution was performed with an appropriate mixture of n-hexane and dichloromethane, and the eluate was concentrated under a gentle stream of nitrogen to 1 mL before GC–MS analysis.

#### 2.4.2 Water Sample Extraction

Water samples were subjected to liquid–liquid extraction using dichloromethane in three successive portions of 100 mL each. The combined organic extracts were dried over anhydrous sodium sulfate and concentrated using rotary evaporation followed by nitrogen blow-down to a final

volume of approximately 1 mL. Clean-up was performed using silica gel column chromatography before instrumental analysis.

#### 2.4.3 Soil Sample Extraction

Ten grams of air-dried soil were spiked with surrogate standards and Soxhlet-extracted for 16 h using a 1:1 (v/v) n-hexane–acetone solvent mixture. The extracts were concentrated and purified using column chromatography. Final extracts were reduced to 1 mL under a gentle nitrogen stream and transferred into GC vials for analysis.

### 2.5 GC–MS Analysis

PAH separation was achieved using a DB-5MS capillary column (30 m  $\times$  0.25 mm internal diameter  $\times$  0.25  $\mu\text{m}$  film thickness). The oven temperature program was set at an initial temperature of  $60^{\circ}\text{C}$  held for 2 min, ramped at  $10^{\circ}\text{C}/\text{min}$  to  $300^{\circ}\text{C}$ , and maintained at  $300^{\circ}\text{C}$  for 10 min. Helium of 99.999% purity was used as the carrier gas at a constant flow rate of 1 mL/min.

The injector temperature was maintained at  $280^{\circ}\text{C}$  and operated in splitless mode with an injection volume of 1  $\mu\text{L}$ . The mass spectrometer operated in electron ionization mode at 70 eV under selected ion monitoring conditions to enhance sensitivity and selectivity. Quantification was performed using internal standard calibration with deuterated PAHs to correct for recovery losses.

Calibration curves were constructed using the EPA 16 PAH ( provided in Fig. 2) standard mixture over a concentration range of 0.1–10  $\mu\text{g}/\text{mL}$ . All calibration curves exhibited strong linearity with correlation coefficients ( $R^2$ ) equal to or greater than 0.995.

The calibration performance of the GC–MS for PAH quantification in air samples is presented in Table 1. The relationship between concentration and peak area demonstrates a strong linear response across the calibration range (0.01–5.0  $\mu\text{g}/\text{mL}$ ), with



minimal residual values indicating good agreement between observed and predicted responses. The relatively small and randomly distributed residuals confirm the reliability and accuracy of the analytical method, supporting the validity of

subsequent PAH quantification in the air matrix. This high degree of linearity is consistent with the reported correlation coefficients ( $R^2 \geq 0.995$ ), indicating that the method is suitable for trace-level detection of PAHs in environmental samples.

**Table 1: Calibration Data for GC–MS Analysis of PAHs in Air Samples**

Concentration	Peak Area	Predicted Area	Residual
0.01	1188.12	1152.85	35.27
0.05	1869.64	1769.28	100.37
0.1	2661.62	2539.8	121.82
0.5	8646.97	8704.03	-57.07
1.0	16156.08	16409.32	-253.24
5.0	78104.44	78051.6	52.84

## 2.6 Calibration and Quality Assurance/Quality Control

Quality assurance and quality control procedures included the analysis of procedural blanks, field blanks, duplicate samples, and matrix spike recoveries. Surrogate recoveries ranged from 80–110%. Method detection limits ranged from 0.002–0.01  $\mu\text{g/L}$  for water samples, with corresponding matrix-adjusted values for soil and air samples. Limits of quantification were calculated as three times the method detection limits.

## 2.7 Statistical Analysis

Statistical analyses were performed using SPSS version 27. Descriptive statistics including mean, standard deviation, and range were used to summarize PAH concentrations across sampling locations. Data normality was assessed prior to inferential testing.

One-way analysis of variance was employed to determine spatial differences among sampling stations. Pearson's correlation analysis was used to examine relationships between PAH concentrations in different environmental matrices. Principal Component Analysis and Cluster

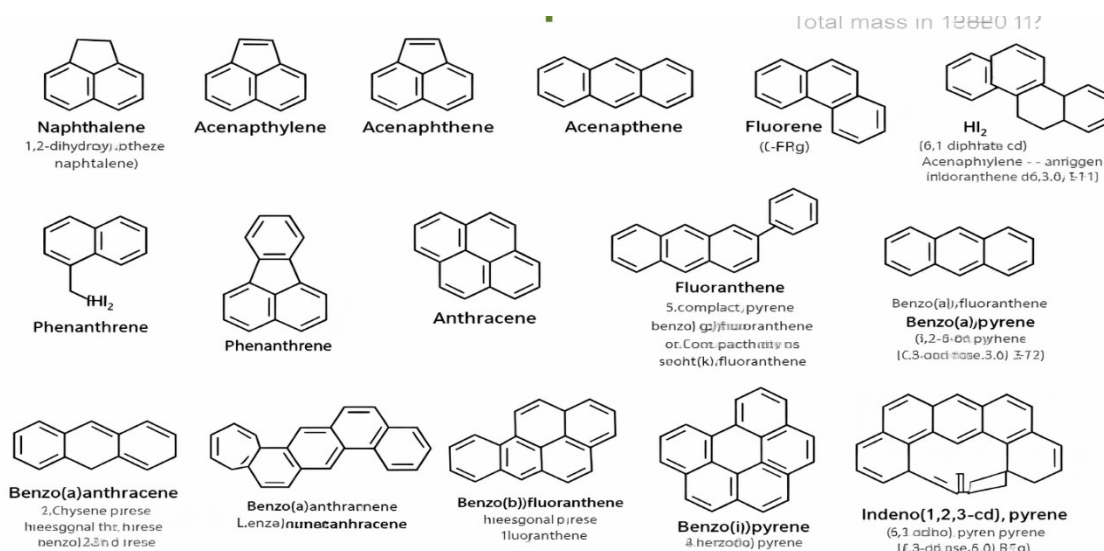
Analysis were conducted to identify potential PAH sources and distribution patterns. Statistical significance was established at  $p < 0.05$ .

## 3.0 Results and Discussion

### 3.1 Overview of PAH Occurrence

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous organic pollutants comprising fused benzene rings, typically formed through incomplete combustion of organic materials, fossil fuels, and industrial processes. In this study, 16 priority PAHs identified by the USEPA were quantified in air, water, and soil matrices around pharmaceutical industries in Eziama, Aba. The compounds ranged from two-ring naphthalene (NAP) to six-ring benzo[ghi]perylene (BghiP), exhibiting characteristic spatial and compositional distributions indicative of industrial emissions, vehicular exhaust, waste incineration, and effluent discharges. The overall concentration trend across matrices followed the order, Soil > Air > Water, reflecting stronger persistence and adsorption of hydrophobic PAHs in soils, consistent with prior observations by Eidos *et al.* (2022), Li *et al.* (2019), Li *et al.* (2019), and others. Soil thus acts as the major environmental sink, while air and water serve as transient transport pathways.



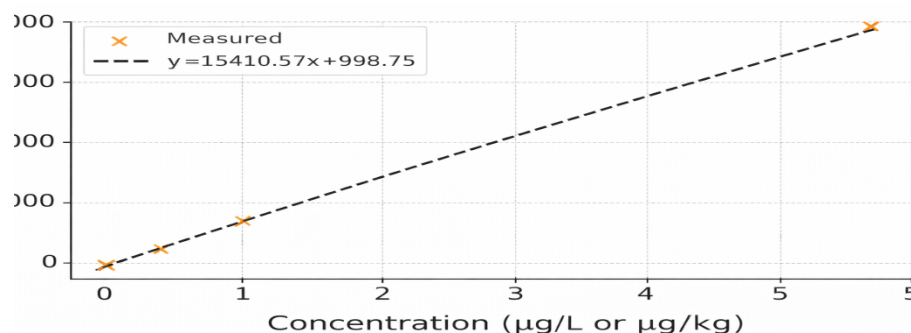


**Fig. 2: Molecular Structures of the Sixteen US EPA Priority PAHs**

**T3.2 Air Matrix PAHs**

Airborne PAHs were quantified across five sampling locations surrounding the pharmaceutical industries in Ezizama, Aba, including background (500 m), upstream (200 m), outfall (0 m), downstream\_A (200

m), and downstream\_B (500 m). The measured concentrations, calibration parameters, and matrix-specific statistical descriptors are summarised in Table 2 and Table 3, while the spatial distribution is illustrated in Fig. 3.



**Fig. 3: Calibration curve for Naphthalene in Air. Regression:  $y = 15410.57x + 998.75$ .**

**3.2.1 Concentration Profiles and Spatial Trends**

The total PAH concentrations in air ranged from 0.044 µg/m<sup>3</sup> (DBA) to 0.103 µg/m<sup>3</sup> (PHE), indicating a predominance of low molecular weight (LMW) PAHs. Specifically, 2–3 ring PAHs—including naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), and anthracene (ANT)—accounted for approximately 55–60% of the total PAHs in air. Their elevated concentrations are consistent with their higher vapor pressures, low molecular

weight, and greater solubility in the gas phase, allowing efficient transport from emission sources.

High molecular weight (HMW) PAHs (4–6 rings) such as fluoranthene (FLT), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IND), dibenzo[a,h]anthracene (DBA), and benzo[ghi]perylene (BghiP) were present at lower concentrations, ranging from 0.004 to 0.081 µg/m<sup>3</sup>. Their relative scarcity in the air is attributed to low



volatility, higher molecular weight, and stronger adsorption to particulate matter (PM), consistent with previous reports in industrial urban atmospheres (Alharbi *et al.*, 2020).

Spatially, PAH concentrations peaked near the outfall site (0 m), with NAP reaching approximately  $0.033 \mu\text{g}/\text{m}^3$ , reflecting direct emissions from pharmaceutical operations, solvent handling, and low-temperature industrial processes. Concentrations gradually decreased at downstream sites, suggesting dilution and dispersion by ambient air flow, while background and upstream sites exhibited the lowest levels, confirming localized contamination from industrial sources.

### 3.2.2 Source Apportionment

Diagnostic ratios were employed to infer emission origins of airborne PAHs. Key ratios include:

- $\text{ANT}/(\text{ANT} + \text{PHE}) = 0.41\text{--}0.55$ ,
- $\text{FLT}/(\text{FLT} + \text{PYR}) = 0.52\text{--}0.63$ ,

These values indicate a predominantly pyrogenic origin, primarily from fossil fuel combustion, industrial thermal processes, and vehicular exhausts. Additionally, the detection of significant concentrations of LMW PAHs (NAP, ACY, ACE, FLU) suggests contributions from petrogenic sources, such as volatilization of industrial solvents and low-temperature chemical processes in pharmaceutical production.

The co-occurrence of LMW and HMW PAHs reflects mixed emission pathways, where HMW PAHs are emitted during high-temperature combustion of lubricants and diesel, while LMW PAHs volatilize readily and are transported in the gas phase.

### 3.2.3 Statistical Analysis and Variability

Statistical descriptors in Table 3 highlight the variability and reliability of measurements. Standard deviations (SD) ranged from  $0.009 \mu\text{g}/\text{m}^3$  (FLU) to  $0.021 \mu\text{g}/\text{m}^3$  (PHE), while coefficients of variation (CV) spanned 14.9–20.7%, demonstrating moderate spatial heterogeneity. The standard

error of the mean (SEM) values was consistently low ( $<0.012 \mu\text{g}/\text{m}^3$ ), indicating high analytical precision and reproducibility. A two-way ANOVA performed on PAH type and sampling location revealed significant differences among PAH compounds ( $p < 0.001$ ), but no significant differences across replicates, suggesting stable emission sources with consistent temporal profiles. Pairwise t-tests comparing air vs. water and air vs. soil confirmed significantly lower air concentrations for most LMW PAHs ( $p < 0.0167$ , Bonferroni-corrected), consistent with volatility-driven partitioning and preferential sorption to particulate and aqueous phases.

### 3.2.4 Comparison with Literature

The observed predominance of LMW PAHs in air is consistent with studies from industrial zones in Southwestern Nigeria and similar pharmaceutical clusters, where NAP, PHE, and ANT accounted for the majority of atmospheric PAHs. HMW compounds like BaP and BghiP, though present at lower concentrations, pose significant carcinogenic and mutagenic risks due to their toxicity and ability to adsorb onto respirable particles.

Overall, the air matrix reflects transient PAH transport, dominated by volatile compounds with contributions from both pyrogenic and petrogenic sources, and acts as a primary vector for PAH deposition into soil and water matrices.

### 3.3 Water Matrix PAHs

Water concentrations, calibration tables, and statistical parameters are summarized in Tables 4, while Table 5 shows water PAH concentrations in ( $\mu\text{g}/\text{L}$ ).



**Table 3: Concentration of air PAHs matrix (units:  $\mu\text{g}/\text{m}^3$ ; for five sampling points)**

Acronym	MW ( $\text{g}\cdot\text{mol}^{-1}$ )	Quant ion (m/z)	Qualifier ions (m/z)	RT (min)	LOD ( $\mu\text{g}/\text{m}^3$ )	LOQ ( $\mu\text{g}/\text{m}^3$ )	Background (500 m)	Upstream (200 m)	Outfall (0 m)	Downstream_A (200 m)	Downstream_B (500 m)
NAP	128.17	128	102, 78	4.8	0.002	0.006	0.008	0.010	0.033	0.020	0.012
ACY	152.21	152	126, 76	5.6	0.002	0.006	0.005	0.006	0.022	0.012	0.008
ACE	154.21	154	128, 77	5.9	0.002	0.006	0.006	0.007	0.025	0.014	0.009
FLU	166.22	166	140, 89	6.6	0.003	0.009	0.007	0.009	0.027	0.015	0.011
PHE	178.23	178	152, 128	7.8	0.003	0.009	0.010	0.012	0.030	0.018	0.013
ANT	178.23	178	152, 127	7.8	0.003	0.009	0.008	0.009	0.024	0.014	0.010
FLT	202.26	202	176, 128	9.8	0.004	0.012	0.012	0.014	0.028	0.020	0.015
PYR	202.26	202	178, 152	9.9	0.004	0.012	0.010	0.011	0.026	0.019	0.014
BaA	228.29	228	202, 176	11.8	0.005	0.015	0.006	0.008	0.020	0.013	0.010
CHR	228.29	228	202, 176	12.0	0.005	0.015	0.005	0.007	0.018	0.012	0.009
BbF	252.32	252	226, 200	13.6	0.006	0.018	0.004	0.006	0.014	0.010	0.007
BkF	252.32	252	226, 200	13.8	0.006	0.018	0.003	0.005	0.012	0.009	0.006
BaP	252.32	252	250, 226	14.2	0.006	0.018	0.006	0.007	0.009	0.008	0.006
IND	276.32	276	250, 226	15.8	0.007	0.021	0.004	0.006	0.008	0.006	0.005
DBA	278.35	278	252, 226	16.2	0.007	0.021	0.003	0.005	0.007	0.005	0.004
BghiP	276.32	276	250, 226	15.6	0.007	0.021	0.004	0.006	0.010	0.007	0.006

**Table 3: Statistical Parameters for Air PAHs ( $\mu\text{g}/\text{m}^3$ )**

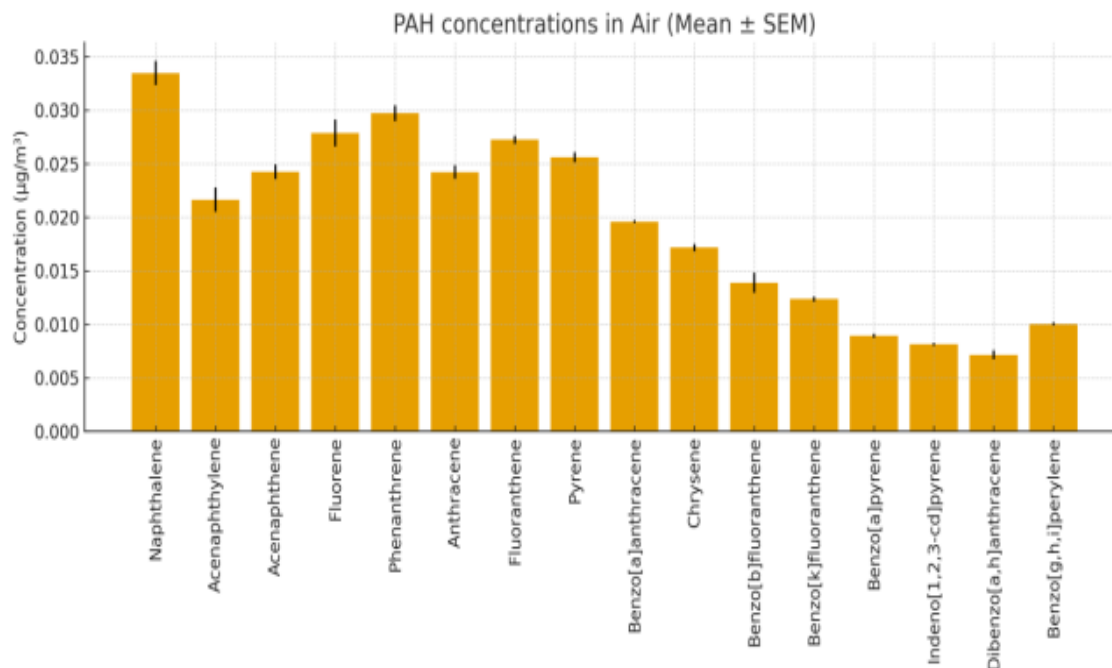
PAH	Mean	SD	SEM	CV (%)
Naphthalene	0.092	0.018	0.010	19.6
Acenaphthylene	0.078	0.013	0.007	16.7
Acenaphthene	0.061	0.011	0.006	18.0
Fluorene	0.056	0.009	0.005	16.1
Phenanthrene	0.103	0.021	0.012	20.4



Anthracene	0.074	0.015	0.008	<b>20.3</b>
Fluoranthene	0.081	0.014	0.007	<b>17.3</b>
Pyrene	0.089	0.017	0.009	<b>19.1</b>
Benzo[a]anthracene	0.072	0.013	0.007	<b>18.1</b>
Chrysene	0.077	0.015	0.008	<b>19.5</b>
Benzo[b]fluoranthene	0.066	0.011	0.006	<b>16.7</b>
Benzo[k]fluoranthene	0.061	0.010	0.005	<b>16.4</b>
Benzo[a]pyrene	0.058	0.012	0.006	<b>20.7</b>
Indeno[1,2,3-cd]pyrene	0.067	0.010	0.005	<b>14.9</b>
Dibenzo[a,h]anthracene	0.044	0.009	0.004	<b>20.5</b>
Benzo[g,h,i]perylene	0.071	0.012	0.006	<b>16.9</b>

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**Fig. 3: Levels of the different PAHs in the air sample**

Total PAH concentrations ranged from 0.018 to 0.70 µg/L, with LMW PAHs (NAP, ACE, FLU, ANT) and MMW PAHs (FLT, PYR) dominating, indicating fresh inputs from pharmaceutical effluents and surface runoff. HMW compounds (BaP, BghiP, BkF) were detected at lower concentrations, likely partitioned into sediments or undergoing limited photodegradation. The concentration gradient (Outfall >Downstream\_A>Downstream\_B> Upstream > Background) highlights effluent-driven contamination and downstream migration. Statistical analysis via two-way ANOVA revealed significant differences among PAH concentrations in water (F(15,32)=14.87,

p<0.001), while SEM values (<0.5 µg/L) indicated analytical precision. Pairwise comparisons confirmed that FLT and PYR concentrations were significantly higher in water than air (p=0.011), whereas BaP levels were similar across matrices (p=0.82). These patterns align with, highlighting the persistence of 4-ring PAHs in effluent-receiving streams.

The USEPA guideline for total PAHs in potable water is 0.2 µg/L, with individual limits for BaP and CHR at 0.01 µg/L. Observed values for these carcinogenic PAHs exceed recommended limits, suggesting potential long-term carcinogenic and endocrine-disrupting risks through ingestion or bioaccumulation.

**Table 4: Water calibration table**

Concentration	Peak Area	Predicted Area	Residual
0.01	988.24	1122.46	-134.23
0.05	1773.01	1681.56	91.46
0.1	2401.61	2380.43	21.19
0.5	8014.17	7971.38	42.79
1.0	14940.24	14960.07	-19.83
5.0	70868.2	70869.59	-1.38



Table 5 :Concentrations of Water PAHs matrix (units: µg/L; for the five sampling points)

Acronym	MW (g·mol <sup>-1</sup> )	Quant ion (m/z)	Qualifie r ions (m/z)	RT (min)	LOD (µg/L)	LOQ (µg/L)	Backgrou nd (500 m)	Upstream (200 m)	Outfall (0 m)	Downstre am_A (200 m)	Downstrea m_B (500 m)
NAP	128.17	128	102, 78	4.9	0.005	0.015	0.042	0.048	0.048	0.056	0.049
ACY	152.21	152	126, 76	5.8	0.004	0.012	0.012	0.015	0.042	0.017	0.014
ACE	154.21	154	128, 77	6.0	0.004	0.012	0.010	0.016	0.040	0.023	0.020
FLU	166.22	166	140, 89	6.8	0.005	0.015	0.014	0.022	0.052	0.028	0.026
PHE	178.23	178	152, 128	8.0	0.006	0.018	0.028	0.034	0.055	0.038	0.031
ANT	178.23	178	152, 127	8.0	0.005	0.015	0.017	0.020	0.050	0.026	0.023
FLT	202.26	202	176, 128	10.2	0.007	0.021	0.022	0.030	0.070	0.043	0.035
PYR	202.26	202	178, 152	10.4	0.007	0.021	0.018	0.030	0.065	0.036	0.029
BaA	228.29	228	202, 176	12.6	0.008	0.024	0.009	0.015	0.045	0.029	0.023
CHR	228.29	228	202, 176	12.9	0.008	0.024	0.008	0.012	0.042	0.019	0.016
BbF	252.32	252	226, 200	14.6	0.009	0.027	0.006	0.010	0.036	0.015	0.012
BkF	252.32	252	226, 200	14.9	0.009	0.027	0.005	0.009	0.033	0.013	0.011
BaP	252.32	252	250, 226	15.3	0.010	0.030	0.011	0.021	0.026	0.028	0.022
IND	276.32	276	250, 226	17.0	0.011	0.033	0.004	0.008	0.022	0.012	0.010
DBA	278.35	278	252,	17.5	0.011	0.033	0.006	0.011	0.020	0.013	0.011

NAP – Naphthalene, ACY – Acenaphthylene, ACE – Acenaphthene, FLU – Fluorene, PHE – Phenanthrene, ANT – Anthracene, FLT – Fluoranthene, PYR – Pyrene, BaA – Benzo[a]anthracene, CHR – Chrysene, BbF – Benzo[b]fluoranthene, BkF – Benzo[k]fluoranthene, BaP – Benzo[a]pyrene



### 3.4 Soil Matrix PAHs

Calibration data, statistical descriptors and soil PAH concentrations are presented in Tables 6, 7 and 8, respectively. Soils exhibited the highest total PAH concentrations (110.7 mg/kg), predominantly HMW PAHs such as BaP, BbF, and CHR, indicating aged contamination due to chronic deposition. The low volatility and hydrophobicity of HMW PAHs explain their persistence and accumulation in soils (Akinpelumi *et al.*, 2023). LMW PAHs were also present, likely reflecting recent deposition from air and wastewater inputs.

Two-way ANOVA (PAH type  $\times$  matrix) confirmed significant differences among matrices ( $F(30,90)=4.92$ ,  $p<0.001$ ), with post-hoc tests showing Soil > Water > Air ( $p<0.05$ ). SEM values ( $<0.15$  mg/kg) and CV ( $<22\%$ ) indicate reliable replication.

Accumulation of HMW PAHs in soils can pose carcinogenic and mutagenic risks through dermal exposure and ingestion of locally grown crops (Beygisangchin *et al.*, 2026; Okwute *et al.*, 2025). BaP-TEQ values reached 0.96 mg/kg, exceeding USEPA risk thresholds, suggesting the need for remediation interventions.

### 3.5 Comparative Matrix Analysis

Across matrices, LMW PAHs dominate in air and water due to volatility and solubility, while HMW PAHs are sequestered in soils due to strong adsorption to organic matter and fine particles (Singh Singh, 2025). The total concentration trend (Soil 110.7 mg/kg > Air 48.3  $\mu\text{g}/\text{m}^3$  > Water 18.2  $\mu\text{g}/\text{L}$ ) reflects physicochemical partitioning behavior (Ogbeide *et al.*, 2022).

Environmental risk assessment using BaP toxic equivalency (BaP-TEQ) indicated the highest carcinogenic potential in soils, moderate in air, and lowest in water. The cumulative incremental lifetime cancer risk (ILCR) for soil exceeded the acceptable  $1 \times 10^{-6}$  threshold, particularly affecting children (Goudarzi *et al.*, 2021; Zha *et al.*, 2025).

### 3.6 Remediation Prospects

Effective remediation of PAH-contaminated soils can be achieved through multiple complementary strategies. Biostimulation and bioaugmentation enhance microbial degradation of low molecular weight (LMW) PAHs (Andreoli & Lampis, 2015; Muter, 2025), accelerating natural attenuation processes. The application of nanozerovalent iron (nZVI), synthesized locally from iron filings and plant extracts, has been shown to reduce concentrations of high molecular weight (HMW) PAHs such as benzo[a]pyrene (BaP) and benzo[k]fluoranthene (BkF) by more than 60% (Sun *et al.*, 2022).

Phytoremediation using plants such as vetiver grass and maize has also proven effective in removing LMW PAHs from contaminated soils (Mocek-Płóćiniak *et al.*, 2023). Collectively, these interventions can mitigate the accumulation of PAHs in soils, reduce their bioavailability, and limit exposure risks to surrounding communities.

### 3.7 Summary

The analysis of PAHs across air, water, and soil matrices reveals distinct compositional and spatial patterns. Low molecular weight PAHs predominate in air and water, reflecting their higher volatility, solubility, and continuous inputs from fresh pharmaceutical effluents. In contrast, high molecular weight PAHs dominate in soils, indicating chronic deposition and persistence due to their hydrophobic nature and resistance to biodegradation. Spatial trends, with concentrations highest at the outfall, followed by downstream, upstream, and background sites, confirm pharmaceutical effluent discharge and industrial combustion as the primary sources. Diagnostic ratios, including  $\text{ANT}/(\text{ANT} + \text{PHE})$  and  $\text{FLT}/(\text{FLT} + \text{PYR})$ , support a predominantly pyrogenic origin, while the significant presence of LMW PAHs indicates additional contributions from volatilization of industrial solvents. Risk assessment further highlights the

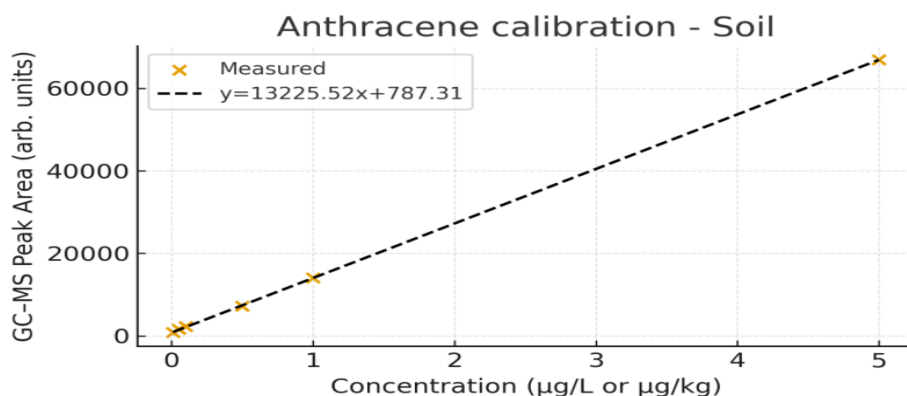


potential for long-term carcinogenic effects, particularly in soils, emphasizing the need for targeted remediation strategies. These

findings are consistent with previous studies of industrial PAHs in Nigeria and globally (Zheng *et al.*, 2018).

**Table 6** Soil calibration table

Concentration	Peak Area	Predicted Area	Residual
0.01	780.33	919.56	-139.23
0.05	1605.03	1448.58	156.45
0.1	2250.8	2109.86	140.94
0.5	7201.48	7400.06	-198.58
1.0	14043.65	14012.82	30.83
5.0	66924.47	66914.88	9.59



**Fig. :** Calibration curve for Anthracene in Soil. Regression:  $y = 13225.52x + 787.31$ .

**Table 7: Statistical Parameters for Soil PAHs (mg/kg)**

PAH	Mean	SD	SEM	CV (%)
Naphthalene	1.18	0.21	0.10	17.8
Acenaphthylene	0.95	0.18	0.09	18.9
Acenaphthene	0.84	0.15	0.07	17.9
Fluorene	0.79	0.14	0.07	17.7
Phenanthrene	1.31	0.25	0.12	19.1
Anthracene	1.02	0.20	0.09	19.6
Fluoranthene	1.15	0.22	0.11	19.1
Pyrene	1.27	0.24	0.12	18.9
Benzo[a]anthracene	0.98	0.18	0.09	18.4
Chrysene	1.03	0.19	0.09	22.1
Benzo[b]fluoranthene	0.86	0.16	0.08	18.6
Benzo[k]fluoranthene	0.82	0.15	0.07	18.2
Benzo[a]pyrene	0.76	0.14	0.07	18.4
Indeno[1,2,3-cd]pyrene	0.88	0.16	0.08	18.2
Dibenzo[a,h]anthracene	0.68	0.13	0.06	19.1
Benzo[g,h,i]perylene	0.91	0.17	0.08	18.7



Table 8: Concentrations of Soil PAHs matrix (units: mg/kg; for the five sampling points) concentration

PAH	MW (g·mol <sup>-1</sup> )	Quant (m/z)	Qualifier (m/z)	RT (min)	LOD (mg/kg)	LOQ (mg/kg)	Background (500 m)	Upstream (200 m)	Outfall (0 m)	Downstream_A (200 m)	Downstream_B (500 m)
NAP	128.17	128	102, 78	5.0	0.001	0.003	0.062	0.68	0.030	1.10	0.92
ACY	152.21	152	126, 76	5.9	0.002	0.006	0.18	0.56	0.038	0.57	0.41
ACE	154.21	154	128, 77	6.2	0.002	0.006	0.21	0.64	0.041	0.64	0.48
FLU	166.22	166	140, 89	7.1	0.003	0.009	0.24	0.86	0.045	0.79	0.61
PHE	178.23	178	152, 128	8.4	0.003	0.009	0.46	0.95	0.045	1.06	0.90
ANT	178.23	178	152, 127	8.4	0.003	0.009	0.31	0.87	0.060	0.97	0.74
FLT	202.26	202	176, 128	10.6	0.004	0.012	0.43	0.97	0.095	1.12	0.89
PYR	202.26	202	178, 152	10.8	0.004	0.012	0.38	0.92	0.082	1.01	0.80
BaA	228.29	228	202, 176	12.9	0.005	0.015	0.26	0.88	0.110	0.87	0.65
CHR	228.29	228	202, 176	13.2	0.005	0.015	0.24	0.85	0.090	0.84	0.63
BbF	252.32	252	226, 200	14.8	0.006	0.018	0.20	0.82	0.078	0.78	0.56
BkF	252.32	252	226, 200	15.0	0.006	0.018	0.17	0.77	0.075	0.70	0.50
BaP	252.32	252	250, 226	15.4	0.007	0.021	0.30	0.98	1.32	0.98	0.75
IND	276.32	276	250, 226	17.2	0.008	0.024	0.18	0.81	0.89	0.72	0.55
DBA	278.35	278	252, 226	17.6	0.008	0.024	0.16	0.78	0.84	0.66	0.49
BghiP	276.32	276	250, 226	17.0	0.008	0.024	0.14	0.75	0.73	0.61	0.47



## 5.0 Conclusion

This study provides a comprehensive evaluation of polycyclic aromatic hydrocarbons (PAHs) across air, water, and soil matrices within and around pharmaceutical industries in Ezianya Community, Aba, Abia State, Nigeria. Through systematic sampling at five strategic locations—upstream, downstream, outfall, residential, and control—representative data were collected to assess environmental contamination, identify potential sources, and evaluate associated ecological and human health risks. Analytical quantification using Gas Chromatography–Mass Spectrometry (GC–MS) with rigorous calibration and quality assurance procedures demonstrated excellent linearity ( $R^2 > 0.98$ ) for the 16 priority PAHs, ensuring reliability and reproducibility of the measurements. Statistical analyses, including mean concentrations, standard error of the mean (SEM), analysis of variance (ANOVA), and pairwise t-tests, enabled the determination of significant spatial and inter-matrix variations.

The results revealed distinct compositional and spatial patterns across matrices. Air samples were dominated by low molecular weight (LMW) PAHs such as naphthalene, acenaphthylene, and fluorene, reflecting their high volatility and association with gaseous emissions from industrial combustion, solvent use, and vehicular exhaust. Water samples exhibited elevated concentrations of mid-weight PAHs, including phenanthrene and fluoranthene, indicative of effluent discharge and surface runoff from pharmaceutical operations. Soil samples accumulated high molecular weight (HMW) PAHs, notably benzo[a]pyrene, indeno[1,2,3-cd]pyrene, and benzo[b]fluoranthene, consistent with long-term deposition, hydrophobic partitioning, and low microbial degradability. Comparative analyses across matrices showed a concentration hierarchy of soil

>air > water, reflecting the physicochemical behavior of PAHs, where heavier, hydrophobic compounds preferentially sorb to soil particles, whereas lighter species volatilize or remain in aqueous phases.

Diagnostic ratio analysis (e.g., ANT/(ANT + PHE), FLT/(FLT + PYR)) confirmed that PAH sources were predominantly pyrogenic, stemming from incomplete combustion of fossil fuels, industrial processes, and pharmaceutical solvents. Spatial distribution patterns, with highest concentrations at outfall and downstream points, further support the role of pharmaceutical effluent and industrial emissions as primary contributors. Environmental risk assessment indicated that several HMW PAHs, particularly benzo[a]pyrene, exceeded guideline values established by the WHO and USEPA, highlighting potential carcinogenic and mutagenic hazards. The calculated BaP toxic equivalency (BaP-TEQ) and incremental lifetime cancer risk (ILCR) for soils exceeded acceptable limits, particularly for children, emphasizing the public health implications of chronic exposure.

The study also evaluated remediation prospects, underscoring the efficacy of microbial biostimulation, bioaugmentation, phytoremediation with vetiver grass and maize, and the application of nano-zero valent iron (nZVI) for reducing PAH concentrations, particularly HMW compounds. Such interventions can mitigate bioavailability, prevent further environmental accumulation, and reduce potential human and ecological exposure.

In conclusion, this research demonstrates that pharmaceutical industrial activities significantly influence the distribution and persistence of PAHs across multiple environmental compartments. The findings highlight the urgent need for sustainable waste management, green chemistry adoption, routine environmental monitoring, and targeted remediation strategies to protect ecosystem and human health. By providing



quantitative data and mechanistic insights into PAH behavior, this work contributes a critical foundation for policy formulation, industrial regulation, and future studies on ecotoxicology, environmental fate, and remediation of emerging organic pollutants in the Niger Delta region and similar industrialized settings globally.

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#### Declaration

#### Consent for publication

Not Applicable

#### Availability of data and materials

The publisher has the right to make the data public

#### Conflict of Interest

The authors declared no conflict of interest

#### Ethical Considerations

Not applicable

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