

Original Article

Source Apportionment of n-Alkanes and Polycyclic Aromatic Hydrocarbons (PAHs) in Surface Sediments of the Makuran Coast (Sirik-Jask), Gulf of Oman: An Integrated Biomarker Approach

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Samar Mortazavi,

Email: mortazavi.s@gmail.com**Abstract**

Introduction: Rising hydrocarbon pollution in coastal sediments raises environmental concerns, but biomarker-based evaluations are still lacking in the Makuran area. Growing industrial development along the Sirik-Jask coastline, including port expansion and petrochemical projects, has raised alarm over sediment pollution, but the origins and extent of hydrocarbon inputs remain largely unknown.

Methods: In this study, surface and intertidal sediment samples were collected from five stations along the Sirik-Jask coast. The samples were freeze-dried, homogenized, and extracted using Soxhlet for n-alkanes (C₁₂-C₃₄) and polycyclic aromatic hydrocarbons (PAHs), followed by GC-MS analysis. Also, Molecular diagnostic ratios and distribution patterns were applied to identify the origin of hydrocarbon pollutants.

Results: n-Alkane concentrations ranged from 3967.48 ± 1290.71 to 28676.33 ± 4046.17 (µg/g dw), while PAHs ranged from 1143.35 ± 156.11 to 8059.95 ± 656.15 (ng/g dw), classifying most stations as highly contaminated. Intertidal sediments consistently accumulated higher hydrocarbon burdens than adjacent coastal zones. Low molecular weight PAHs accounted for over 80% of the total PAH burden, and diagnostic biomarker ratios across all samples converged on a predominant petrogenic origin for sedimentary hydrocarbons throughout the study area.

Conclusion: According to established sediment quality guidelines, most stations showed high levels of contamination. This research provides the initial biomarker-based baseline information on hydrocarbon pollution in Makuran coastal sediments. The results reveal the region's exposure to oil-derived contaminants and reinforce the importance of continuous environmental monitoring and strategic mitigation efforts to preserve marine ecosystems.

Keywords: Hydrocarbon biomarkers, Surface sediments, n-Alkanes, PAHs, Makuran

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Introduction

Sediments are recognized as major sinks for pollutants such as hydrocarbons and can function as long-term reservoirs as well as secondary sources of contamination. As a result, hydrocarbon accumulation in marine sediments has become an important environmental concern, particularly in coastal ecosystems exposed to continuous anthropogenic activities.¹ Among these contaminants, polycyclic aromatic hydrocarbons (PAHs) have attracted considerable attention because of their environmental persistence, tendency for bioaccumulation, and moderate to high toxicity, including carcinogenic effects on marine organisms and humans.² In addition, aliphatic

hydrocarbons detected in sediments originate from a mixture of biogenic, diagenetic, petrogenic, and pyrogenic inputs.³ Petroleum-derived hydrocarbons are therefore considered among the most important contaminants in marine environments due to their complex origins and long-term persistence in sediments.² The introduction of hydrocarbons into marine ecosystems occurs through both natural processes and human activities. However, large-scale oil contamination events have played a major role in increasing hydrocarbon burdens in the Gulf region. One of the most substantial petroleum contamination incidents occurred during the 1991 Gulf War, when approximately 11 million barrels of oil were released



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into the Gulf environment.⁴ Besides catastrophic spill events, continuous low-level contamination associated with port activities, shipping operations, urban runoff, and industrial discharge also contributes significantly to sediment pollution.⁵ In port areas, concentrations of light hydrocarbons in sediment samples ranged from 648 to approximately 1185 $\mu\text{g/g}$, which was nearly ten times higher than previously reported levels. This increase indicates that contamination by aliphatic hydrocarbons and PAHs has become an increasingly important environmental problem in many urban ports.¹ Similarly, urban runoff is considered a major pathway for transferring a broad spectrum of pollutants into receiving environments, while hydrocarbons are among the most common contaminants transported together with suspended particles and sediments.⁶ Consequently, long-term environmental monitoring is essential for preserving marine ecosystem quality in regions experiencing rapid economic expansion and population growth, conditions that intensify pollutant emissions into marine environments.² Given the complexity of hydrocarbon contamination, identifying pollutant origins is a critical step toward effective environmental assessment and management. Source characterization of PAHs provides a reliable approach for linking contaminants to their origins, and these compounds are generally classified according to either their formation temperature or source type, including pyrogenic PAHs generated through combustion processes and petrogenic PAHs associated with petroleum-related inputs. To achieve this objective, molecular diagnostic indices have been widely developed to distinguish among different PAH sources, while the use of PAH ratios helps reduce uncertainties associated with variations in volatility, solubility, and adsorption behavior.⁷ Among these indices, the carbon preference index (CPI) is a molecular ratio often used to assign the biological source and maturity of organic matter, and different methods for calculating CPI can lead to significantly different interpretations. The phenanthrene/anthracene (Phe/Ant) ratio has been extensively applied to differentiate petrogenic from pyrogenic PAH contamination in sediments, whereas the fluoranthene/pyrene (Flt/Pyr) ratio is strongly associated with formation temperature.^{7,8} Since no single ratio can provide a fully reliable interpretation, evaluating several PAH diagnostic indices covering different molecular weight ranges is considered necessary for robust source identification.⁸ Despite growing environmental concerns in Iran's southern marine waters, most previous studies have concentrated on the Persian Gulf, whereas the Gulf of Oman has received considerably less scientific attention. The Makuran coast, located along the Gulf of Oman, is increasingly exposed to industrial wastewater discharge and petroleum-related contamination enriched with organic pollutants, emphasizing the necessity of continuous petroleum pollution monitoring in this region.⁹ Rapid industrial development, expanding maritime activities, and increasing coastal infrastructure

may further intensify hydrocarbon inputs into this vulnerable marine ecosystem. The presence of unresolved complex mixture (UCM) hydrocarbons and petrogenic steranes and hopanes in seafloor sediments suggests that petrogenic sourced hydrocarbon inputs were present in coastal areas.¹⁰ Weathering processes, including evaporation, dissolution, and microbial biodegradation, cause significant changes in the chemical composition of spilled oil and can strongly modify hydrocarbon fingerprints.¹¹ Nevertheless, additional studies are still required to clarify the compositional characteristics and exact origins of hydrocarbons in the aquatic environments of the Makuran coast.⁹ Therefore, the current study aimed to (i) quantify the concentration, origin, and compositional patterns of PAHs and n-alkanes in surface sediments collected from coastal and intertidal zones of Makuran (Sirik-Jask), (ii) identify hydrocarbon sources using diagnostic ratios, and (iii) discuss the implications for coastal environmental management. The findings will provide baseline data for future monitoring and inform mitigation strategies to protect marine ecosystems in the Gulf of Oman.

Materials and Methods

Study Area

The Makuran coastal region lies along the northern shores of the Gulf of Oman, spanning southeastern Iran and southwestern Pakistan. It stretches from Ras-e-Kuh near Jask in Iran to Lasbela in Balochistan, Pakistan, covering approximately 1500 km of coastline. The Iranian portion includes the counties of Chabahar, Konarak, Jask, Sirik, and Minab, located in the provinces of Sistan and Baluchistan and Hormozgan. This study focused on a 155-km stretch between Sirik and Jask, encompassing key sampling stations such as Sirik Port, the Khor Azini mangrove forests, Mangeli fishing docks, the Kuh Mobarak protected area, and Jask Port.

Sampling and Initial Preparation

Five sampling stations were selected along the Iranian coast of the Oman Sea in 2023. The stations were spaced approximately 30–40 km apart, covering a total distance of around 150 km. The selection of sampling sites was based on ecological characteristics, spatial distribution, proximity to pollutant sources, and the presence of areas of industrial significance (see Figure 1 for sampling locations on Makuran beaches). Station selection was guided by high-resolution satellite imagery and field reconnaissance surveys conducted during low tide. Criteria included proximity to anthropogenic activities (ports, fishing areas, and mangrove zones), sediment texture, visual evidence of surface oil residues, and the presence of the Goreh-Jask oil pipeline. This stratified approach ensured representative coverage of both coastal and intertidal environments. Surface sediment samples were collected from both coastal and intertidal zones using a stainless-steel scoop from depths of 0 to 5 cm. At each station, three replicate samples were collected to ensure reproducibility

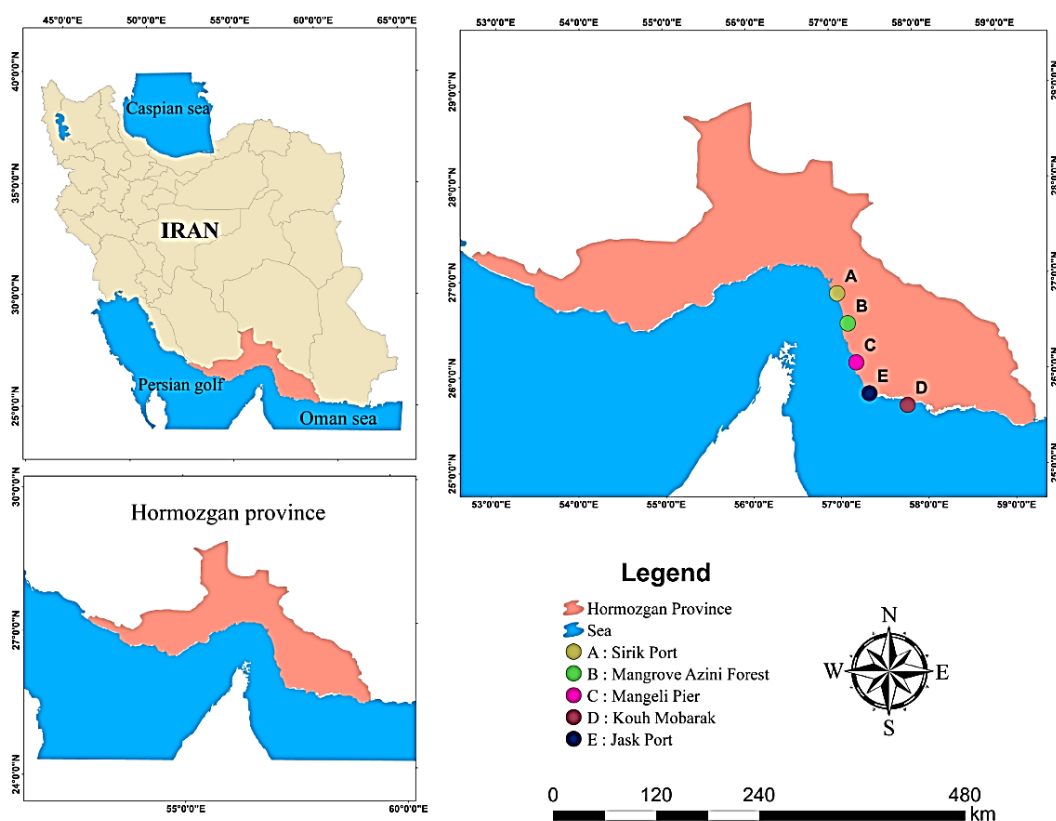


Figure 1. Sampling Locations on Makuran Beaches (A: Sirik Port, B: Azini Mangrove Forest, C: Mangeli Pier, D: Kuh Mobarak, E: Jask Port)

and statistical reliability. Shells, debris, and biological materials were carefully removed from the samples. Each sample was wrapped in aluminum foil and stored in coolers with dry ice for transport to the laboratory. Upon arrival, samples were stored at -20°C until further analysis. GPS coordinates and site characteristics were recorded for each sampling location. For each parameter measured, the mean value of the triplicates was calculated and reported per station. Standard deviations (SD) were also computed to reflect data variability and are presented in the corresponding tables. At Mangeli, Kuh Mobarak, and Khor Azini stations, only coastal surface sediments were collected because the intertidal zones in these areas are either narrow, rocky, or exposed to strong wave and current action, preventing the accumulation of soft intertidal deposits.

Sample preparation and Extraction

The Makuran coast includes two distinct depositional environments, coastal and intertidal sediments, which differ markedly in texture, hydrodynamic energy, and organic matter content. The coastal sediments are predominantly sandy with higher permeability, allowing faster pollutant dispersion. In contrast, intertidal sediments are fine-grained (silt-clay rich), with prolonged exposure to tidal fluctuations, promoting the accumulation and retention of hydrocarbons. Therefore, these two environments were analyzed separately to better evaluate source variations and contamination intensity. The sediment samples were freeze-dried and homogenized. A 10 g portion of each dried sample was

weighed for extraction. To monitor recovery and ensure analytical accuracy, surrogate standards containing deuterated PAHs (naphthalene-d₈, anthracene-d₁₀, chrysene-d₁₂, perylene-d₁₂) were added to each sample at concentrations of 100 ng/g.

Organic compounds were extracted using a Soxhlet apparatus with 100 mL of dichloromethane over a 10-hour period. To remove sulfur interference, activated copper was added to the sediment extracts. Copper activation involved immersion in 6N hydrochloric acid followed by rinsing with deionized water, methanol, hexane, and dichloromethane. If the copper lost its luster after 24 hours of refrigeration, it was reactivated and reintroduced until sulfur was fully removed. The extracts were concentrated using a rotary evaporator and gentle nitrogen flow to a final volume of 2-3 mL. The concentrated extracts were then purified using a silica gel column (5% deactivated with water). Hydrocarbon fractions, including alkanes and PAHs, were eluted with 20 mL of a dichloromethane/hexane mixture (1:3 v/v). The normal alkanes were further separated using 4 mL of hexane and concentrated again to 1 mL using rotary evaporation and nitrogen gas. Also, analysis of normal alkanes and PAHs was performed using a gas chromatograph (Agilent 7890A) coupled with a mass spectrometer (Agilent 5975C). The GC column was a 30 m capillary column with an internal diameter of 0.25 mm and a stationary phase thickness of 0.25 μm . Helium (99.999% purity) was used as the carrier gas, and ionization was achieved at 70 eV. The injection port was maintained at 310 $^{\circ}\text{C}$. The initial column temperature was set at 70 $^{\circ}\text{C}$ and held for 1 min. The temperature

was then ramped at 30 °C/min to 150 °C, followed by a slower ramp of 5 °C/min to a final temperature of 310 °C. The column was held isothermally at 310 °C for 10 min. Aliphatic compounds were detected using the scan mode with a mass-to-charge ratio (m/z) of 85. For PAH analysis, the injection temperature was also set at 310 °C. The column temperature began at 70 °C and was held for 2 min. It was then increased at 30 °C/min to 150 °C, followed by a ramp of 4 °C/min to 310 °C. The final temperature was held isothermally for 10 min. To enhance sensitivity and resolution, selected ion monitoring (SIM) mode was used. SIM parameters were optimized based on the mass-to-charge ratios and retention times of target PAHs, as reported in previous studies.⁹

Data Analysis and Diagnostic Ratios

CPI was calculated to distinguish between biogenic and petrogenic sources of n-alkanes. This index is based on the principle that odd-carbon-numbered n-alkanes typically dominate in biogenic sources (e.g., terrestrial plant waxes), whereas petroleum-derived n-alkanes show no odd-even predominance. CPI was computed using Equation 1:

$$\text{Overall CPI}_{15-35} = \frac{\sum \text{Odd } C_{15-35}}{\sum \text{Even } C_{16-34}} \quad (1)$$

where $\sum$ odd represents the sum of concentrations of odd-carbon-numbered n-alkanes and $\sum$ even represents the sum of even-carbon-numbered n-alkanes within the specified carbon range.^{1,3} Furthermore, several molecular diagnostic ratios were calculated for source apportionment, including pristane/phytane (Pr/Ph), n-C₁₇/pristane (n-C₁₇/Pr), n-C₁₈/phytane (n-C₁₈/Ph), and the low-to-high molecular weight PAH ratio (LMW/HMW). For PAHs, source-specific isomer ratios were computed, including phenanthrene/anthracene (Phe/Ant), fluoranthene/pyrene (Flt/Pyr), anthracene/(anthracene+phenanthrene) [Ant/(Ant+Phe)], fluoranthene/(fluoranthene+pyrene) [Flu/(Flu+Pyr)], and methyl phenanthrenes/phenanthrene (MP/P). Calculating these ratios minimizes the risk of misclassification and increases the reliability of distinguishing petrogenic from pyrolytic inputs in complex coastal sedimentary environments.^{6,12}

Quality Control

To ensure analytical reliability, surrogate standards were added at the beginning of the extraction process to estimate analyte recovery. Internal standards, including 100 µL of iso-octane for alkanes and 100 µL of p-terphenyl-d14 for PAHs, were added prior to GC-MS injection. The blank samples were processed alongside every 10 sediment samples to monitor contamination and background noise. Instrument detection limits were calculated based on the slope of calibration curves and the standard deviation of blank sample injections.

Results and Discussion

Distribution of n-Alkanes

Concentrations and Spatial Distribution

Total n-alkane concentrations (n-C₁₂–n-C₃₄) in surface sediments from the Sirik–Jask coastline ranged from 2,479.69 to 33,054.71 µg/g dw (Table S1). The highest concentrations were recorded in intertidal sediments at Sirik Port (25,075.10–33,054.71 µg/g dw), followed by coastal sediments at the same station (17,898.39–20,371.22 µg/g dw) and coastal sediments at Kuh Mobarak (13,951.37–16,105.79 µg/g dw). Moderate levels were observed at Jask Port (intertidal: 6,938.3–9,368.3 µg/g dw; coastal: 3,590.5–4,684.8 µg/g dw) and Mangeli (4,693.0–7,686.1 µg/g dw). The lowest concentrations were found in the coastal sediments of the Khor Azini mangrove area (2,479.69–4,784.44 µg/g dw). A consistent spatial pattern emerged across all stations where paired samples were available: intertidal sediments accumulated substantially higher n-alkane loads than their adjacent coastal counterparts. The molecular composition in all samples was dominated by mid- to high-molecular-weight homologues (n-C₂₂–n-C₂₉), with an even-over-odd predominance evident in the n-C₁₆–n-C₂₆ range. Pristane (Pr) and phytane (Ph) were detected in all samples, with concentrations ranging from 17.21 to 51.72 µg/g dw and 31.05 to 81.33 µg/g dw, respectively. Figure 2 provides a comparative overview of the mean n-alkane concentrations in coastal and intertidal sediments across all sampling stations.

According to global sediment pollution benchmarks (<10 µg/g = unpolluted; >500 µg/g = highly polluted),¹³ all seven sampling stations along the Sirik–Jask coastal stretch are classified as highly polluted, particularly at Sirik and Kouh Mobarak, where intense port and fishing activities occur. It is worth noting that the n-alkane levels recorded in this study are comparable to values recorded in other coastal systems affected by oil, such as the Nile Delta (Mediterranean Sea) and the Gulf of Mannar (India).^{14,15} Such elevated concentrations are consistent with the region's exposure to intensive maritime traffic, fishing port operations, and small-scale oil handling activities. The pronounced spatial variability in n-alkane burdens, with Sirik and Kuh Mobarak exhibiting the highest concentrations and Khor Azini the lowest, points to the dominant influence of localized anthropogenic sources rather than diffuse background inputs. Sirik Port is a hub of intense fishing and docking activities, while Kuh Mobarak lies in close proximity to oil transport infrastructure, including the Gore–Jask pipeline. In contrast, the comparatively lower concentrations at Khor Azini may reflect either reduced direct hydrocarbon inputs or enhanced dispersion and dilution associated with the complex hydrodynamics of this semi-enclosed mangrove wetland system. In the studied intertidal zones, fine-grained sediments (silt-clay) with high organic matter content provide greater surface area and adsorption capacity for hydrophobic compounds. Compared to adjacent coastal areas, these sediments contain higher amounts of n-alkanes, which

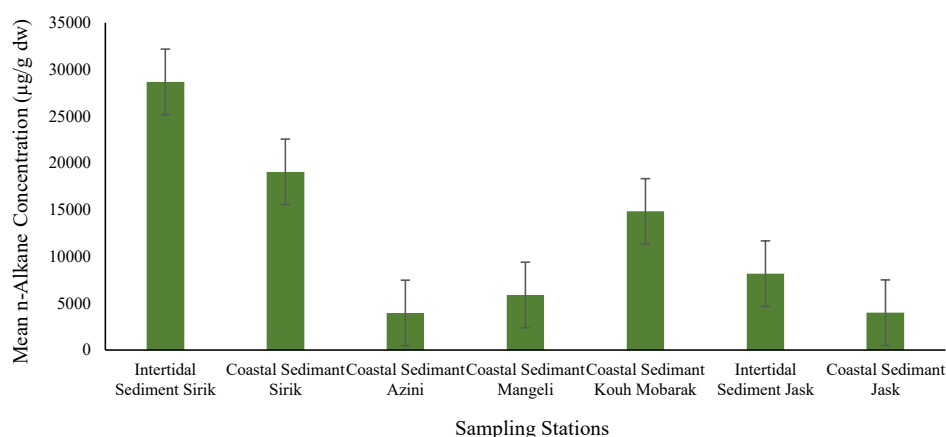


Figure 2. Comparison of the Mean Concentrations Of N-Alkanes in Coastal and Intertidal Sediment Samples from the Makuran Coasts

results from stronger hydrocarbon preservation under fluctuating redox conditions and continuous exposure to pollutants. Periodic tidal flooding also inhibits aerobic microbial degradation, thereby enhancing the long-term sequestration of petroleum-derived hydrocarbons.⁹ This pattern is consistent with previous findings that identify organic-rich, fine-grained intertidal zones as the main reservoirs of petroleum-derived hydrocarbons.^{16, 17} The observed dominance of even-numbered n-alkanes ($n\text{-C}_{16}\text{--}n\text{-C}_{26}$) further suggests bacterial re-working of hydrocarbons. Collectively, these biomarker indicators reveal that petrogenic inputs are predominant, supplemented by limited bacterial alteration and natural organic contributions.¹⁰

Diagnostic Ratios

The diagnostic ratios derived from n-alkane profiles provide multiple, convergent lines of evidence for a predominant petrogenic origin of aliphatic hydrocarbons throughout the Sirik–Jask coastal system. CPI values tightly clustered around unity (0.94–1.04) represent a classic molecular signature of thermally mature organic matter derived from petroleum and its refined products, including diesel and lubricant residues. This near-unity CPI stands in stark contrast to the odd-over-even predominance ($\text{CPI} > 3$) characteristic of fresh terrestrial plant waxes, indicating negligible biogenic contributions from vascular plants in the study area. The petrogenic assignment is independently corroborated by Pr/Ph ratios consistently below 1.0 (0.40–0.95), a hallmark of crude oil and its derivatives deposited under sub-oxic to anoxic conditions.^{9, 10} The co-occurrence of $\text{CPI} \approx 1$ and $\text{Pr/Ph} < 1$ observed here mirrors the biomarker signature widely documented in sediments of the Persian Gulf and the Nile Delta.^{18, 19} The $n\text{-C}_{17}/\text{Pr}$ and $n\text{-C}_{18}/\text{Ph}$ ratios offer further insight into the nature and diagenetic state of the petroleum contamination. The predominance of $n\text{-C}_{17}/\text{Pr}$ values above 1.0 (1.26–2.05) suggests the presence of moderately fresh to slightly biodegraded oil residues, with a possible minor planktonic contribution, as pristine biogenic n-alkanes typically yield elevated $n\text{-C}_{17}/\text{Pr}$ ratios. Conversely, $n\text{-C}_{18}/\text{Ph}$ ratios approaching or falling

below 1.0 (0.84–1.50) indicate that biodegradation has progressed sufficiently in some samples to preferentially remove n-alkanes relative to the more recalcitrant isoprenoid phytane.^{6, 11} This interpretation is further supported by the strong even-over-odd predominance observed in the mid-chain n-alkanes ($n\text{-C}_{16}\text{--}n\text{-C}_{26}$), a molecular feature that provides independent evidence for extensive in situ bacterial reworking of the sedimentary hydrocarbon pool. Collectively, these biomarker indicators reveal that petrogenic inputs are predominant, supplemented by limited bacterial alteration and natural organic contributions.¹⁰ The very low LMW/HMW ratios (0.04–0.09) reflect the pronounced dominance of higher molecular weight homologues ($n\text{-C}_{22}\text{--}n\text{-C}_{29}$), consistent with a weathered petroleum signature where lighter n-alkanes have been preferentially lost. This interpretation aligns with the findings of Li et al, who reported that the weathering loss of n-alkanes is directly related to molecular weight, with low-molecular-weight homologues exhibiting higher volatility.²⁰ Collectively, these biomarker indicators establish that petrogenic inputs overwhelmingly dominate the aliphatic hydrocarbon signature throughout the Sirik–Jask coastal stretch. Biogenic contributions from terrestrial plants or fresh planktonic material appear to be minor and are largely masked by the overwhelming petrogenic signal and subsequent microbial alteration within the sediment column. These findings are consistent with the results of the study by Malekolkalami et al.⁹

Distribution of PAHs

Concentrations and Spatial Distribution

A total of 30 PAHs, including the 16 priority compounds identified by the U.S. EPA, were quantified in coastal and intertidal sediments from five stations along the Makuran coast. Total PAH concentrations ranged from 363.58 to 8,690.49 ng g^{-1} dw, with intertidal sediments exhibiting distinctly higher levels than their coastal counterparts (Table S2). The highest concentrations were observed in intertidal sediments at the Sirik station, while the lowest were recorded in coastal sediments at the Jask station. Overall, intertidal sediments exhibited

higher PAH concentrations than coastal sediments (Figure 3). Compositionally, LMW PAHs accounted for more than 80% of the total, with acenaphthene and benzo(a)anthracene dominating most samples.

The gradient of higher PAH concentrations in intertidal sediments highlights the greater capacity of these zones to retain hydrophobic organic compounds. This retention is attributed to periodic immersion, fine-grained texture, and high organic matter content. This pattern aligns with the n-alkane distribution observed in this study and reinforces the role of intertidal areas as depositional sinks for petroleum-derived contaminants. Similar intertidal enrichment of PAHs has been documented in other coastal systems, including the Persian Gulf reported by Jafarabadi et al,²¹ and the Xiaoqing River estuary, China reported by Zhang et al,²² where fine-grained sediments with elevated organic carbon content exhibited significantly higher PAH burdens than adjacent sandy coastal deposits. The severity of PAH contamination varied considerably across the study area. According to the classification by Baumard et al, PAH levels exceeding 5,000 ng/g represent very high contamination.²³ By this standard, sediments at Sirik and Kouh Mobarak fell within the severe contamination category, consistent with the influence of port activity, shipping, and small-scale oil handling. In contrast, the sediments from Jask and Khor Azini showed moderate concentrations, reflecting lower anthropogenic inputs. This spatial variability, with peak contamination centered on active port areas, mirrors the pattern reported in Samsun, Turkey, where the highest PAH levels were recorded near harbors and vessel traffic zones.²⁴ Comparatively, PAH concentrations in the Makuran sediments (449-8690 ng/g dw) exceed those reported from relatively pristine coastal zones (e.g., East China Sea), but are comparable to heavily industrialized or oil-transport regions, such as the Gulf of Mannar and Egyptian deltaic coasts. The maximum concentration of PAHs in Sirik Port (8,059.95 ng/g dw) is significantly higher than the levels reported for the Nile Delta coast (range: 4.55 to 207.48 ng/g dw), but it remains lower than the very high concentrations reported in some Persian Gulf sediments affected by chronic oil spills and industrial

and petrochemical activities. This finding emphasizes that heavy maritime activity, fishing harbors, and limited water exchange collectively enhance the retention of oil residues in nearshore areas along the Gulf of Oman.^{14, 15, 21}

Diagnostic Ratios

To determine the origin of PAHs, several molecular diagnostic ratios between specific isomers and alkylated compounds were evaluated. Owing to differences in reactivity, solubility, and thermodynamic properties, these ratios serve as effective indicators for distinguishing between petrogenic (petroleum-derived) and pyrogenic (combustion-derived) sources. The calculated ratios included MP/P, Phe/Ant, Flt/Pyr, Ant/(Ant+Phe), and Flu/(Flu+Pyr), which enable the identification of both the origin and the degree of transformation of PAHs. The diagnostic ratios calculated for all samples, together with the threshold values used for source discrimination, are summarized in Table 1. The LMW/HMW ratio consistently exceeded unity across all samples. The Phe/Ant and Flt/Pyr ratios fell within ranges characteristic of petrogenic sources, while Ant/(Ant+Phe) and Flu/(Flu+Pyr) values remained below their respective pyrogenic thresholds at all stations. The MP/P ratio indicated a predominance of alkylated over parent PAHs in most samples. Compositionally, low-molecular-weight PAHs accounted for more than 80% of the total PAH burden.

Environmental interpretation of diagnostic ratios clearly indicates predominantly petrogenic origins for PAHs in the Makuran sediments. The LMW/HMW ratios (1.03-2.7), Phe/Ant values (10.14-41.84), and Flt/Pyr ratios (0.17-0.97) all fall within the petroleum-source domain. Ant/(Ant + Phe) < 0.1 and Flu/(Flu + Pyr) < 0.4 likewise confirm strong petroleum influence with minimal pyrogenic input.⁸ The molecular composition of PAHs provides further support for this petrogenic assignment. Low-molecular-weight PAHs containing 2-4 aromatic rings are typical indicators of petrogenic origin, whereas high-molecular-weight PAHs (≥ 5 rings) reflect pyrogenic sources associated with combustion. In the present study, LMW compounds contributed over 80% of

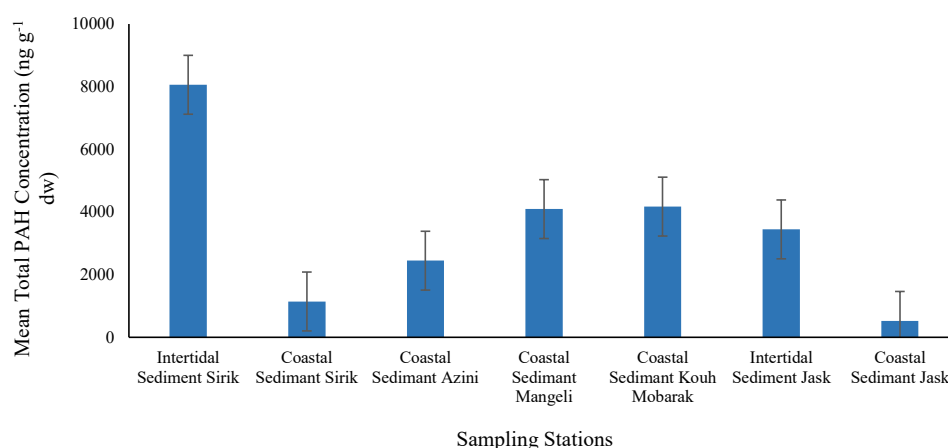


Figure 3. Comparison of the Mean Concentrations of 30-Total PAH in Coastal and Intertidal Sediment Samples from the Makuran Coasts

Table 1. Summary of Ratios Used for Source Identification of Organic Compounds Based on PAH Compounds

Diagnostic Ratio	Pyrogenic Threshold	Petrogenic Threshold	Present Study (Range)	Predominant Source Identified	References
MP/P	1 >	1 <	1-6.45	Petrogenic	25
Ant/ (Ant + Phe)	0.1 <	0.1 >	0.02-0.09	Petrogenic	8
Flu/ (Flu + Pyr)	0.5 <	0.4 >	0.01-0.37	Petrogenic	8
LMW/HMW	Low (<1)	High (>1)	1.03-2.7	Petrogenic	26
Phe/Ant	10 >	15 <	10.14-41.84	Petrogenic	23

total PAHs across all sampling stations, and the LMW/HMW ratio ranged from 1.03 to 2.7. Values exceeding unity are consistent with chronic oil inputs rather than combustion-derived PAHs, as confirmed by previous studies from the Gulf of Mannar (Behera et al) and the Klang River estuary, Malaysia (Bakhtiari et al). This clear predominance of LMW species confirms that petrogenic inputs are the major contributors to sedimentary PAHs along the Makuran coast.^{14, 15, 27} The dominance of petrogenic PAHs observed here is consistent with findings from other semi-enclosed seas in the region, such as the Persian Gulf, where alkylated PAHs and low molecular weight compounds similarly dominate sedimentary PAH profiles.^{2, 21}

Individual diagnostic ratios provided convergent evidence for this interpretation. The MP/P ratio ranged from 1 to 6.45, suggesting that most sediments are dominated by petroleum-derived PAHs ($MP/P > 1$), while a few samples exhibiting values below 1 indicate minor pyrogenic contributions.² These results align closely with patterns observed in the Nile Delta (Hassan et al),¹⁴ and Persian Gulf (Jafarabadi et al),²¹ where mixed but predominantly petrogenic inputs were also reported. Unlike some Mediterranean ports, including the western port of Alexandria, where combustion processes were identified as the dominant sources of PAHs through runoff, industrial wastewater discharge and atmospheric input (Mostafa et al),²⁸ the high MP/P ratios recorded here reinforce the distinct petrogenic character of the study area. Similarly, the Phe/Ant ratio ranged between 10.14 and 41.84, clearly exceeding the pyrogenic threshold (< 10), thus reinforcing the petrogenic character of the hydrocarbons. The Flt/Pyr ratio ranged between 0.17 and 0.97, and Flu/(Flu+Pyr) between 0.01 and 0.37, both falling within the petroleum-source domain (< 1 and < 0.4 , respectively). Moreover, the Ant/(Ant+Phe) ratio (0.02-0.09) provides additional confirmation of a strong petrogenic influence, as values below 0.1 typically denote petroleum contamination. These diagnostic results collectively demonstrate that petrogenic PAHs dominate in both coastal and intertidal sediments of the Makuran region, with only minor pyrogenic input possibly linked to limited local combustion sources or weathered residues.^{8, 9, 23} The results reported by Alidoust et al. confirmed the findings of the present study.² The relatively high abundance of two- and four-ring PAHs (e.g., naphthalene, phenanthrene, fluoranthene) compared with five- and six-ring compounds suggests inputs of fresh petroleum

residues with limited atmospheric combustion influence. The volatility and solubility of these lighter PAHs also promote their persistence in fine-grained, organic-rich sediments, confirming that petroleum discharges and shipping-related emissions are the dominant regional sources of PAHs. Cross-plot analyses of these ratios (Figures 4–6) revealed clustering of intertidal samples within the petrogenic domain, providing visual confirmation of persistent oil-derived contamination likely associated with shipping lanes, fishing harbors, and local oil transportation.

The complete convergence of results obtained from four binary cross-plots (Figures 4–6), which employed different combinations of PAH diagnostic ratios, clearly demonstrates that all coastal and intertidal sediments of the Makuran region (from Sirik to Jask) are dominated by a strong and nearly uniform burden of aromatic hydrocarbons of petrogenic origin. In Figure 4, the simultaneous positioning of all samples in the $LMW/HMW > 1$ and $MP/P > 1$ domain indicates that not only do low molecular weight compounds dominate the pollution load, but the abundance of MP/P also exceeds the petrogenic-pyrogenic discrimination threshold. Figure 4 complements this pattern by showing $Flt/Pyr < 1$ alongside $LMW/HMW > 1$; although intertidal samples exhibited slightly higher Flt/Pyr values compared to coastal samples, they never exceeded the petrogenic threshold, suggesting a minor influence of weathering or biodegradation without altering the source signature. Figure 5 provides the most decisive evidence, with all points clustered in the $Phe/Ant > 15$ (and in some samples up to 41) and $Flt/Pyr < 1$ domain, which reveals a severe depletion of Phe/Ant and completely rules out the input of pyrogenic sources such as fuel combustion. Finally, in Figure 6, which presents the most reliable ratio combination ($Flu/(Flu+Pyr)$ versus $Ant/(Ant+Phe)$), all samples without exception fall within the $Ant/(Ant+Phe) < 0.1$ and $Flu/(Flu+Pyr) < 0.4$ domain, consistent with international standards for petrogenic hydrocarbons; only the Jask intertidal samples were very close to the petrogenic-pyrogenic boundary, which is possibly due to mixing with negligible amounts of surface runoff. In summary, the complete convergence of these four independent cross-plots confirms that the dominant source of PAHs throughout the study area is related to oil and gas industries, maritime transportation, and fishing port activities, while the contribution of combustion sources is negligible. The findings from the cross-plots of the present study were compared with three

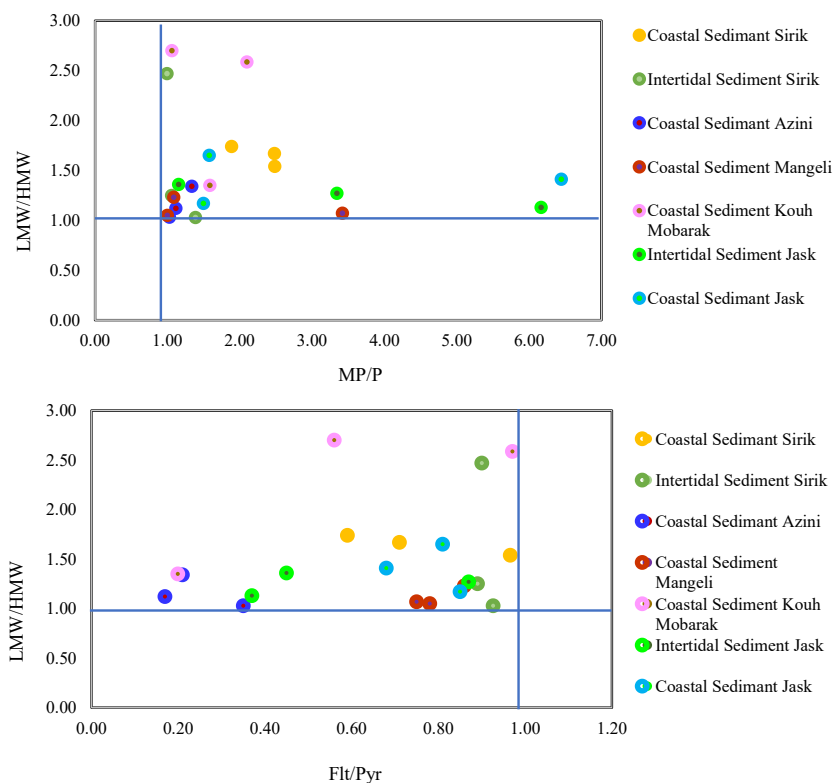


Figure 4. Cross-Plots of Diagnostic Ratios LMW/HMW Versus MP/P and Flt/Pyr in Coastal and Intertidal Sediments of the Makuran Coasts

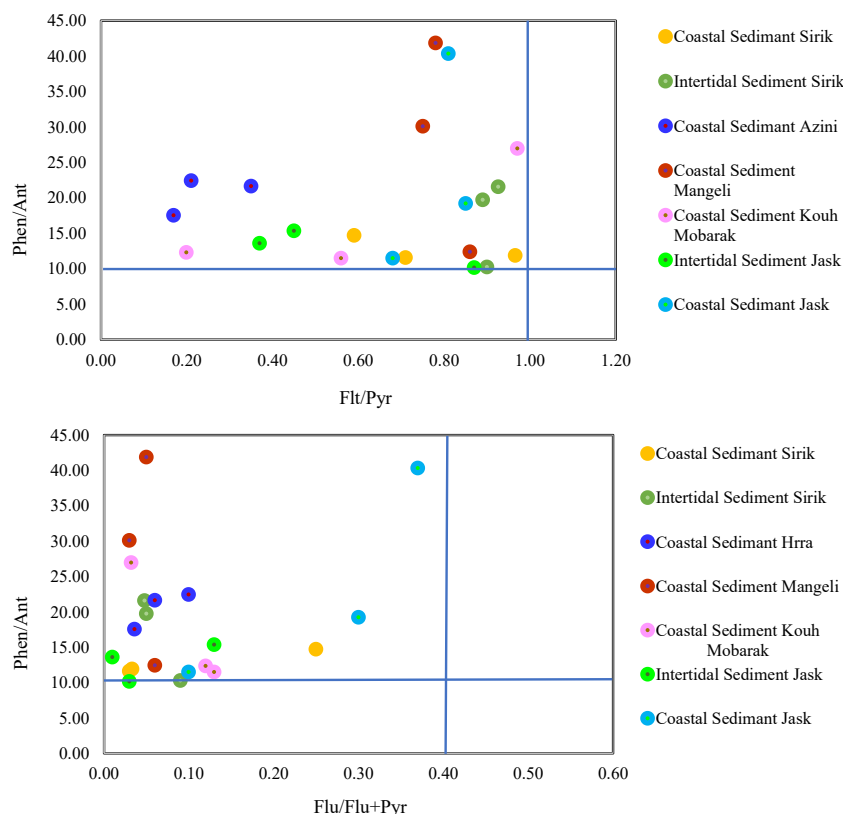


Figure 5. Cross-Plots of Diagnostic Ratios Phe/Ant Versus Flt/Pyr and Flu/Flu+Pyr in Coastal and Intertidal Sediments of the Makuran Coasts

previous studies from different regions of the world. The benchmark study of Soclo et al. in Benin and France, using six molecular indices (Phe/Ant, Flt/Pyr, LMW/HMW), demonstrated that the sediments of the Cotonou lagoon were predominantly of petrogenic origin, resulting from

small-scale petroleum trade and waste oils. This pattern is consistent with the present study, where all cross-plots (Figures 4–6) placed samples exclusively within the petrogenic domain ($LMW/HMW > 1$, $MP/P > 1$, $Ant/(Ant+Phe) < 0.1$, and $Flu/(Flu+Pyr) < 0.4$).²⁹ A

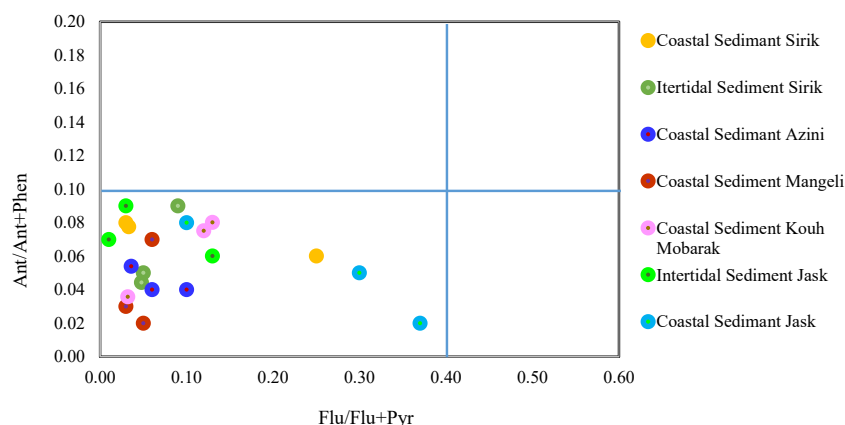


Figure 6. Cross-Plots of Diagnostic Ratios Flu/Flu+Pyr and Ant/Ant+Phe in Coastal and Intertidal Sediments of the Makuran Coasts

comparison with the study of Barakat et al. along the Mediterranean coast of Egypt reveals an important distinction. In that study, PAH sources were spatially variable, including petrogenic, pyrogenic, and mixed origins. In contrast, the present study along the Makuran coast exhibited a uniform and exclusively petrogenic signature at all stations, indicating a continuous linear source (likely the Gore-Jask pipeline) rather than site-dependent mixed sources.³⁰ Overall, the combination of diagnostic ratios ($LMW/HMW > 1$, $Ant/(Ant+Phe) < 0.1$, $Flu/(Flu+Pyr) < 0.4$) and molecular composition confirms a clear petrogenic signature for PAHs in the Makuran sediments. These findings indicate that petroleum-derived hydrocarbons are the dominant pollutants, while minor deviations among stations likely reflect localized pyrogenic contributions and post-depositional weathering. This pattern aligns with regional observations across the northwestern Indian Ocean, highlighting a shared contamination fingerprint among oil-transport corridors connecting Iran, India, and Egypt.^{14, 15}

Conclusion

This study provided the first integrated assessment of n-alkanes and PAHs to trace the origin of hydrocarbon pollutants in surface sediments from the Makuran coastal region (Sirik–Jask). The results consistently indicate a predominance of petrogenic inputs, as evidenced by low CPI values, even-to-odd carbon ratios, and PAH diagnostic ratios dominated by low molecular weight compounds. The combined use of both biomarker classes enhanced the reliability of source identification, with n-alkanes offering complementary insights to PAHs in distinguishing petroleum-derived contamination from natural organic matter. Intertidal sediments consistently accumulated higher hydrocarbon loads than adjacent coastal deposits, identifying these fine-grained, organic-rich zones as primary depositional sinks. These findings underscore the vulnerability of Makuran coastal sediments to hydrocarbon pollution, particularly in light of expanding maritime and industrial activities. The study highlights the need for routine environmental monitoring, adoption of integrated biomarker approaches, and the development of targeted mitigation strategies to protect the ecologically

sensitive habitats of the Gulf of Oman. Moreover, the results provide a valuable baseline for future ecological risk assessments and support evidence-based coastal management. Further research incorporating seasonal sampling and direct toxicity assessments is recommended to better constrain the ecological risks associated with the documented contamination.

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Competing Interests

The authors declared that there is no conflict of interest.

Ethical Approval

Not applicable.

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Supplementary Files

Supplementary file 1 contains Tables S1 and S2.

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