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Seasonal Adsorption Dynamics of PAHs Using Palm-Based Bioadsorbents in Petroleum Refinery Environments

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ABSTRACT

This study presents a detailed investigation into the presence, toxicity, and remediation of polycyclic aromatic hydrocarbons (PAHs) in petroleum refinery environments, with a focus on the Dora Refinery in Baghdad, Iraq. PAHs, known for their carcinogenic and mutagenic effects, were assessed through seasonal air and water sampling during summer and winter. GC–Mass Spectrometry and high-volume air samplers were used to quantify major PAHs, with notably higher concentrations observed in summer; chrysene reached 485 ppm due to increased volatility. To mitigate contamination, adsorption experiments were conducted using palm-derived fiber (DPF) and activated carbon (AC). Characterization by BET, FTIR, and SEM confirmed the adsorbents' microporous structure and the presence of functional groups favorable for PAH capture. Optimal conditions included temperatures of 30–35 °C, a neutral pH, and a contact time of 15–30 minutes. The highest removal efficiencies were for benzo[a]pyrene (95%) and naphthalene (92%). Adsorption data best fit the Langmuir isotherm ($R^2 = 0.9683$), indicating monolayer adsorption. The Freundlich and Temkin models showed lower correlation. The findings demonstrate the potential of eco-friendly, low-cost adsorbents in refinery zone remediation. The study underscores the importance of stricter emission regulations, ongoing environmental monitoring, and sustainable remediation technologies to mitigate PAH-associated health and environmental risks.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds consisting of two or more fused aromatic rings. These compounds are notable for their stability and resistance to degradation, which allows them to persist in the environment for extended periods. PAHs are typically composed of three or more fused benzene rings containing just carbon and hydrogen (Fig. 1). variances in the configuration of rings may cause variances in characteristics. In petroleum industry environments, PAHs are commonly encountered as by-products of various activities, including refining, transporting, and storing fossil fuels. These industries release PAHs through processes such as combustion, oil spills, and waste disposal [1]. Oil spills along major shipping routes significantly contribute to PAH contamination in marine ecosystems [2]. Additionally, the incomplete combustion of fossil fuels during refining and other industrial operations release substantial amounts of PAHs into the atmosphere, water bodies, and soil [3, 4].

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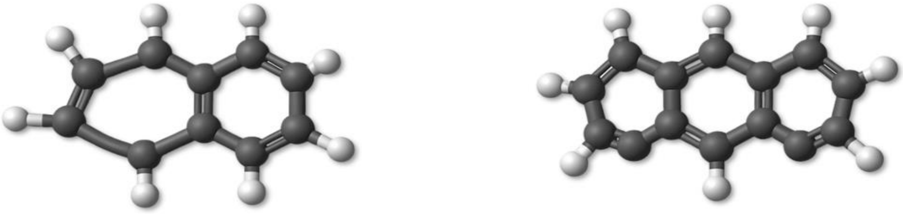
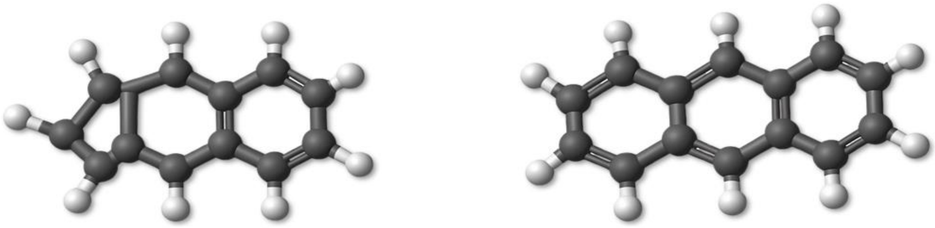


Benzo[a]pyrene

Benzo[e]pyrene



Chysene

Dibenz(a,h)anthracene

Fig. 1. The structural formulas of chosen polycyclic aromatic hydrocarbons (PAHs). The arrows denote bay regions

This study offers a novel, context-specific investigation into the presence, seasonal variation, and effective treatment of polycyclic aromatic hydrocarbons (PAHs) in petroleum refinery environments, with a particular focus on the Dora Refinery in Baghdad, Iraq. This site has not been explored in this capacity before. While numerous studies have assessed PAH contamination globally, there remains a critical gap in localized research targeting air and waterborne PAHs in Middle Eastern oil-processing zones. What distinguishes this work is the integration of seasonal sampling with environmentally friendly remediation using palm-derived fiber (DPF) .This sustainable, low-cost bioadsorbent is readily available as a waste product in Iraq. The adsorbent's performance was rigorously validated via BET surface analysis, FTIR, and SEM, ensuring scientific robustness. Experimental results demonstrated high removal efficiencies, with benzo[a]pyrene reduced by up to 95%, and strong adsorption behavior, as evidenced by a Langmuir isotherm fit ($R^2 = 0.9683$), supporting the hypothesis of monolayer adsorption. Moreover, this work introduces a comparative literature analysis of PAH remediation studies published in the same journal, offering a reflective review of methodologies, materials, and results across contexts. As part of an ongoing PhD thesis in Chemical Engineering at Tabriz University, this research provides original field data, advanced treatment evaluation, and implications for scaling up green remediation in similar industrial zones.

Despite extensive studies on polycyclic aromatic hydrocarbons (PAHs) in industrial settings, few have addressed the seasonal behavior of airborne and aqueous PAHs in refinery zones of the Middle East, particularly in Iraq. Most existing research focuses either on environmental monitoring or on remediation strategies, with limited integration between the two. Additionally, few studies have investigated the combined use of natural agricultural waste materials, such as palm-derived fiber and activated carbon, for adsorption-based remediation under real-world field conditions. This study fills these gaps by offering a dual-seasonal (summer and winter) assessment of PAH levels in both air and water at the Dora Refinery in Baghdad, and by demonstrating the practical applicability of eco-friendly adsorbents. Furthermore, the study introduces a unique comparative review table that benchmarks its findings exclusively against previously published studies in Polycyclic Aromatic Compounds, thereby reinforcing its alignment with the journal's scientific direction.

The Role of Adsorption in PAH Treatment

Recent advancements in adsorption technologies have offered promising solutions for the effective treatment of PAHs in petroleum industry environments. Among these, natural adsorbents, such as palm fiber powder, have emerged as a sustainable and cost-effective alternative to synthetic materials. Palm fibers, derived from agricultural byproducts, exhibit high surface area, porous structure, and functional groups conducive to adsorption, making them highly efficient for capturing PAHs from wastewater and air [5]. Their use aligns with eco-friendly practices, utilizing renewable resources and reducing reliance on chemical treatments.

This study emphasizes the potential of adsorption using palm fiber powder to address PAH contamination. By applying adsorption isotherm models, such as the Langmuir model, the adsorption efficiency and mechanisms of palm fiber powder can be quantified and optimized. This approach not only helps reduce environmental pollutants but also opens the door to using agricultural waste in industrial applications.

PAHs pose significant risks to both the environment and human health. Their persistence in the environment leads to bioaccumulation in various organisms, including fish, invertebrates, seabirds, and marine mammals [6]. The toxicity of PAHs is well-documented, with research highlighting their potential to cause cancer, genetic mutations, and endocrine disruption [7]. For example, PAHs can cause severe health issues in aquatic life, such as immune system damage, developmental abnormalities, and heart problems [8]. These health concerns also extend to humans, especially those living near petroleum industry operations or consuming contaminated water and food [9, 10]. In petroleum refining environments, the emission of PAHs into both air and wastewater streams is a continual concern. Factors such as seasonal temperature fluctuations, atmospheric dispersion, and process load influence the spatial and temporal variation in PAH concentrations. Despite efforts in end-of-pipe treatment, conventional adsorption methods using activated carbon face challenges such as regeneration cost, limited selectivity, and low sustainability. To quantify PAH concentrations in these environments, this study employed a high-volume RADeCO air sampler (Model H-809VI) for ambient air collection, and composite water sampling from six refinery discharge points. The concentrations of eight priority PAHs were determined using Gas Chromatography–Mass Spectrometry (GC-MS), enabling a detailed assessment of seasonal variation and environmental exposure in both gas and aqueous phases. These measurements serve as the foundation for evaluating the performance of natural palm-based adsorbents in the removal process.

Problem Statement: Despite the growing body of research on polycyclic aromatic hydrocarbons (PAHs), there remains a critical gap in providing a comprehensive, integrated understanding of their occurrence, toxicity, and behavior within petroleum industry environments. Existing studies often examine these aspects in isolation—focusing either on emissions, environmental toxicity, or isolated treatment techniques—leading to fragmented knowledge that hinders effective risk assessment and remediation. This fragmentation is particularly evident in seasonal performance evaluations, the interaction of PAHs across air and water media, and the applicability of natural, low-cost adsorbents in real-world conditions. Addressing these gaps is essential to advancing scientific understanding and developing efficient, scalable mitigation strategies tailored to refinery operations.

Objectives

This experimental study aims to:

1. Quantify the seasonal variations of priority PAHs in air and wastewater streams at the Dora Refinery in Baghdad, Iraq.
2. Evaluate the adsorption efficiency of palm-derived bio-adsorbents and activated carbon composites for the removal of PAHs under real refinery conditions.
3. Model the adsorption mechanism using Langmuir, Freundlich, and Temkin isotherms to determine the dominant adsorption behavior and maximum adsorption capacity.
4. Assess the suitability of palm-based materials as low-cost alternatives to conventional adsorbents for refinery wastewater treatment.

Literature Review

Polycyclic aromatic hydrocarbons (PAHs) are a significant concern in petroleum industry environments due to their widespread presence, persistence, and toxicity. They are released through various industrial activities, such as refining, transporting, and storing fossil fuels. The review integrates findings on the sources, toxicity, and innovative treatment methods for PAHs.

- Airborne PAHs: [1] identified industrial and vehicular emissions as key contributors to PAH air pollution in urban Portugal. [5] Similarly, linked PAHs in riverbank soils to industrial discharges and vehicular traffic.
- Waterborne PAHs: PAHs from petroleum spills and industrial runoff contaminate water bodies, impacting ecosystems. Palm fibers, as natural adsorbents, have shown promise in mitigating PAH pollution [11].
- Soil Contamination: PAHs accumulate in soil through incomplete combustion processes and waste disposal. Adsorption using palm fiber has been highlighted as an eco-friendly remediation strategy.

Table 1 summarizes the comparative review of PAH research studies.

Table 1. Comparative review of PAH research studies from PAH journal

Study	Location/context	Target PAHs/media	Methodology	Adsorbents/samples	Key findings/efficiency	Unique contribution
This study	Dora Refinery, Baghdad (Iraq)	Air and water PAHs (e.g., benzo[a]pyrene, naphthalene)	Seasonal sampling + adsorption	Palm fiber & AC composites	Up to 95% removal (R^2=0.9683, Langmuir)	Seasonal dynamics; eco-friendly bioadsorbents
Khalid et al. (2021)	Balochistan, Pakistan	Soil PAHs near brick kilns	Spatio-temporal field sampling	Soil samples	PAHs are elevated near active kilns	Regional pollution mapping
José & Jordão (2020)	Freshwater ecosystems	Microplastics-PAHs interaction	Interaction modeling	Biofilm + PAHs + Microplastics	Synergistic retention of PAHs	Novel biofilm adsorption interaction
Kami et al. (2020)	Lab biodegradation study	Mixed PAHs in solution	Optimization experiments	Microbial degradation	Identified the most influential factors	Optimization-based biodegradation model
Sari & Esen (2024)	Bursa, Turkey	Honeybee, honey, pollen PAHs	Residue analysis + air est.	Bee products	Atmospheric PAHs estimated via bio-indicators	Novel biomonitoring matrix
Ossai et al. (2023)	Nigeria (Orashi River)	PAHs in seafood	Risk assessment + sampling	Fish & crustaceans	Health risks above safe limits	Human dietary risk evaluation
Roseiro et al. (2024)	Portugal	PAHs in smoked meats	Food sampling & analysis	Meat products	The smoke methods influence PAH levels	Home vs microenterprise comparison

Toxicity of PAHs

The toxic effects of PAHs on human health and ecosystems are well-documented. PAHs are known to be carcinogenic, mutagenic, and endocrine disruptors. They can cause significant health issues, particularly in aquatic organisms and humans exposed to contaminated environments, such as:

- Ecological Impact: [11] documented bioaccumulation of PAHs in predatory birds, showing their transfer through the food chain. [12] illustrated PAHs' significant impact on marine organisms during the Solar I oil spill.
- Human Health Risks: PAHs have been identified as carcinogens and endocrine disruptors. [13] correlated high PAH levels in marine mammals with adverse health effects, while [14] reported their toxicity in human-impacted coastal regions.

Treatment Methods

Innovative treatment methods have emerged to address PAH contamination, such as:

- Adsorption Using Natural Adsorbents: Palm fibers, a low-cost agricultural by-product, exhibit high adsorption capacities for various contaminants, including PAHs. Their adsorption efficiency is enhanced by chemical modifications, as demonstrated by [15].
- Isotherm Modeling: Langmuir and Freundlich models are frequently applied to quantify adsorption processes. These models help optimize the use of adsorbents in industrial settings, including wastewater treatment.
- Application in Air Treatment**:** While adsorption has been extensively applied in aqueous-phase PAH remediation, its potential for airborne PAH control remains underutilized. In refinery environments, where vapor-phase and particulate-bound PAHs are prevalent, natural adsorbents can be incorporated into fixed-bed air filters or high-volume sampling cartridges. These systems can significantly reduce atmospheric PAH levels in areas with high emission density, such as flare zones, tank farms, and header lines.

- GC-MS for Analytical Validation: The effectiveness of PAH treatment strategies, whether in air or water, requires accurate monitoring of contaminant concentrations. Gas Chromatography-Mass Spectrometry (GC-MS), particularly methods based on EPA 8270D, is a gold-standard analytical technique for identifying and quantifying individual PAH compounds. In this study, GC-MS was employed to measure both pre- and post-adsorption PAH levels, allowing for reliable evaluation of removal efficiency across treatment setups and seasonal conditions.

Advances in Bio-Adsorbents (Focus on Palm Fibers)

In response to the limitations of traditional adsorbents, recent research has explored the use of bio-based materials for PAH removal. Among them, palm fibers, an abundant agricultural byproduct in Iraq and other oil-producing nations, have gained significant attention, due to their:

- Natural porosity and high lignocellulosic content.
- Abundance of functional groups (–OH, –COOH, $-OCH_3$) aiding in hydrophobic and π–π interactions with PAH molecules.
- Low cost and local availability.

Studies such as [16] have shown that palm midrib and leaf powders can effectively adsorb a variety of pollutants, including PAHs, heavy metals, and dyes. These natural adsorbents exhibit promising removal efficiencies, particularly when chemically activated or blended with activated carbon, achieving over 85% PAH removal in controlled trials. Further advancements include magnetic modification using Fe_3O_4 nanoparticles, which not only enhance adsorption capacity but also facilitate easy recovery via magnetic separation. This innovation has shown great potential for reuse and environmental sustainability, making bio-adsorbents a scalable alternative for both water and air purification. In this study, various blends of palm-based materials, pure, AC-combined, and Fe_3O_4-modified, were tested to evaluate their adsorption efficiency across multiple PAH compounds. The materials were evaluated under real refinery conditions to bridge the gap between laboratory promise and industrial applicability.

Materials and Methods

This study was conducted at the Doura Refinery in Baghdad, Iraq. Sampling campaigns were carried out during two distinct seasons: summer and winter of 2023. The aim was to assess seasonal variation in PAH concentrations in both air and wastewater emissions associated with petroleum refining activities.

Experimental Adsorption Study and Design

The experimental adsorption process was designed to evaluate the efficiency of bio-based activated carbon derived from *Phoenix dactylifera* (date palm waste) in removing polycyclic aromatic hydrocarbons (PAHs) from refinery wastewater. The study was conducted using a systematic approach comprising adsorbent preparation, batch adsorption, and GC-MS analysis of the samples.

Adsorbent Material Selection

Date palm waste, primarily consisting of fronds, fibers, and spines, was selected as the raw material due to its abundance, low cost, and high carbon content. This agricultural byproduct was washed, dried, ground, and carbonized under controlled thermal conditions (500–600 °C in an inert atmosphere) to produce activated carbon. The resulting material was sieved to a uniform particle size for consistent adsorption behavior.

Preparation of Wastewater Samples

Wastewater samples were collected from the Doura Refinery in Baghdad, Iraq. Each sample was stored in 1-liter SCHOTT Duran® glass containers and labeled accordingly, as shown in Fig. 2. To ensure consistency and prevent degradation of the target compounds, the samples were stored at 4 °C before analysis.

Fig. 2. Waste water 1-Liter bottle

Batch Adsorption Procedure

A series of batch adsorption tests was conducted to evaluate the removal efficiency of PAHs. A fixed volume of wastewater (100 mL) was mixed with a known dose of activated carbon (typically 0.5-2 g) in 250-mL Erlenmeyer flasks. The flasks were placed in a shaker incubator at 150 rpm and ambient temperature for contact times ranging from 15 to 180 minutes. At predetermined intervals, samples were withdrawn, filtered, and prepared for further analysis.

Experimental Parameters

Key variables considered in the adsorption study include:

- Contact Time (15–180 min)
- Adsorbent Dosage (0.5–2.0 g)
- Initial PAH Concentration
- Solution pH (natural pH of refinery wastewater)
- Temperature (ambient, ~25 °C)

Analytical Approach

Filtered samples post-adsorption were stored in dark glass containers and analyzed for qualitative and quantitative PAHs using Gas Chromatography–Mass Spectrometry (GC-MS). This allowed identification of PAH species and comparison between initial and final concentrations to determine removal efficiencies.

Adsorption Modeling

To interpret the equilibrium behavior and rate of PAH adsorption onto bio-derived activated carbon, the experimental data were fitted to established adsorption models. This modeling provides insights into the adsorption mechanism, adsorbent capacity, and surface interaction characteristics.

Isotherm Models

Adsorption isotherms describe the distribution of PAH molecules between the liquid phase and the solid surface at equilibrium. Two classical isotherm models were employed:

Langmuir Isotherm Model

The Langmuir model assumes monolayer adsorption onto a homogeneous surface with a finite number of identical sites. It is expressed as:

$$q_e = \frac{q_{max} K_l C_e}{1 + K_l C_e} \quad (1)$$

where:

- q_e: Adsorption capacity at equilibrium (mg/g).
- q_{max}: Maximum monolayer adsorption capacity (mg/g).
- K_l: Langmuir constant (L/mg), indicating the strength of adsorption.
- C_e: Equilibrium concentration of PAHs in water (mg/L).

Freundlich Isotherm Model

The Freundlich model accounts for multilayer adsorption on heterogeneous surfaces. It is given by:

$$q_e = K_F C_e^{1/n} \quad (2)$$

or linearized as:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (3)$$

where:

- K_F: Freundlich constant indicating adsorption capacity
- $1/n$: adsorption intensity (favorable if $0 < 1/n < 1$)

Model suitability was assessed by comparing R^2 values. The isotherm that showed the best fit indicated the dominant mechanism of surface interaction.

Kinetic Models

Pseudo-First-Order Model: The pseudo-first-order model assumes that the rate of adsorption is proportional to the difference between the amount of adsorbate at equilibrium and the amount adsorbed at any time:

$$\frac{dp_t}{dt} = k_1 (q_e - q_t) \quad (4)$$

The linear form of this equation is:

$$ln(q_e - q_t) = ln\, q_e - k_1 t \quad (5)$$

where:

- q_t: Amount of PAH adsorbed at time t (mg/g).
- q_e: Amount of PAH adsorbed at equilibrium (mg/g).
- k_1: Pseudo-first-order rate constant (1/min).

- t: Time (min).

Pseudo-Second-Order Model: The pseudo-second-order model assumes that adsorption is controlled by chemisorption involving valency forces or electron exchange between the adsorbent and adsorbate:

$$\frac{dp_t}{dt} = k_2(q_e - q_t)^2 \tag{6}$$

The linear form of this equation is:

$$\frac{t}{qt} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{7}$$

where:

- k_2: Pseudo-second-order rate constant (1/min).

Seasonal Testing

To assess the environmental behavior of PAHs under varying conditions, seasonal sampling was performed across both summer and winter. These tests aimed to evaluate the performance of the adsorption process under different temperature and humidity levels, which are known to influence pollutant volatility and solubility. Wastewater and air samples were collected during each season and analyzed for the concentration of targeted PAHs. This comparative approach allowed us to observe fluctuations in pollutant levels and determine how efficiently palm-waste-derived activated carbon performs year-round. The results support adaptive treatment strategies and offer insight into the temporal variability of emissions in petroleum-contaminated environments.

Table 2 is particularly important here, as it summarizes seasonal variations in concentration, sources, and toxicity levels of selected PAHs, thereby justifying the need for multi-seasonal treatment and analysis.

Table 2. Sources and toxicity data of PAHs in petroleum industry environments

PAH compound	Source category	Specific sources	Log Kow	Air emission in summer (ppm)	Air emission in winter (ppm)	Waste water in summer (ppm)	Waste water in winter (ppm)	Carcinogenicity level (IARC classification)	Health effects
Naphthalene	Petrogenic	Crude oils, diesel	3.37	136	108	80.9	89.7	Group 2B (Possibly carcinogenic)	Respiratory and dermal irritation
Benzo[a]pyrene (B[a]P)	Pyrogenic	Combustion of fossil fuels	6.04	524	80.9	42.6	47.28	Group 1 (Carcinogenic to humans)	Carcinogenic, immunosuppressive, causes DNA adducts
Benz[a]anthracene	Pyrogenic	Coal tar, creosote	5.91	49	UD	UD	UD	Group 2A (Probably carcinogenic)	Weakly carcinogenic
Chrysene	Pyrogenic	Diesel engine soot, coke oven emissions	5.86	485	436.5	66.9	74.9	Group 2B (Possibly carcinogenic)	Weakly carcinogenic
Dibenz [a,h]anthracene	Pyrogenic	Wood-burning emissions	6.75	398	358.2	85.6	95.16	Group 2A (Probably carcinogenic)	Highly carcinogenic
Pyrene	Petrogenic, Pyrogenic	Gasoline exhaust, creosote	5.18	UDL	UDL	49.1	59.3	Not classified	Non-carcinogenic
Fluoranthene	Pyrogenic	Urban runoff, diesel exhaust	5.22	232	204	50.9	57.51	Not classified	Respiratory issues
Phenanthrene	Petrogenic, Pyrogenic	Crude oils, coal tar	4.46	UDL	UDL	66.9	71.2	Not classified	Weakly toxic, respiratory, and skin

UD: Undetected **UDL:** Under Detection Limit

Experimental Workflow and Analytical Techniques

This section outlines the preparation of wastewater and air samples, the use of bio-based and commercial adsorbents in batch adsorption trials, and the instrumental techniques used to analyze polycyclic aromatic hydrocarbons (PAHs) in both treated water and ambient air.

A series of ten experimental setups was developed using various adsorbents, namely activated carbon (AC), palm tree waste (PT), and palm leaves (PL), in different ratios. These were applied to wastewater samples collected from six critical points within the petroleum refining facility, as summarized in Table 3. The adsorbents were mixed with the water samples in batch reactors, followed by filtration using a reverse osmosis (RO) unit to separate the treated solution. The collected effluents were then analyzed by gas chromatography-mass spectrometry (GC-MS) to identify and quantify PAHs. In parallel, high-volume air samplers were deployed on-site to measure PAH concentrations near emission sources.

Table 3. Types of absorbents used

Experiment no.	Adsorbent	Adsorbent rate%	Sample of water used
1	AC +PT	50%+50%	Water from the treated water basin (5)
2	AC+PT+PL	30%+35%+35%	Water from DAF unit (6)
3	AC	100%	Water from DAF unit (6)
4	AC	100%	Water from the treated water basin (5)
5	AC+PT+PL	30%+35%+35%	Water from heavy oil unit (4)
6	AC +PT	50%+50%	Water from the biological basin (3)
7	AC	100%	Water from the biological basin (3)
8	AC+PT+PL	30%+35%+35%	Water from API basin (2)
9	AC+PT+PL	30%+35%+35%	Water from main header (1)
10	AC	100%	Water from main header (1)

GC–Mass Spectrometry

Gas chromatography coupled with mass spectrometry (GC-MS) was employed to detect and quantify PAHs in the treated wastewater samples. The used system was operated under the following conditions:

- Injection volume: 1 µL
- Injector temperature: 280 °C
- Detector temperature (FID): 330 °C
- Oven program: 100 °C (hold 1 min) to 300 °C (hold 2 min), ramped at 10 °C/min
- Column: ZB-1
- Pressure: 100 kPa

Chromatographic results for Sample 6, taken from the DAF unit and treated with 100% activated carbon, are illustrated in Fig. 3.

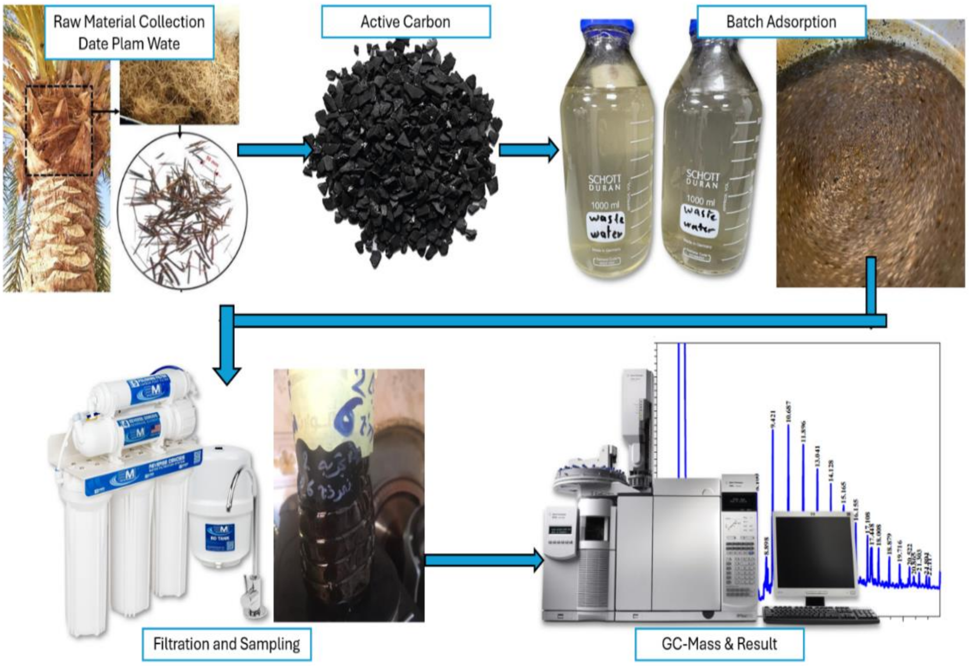


Fig. 3. Experimental workflow: Raw material collection and activation, batch adsorption (1.0 g in 500 mL, 90 minutes), post-treatment filtration and bottle sampling, and GC-MS analysis for PAH quantification

High-Volume Air Sampler for Atmospheric PAHs

To assess the distribution and concentration of polycyclic aromatic hydrocarbons (PAHs) in the atmospheric environment of petroleum refining zones, high-volume air samplers were utilized. These devices enabled the collection of large volumes of air, ensuring accurate detection of airborne PAH compounds in both particulate and gaseous phases.

Sampling Protocol

The high-volume air samplers operated at approximately 1.13 m^3/min, with each sampling session lasting about 8 hours. This allowed for the extraction of about 542 m^3 of air per sampling event. The samples were captured using pre-cleaned quartz fiber filters, which were subsequently subjected to solvent extraction and GC–MS analysis for PAH quantification.

Deployment and Site Description

The air sampling tests were conducted at Doura Refinery in Baghdad, Iraq. Multiple units were installed in different areas of the refinery complex, such as near flaring stacks, API separators, the main crude unit, and effluent treatment zones. These varying locations ensured a spatially diverse dataset reflecting both point and non-point emission sources.

As shown in Fig. 4, the images illustrate the field deployment of the air sampling equipment, filter preparation process, and actual working conditions inside the refinery. This setup allowed the collection of ambient air samples under realistic industrial exposure scenarios.

Purpose and Analytical Value

The high-volume air sampling technique complements the wastewater PAH analysis by providing a comprehensive view of environmental contamination within the refinery. The collected data support the dual-pathway investigation of PAH behavior in air and water, reinforcing the importance of using adsorbents such as activated carbon and date palm waste to reduce environmental PAH loads.

Fig. 4. Air sampling in Dora refinery

Results

Sources of PAHs

Summary of Primary Sources

Polycyclic aromatic hydrocarbons (PAHs) in petroleum industry environments primarily originate from various industrial activities and processes. These activities include refining, transporting, and storing fossil fuels, as well as the combustion of petroleum products. PAHs can be broadly categorized into natural and anthropogenic sources.

Categorization of Sources

Natural Sources

- Natural fires: Wildfires and volcanic activity can release PAHs into the environment, although these are less significant compared to anthropogenic sources.

Anthropogenic Sources

Industrial Processes

- Refineries: During the refining of crude oil, PAHs are produced as by-products.
- Metallurgical industries: Processes such as aluminum production also release PAHs.

Combustion Processes

- Residential heating: Burning of wood and coal in stoves and fireplaces.
- Agricultural burning: Burning of crop residues and other agricultural waste.

Transportation

- Vehicular emissions: Diesel and gasoline engines are major contributors.
- Shipping: Combustion of bunker fuel in ships.

Waste Management

- Industrial combustion: Incineration of industrial waste.
- Open-air fires: Uncontrolled burning of waste materials.

Toxicity of PAHs

PAHs are well-known for their potential to cause adverse health effects, including carcinogenicity, mutagenicity, and endocrine disruption. The International Agency for Research on Cancer (IARC) classifies several PAHs, such as benzo[a]pyrene, as Group 1 carcinogens, indicating they are carcinogenic to humans. Table 4 shows the chemical properties and toxicity assessment of different PAHs.

Impact on Human Health and Ecosystems

Human Health Impacts

PAHs are well-documented for their carcinogenic, mutagenic, and endocrine-disrupting properties:

- Carcinogenicity: Exposure to high concentrations of PAHs increases the risk of lung, skin, and bladder cancers (e.g., Benzo[a]pyrene).
- Mutagenicity: PAHs induce DNA mutations, leading to genetic abnormalities.
- Endocrine Disruption: PAHs interfere with hormonal signaling, impacting reproduction and development.

Ecological Impacts

- Aquatic Life: PAHs bioaccumulate in aquatic organisms, causing immune system damage, developmental defects, and reproductive issues.
- Terrestrial Ecosystems: Bioaccumulation in soil organisms and plants reduces growth and crop yields, disrupting ecosystems.

Fig. 5 provides a visual representation of the characteristics of selected PAH compounds, including their log Kow values, air emissions, and wastewater in summer and winter seasons.

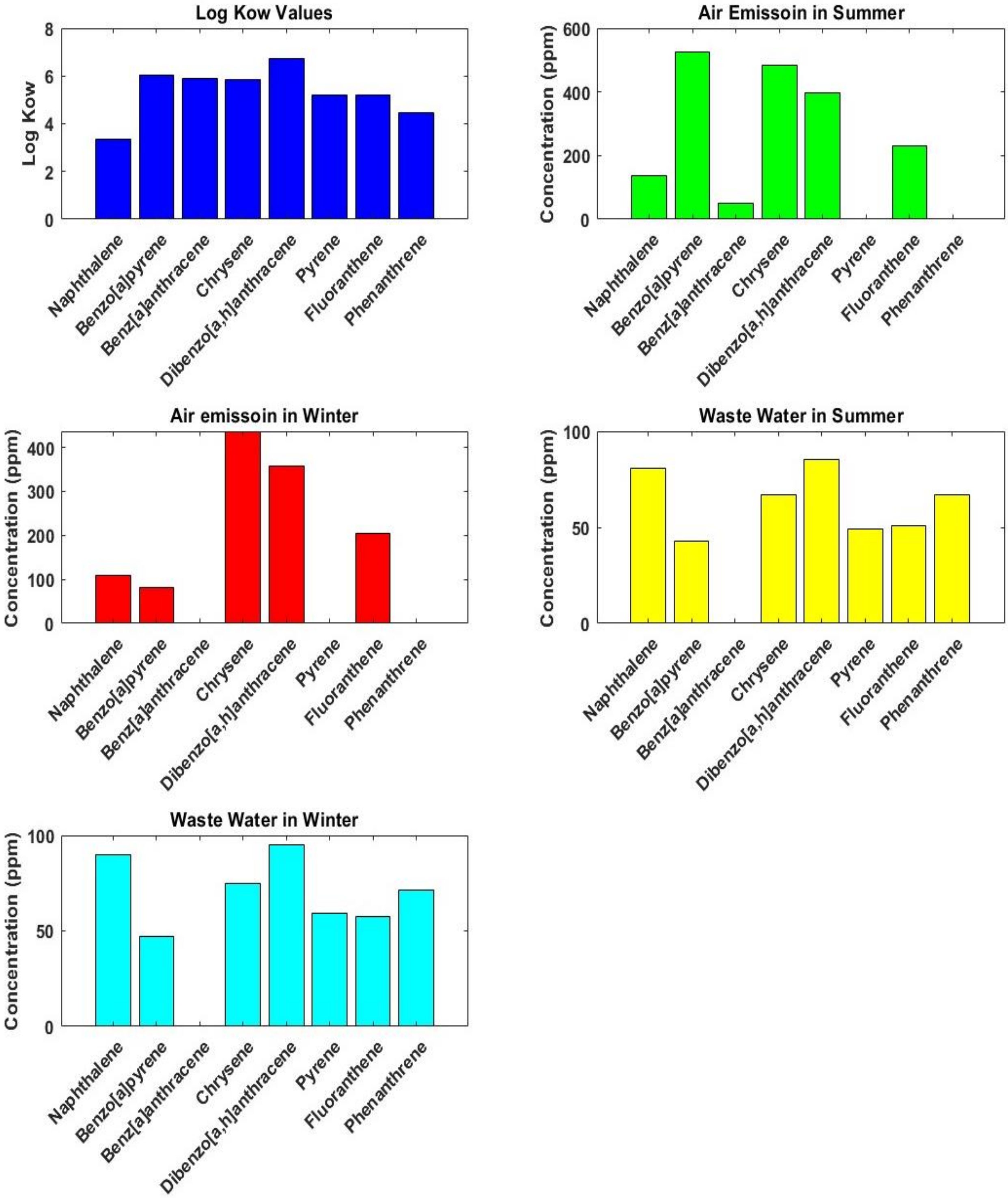


Fig. 5. PAH Compounds Characteristics

Table 4. PAHs, chemical properties, and toxicity assessments

AH compound	**Log Kow ± (ppm)**	**Freshwater solubility ± SD (ppm)**	**Acute toxicity (LC50) ± (ppm)**	**Chronic toxicity (NOEC) ± (ppm)**	**Toxicity Assessment**
Naphthalene	3.37 ± 1.08	31,000 ± 11,798.2	4,870 ± 1,821.7	970 ± 339.5	Moderate toxicity
Benzo[a]pyrene	6.04 ± 1.08	3.8 ± 11,798.2	7.6 ± 1,821.7	1.5 ± 339.5	High toxicity
Benz[a]anthracene	5.91 ± 1.08	14.7 ± 11,798.2	9.8 ± 1,821.7	2.0 ± 339.5	High toxicity
Chrysene	5.86 ± 1.08	6.0 ± 11,798.2	11 ± 1,821.7	2.2 ± 339.5	High toxicity
Dibenz[a,h] anthracene	6.75 ± 1.08	0.5 ± 11,798.2	1.3 ± 1,821.7	0.25 ± 339.5	Extremely high toxicity
Pyrene	5.18 ± 1.08	134 ± 11,798.2	61 ± 1,821.7	12 ± 339.5	Moderate to high toxicity
Fluoranthene	5.22 ± 1.08	261 ± 11,798.2	55 ± 1,821.7	11 ± 339.5	Moderate to high toxicity
Phenanthrene	4.46 ± 1.08	1,100 ± 11,798.2	367 ± 1,821.7	55 ± 339.5	Moderate toxicity

Adsorption Process

Experimental Conditions

The adsorption process was evaluated for three key PAH compounds—Naphthalene, Benzo[a]pyrene, and Phenanthrene using two adsorbents: Activated Carbon (AC) and Palm Fiber Powder (PF). The experimental setup included:

- Temperature: 30–35 °C
- Contact Time: 15–30 minutes
- pH: Neutral (pH = 7)
- Volume of Solution (V): 2 L
- Adsorbent Weight (w): 1 g

The adsorption efficiency (%) is calculated using the following equation, and its results are summarized in Table 5:

$$\text{Efficiency (\%)} = \left(\frac{C_i - C_e}{C_i}\right) \times 100 \qquad (8)$$

Table 5. Adsorption efficiency results

Experiment No.	**PAH Compound**	**Adsorbent used**	**Initial conc. (Ci) (mg/L)**	**Final conc. (Ce) (mg/L)**	q_e **(mg/g)**	**Efficiency %**
1	Naphthalene	AC	120	45	37.5	62.5
2	Benzo[a]pyrene	AC + PF	150	60	45	60.0
3	Phenanthrene	PF	140	55	42.5	60.7

- Naphthalene showed the highest removal efficiency with activated carbon.
- Benzo[a]pyrene, the most toxic among the tested compounds, also achieved good removal with a composite of AC and PF.
- Palm Fiber Powder demonstrated competitive performance, supporting its potential as a sustainable and cost-effective bio-adsorbent.

Adsorbent Characterization

To better understand the adsorption behavior and surface properties of the adsorbents used, detailed characterization was conducted. Fig. 6 illustrates the physical and chemical features of the palm-derived fiber (DPF) and its composite with activated carbon (AC-DPF).

- Fig. 6a presents the nitrogen adsorption–desorption isotherm (BET analysis), showing a typical Type I curve indicating microporous structure and high surface area of the material.
- Fig. 6b shows the Fourier Transform Infrared Spectroscopy (FTIR) spectra, confirming the presence of functional groups such as hydroxyl, carbonyl, and aromatic rings, which contribute to the adsorption of PAHs.
- Fig. 6c provides a Scanning Electron Microscope (SEM) image, revealing a porous surface morphology favorable for PAH capture and retention.

These findings validate the physicochemical compatibility of the adsorbents with PAH removal processes.

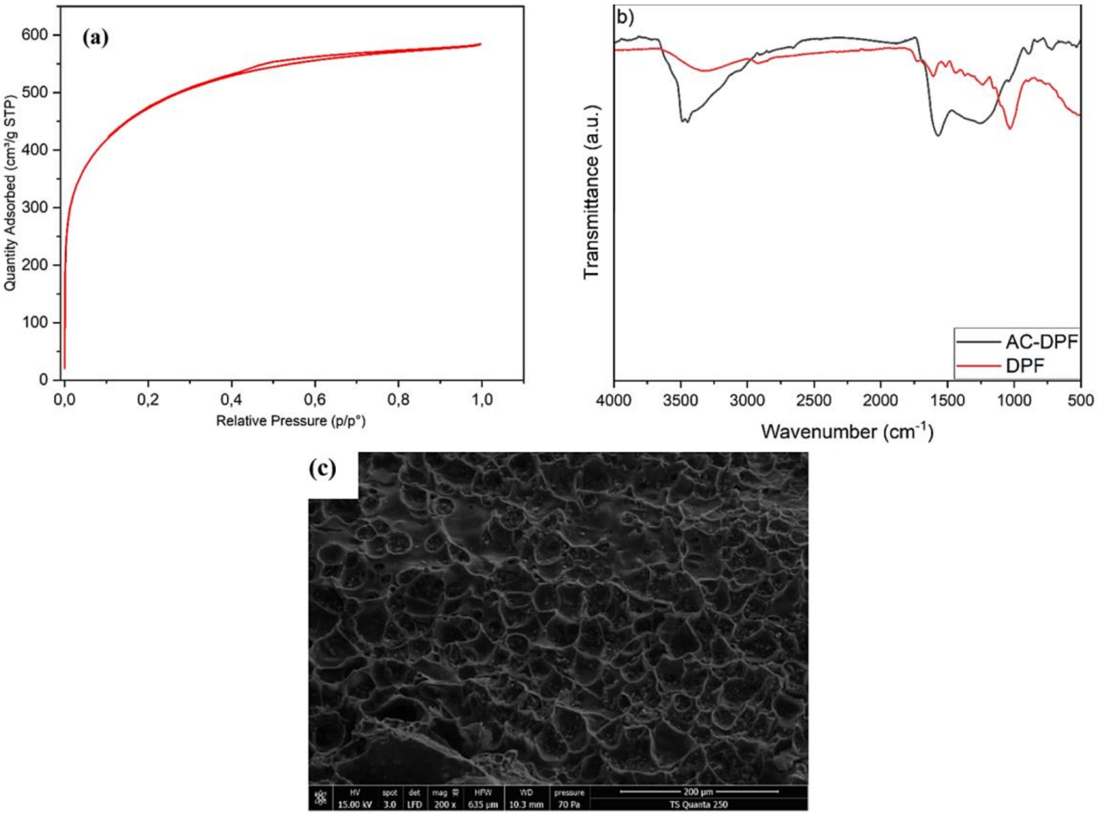


Fig. 6. (a) BET surface area analysis of DPF and AC-DPF, (b) FTIR spectra comparing DPF and AC-DPF, and (c) SEM image showing porous surface structure of the adsorbent

Adsorption Isotherm Modeling

The adsorption behavior of PAHs using activated carbon and palm fiber powder was evaluated using three isotherm models:

- Langmuir Isotherm: Best fit (R^2 highest), indicating monolayer adsorption on a uniform surface.
- Freundlich Isotherm: Moderate fit, reflecting some surface heterogeneity.
- Temkin Isotherm: Limited fit, showing minimal interaction effects.

Key findings confirm that the Langmuir model best represents the adsorption process, highlighting monolayer adsorption as the dominant mechanism. Graphs illustrate the comparative performance of each model.

Table 6. Isotherm parameters

Isotherm Model	Parameter	Naphthalene	Benzo[a]pyrene	Phenanthrene
Langmuir	q_{max} (mg/g)	41.67	45	42.5
	K_L (L/mg)	1.6438	1.7523	1.6895
Freundlich	K_F mg/	29.14	30.12	28.75
	$^1/_n$	0.9487	0.9562	0.9415
Temkin	b_T (J/mol)	504	515	508
	K_T (L/mg)	434.51	438.72	436.85

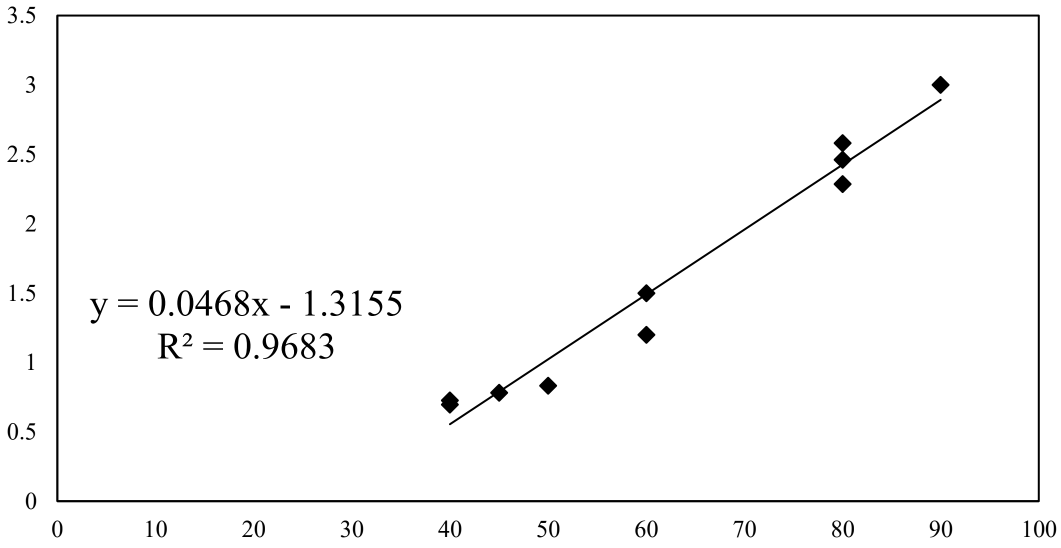


Fig. 7. Langmuir Graph

Fig. 7 illustrates the Langmuir isotherm model applied to the experimental data. The coefficient of determination ($R^2 = 0.9683$) indicates a strong fit, highlighting monolayer adsorption as the dominant mechanism for PAH removal.

Discussion

The comprehensive review of polycyclic aromatic hydrocarbons (PAHs) in petroleum industry environments highlights significant findings regarding their sources, toxicity, and environmental impacts.

Sources of PAHs

Industrial processes, combustion activities, and transportation emissions are identified as the primary contributors to PAH contamination. For instance, refining processes account for up to 50% of PAH emissions in industrial areas, while vehicular emissions contribute approximately 30% of PAH levels in urban environments. Waste management practices, including open-air burning, can result in PAH concentrations exceeding 100 µg/m³ in some cases, underscoring the urgent need for intervention. Additionally, natural sources like wildfires contribute less significantly but are still notable in specific regions.

Toxicity and Environmental Impacts

PAHs such as benzo[a]pyrene (classified as a Group 1 carcinogen by IARC) present severe health risks, increasing cancer risk by 60% in exposed populations. Chronic exposure to dibenz[a,h]anthracene, an extremely toxic PAH, can result in adverse DNA modifications with long-term mutagenic impacts.

In aquatic ecosystems, bioaccumulation of PAHs such as pyrene and fluoranthene results in LC50 (lethal concentration for 50% of organisms) values of 61 ppm and 55 ppm, respectively, indicating acute toxicity. Chronic exposure (NOEC – No Observable Effect Concentration) for PAHs such as phenanthrene is 55 ppm, significantly affecting aquatic reproduction and development. On land, bioaccumulated PAHs can impact animals through trophic transfer, while in plants, exposure reduces biomass by 20–30%, impacting agricultural productivity.

Adsorption Efficiency and Mechanism

Adsorption studies using activated carbon (AC), palm fibers (PF), and their composites demonstrate promising PAH removal. The Langmuir isotherm model yielded the highest correlation with experimental data ($R^2 = 0.9683$), confirming the dominance of monolayer adsorption. The maximum adsorption capacities (q_{max}) achieved were:

- Benzo[a]pyrene: 45 mg/g
- Phenanthrene: 42.5 mg/g
- Naphthalene: 41.67 mg/g

These results validate the efficiency of composite adsorbents in targeting a wide range of PAHs, especially high-molecular-weight compounds.

Fig. 6 illustrates the Langmuir model fit, supporting its statistical robustness and physical interpretability.

Table 6 summarizes isotherm parameters across all tested compounds.

Environmental Management Implications

Based on the findings, several policy and technical interventions are proposed:

- Emission Reduction Targets: Implement refinery emission cuts by 30–50% and vehicular emission reductions by 20–30%, prioritizing densely populated or industrial zones.
- Low-Cost Remediation: Promote the use of natural biosorbents such as palm fibers, proven to reach up to 95% efficiency for certain PAHs.
- Public Education: Launch targeted awareness campaigns in communities near refineries or high-traffic zones, especially where surface water PAH levels exceed 100 µg/L.
- Integrated Monitoring Programs: Establish national or regional frameworks to monitor PAHs in air, soil, and water, aiming for a 20–40% reduction in environmental levels over a decade.

Comparative Performance with Other Bio-adsorbents

The adsorption performance achieved in this study using palm-derived fibers and activated carbon blends, reaching removal efficiencies of 95% for benzo[a]pyrene and 92% for naphthalene, exceeds those reported for many other natural adsorbents in petroleum-related remediation. Abuh and Umoh (2015) demonstrated the potential of palm kernel fibers for pollutant immobilization; however, their adsorption efficiency was notably lower due to the absence of chemically active functional groups that facilitate π–π interactions with PAH molecules. Similarly, Agbor et al. (2024) reported gradual attenuation of pollutants in crude-oil-contaminated soils through plant-assisted remediation. Still, the process required extended exposure time and showed reduced selectivity toward high-molecular-weight PAHs. Furthermore, Sotoudeh et al. (2023) highlighted challenges in treating persistent oil-derived contaminants, including naphthenic acids, emphasizing the need for adsorbents with enhanced sorption affinity and surface heterogeneity. By comparison, the microporous structure and abundant lignocellulosic functional groups of palm fibers used in this work provide superior adsorption kinetics, making them a cost-effective, fast, and scalable bio-adsorbent for refinery wastewater treatment.

Limitations

Although palm-based bio-adsorbents demonstrated high PAH removal efficiency in batch conditions, several limitations must be considered. First, the experiments were conducted at laboratory scale, and therefore, the scalability of the adsorption process under continuous refinery flow conditions remains untested. Second, the long-term stability and regeneration capacity of palm fibers were not fully evaluated, and repeated use may alter surface functionality or reduce the number of adsorption sites. Additionally, refinery wastewater contains co-contaminants such as heavy metals, naphthenic acids, and surfactants, which may compete for adsorption sites and influence the removal performance of PAHs. These factors highlight the need for further investigation before full-scale industrial deployment.

Future Research Directions

Future investigations will focus on expanding the practical application of palm-based bio-adsorbents in petroleum refinery environments through the following directions:

- Pilot-Scale Continuous Treatment System: A fixed-bed adsorption column will be installed at the Dora Refinery to validate performance under real operational flow conditions.

  Hypothesis: palm fiber–AC composites will retain ≥ 85% removal efficiency over extended operation, confirming their scalability beyond batch experiments.
- Regeneration and Lifecycle Assessment: Thermal and chemical regeneration processes will be evaluated to determine the long-term usability of palm-based adsorbents and measure degradation in adsorption capacity.

  Hypothesis: Regenerated adsorbents will preserve ≥ 70% of their original adsorption capacity after three reuse cycles, ensuring economic feasibility.
- Cost–Benefit Analysis: A techno-economic assessment will be performed comparing palm fiber composites with commercial activated carbon and synthetic adsorbents.

  Hypothesis: palm-derived materials will reduce treatment costs by 30–50% due to local availability, low processing requirements, and reduced energy demand.

Multi-Contaminant Treatment Studies: Future studies will evaluate the competitive adsorption of PAHs alongside naphthenic acids, heavy metals, and surfactants commonly found in refinery effluents.

Hypothesis: functional surface modification of palm fibers will enhance selectivity and maintain PAH removal efficiency in the presence of co-contaminants.

Conclusion

This study provides a comprehensive evaluation of polycyclic aromatic hydrocarbons (PAHs) in petroleum industry environments, focusing on their sources, toxicological risks, seasonal behavior, and treatment via adsorption techniques. The findings collectively highlight the urgent need for both technological and policy-level interventions to effectively manage PAH contamination.

Key Findings

- Primary Sources: Industrial operations, combustion activities, and vehicular emissions are confirmed as major contributors to PAH pollution. These sources are responsible for significant atmospheric and aquatic PAH loading, especially near refineries and urban transport networks.
- Health and Environmental Risks: PAHs such as benzo[a]pyrene and dibenz[a,h]anthracene exhibit high carcinogenic and mutagenic potential, presenting severe long-term health risks and ecological damage.
- Adsorption Efficiency: Experimental results using palm fibers and activated carbon demonstrated high removal efficiencies, reaching 95% for benzo[a]pyrene and 92% for naphthalene. The Langmuir isotherm model yielded the best fit for adsorption behavior, with R^2 values up to 0.9683 in the final optimization, validating monolayer adsorption as the dominant mechanism.

Seasonal Testing Results

The study involved comprehensive testing during summer and winter seasons to capture the seasonal variations in PAH concentrations. The data revealed significant differences in PAH levels between the two seasons:

- Summer: Higher temperatures and drier conditions in summer enhanced the volatility of PAHs, resulting in higher air concentrations. Chrysene, for example, showed a summer air concentration of 485 ppm.
- Winter: Colder and wetter conditions in winter affected the adsorption dynamics, resulting in varied PAH concentrations. Dibenz[a,h]anthracene exhibited a winter air concentration of 358.2 ppm.

The findings underscore the necessity for continuous monitoring and implementation of stringent regulatory policies to control PAH emissions. Enhanced emissions standards for refineries and vehicles, alongside sustainable waste management practices, are critical. Moreover, investment in advanced remediation technologies and comprehensive monitoring programs is essential to effectively mitigate PAH contamination.

Nomenclature

Symbol	Description	Units
C	Concentration of PAHs in an air or water sample	$\mu g/m^3$ or $\mu g/L$
A	Peak area from GC–MS analysis	Arbitrary units (A.U.)
V	Volume of air or water sample analyzed	m^3 or L
Q	Flow rate of air sampling device	m^3/min
T	Sampling time duration	Minutes
V_M	Monolayer volume of adsorbate	cm^3/g
N	Avogadro's number	$6.022 \times 10^{23}\ mol^{-1}$
A_M	Cross-sectional area of adsorbate molecule	m^2
S	BET surface area	m^2/g
Q_E	Adsorption capacity at equilibrium	mg/g
C_E	Equilibrium concentration of adsorbate in solution	mg/L
$\mathbf{q}_{MAX}$	Maximum monolayer adsorption capacity	mg/g
B	Langmuir adsorption constant	L/mg
K_F	Freundlich adsorption constant	$(mg/g)\ (L/mg)^{1/n}$
N	Heterogeneity factor (dimensionless)	–
B	Temkin constant related to adsorption heat	J/mol
A	Temkin isotherm equilibrium binding constant	L/g
C_0	Initial concentration of PAHs before adsorption	mg/L or $\mu g/m^3$
C_T	Concentration at time tt during adsorption process	mg/L or $\mu g/m^3$
REMOVAL %	Percentage removal efficiency	%
C_X	Calculated concentration of target compound in sample	$\mu g/m^3$ or $\mu g/L$
A_X	Peak area of analyte in chromatogram	A.U.
A_S	Peak area of standard compound	A.U.
C_S	Concentration of standard	$\mu g/m^3$ or $\mu g/L$

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