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Evaluation of the Contents of Aliphatic and Aromatic Hydrocarbons in Sediment from Zwitina Harbor Coast (Libya), as Indicator of Petroleum Pollution

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ABSTRACT

In the current study, sediment samples from Zwitina Harbor in northeastern Libya were analyzed using a GC-Mass apparatus. The objective was to evaluate how the region's activities related to petroleum use affected the environment. The findings indicated the existence of several hydrocarbon compounds that might have an impact on marine life. Numerous aromatic compounds and aliphatic hydrocarbons (C9 to C20) were found. Several polyaromatic hydrocarbons were detected, including Benzo(g, h, i) perylene, Acenaphthylene, Phenanthrene, Fluorene, Anthracene, Pvrene. Benzo(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)Pyrene, Indeno(1,2,3-cd)pyrene, and Dibenzo(a,b)anthracene. The study deduced that sediment samples showed the effects of activities related to the petroleum sector and that the majority of petroleum compounds in the area come from pyrolytic and petrogenic sources.

INTRODUCTION

Scopus

Industrial operations in the coastal region of Libya are mostly concentrated in major urban centers, such as Tripoli, Misurata, and Benghazi. These cities serve as hubs for several specialized industrial complexes, including those involved in petrochemicals, oil and gas refineries, as well as the steel sector. According to **Hamza (2022)**, the petrochemical complex in Abukammash, which is near Farwa Lagoon in the western region, poses a persistent threat to the marine ecosystem. Although there have been no documented instances of pollution crises thus far, the potential risks associated with this complex remain a concern.

An additional plausible contributor to oil contamination is represented by the coastal oil refineries, such as those situated in Zawiya, RasLanouf, Brega, Zwitina, and

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Tobruk, alongside the steel and iron facility in Misuratah. Over the course of the past two decades, a significant number of state-owned enterprises have collaborated with the EGA and Mediterranean Action Plan (MAP) to develop contingency strategies aimed at addressing inadvertent pollution incidents. Several investigations have been conducted worldwide to determine the presence and characteristics of hydrocarbons (Saleh, 2012; El-Fergani *et al.* 2023).

Prior research has demonstrated that oil pollution originating from refineries and industrial plants can have significant adverse effects on marine ecosystems and communities residing along coastlines. Alongside the endeavors of state-owned enterprises, the United Nations Environment Programme (UNEP) and other international organizations have actively participated in the surveillance and mitigation of oil pollution in the Mediterranean region (El-Hajaji, 2018; Bennett *et al.*, 2020; Naryono, 2023; Zhuang, 2023).

Hydrocarbon resources, comprising hydrogen and carbon, constitute the primary constituents of fossil fuels, such as coal, oil, and natural gas. Renewable energy sources are the predominant contributors to the world's primary energy supply, accounting for more than 85% of the total. Hydrocarbons are classified into aliphatic and aromatic substances according to their origins and characteristics. Aliphatic chemicals have the property of being either cyclic or acyclic, and possess a lower density compared to water, hence resulting in their ability to float on the surface. Aromatic hydrocarbons, which only consist of carbon and hydrogen atoms, demonstrate aromaticity and find applications in both biological and synthetic processes (Amin et al., 2022). Polycyclic aromatic hydrocarbons (PAHs), which consist of two or more benzene rings fused together, are organic pollutants of anthropogenic origin. Certain PAHs have been recognized as carcinogenic and mutagenic substances. Polycyclic aromatic hydrocarbons (PAHs) are organic compounds originating from the utilization of petroleum-based substances, as well as the incomplete combustion of fossil fuels or biomass. Examinations of polycyclic aromatic hydrocarbons (PAHs) in sediment samples collected from rivers, lakes, estuaries, and coastal regions provide insights into the spatial patterns and relative proportions of PAHs originating from petroleum-related and combustion-related sources (Patel et al., 2020).

Polynuclear aromatic hydrocarbons (PAHs), specifically 3, 4-benzopyrene, are widely recognized as carcinogenic agents, capable of inducing cancer. Crude oil contains trace levels of these compounds. Marine species can uptake polycyclic aromatic hydrocarbons (PAHs), which can subsequently be transferred up the food chain to humans. This raises significant concerns regarding human health (**Heo** *et al.*, **2019; Fu** *et al.*, **2022**).

The objective of this study was to assess the environmental impact of the petroleum industry on the coastal region of Zwitina in Libya. The evaluation of the contents of aliphatic and aromatic hydrocarbons in sediment from Zwitina Harbor Coast in Libya serves as a valuable indicator for assessing petroleum pollution levels. By analyzing the presence and concentration of these hydrocarbons, researchers can gain insights into the extent of contamination and potential environmental risks posed by petroleum-based pollutants. This evaluation would aid in understanding the impact on marine ecosystems and help formulate effective mitigation strategies to safeguard the coastal environment.

MATERIALS AND METHODS

1. Description of the studied area

The study area is in the Zueitina region, located west of the city of Benghazi at a distance of about 140km, and northeast of the city of Ajdabiya at a distance of about 20km at coordinates 30° 54' 54.30" N and 30° 05' 30.36" E. Samples were gathered for the investigation from five different locations on the sea during the winter and summer of 2023. GPS was used to determine the locations of each research sample, which were approximately spaced five kilometers apart. After being gathered, the samples were placed in plastic bags and brought to the laboratory. The Zwitina area is home to Libya's most significant fishing hub, one of the nation's busiest seaports and the biggest city in Cyrenaica. The Mediterranean Sea region has a temperate climate with mild winter temperature drops too much. The climate's most notable feature is that summers are hot and dry, while rainy seasons are characterized by variation in temperature (**Elemam & Eldeeb, 2023**).



Fig. 1. Geographic map of the study area (Google earth 2023).

1.1. Aliphatic and polycyclic aromatic hydrocarbons analysis

Aliphatic and PAHs analysis (The polycyclic aromatic hydrocarbons) was carried out following the techniques given by **UNEP** (1992). The compounds were analyzed by GC/ MS. Identification of each compound in the extract was made by comparing the retention times and its spectrum with those taken from HP memory and with the EPA standard at the Desert Research Center (Cairo, Egypt). The samples were analyzed for PAHs following different steps, including extraction, cleaning up, fractionation, instrumental analysis, and analytical quality control.

RESULTS AND DISCUSSIONS

1. Aliphatic hydrocarbons

The concentrations of aliphatic hydrocarbons in marine sediment samples are shown in Table (1). The results showed the types and n-alkane compounds containing the aliphatic hydrocarbons of C9 to C20. The average concentrations of hydrocarbon compounds in sediment samples fluctuated between 8.69 and $684.46\mu g/g$. This value is lower than those recorded in some Alexandria organisms ($451-1148\mu g/g$). Additionally, it is lower than the values reported by **El-Sikaily** *et al.* (2002) on some Mediterranean coasts ($180\mu g/g$). The results are in harmony with the results of the Neussrey (2013) study, where a concentration of $100\mu g/g$ was reported. However, it is higher than the data recorded by Hassaan *et al.* (2023) for the sediment of Bardaweel Lagoon along the Mediterranean ($51.98\mu g/g$).

It is worthy to mention that, the high concentrations of the detected aliphatic hydrocarbons (755.01g/g) are related to the C₉ compound in the sediment (**Asare** *et al.*, **2021**). This was less than the recorded levels for clean urban sites in Scotland, UK, with an average value of $3003\mu g/g$ (wet weight) (**Mackie** *et al.*, **1980**). However, it is less than the recorded level for the Black Sea, which ranged between 1200 to 24000 $\mu g/g$ of the sediment (**Readman** *et al.*, **2002**).

Commendatore and Esteves (2007) classified the TPH (Total Petroleum Hydrocarbon) concentration levels for oil pollution in coastal sediment into three levels: low (< $10\mu g/g$), low to moderate (10 - $100\mu g/g$), and moderate to high (100– $1000\mu g/g$). On the other hand, **Readman** *et al.* (2002) considered the sediment with a TPH concentration above $100\mu g/g$ as polluted. While, **Tolosa** *et al.* (2004) considered TPH concentrations higher than $500\mu g/g$ as a significant indicative of pollution and values below $10\mu g/g$ as unpolluted sediment. A significant proportion of the study's samples are categorized as polluted since they fall within the range of high TPH concentrations.

Station						
	WS1	WS2	ES1	ES2	ES3	Average ± SD
Compound						
C-9	755.01	645	662.99	689.80	667	684.46 ± 40.67
C-10	32.10	32.30	26.77	27.14	28.19	29.11 ± 2.61
C-11	12.33	8.85	9.31	11.10	12.50	10.63 ± 1.24
C-12	56.32	90.18	64.17	62.70	79.68	$68.83{\pm}10.40$
C-13	36.54	37.53	23.16	41.17	37.39	$36.54{\pm}7.72$
C-14	60.65	67.90	74.09	57.89	65.80	$63.57{\pm}~6.74$
C-15	20.87	26.50	24.05	20.45	26.70	$23.59{\pm}~2.24$
C-16	39.80	45.70	50.28	47.13	49.65	$47.6{\pm}~3.32$
C-17	14.60	16.80	16.49	15.56	16.87	$15.82{\pm}~0.83$
C-18	26.50	33.18	30.11	29.40	31.80	$30.69{\pm}~2.98$
C-19	7.85	8.26	8.1	9.12	8.55	$8.69{\pm}~0.48$
C-20	15.4	19.44	16.01	15.87	16.65	$16.51{\pm}~0.65$
ТРН	1077.97	1031.64	1005.53	1027.33	1040.78	1036.65 ± 26.4

Table 1. The concentration ($\mu g/g$) of aliphatic hydrocarbons in the marine sediment from the studied regions

* TPH \rightarrow Total aliphatic hydrocarbon.

1.1. Poly aromatic hydrocarbons (PAHs)

The poly aromatic hydrocarbons that were obtained and detected by the GCmass instrument in this study include Benzo(g, h, i)perylene, Acenaphthylene, Fluorene, Phenanthrene, Anthracene, Pyrene, Benzo(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Indeno (1,2,3-cd) pyrene, and Dibenzo(a,b)anthracene. The concentrations of PAHs fluctuated between 0.108 and 3.670µg/ g in sediment samples. Furthermore, the maximum value recorded for compound Indeno (1,2,3-cd) pyrene in the samples was 8.18µg/ (Table 2).

In general, the PAH contents recorded in this study were lower than those recorded for Derna coasts (Neussery, 2013). The concentrations of carcinogenic Benz(a) pyrene were also low in the studied samples (Table 2). This value is significantly below the average values of 5.59 and $0.86\mu g/g$ obtained from the Gulf of Suez and Gulf of Aqaba, respectively (Said & Hamed, 2006; Soliman *et al.*, 2023).

The present study recorded that the values of PAHs were 8.2029, which is to some extent safe and will have a weak but not harmful effect on marine sediment. The present investigation briefly concludes that the sources of PAHs in the studied area are mainly from incomplete combustion at high temperatures of recent and fossil organic matter (**Huang** *et al.*, 2023). Pyrolytic origins with little evidence of petrogenic origins atmospheric deposition, industrial discharges, and land runoff water are the main factors

responsible for pyrolytic PAHs. Various PAH concentration diagnostic ratios have been used to identify and quantify the contribution of each source of pollution to the specified environmental regions. The fluoranthene/pyrene (Fluo/ Pyr) ratio indicated the origin of PAHs. The significant aromatic fraction of the organic matter is responsible for the high PAH partitioning to sedimentary organic matter. Since sedimentary organic matter is a naturally occurring "heterogeneous polymer," it interacts with PAHs more favorably in the aromatic regions (**Hwang** *et al.*, **2003**; **Hasan** *et al.*, **2022**).

According to **Sicre** *et al.* (1997), the ratio of (flu/ Pyr) < 1 was linked to petrogenic origins, whereas the ratio of (flu/ Pyr) > 1 is associated with pyrolytic origins. The combustion of coal and wood produced (fluo/ Pyr) ratios of 2.180 and 2.632µg/ g, respectively. While crude oil and fuel oil had values of 0.6– 0.9 (Hasan *et al.* 2022); in the present study, all sites had a (fluo/ Pyr), ratio < 1, reflecting petrogenic origin. The ratio of major combustion-specific compounds Σ COMB = (Flu, Pyr, BaA, Chr, BbF, BkF, BaP, InP, and BghiP) to the sum of total PAHs (Σ COMB / Σ PAHs) were 0.7994 of the sediment marines in the study area, and the Σ COMB concentrations displayed value of 6.71 for sediment marine. The sum of six carcinogenic PAHs, Σ PAHs CARC, (Σ PAHs CARC = Flu, BaA, BbF, BkF, BaP, and BghiP) varied in marine sediment with a concentration of 3.262 for the studied samples.

Station	WC1	WGO	EC1	ECO	EGO	
Compound	W51	W 52	E91	E52	E93	
Benzo(g,h,i)perylene	0.007	0.006	0.009	0.001	0.0023	
Acenaphthylene	0.318	0.306	0.32	0.325	0.319	
Fluorene	0.278	0.297	0.30	0.304	0.307	
Phenanthrene	0.543	0.518	0.526	0.525	0.527	
Anthracene	0.390	0.406	0.422	0.419	0.408	
Pyrene	0.108	0.111	0.114	0.111	0.111	
Benzo(a)anthracene	0.689	0.704	0.716	0.716	0.718	
Chrysene	0.485	0.480	0.496	0.496	0.487	
Benzo(b)fluoranthene	0.372	0.369	0.363	0.363	0.372	
Benzo(k)fluoranthene	0.384	0.380	0.357	0.357	0.367	
Benzo(a)Pyrene	0.671	0.709	0.699	0.700	0.708	
Indeno(1,2,3-cd)pyrene	3.672	3.71	3.663	3.663	3.670	
Dibenzo(a,b)anthracene	0.208	0.204	0.205	0.205	0.206	
∑PAHs	8.125	8.201	8.191	8.189	8.203	
A viene and SD	$0.625 \pm$	0.631±	$0.630\pm$	$0.629 \pm$	0.631±	
Average±SD	0.936	0.947	0.934	0.934	0.936	

Table 2. The concentration $(\mu g/g)$ of poly aromatic hydrocarbons in the marine sediment from the studied regions

1.2. The Low Molecular Weight/High Molecular Weight ratio

The ratio of low molecular weight to high molecular weight compounds serves as an indicator, with low molecular weight representing polyaromatic hydrocarbon compounds with 2- 3 rings and high molecular weight representing polyaromatic hydrocarbon compounds with 4- 5 rings. This ratio provides insights into the composition and characteristics of the detected polyaromatic hydrocarbons in the samples (**Abdulla** *et al.*, **2023**).

The ratio of LMW/ HMW (Naphthalene + Acenaphthylene + Acenaphthene + Fluorene + Phenanthrene + Anthracene / Fluoranthene + Pyrene + Benzo(a)anthracene + Chrysene + Benzo(b)fluoranthene + Benzo(k)fluoranthene + Benzo(a)pyrene + Dibenzo(a,h)anthracene + Benzo(ghi)perylene + ndeno(1,2,3-cd) Pyrene) in sediment marine samples was > 1. This ratio indicates that the source of TPAHs is related to a petrogenic origin (**Hasan** *et al.*, 2022).

In general, the PAHs accumulated in the Mediterranean Sea sediments may be originated from different sources, such as sewage discharge from nearby human activity and fuel combustion emissions. The degree of sediment contamination by PAHs in this study is moderate in comparison with other aquatic systems in other countries. The difficulty in identifying PAHs origins is traced back to the possible coexistence of many contamination sources and the information processes that PAHs can diffuse in the air, water, or wastes before deposition in the analyzed sediments. Some compounds could exhibit comparable evolution kinetics that could be used to identify the origin of organic matter in the environment (Soclo *et al.*, 2000). Phenanthrene and Benzo(b)fluoranthene are components of fossil fuels, and a portion of them is associated with their combustion. Benzo(a)pyrene is usually emitted from catalysts, non-catalysts, and automobiles. Benzo(a)anthracene and Chrysene are often produced from the combustion of both diesel and natural gas (**El-Maradny** *et al.*, 2023).

Petroleum often contains a higher proportion of phenanthrene relative to anthracene since phenanthrene is a more thermodynamically stable tricyclic aromatic isomer than anthracene (Chokor & Achugwo, 2022). Hence, a Phe/ Ant ratio is observed to be very high in PAH petrogenic pollution, but low ratio in pyrolytic contamination cases (Ameur *et al.*, 2023; Areguamen *et al.*, 2023). Crude oil had a Phe/ Ant ratio of around 50, and motor vehicle exhaust had a ratio of around four (Yang, 2000), low Phe/ Ant ratio values (less than 10) indicate that the major PAH input was from the combustion of fossil fuel (Gschwend & Bites, 1981). Budzinski *et al.* (1997) suggested that sediments with Phe/ Ant > 10 were mainly contaminated by petrogenic inputs, and Phe/ Ant < 10 was typical of pyrolytic sources. In the present study, the ratio of individual PAH compounds in sediment samples reveals that the sources of PAH contamination might be different. Low Phe/ Ant ratios (< 10) were found in all locations in sediment samples, anticipating that they were pyrolytic-derived PAHs. However, the different Phe/Ant ratio values might be related to weathering, such as photo-degradation,

chemical degradation, or biodegradation, as well as the composition (for sediment samples). The (Flu/ Pyr) ratio can indicate the origin of PAHs. In the present study, the samples had (Flu/ Pyr) ratio values more than 1.

Other six diagnostic ratios between individual PAHs concentrations were calculated and used to make the identification of the PAHs origin more precisely as follows:

R-l = InP((lnP + BghiP); R-2 = Flu/(Flu + Pyr); R-3 = BaP/(BaP + Chr); R-4 = Phe(Phe)+ Ant), R-5 = BbF BkF and R-6 = Ant (Ant+Phe) (Mandalakis *et al.*, 2002). R-1 ratio is less than 0.4 for petroleum and > 0.5 for petroleum/ combustion mixture. Literature values of R-1 ratio are above 0.5 for grass combustion, wood soot and creosote (Yunker et al., 2002). Furthermore, R-3 ratio values are less than 0.2 for combustion and between 0.6 and 0.9 for petroleum, while ratios between 0.1 and 0.6 are due to petroleum/ combustion mixture. R-6 ratio < 0.1 is usually taken as an indicator of petroleum, while a ratio> 0.1 indicates a dominance of combustion. R-6 ratios > 0.1 represent the combustion of diesel oil, shale oil, coal, and some crude oil samples. The R-2 ratio of 0.4 is usually defined as petroleum, while a ratio of 0.5 is considered the transitional point between petroleum and combustion. However, this boundary appears to be less definitive than 0.1 for R-6. Generally, for the majority of petroleum samples, the R-2 ratio is usually below 0.4, but it is above 0.5 for kerosene and grass. Ratios of 0.4 to 0.5 are typical for most samples of coal and wood combustion, as well as creosote. This range is frequently linked to the combustion of liquid fossil fuels, such as gasoline, diesel, fuel oil, and crude oil, which are frequently detected in the emissions of diesel-powered vehicles and trucks (Hasan et al., 2022). On applying the results obtained from the above ratios, three groups emerged, each indicating a different source. The first group represents the samples of petrogenic sources, the second group represents the stations of pyrolytic sources (most ratios in the studied samples), and the third represents stations from a mixture of petrogenic and pyrolytic sources (Song et al., 2020; Grmasha et al., 2022).

CONCLUSION

Aliphatic and aromatic hydrocarbons found in the sediments of the Mediterranean Sea, particularly those along the coast of Zwitina Harbor in Libya, may have originated from human activities, the petroleum industry, or emissions from fuel combustion. PAHs can be identified based on their composition, as well as the patterns of distribution they exhibit, with pyrolytic contamination coming from the combustion of fuel and petrogenic contamination coming from crude oil. Diagnostic ratios are useful for identifying sources; they can point to one of three categories: petrogenic, pyrolytic, or mixed. According to the findings of this research, certain samples of sediment were found to include a variety of aliphatic and aromatic hydrocarbons. These hydrocarbons were shown to be the primary sources of the identified hydrocarbon compounds, mostly because of the effect that activities in the petroleum industry had. When compared to the levels of PAH pollution found in sediment in other aquatic systems in other countries, the levels found in this study are considered to be moderate. In general, the indications of pollution point to a low degree of risk to species. Nonetheless, in order to effectively implement steps to minimize pollution, it is essential to carry out additional monitoring of both the water and the sediment in this area.

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Conflicts of Interest:

All authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.

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