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Levels, distribution, and potential sources of polycyclic aromatic hydrocarbons in surface sediments of Petrolifera and Porto Romano ports in Albania

ABSTRACT

This study reports the levels, distribution,and sources of polycyclic aromatic hydrocarbons (PAHs) analyzed in surface sediment samples from Petrolifera and Porto Romano ports. They are well known for their industrial activity and good transportation.A total of 17 samples were collected from both ports in December 2023. A gas chromatography/flame ionization detector (GC/FID) was used to analyse the 13PAHs. The total concentrations of PAHs in sediments at PetroliferaPort varied from 102.9ngg^{-1} to 358.3ng g^{-1}, with an average value of 226.6ng g^{-1}. Similarly, the quantities in sediments from Porto Romano Port varied from 135ng g^{-1}to 1241ng g^{-1}, with an average value of 556ng g^{-1}. Sites within the ports have the highest concentrations of PAHs. Approximately 40% of the Σ13PAHs in Petrolifera and 39% in Porto Romano were attributed to 3-ringed PAHs within the analyzed areas. About 60% of the PAHs with rings between 4 and 6 were found in Petrolifera Port, and 61% in Porto Romano Port. The isomeric diagnostic ratio showed that PAHs in surface sediments in both areas originated from both sources, but mainly dominatedby pyrogenic sources. The results were compared with different sediment quality guidelines.

***Keywords**: port, PAHs, 3-ring, sediments, diagnostic ratio*

1. INTRODUCTION

Increasing urbanization and industrialization, the import and export of petroleum and related goods, and the positioning of cities close to ports are thought to be the primary causes of the increasing levels of organic pollutants in port areas [1]. Ports are very important to the development of country's trade and economy [2]. Their development has led to an increase of several contaminants [3]. Albania's major industrial ports are Petrolifera and Porto Romano, which are situated in Vlora and Durrësi Bay, respectively. Petrolifera Port is known for handling and transportation of crude oil and refined petroleum products. The port is equipped with specialized infrastructure for the loading and unloading of oil tankers, including storage tanks and pipelines for efficient transfer.

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The proximity to refineries in Greece and Italy is its main advantage. Porto Romano serves as a vital port for shipping and trade, facilitating the movement of goods (construction materials, consumer goods, agricultural products, fisheries), in and out of Albania. This port is involved in handling liquefied natural gas (LNG) and byproducts from the oil and gas industry along the coast. The construction of Porto Romano seaport is now underway. One class of organic compounds commonly analyzed in marine sediments is polycyclic aromatic hydrocarbons or PAHs[4,5]. Due to hydrophobicity, low solubility in water, and high vapor pressure, the sediment samples are considered the last sink of PAH[6].Numerous literature have reported that carcinogenic and mutagenic PAHs are widely present in marine environments [7,8].Therefore, seawater contamination may be reflected in the PAH quantities found in sediments over a prolonged time. Many sources, including marine fuels and oils, vessel operations, cargo handling, stormwater runoff, industrial activities, sediment resuspension, atmospheric deposition, and waste disposalreleased PAHs in

the marine environment [2,4,5]. Their anthropogenic sources are divided intopetroleum-associatedPAHs and pyrogenic-associated PAHs[9,10]. Petroleum-associated hydrocarbons are produced during crude oil maturation.They can be present to port areas during their storage, transportation, utilization, production, and leakage. Pyrogenic-associated PAHs are mainly produced during the incomplete combustion of petroleum fuels, coal, biomass, and other hydrocarbons. Also, they can be transported to port areas via different pathways, including atmospheric transport, wet and dry deposition, surface runoff, and river discharges [11,12]. Referred to other studies, the origin of PAHs is defined using different diagnostic ratios. The ratios of ‘‘anthracene/anthracene + phenanthrene, benz(a)anthracene/ benz(a)anthracene+ chrysene, and indeno(1,2,3-c,d)-pyrene/indeno (1,2,3-c,d)+benzo[g,h,i]perylene are the most used in literature[13]. The main objectives of this study were: i) to determine the levels of PAHs in both areas; ii) to define the potential sources of PAHs and evaluate the results using sediment quality guidelines (SQGs).

2. MATERIALS AND METHODS

Sampling sites

A total of 17 stations were established throughout the two industrial ports, in Albania, with 8 stations atPertoliferaPort and 9 stations at Porto Romano Port, during May 2023. Eight surface sediment samples were taken from various stations, both inside and outside Petrolifera Port, labeled SP-1 to SP-8. Samples SP-1, SP-2, SP-7, and SP-8 were collected from outside the port, while the remaining samples were gathered within the port area. The same methodology was applied to the nine sediment samples collected in Porto Romano Port. The samples (SPR-1, SPR-2, SPR-8, and SPR-9) were collected outside the port. All sampling stationswere selected to cover all the areas impacted by shipping and industrialactivities. The locations of the sampling sites are illustrated in Fig. 1.

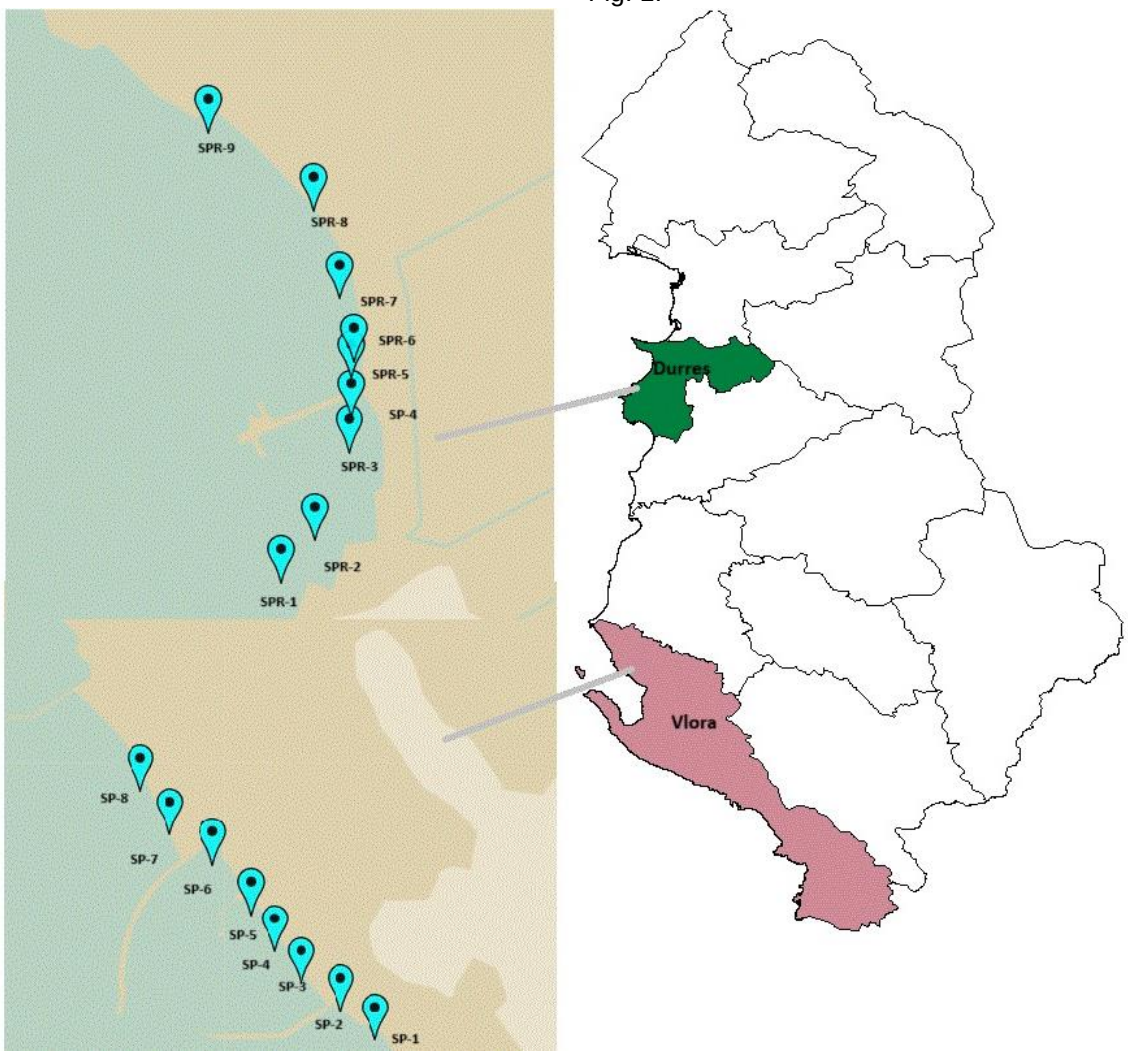

Figure 1. Map of sampling area and the sampling station: a)Porto Romano Port, and b) Petrolifera Port

Sample treatment

A van grab was used to collect the sediment samples in both sites. After the sampling, all the samples were kept in ice boxes for further analysis in the laboratory. The sediment samples were air dried, crushed with a mortar, andsieved through a 63 mesh to remove debris and obtain awell-mixed sample.The detailed procedure ofextraction and qualitative and quantitative analysis of PAHs in sediment samples is describedbyNuro et al. 2024 [14]. In this study, theindividual PAH analyzed in the sediment samples were anthracene (ANT), acenaphthylene (ANP), phenanthrene (PHE), fluorene (FLU), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), perylene, benzo[b]fluoranthene (BbF), benzo[k] fluoranthene (BkF), indene[1,2,3-cd]pyrene (IcP), dibenzo[a,h] anthracene (DhA),benzo[ghi]perylene (BgP).A gas chromatography/flame ionization detector (GC/FID) was used to analyse the 13PAHs.

3. RESULTS AND DISCUSSION

Descriptive analyses were performed to determine the distributions of the data. Some individual PAHs were not detected, and during the descriptive analysis their value was set to zero. All the statistical analysis was done using MiniTab (version 22) and Excel software.

Concentration of PAHs in sediment samples

The measured concentrations of 13 priority PAHs and the total sum (Σ13PAHs) analyzed in the surface sediments from Petrolifera and Porto Romano Ports are shown in Tab. 1. The level of individual PAH ranged widely in both investigated areas. Sediment samples from PetroliferaPort showed thatthe total concentration of 13 PAHs ranged from 102.9 to 358.3 ng g^{-1}, with a mean value of 226.6 ng g^{-1}. In Porto Romano, the total PAH concentration ranged from 135 to 1241 ng g^{-1}, with a mean value of 556 ng g^{-1}. The PAH concentration in sedimentscollected inside ports showed higher levels than those analyzed outside the areas. The same finding was reported in other studies [15,16].Sediment from Porto Romano Port recorded the highest total PAHs concentration, which could be due to the intense activities of ships.

Table 1. The mean, median, minimum, and maximum of individual PAHs and the total PAHs levels in Petrolifera and Porto Romano ports. The values used as guidelines according to CCME

Analytes	Petrolifera Port				Porto Romano Port				CCME	
	Mean ng/g	Median ng/g	Min ng/g	Max ng/g	Mean ng/g	Median ng/g	Min	Max ng/g	ISQGs	PEL
Acenaphthylene (ANP)	17.84	18.43	n.d	51.30	**144**	1.10	n.d	1098	6.71	88.9
Fluorene (FLU)	16.23	18.45	0.25	37.90	**106**	39.85	n.d	652	21.2	144
Phenanthrene (PHE)	20.84	23.50	2.10	34.90	0.613	0.181	n.d	3.535	86.7	544
Anthracene (ANT)	**31.3**	26.6	0.7	100.3	**0.704**	0.280	n.d	4.022	46.9	245
Pyrene (PYR)	17.78	10.13	n.d	48.50	**74.7**	4.10	n.d	599	153	1398
Benzo(a)anthracene, (BaA)	**18.79**	16.27	n.d	37.95	**63.7**	40.10	n.d	332	74.8	63
Chrysene (CHR)	16.92	10.10	n.d	46.90	28.5	n.d	n.d	179	108	846
Perylene	4.29	n.d	n.d	33.55	5.817	n.d	n.d	28.55	-	-
Benzo[b]fluoranthene(BbF)	21.19	12.07	n.d	65.60	5.522	n.d	n.d	35.80	-	-
Benzo[k]fluoranthene(BkF)	21.51	25.19	n.d	41.80	**49.30**	n.d	n.d	287.6	-	-
Indeno[1,2,3-cd]pyrene (IcP)	**22.45**	16.32	n.d	79.35	7.36	n.d	n.d	37.50	-	-
Dibenzo[a,h] anthracene (DhA)	12.44	0.0	n.d	68.85	4.100	n.d	n.d	36.90	6.22	135
Benzo[ghi]perylene (BgP)	4.98	0.0	n.d	39.80	n.d	n.d	n.d	n.d	-	-
ΣPAHs	**226.6**	**195.2**	**102.9**	**358.3**	**491.4**	**422**	**84.6**	**1237**	**-**	**16700**

n.d –no detected, CCME- Canadian Council of Ministers of the Environment, ISQGs-Canadian interim marine sediment quality guidelines, PEL-Probable Effects Level

In both areas,Perylene, Dibenzo [a,h] anthracene, and Benzo[g,h,i]perylene represented in most stations were non-detected. The highest concentration of PAH was found for ANT in station SP-1 (Petrolifera) and ANP was found with the highest concentration in SPR-7 (Porto Romano). The high abundance of low molecular weight PAHs in some stations suggests relatively recent local PAH sources that entered the seawater[17].

The findings revealed that the light PAHs were present in both areas, whereas the heavy PAHs were mostly present in Porto Romano Port. Numerous sources, including accidental oil spills, ship activities, emissions from fuel burning, discharged waters, closeness to the cities, and human activities can contribute to the difference in PAH content along the examined locations.

The Canadian interim marine sediment quality guidelines (ISQGs) is widely applied, to find out the PAH pollution in marine environment. (Tab. 1). The results from Petrolifera samples showed that some of the PAHs contents in sediments of this port were lower than the levels reported by CCME. Acenaphthylene and Dibenzo[a,h]anthracene were found in higher levels than those reported in ISQGs. The results from Porto Romano samples showed that Acenaphthylene and Fluorene were found at higher levels than those reported from ISQGs. Based on this guideline, the sediment samples from both areas have less potential for negative effects. The results found in both areas are lower than the values of Probable-Effects Level (PEL).

The levels of PAHs in sediments are frequently used to classify an area as polluted or not. In Tab. 2, are given the range of PAHs quantities, indicating the level of pollution in sediments.

Table 2. The range values for the classification of pollution [18]

PAHs quantities (range)	Level of pollution
0–100 ng/g	light pollution
100–1000 ng/g	moderate pollution
1000–5000 ng/g	high pollution
> 5000 ng/g	extreme pollution

The range of PAHs content in the sediments from Petrolifera Port (ΣPAHs 102.9-358 ng g^{-1}) and Porto Romano Port (ΣPAHs 105.4-1241 ng g^{-1}) indicated light to moderate pollution,except the station SPR-6 and SPR-7 that were considered as highly polluted, according to the total PAHs concentration. The main factors that have increased the concentration of PAHs in both areas are their geographical location, export of goods, ship repair, and incoming fuel from ships. The individual plot of PAH concentration in both areas is shown in Fig. 2.

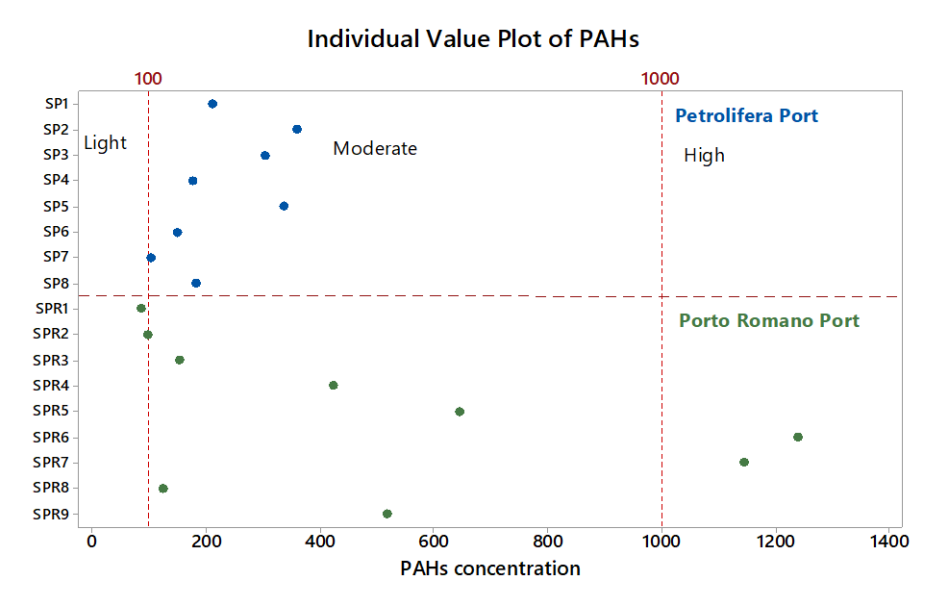

Figure 2. The plot of individual PAHs in both areas

According to USEPA [19], the possible carcinogenic PAHsare BaA,CHR,BaP, BbF, BkF, Ba, hA, BgP, and IcP. The sum of carcinogenic PAHs labeled as Carc_PAHs, was calculated. BaP is not analyzed, and the sum is calculated for the rest of the PAHs. In sediments from Petrolifera Port, Carc_PAHsranged from 5.2 to 232 ng g^{-1}, and in Porto Romano Port ranged from 5.2 to 577 ng g^{-1}. The ratio of ΣCarc/ΣPAHs expressed in %, ranged from 3% to 76 % for samples in Petrolifera Port and

1% to 76 % for Porto Romano. Another parameter that has been calculated in this study is the total amount of the main combustion-specific PAH. The Comb_PAH (BbF; BaA; CHY; PYR; FLU; BgP; BaP; IcP; and BkF), ranged from 63 to 239 ng g^{-1} and 21.9 to 1273 ng g^{-1} in Petrolifera and Porto Romano,respectively.The percent of the ratio Comb_PAH to ΣPAHs ranged from 36% to 76%, and in PortoRomano ranged from 2% to 99%.

Distribution of PAHs

The composition pattern of PAHs by ring size in sediment samples from Petrolifera and Porto Romano Ports are shown in Fig. 3. The compositionof the sediment samples from Petrolifera Port were found in the order; 3-ring PAHs > 4-ring PAHs > 5-ring PAHs >6-ring PAHs (Figure 3).On average, the 3-ring PAHs were the most predominant compound accounting 40 % and 4-ring PAHs accounting 26 % of the total PAHs. In Porto Romano Port the distribution were found in the order: 4-ring PAHs > 3-ring PAHs > 5-ring PAHs >6-ring PAHs. The 4-ring PAHs contributed with 44 % and 3-ring PAHs with 40% of the total PAH concentrations.

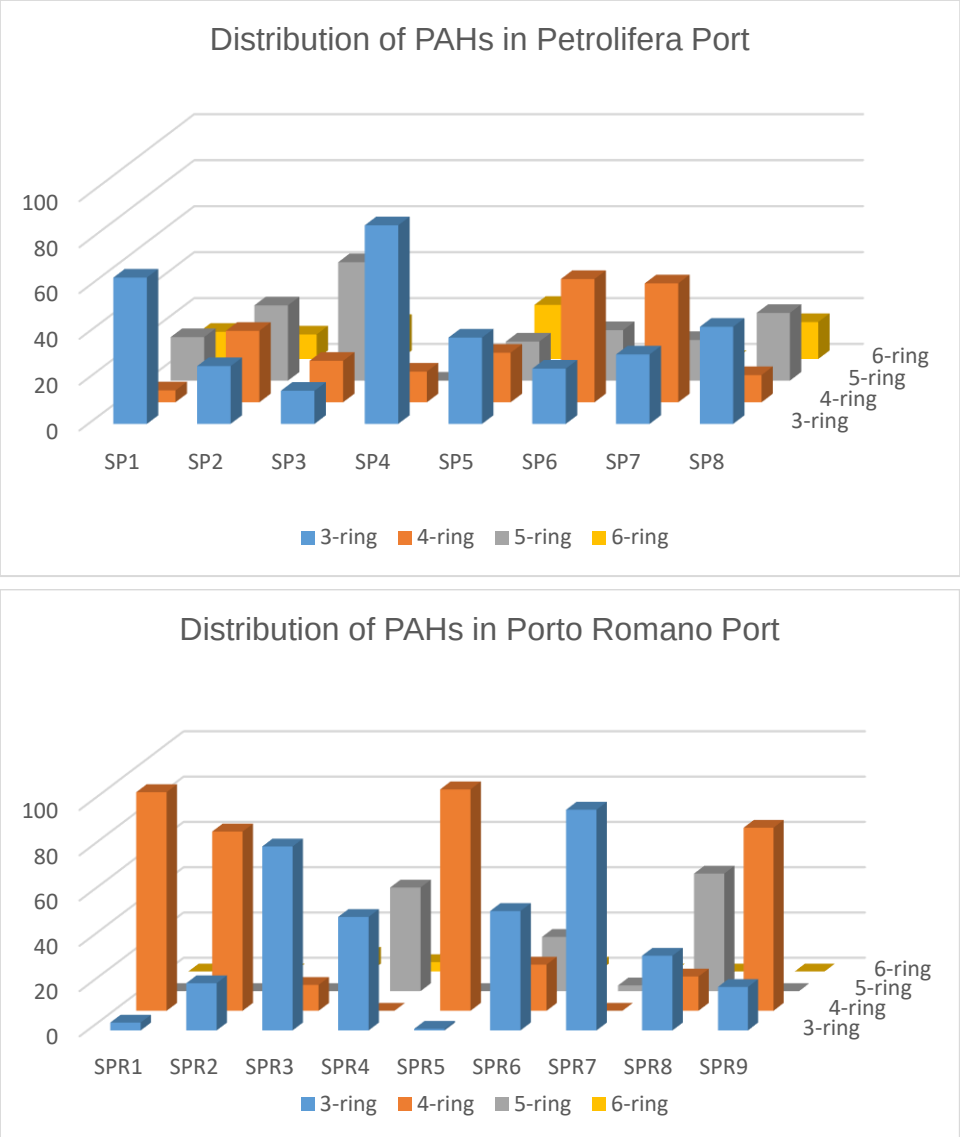

Figure 3. The composition profile of different ring numbers in surface sediments from Petrolifera and Porto Romano Ports

PAHs sources using diagnostic ratio

The measured PAHs were categorized using Low molecular weight rings (LMW, up to 3-rings, MW<202) and Heavy molecular weight rings (HMW, 4-rings and above, MW>202). In Petrolifera Port, the LMW PAHs dominated the surface sediment sampleswith 41 %, whereas in Porto Romano, the LMW PAHs dominated the sediment samples with 40 %. The source of PAH fractions can be identified by comparing the ratio between

Low molecular weight rings (LMW) and heavy molecular weight rings (HMW). The ratio of LMW to HMW >1 indicates dominance of petrogenic sources and lower than 1, indicates pyrolytic sources. The sediment samples from both areas indicated the dominance of pyrolytic sources.

To investigate the potential sources of PAHs were calculated the most used ratio of BaA/(BaA + CHR) and ANT/(ANT + PHE), and IcP/(IcP + BgP)[13]. The distribution of the ratios BaA/(BaA + CHR) versus ANT/(ANT + PHE), and IcP/(IcP + BgP) versus ANT/(ANT + PHE) are shown in Fig.4. In both areas, the ratios of ANT/(ANT+ PHE) were higher than 0.1 with a mean value of 0.5 in Pertolifera and 0.43 in Porto Romano, indicating that PAHs derived from different combustion sources (pyrolytic sources). In this study, the BaA/(BaA+CHY) fluctuated between 0 and 1 with an average value of 0.56 which indicates petroleum combustion (0.20–0.50), and fossil fuel combustion (>0.35). Our results are shown in Fig.4, where the major stations pointedto pyrolytic sources.

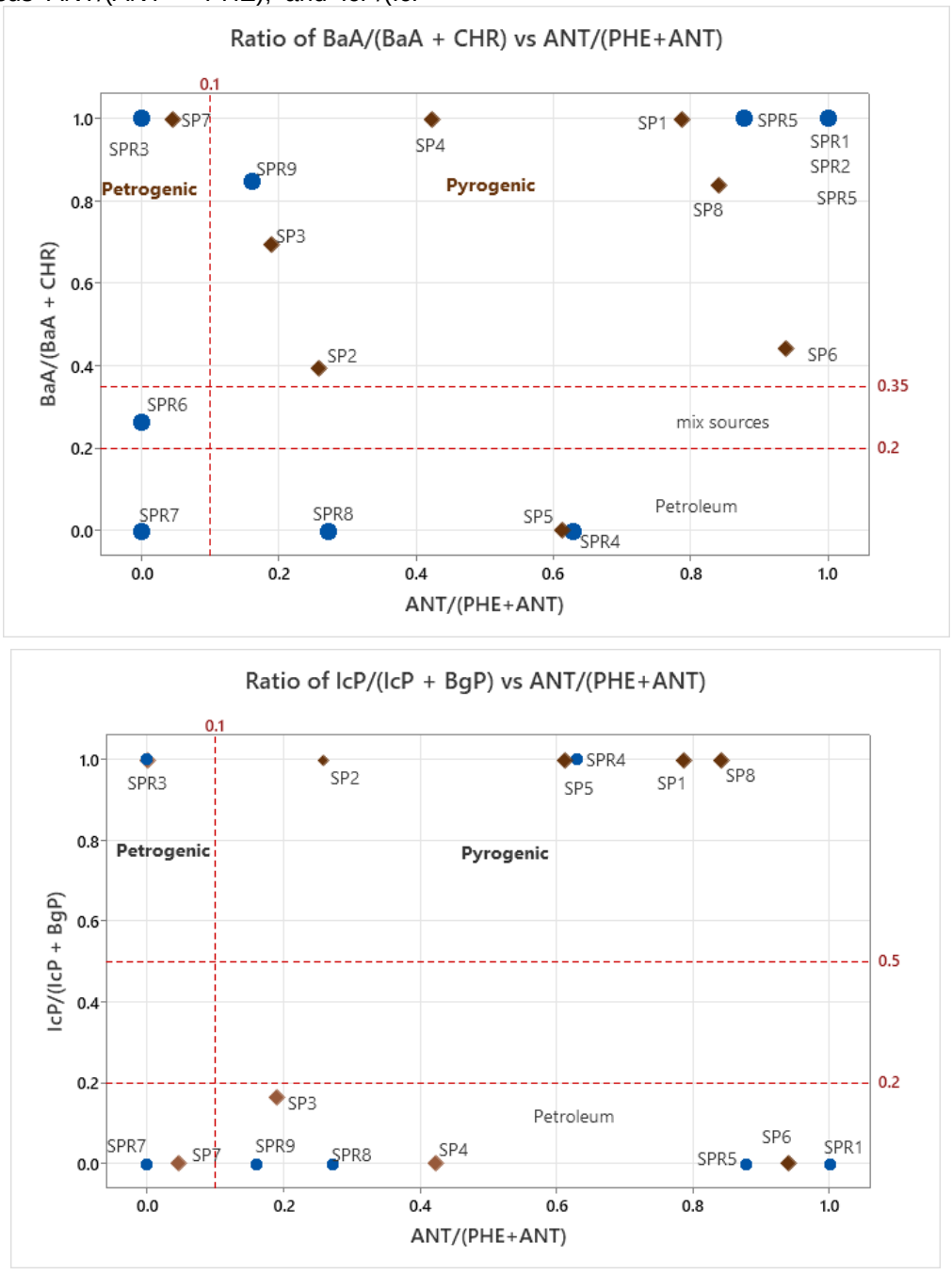

Figure 4. Possible sources of PAHs, identified using diagnostic ratio of BaA/(BaA + CHY) a and IcP/(IcP + BgP) versus ANT/(ANT + PHE) in both areas.

The distribution of the ANT/(ANT + PHE) and BaA/(BaA + CHR) ratios showed that the PAHs in the sediments of both areas originated mainly from combustion and/or pyrolysis sources.The ratios of ANT/(ANT + PHE) and IcP/(IcP + BgP) showed that PAHs in the sediments were mainly contributed by the combustion of petroleum. PetrogenicPAHs have frequently entered the environment through the usage, storage, and transportation of crude oil and its derivatives, storage tank breaches, oil spills, motor oil and gasoline leaks, and small gasoline.All release petroleum-based PAHs into the marine environment. Pyrogenic PAHs are formed by incomplete combustion or pyrolysis of the organic matter at very high temperatures. The main contributors are the incomplete combustion of carbonaceous matter, burning and pyrolysis of coal, oil, gas, trash, wood, vehicular emissions, and industrial processes. The dominance of 3- and 4-ring PAHs is indicative of the potential sources of low-temperature combustion/pyrolysis of biomass (coal, wood, and straw) or petroleum-related fuels [20,21].

Ecosystem Risk Assessment

To assess the ecological risk of PAHs in sediments from Petrolifera and Porto Romano Ports, total and individual PAH levels were compared to effects-based guidelines, including two target values: the effects range-low (ERL) and effects range-median (ERM) [22]. Total PAH concentrations at study sites were below the sediment quality guidelines (SQGs) for both the ERL (3442 ng g−1) and ERM (24290 ng g−1), suggesting that the sediments were not toxic to local organisms. However, some stations showed elevated levels of Acenaphthylene, Fluorene, Anthracene, and Benzo(a)anthracene, exceeding the ERL but remaining below the ERM values. This indicates that occasional biological effects could occur. The sediment quality guidelines values for PAHs and the relative percentage of samples within each SQG range are summarized in Table 3.Although some stations had individual PAH concentrations exceeding the ERL but still below the ERM, the overall results indicate no significant toxic effects. However, these elevated levels at certain stations could lead to occasional negative impacts on the ecosystem.

Table 3. Sediment quality guidelines values for PAHs and relative percentage of samples amongst ranges of ERL and ERM values.

PAH	SQGs		% of stations			% of stations		
	ERL ng g^{-1}	ERM ng g^{-1}	Petrolifera Port			Porto Romano Port		
			<ERL	ERL-ERM	>ERM	<ERL	ERL-ERM	>ERM
NP	44	500	87.5	12.5	-	66.7	33.3	-
FLU	19	540	75	25	-	44.4	55.6	-
PHE	240	1500	100	-	-	100	-	-
ANT	85.3	1100	87.5	12.5	-	100	-	-
PYR	665	2600	100	-	-	100	-	-
BaA	261	1600	100	-	-	88.9	11.1	-
CHR	385	2800	100	-	-	100	-	-
Perylene	-	-	-	-	-	-	-	-
BbF	-	-	-	-	-	-	-	-
BkF	-	-	-	-	-	-	-	-
IcP	240	950	100	-	-	100	-	-
DhA	63.4	260	100	-	-	100	-	-
BgP	85	330	100	-	-	100	-	-

Concentration <ERL- Biological effects are rare

ERL ≤Concentration ≤ ERM -Biological effects may occasionally occur

Concentration >ERM -Negative biological effects are frequent

4. CONCLUSIONS

This study reports the levels, distribution, and potential sources of PAHs in the surface sediments from 17 stations in total, from two major industrial ports in Albania, Petrolifera and Porto Romano. The sediment samples from Petrolifera Port showed light pollution compared with samples from Porto Romano, which showed light to moderate pollution, except two stations that showed a high level of pollution. The primary sources of PAHs in the sediments of both ports were found to be pyrogenic, such as from incomplete and complete

combustion of petroleum products, including emissions from boats, ships, and vehicle engines. Total PAH concentrations in the sediments were found to be below the Effects Range-Low (ERL) and Effects Range-Median (ERM) values, suggesting that there is no significant toxicity to the local organisms.The study offers useful baseline information on PAH pollution in these ports, emphasizing the need for ongoing observation to better understand the situation and inform future environmental management strategies.

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IZVOD

NIVOI, DISTRIBUCIJA I POTENCIJALNI IZVORI POLICIKLIČNIH AROMATIČNIH UGLJOVODONIKA U POVRŠINSKIM SEDIMENTIMA LUKA PETROLIFERA I PORTO ROMANO U ALBANIJI

Ova studija prikazuje nivoe, distribuciju i izvore policikličnih aromatičnih ugljovodonika (PAH) analiziranih u uzorcima površinskih sedimenata iz luka Petrolifera i Porto Romano. Poznati su po svojoj industrijskoj aktivnosti i dobrom saobraćaju. Ukupno 17 uzoraka je prikupljeno iz obe luke u decembru 2023. Za analizu 13 PAH-ova korišćen je gasna hromatografija/detektor plamene jonizacije (GC/FID). Ukupne koncentracije PAH u sedimentima u luci Petrolifera varirale su od 102,9 ng g^{-1} do 358,3 ng g^{-1}, sa prosečnom vrednošću od 226,6 ng g^{-1}. Slično tome, količine u sedimentima iz luke Porto Romano varirale su od 135 ng g^{-1} do 1241 ng g^{-1}, sa prosečnom vrednošću od 556 ng g^{-1}. Lokacije unutar luka imaju najveću koncentraciju PAH. Približno 40% S13PAH-a u Petroliferi i 39% u Porto Romanu pripisano je PAH-ovimasa 3 prstena unutar analiziranih oblasti. Oko 60% PAH-ova sa prstenovima između 4 i 6 pronađeno je u luci Petrolifera, a 61% u luci Porto Romano. Izomerni dijagnostički odnos je pokazao da PAH u površinskim sedimentima u oba područja potiču iz oba izvora, ali uglavnom dominiraju pirogeni izvori. Rezultati su upoređeni sa različitim smernicama za kvalitet sedimenta.
Ključnereči: *luka, PAH, 3-prsten, sedimenti, dijagnostički odnos*

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