



# Seasonal variations of polycyclic aromatic hydrocarbons in coastal sediments of a marine resource hot spot: the case of pars special economic energy zone, Iran

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**Abstract** Polycyclic aromatic hydrocarbons (PAHs) are an important group of compounds of major environmental concern, which are in the class of persistent organic pollutants. Therefore, the key purpose of this research was to analyze seasonal fluctuations and to determine the probability of polycyclic aromatic hydrocarbons in coastal sediments of the Iranian Marine Resource Center based on the evaluation of 16 US-EPA important PAH compounds. These compounds have been collected from intertidal sediments located in the marine resources of southern Iran in different seasons. These samples of the surficial sediment were collected at the PSEEZ area using a stainless steel grab sampler in four

seasons, from depths between 0.5 and 30 m. Surface sediment samples were removed by spoons and carefully placed in an aluminum foil; they were taken to the laboratory on ice and held at 20° C until their study. After extraction, by using a rotary evaporator apparatus, samples were condensed. The assay was added to roughly 2 g of activated copper flasks in the refrigerator for 36 h for desulfurization. Among different seasons, the highest concentration was observed in winter, with a mean of 281.3 ng g<sup>-1</sup>. According to ecological risk assessment (concentrations of possible effects, low effect range, degree of threshold effects, and median effect range), PAH risks in surface sediments of PSEEZ were lower than the

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threshold results levels (TEL), possible effects levels (PEL), low range of effects (ERL), and median range of effects (ERM), indicating that a biological effect would rarely occur. The dry weight scale of the concentration of  $\sum$ PAHs ranges from 145.7 to 348.42 ng g<sup>-1</sup> with a mean quantity of 260.52 ng g<sup>-1</sup>. Therefore, according to the amount of  $\sum$ PAH concentration, the sediments in the PSEEZ area indicated moderate to heavy pollutions. In this way, the sedimentary surface ecosystems of the Persian Gulf were considered as moderately polluted compared with other ecosystems worldwide. Our study highlighted some of the research gaps in PAH contamination studies and the level of PAH contamination. Therefore, this study will provide a scientific background, planning, and policies for PAH pollution control and environmental protection in Iran and similar regions around the world.

**Keywords** Environmental risk assessment · Diagnostic ratios · Sediments pollution · Petroleum industries

## Introduction

The Persian Gulf is a low, deepwater region with a mean depth of 35 m and an area of 240,000 km<sup>2</sup> (Agah et al., 2009; Akhbarizadeh et al., 2019), which is located in the south of Iran and is connected to the international waters via Hormuz strait. Intense human activity is observed in this region due to the extremely rich reserves of oil, gas, and minerals. Rapid industrialization, urbanization, transportation, and development of oil refineries and petrochemical industries, along with tourism and aquatic activities, are commonplace activities in the region (Gevao et al., 2016; Lübeck et al., 2016). As a result of such an extensive development, various types of pollutants, including organic and inorganic ones like nutrients, toxic metals, and volatile organic compounds (VOCs (PAHs)), have been massively released into marine sediments (Davoodi et al., 2017). The activities may lead to an increase in the amounts of PAHs in coastal areas. According to Zhang, Wang, et al. (2020), Zhang, Pan, et al. (2020) and Mehr et al. (2020), PAHs commonly come to the coastal zone as a result of municipal and industrial wastewater dump, urban runoff, combustion

processes associated with petroleum industries projects, and urban activities (including transportation and other municipal activities) (Mehr et al., 2020; Zhang, Pan, et al., 2020; Zhang, Wang, et al., 2020).

As a result of the widespread development of human activities, various types of pollutants, including organic and inorganic pollutants such as nutrients and PAHs, have been significantly released in coastal sediments, which have destructive effects. This release will affect human health and ecosystems and other organisms (Sharma & Chatterjee, 2017; Smith et al., 2018). Combustion of multiple fossil fuels and penetration of refined products and oils are widely known as mutants, carcinogens, and teratogens, and therefore endanger human health. The physicochemical properties of PAHs make them highly transportable materials in the environment and allow them to be distributed through water, soil, and air ecosystems. As a result, humans, as living organisms that depend on other living organisms for their survival, including aquatic organisms, will be affected by the adverse effects of this phenomenon. Therefore, awareness of the dangerous effects of these toxic substances on deep-sea creatures is essential to maintain the well-being of human beings (Karbalaei et al., 2018; Laffoley et al., 2019; Lu et al., 2019). PAHs are thus an essential category of compounds of considerable environmental significance. PAHs are highly mutagenic and humanly carcinogenic. The main cause of human susceptibility to PAHs is food exposure. As a result of food processing methods such as curing, drying, burning, roasting, grilling, barbecuing, and refining, PAHs are found in food (Adeyeye, 2020; Tongo et al., 2017). It is understood that these food processing steps produce and increase the amount of PAHs in the food. Sea food is one major food source of PAHs. In developed countries, most diets contain fish and other aquatic foods as a common source of protein. Smoke not only provides a special flavor and fragrance to these foods but also increases preservation because of its dehydrating and bactericidal effects. Smoke, however, includes PAHs, many of which are carcinogenic (Bauer et al., 2018; Malarut & Vangnai, 2020; Sun et al., 2019). Smoking is the most common process used to conserve seafood in developing countries such as Iran (south of Iran), and smoked food items are the most available source of food for consumption. Thus, the risk of pollution of PAHs by ingestion has increased. Food protection is of

rising concern worldwide and PAH residues could raise significant public health issues if they are found in sea foods above the prescribed thresholds. The high concentration of benzo[a]pyrene in *C. gariepinus* and *E. fimbriata* above the guideline value of 0.05 mg/kg calls for serious health concern (Yusuf et al., 2015). As a result, if the PAH4 levels in food are higher than recommended, there will be serious concerns for human health. The findings of this study reveal that there are significant toxicity and potential carcinogenic risk to human health from the ingestion of Southern Iranian species of seafood.

PAH compounds are a type of organic chemical materials that comprise two to seven molten aromatic carbon rings with an angular linear and cluster arrangement. These substances are categorized into high molecular weight (HMW) and low molecular weight compounds, depending on their molecular weight (LMW) (containing 4–6 combined rings) (containing two or three benzene rings) compounds (Kim et al., 2013; Konstantinova et al., 2020; Oracz & Zyzelewicz, 2019; Tolosa et al., 2005). PAH compounds are also one of the most substantial groups of hazardous pollutants that are generated from both anthropogenic and natural origins (Darilmaz et al., 2013). The burning of numerous fossil oils and line oil and refined products penetration provide the anthropogenic basis (Liu et al., 2012; Yun et al., 2016). Some of these compounds are widely known as mutagens, carcinogens, and teratogens, and therefore endanger crucially the health of living organisms (Taghvaei et al., 2018). The physicochemical traits of PAHs make them highly moveable materials in the environment, permitting them to distribute through water, soil, and air ecosystem where their presence is ubiquitous (Baklanov et al., 2007; McCubbin et al., 2019; Qiao et al., 2006; Sverdrup et al., 2002). In aquatic ecosystems, PAH compounds bind to suspended solids and are deposited (Hu et al., 2014; Kesavvarzifard et al., 2017). Consequently, the impurity of hydrocarbons in sediments will endanger deep-sea creatures. Therefore, as a result of consuming them as a food source, it may lead to acute or chronic poisoning in humans (Bellasi et al., 2020; Ramzi et al., 2017; Zeng et al., 2013; Zhang, Pan, et al., 2020a, b).

Human actions increase the concentration of PAHs in waters and sediments in aqueous environments. The

general global issues resulting from the growth of industrial and agricultural operations are water pollution and the lack of access to safe water (Wang et al., 2017). In recent decades, chemical compounds such as polycyclic aromatic hydrocarbons (PAHs) have been detected extensively in aquatic environments. Moreover, the number of new organic compounds entering the world market is growing significantly every year, and most of these compounds, including pharmaceuticals, pesticides, personal care goods, and PAH surfactants, are used in vast amounts in farming operations worldwide, after which they are released into various water sources where they can live, creating extreme heaven (Zambianchi et al., 2017). Recent studies have stated the occurrence of organic pollutants, such as PAHs, in various aquatic systems such as influent and effluent from wastewater treatment plants, groundwater, surface waters, or seawater (Grandclément et al., 2017). The discovery of organic compounds has been confirmed by researchers in America (Gilliom, 2007), Africa (Edokpayi et al., 2017), and Asia (Lin et al., 2018). PAHs have been recognized as soil mixtures, water sources, and crustacean drainage and usually co-occur with other contaminants (Jaward et al., 2012; Ozaki et al., 2019). PAHs are mostly released into the atmosphere as by-products of the combustion of fuels, but agricultural explosions, industrial waste, and cooking can increase the discharge of these dangerous chemicals. The solubility of PAHs in water typically reduces as the molecular weight increases as their boiling and melting point increases (Adeniji et al., 2018). Adeniji et al. (2018) said that four-ring and five-ring PAHs, such as chrysene and benzo[a]pyrene, are practically insoluble in water. Because of this characteristic, particulate matter can be applied to the surface, and this mechanism defines the primary transport path of PAHs from land and air to marine habitats as well as from the water surface to lower depths (Vagge et al., 2018). Karyab et al. (2013) proposed that PAHs usually infiltrate water sources by dry and wet deposition, road runoff, leaching from creosote impregnated wood, industrial waste water, chemical spills, and fossil fuel combustion. Many researchers have thus verified PAHs in the contamination of marine ecosystems. In regions with a manifold of urban and industrial activities, sediments comprise

high levels of these pollutants (Abdollahi et al., 2013; Birch, 1996; de León-Gómez et al., 2020). Sediments contain the highest amount of PAH accumulation in aquatic ecosystems (Gagnon & Fisher, 1997; Yan et al., 2014). The major resources of organic and inorganic contaminants in the coasts of the Persian Gulf are numerous industrial operations such as shipping, fishing, sewage, and wastewater discharges, urban runoff, and landfill leachate (Abdollahi et al., 2013; Chen et al., 2013; Huang et al., 2012; Mostafa et al., 2009; Readman et al., 2002).

According to the above-mentioned explanations, massive industrial activities (e.g., oil industries, transportation, fishing, sewage and wastewater discharges, urban runoff, and landfill leachate) in the Persian Gulf coast can result in two major hypotheses:

- i) The coastal sediments of the Gulf have been polluted by PAHs and their main source is petrogenic.
- ii) There is a difference in the amount of PAHs in various seasons.

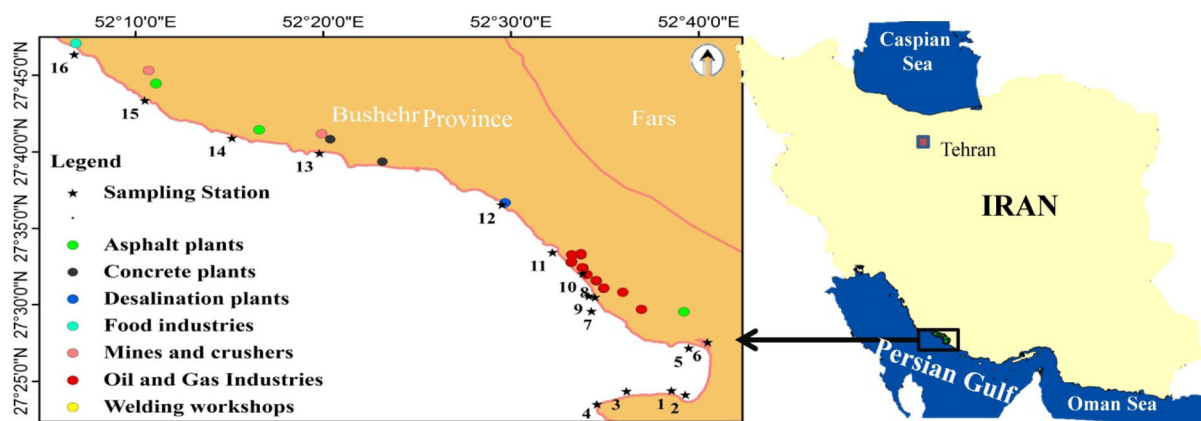
Therefore, the key aim of this study is the identification of the concentrations and distribution of sixteen main PAHs and sediments of PSEEZ in the Persian Gulf, as well as identifying their potential sources and effects on the bio-resources.

## Materials and methods

In the present study, PSEEZ was the study area (Fig. 1). This site is an industrial area containing petrochemical industries (Asaluyeh) and human activities such as fishing and oil transport. Consequently,

the effluent of untreated municipal wastewaters and hospital sewages are discharged into this site. In this study, point stations were selected, first, to cover all case study (PSEEZ) coasts, and second, to distinguish all probable local sources, industries, and human activities that cause high pressures with respect to the levels of PAHs in this region. Samples were collected within different seasons in 2013 from 16 sampling sites in the coastal area of the Persian Gulf at the PSEEZ (Fig. 1); three subsamples were taken at each site.

Samples of the surficial sediment were collected at the PSEEZ area using a stainless steel grab sampler in four seasons, from depths between 0.5 and 30 m (Fig. 1). Surface sediment samples were removed by spoons and carefully placed in an aluminum foil; they were then taken to the laboratory on ice and held at 20 °C until their study. In the laboratory, each sample was homogenized and freeze-dried (− 20 °C) for PAH analysis. Before the extraction, with 300 ng.g<sup>−1</sup> of deuterated surrogates, 20 g of homogenized sediment until extraction, the sample was spiked to analyze the methodological recovery (acenaphthene-d10, phenanthrene-d10, naphthalene-d8, chrysene-d12, and perylene-d12). Soxhlet extraction was conducted for the extraction of the sixteen essential and priority PAHs of the US-EPA: (Naphthalene = Naph), (Fluorene = Fl), (Acenaphthene = Ace), (Acenaphthylene = Acy), (Phenanthrene = Phen), (Fluoranthene = Fluo), (Anthracene = Anthr), (Aenzo[a]pyrene = BaP), (Benzo[a]anthracene = BaA), (Pyrene = Py), (Benzo[b]fluoranthene = BbF), (Chrysene = Chry), (Dibenz[a,h]anthracene = DahA), (Benzo[g,h,i] perylene =



**Fig. 1** Place and sampling site of the case study area in PSEEZ, Persian Gulf, Iran

BghiP), and (Indeno [1,2,3-cd] pyrene = IDP) (Benzo [a] anthracene = BaA). Five grams of sediment sample were weighed and placed in the extractor, and a 250 ml blend of dichloromethane and n-hexane (1:1, v/v) was obtained for 72 h by Soxhlet. Samples were condensed to a volume of 5 ml after extraction using a rotary evaporator apparatus. In the sample flasks for desulfurization (removal of elemental sulfur) for 36 h in the refrigerator, 2 g of activated copper was applied. Samples were then purified and condensed until the volume of the rotary evaporator method was reduced to 2 ml and the main volume was reduced to 2 ml. The solvent was substituted for hexane. With solid phase extraction (column of alumina/silica gel (300 mm $\times$  10 mm i.d)), the hexane extract was distilled and then reduced to 1 ml using a purified nitrogen stream for instrumental analysis. 100  $\mu$ l of terphenyld14 was put as an internal norm on each sample prior to the instrumental analysis (GC–MS). By the internal standard procedure, target analytes were quantified. Organic material content was detected through (LOI) according to the guidelines (Heiri et al., 2001). For data analyses, mean and standard deviation levels of PAHs were measured and the correlation analysis test was executed using SPSS 21.0 software package.

Sediments are an inherent aspect of the marine ecosystem and, due to their inadequate nitrogen loading, in nurturing floodplains and farm fields; they have been seen to be helpful. Nevertheless, the harmful impacts of the sediments are very alarming: (a) they include benthic marine population smothering (Hull et al., 2014; Mocq & Hare, 2018); (b) the reduction in the capability of shipping channels impedes navigation and thus has a negative impact on the country's economy; (c) they cause the capacity reduction of dams and reservoirs; (d) their presence leads to hydroelectric turbine obstruction (Yun et al., 2016); (e) they cause a reduction in water treatment plant and lagoon capability (Collivignarelli et al., 2018; de Magalhães et al., 2019; Fabião et al., 2016); and (f) the scenic and leisure importance of the rivers is declining. This shows that sediments pose a significant threat to experts and scientists interested in the sectors of water supply, wastewater, environmental cleaning, and dredging (de Magalhães et al., 2019). Sediments will serve as a drain or outlet for pollutants situated at the bottom or in the bed of the water body. This is an ongoing challenge for the

contemporary society and needs particular consideration from planners, scientists, engineers, and other players of society. This is a continuing challenge for mainstream society and needs relevant consideration from planners, scientists, engineers, and other social actors. However, only a few attempts by civil engineers have been made to characterize these sediments as a potential raw material for successful mining, safe disposal, and use for sustainable growth.

More specifically, in order to meet the aims of using sediments as a resource, they should be defined on the basis of their general characteristics as an intrinsic part of the body of water (i.e., physical, morphological, geological, geotechnical, bacteriological, and toxic characteristics) which can be changed by the presence of benthic fauna. It should be remembered here that their sampling may be very intricate and advanced for such an elaboration of the classification of sediments (Collivignarelli et al., 2018; de Magalhães et al., 2019).

Quality control and quality assurance data over the years

The rigorous process-oriented quality assurance (QA) model has been developed and applied by ONC in combination with a product-oriented data quality management (QC) model. This model regularly intercepts and analyzes the instruments and the data source at various points of the instrument operation to ensure long-term data quality control and assurance. Five separate phases have been established by the QAQC model data that include data quality management and assurance measures during the life cycle of an instrument (Cowger and Gray 2020; Loh et al., 2017; D'Amore et al., 2015; Porter et al., 2012). Pre-deployment tracking covers all QAQC data/metadata checks conducted up to final deployment at the pre-deployment testing stage of an instrument. Post-deployment commissioning requires all QAQC data/metadata checks, from actual deployment to commissioning of the data as effective or compromised. Automated quality checking covers all real-time or delayed QAQC-related data checks performed when the tool is implemented through automated quality control procedures.

Manual quality management methods contain all QAQC data checks conducted by detailed manual data assessments and routines for annotation. Post-



recovery monitoring includes all post-calibration checks undertaken during the post-recovery and restoration phases of the instrument. These phases describe a workflow of QAQC that minimizes individual and/or systemic errors during the life cycle of an instrument to ensure high-quality data. QC and QA monitor the efficiency of measurement devices that essentially lead to the scheduling of maintenance expeditions and instrument platform calibrations. The research and development of enhanced and emerging control systems were complementary to these two processes.

#### *Quality assurance data*

Data QA procedures are proactive steps to reduce data source problems and inaccuracies while restricting corrective measures to increase the accuracy of data. The QA data portion requires procedures to ensure that the instrument sensor network's protocols are correctly specified and adhered to. Periodic manual data processing by ONC data professionals, incorporation of annotations for data validation, and end-to-end workflow execution activities are examples of QA procedures currently in operation (Abatamarco et al., 2018; Abeysirigunawardena et al., 2015; Integrated, 2015).

#### *Quality control data*

QC data quality management is a product-oriented method for the identification and flagging of questionable information after it has been produced. QC requires both automatic and manual procedures to verify whether the information satisfies the consistency specifications outlined by the end users.

Three components compose the QC of information. The first part dynamically tests real-time data before parsing the data into the database. Using delayed-mode automatic testing, the second factor tests near-real-time or archived results. Manual inspection is the third part, where a specialist inspects the consistency data for problems (Abeysirigunawardena et al., 2015).

The method detection limit (MDL) was measured by the analysis of five sediment samples through the injection of low concentrations (lower than the concentration level on the calibration curve resulting in a detectable peak) of the target analytes until a small peak at  $\sim 3:1$  (signal: noise ratio) was achieved. The

MDL was calculated by the standard deviation of the five replicates and the value varied between 0.02 and  $0.14 \text{ ng g}^{-1}$  (mean  $0.06 \text{ ng g}^{-1}$ ). Concentrations lower than MDLs have been defined as non-detected (N.D.). Target analytes were also quantified using the internal standard method. The practical quantification limits (PQL) for all the sixteen PAH compounds were lower than  $0.15 \text{ ng g}^{-1}$ .

## **Results**

The values, mean  $\pm$  SD, and range of sixteen essential and priority PAHs from the US-EPA compounds in sediment samples are represented in Tables 1, 2, 3, and 4 ( $\text{ng g}^{-1}$ -dry weight). As illustrated in Table 1, the maximum  $\sum$ PAH content of  $297.1 \text{ ng g}^{-1}$ -dry weight was found at station 9 in the spring season. Based on the results of Table 2, the maximum  $\sum$ PAH content of  $328.28 \text{ ng g}^{-1}$ -dry weight was found at station 10 in the summer season. As shown in Table 3, the maximum  $\sum$ PAH content of  $343.63 \text{ ng g}^{-1}$ -dry weight was found at station 10 in the autumn season. As shown in Table 4, the maximum  $\sum$ PAH content of  $348.42 \text{ ng g}^{-1}$ -dry weight was found at station 10 in the spring season. Therefore, the maximum  $\sum$ PAH content was found at station 9 in the spring season. This station is located near petrochemical industries and refinery areas (near the phases of the South Pars petrochemical plant), with heavy traffic of ships, loaded trains, and urban and industrial wastewater discharge. The maximum levels of total PAH concentrations were measured at station 10 in summer, autumn, and winter, respectively. This subject can be attributed to their location near industrial activities, active oil locations, petrochemical activities, and refined oil products, which lead to high transportation of vehicles such as ships, boats, and petroleum transfer buoys and crossing of oil tankers and human activities.

The average of  $\sum$ PAH content in each season is estimated to be  $244.59 \text{ ng g}^{-1}$  for spring ( $145.7\text{--}289.8 \text{ ng g}^{-1}$ ),  $260.11 \text{ ng g}^{-1}$  for summer ( $179.22\text{--}328.28 \text{ ng g}^{-1}$ ),  $276.10 \text{ ng g}^{-1}$  for autumn ( $199.62\text{--}343.63 \text{ ng g}^{-1}$ ), and  $281.3$  for winter ( $205.85\text{--}348.43 \text{ ng g}^{-1}$ ) (Tables 1, 2, 3, and 4).

Based on ANOVA test (Fig. 2), total concentrations in winter were  $246 \text{ ng g}^{-1}$ . In autumn and summer, the overall concentrations were between 246 and  $296 \text{ ng g}^{-1}$ , and total concentrations in spring

**Table 1** Concentrations of 16 US-EPA important and priority PAHs in sediment samples (ng g<sup>-1</sup>-dw) from the PSEEZ of Persian Gulf in spring

Station	NAP	ACL	AC	FLA	ANT	PHE	Flu	PY	BaA	CHR	BbFA	BkFA	BaP	DBahA	IP	BghiP	ΣPAHs	LWM/HWM
1	8.89	5.01	3.14	4.15	3.45	40.32	19.82	18.79	19.45	8.71	12.51	11.34	27.34	2.54	5.32	4.13	194.91	0.833
2	7.81	4.91	3.16	3.73	4.98	37.56	21.34	20.65	18.19	9.45	13.24	11.87	28.12	2.91	6.18	4.18	198.28	0.761
3	7.51	6.05	2.15	4.92	4.18	38.19	18.76	17.01	19.23	7.66	13.88	10.24	28.79	3.82	5.47	4.57	192.43	0.811
4	5.8	4.9	1.14	4.53	1.81	22.89	17.23	15.39	16.54	6.52	8.5	8.74	20.2	2.65	4.66	4.2	145.7	0.654
5	9.15	7.11	2.17	5.12	3.42	37.85	17.98	16.49	17.91	7.48	10.35	9.69	25.64	4.51	5.46	3.98	184.31	0.904
6	9.82	6.14	2.16	4.98	3.16	37.25	20.71	20.34	19.31	7.99	11.27	10.46	26.57	4.57	5.19	4.12	194.04	0.811
7	10.23	8.56	2.89	5.25	4.16	38.79	21.63	20.27	19.87	8.34	12.49	11.35	28.96	5.26	6.92	4.87	209.84	0.832
8	12.62	9.11	3.67	5.96	4.57	89.68	19.8	24.42	22.65	8.49	13.06	14.76	29.18	5.94	7.15	3.56	274.62	1.405
9	13.96	9.89	6.57	6.62	4.06	99.51	19.5	25.85	23.38	8.85	14.96	15.99	29.71	6.84	7.85	3.56	297.1	1.498
10	13.46	8.76	5.12	6.29	5.03	94.57	20.86	24.92	23.17	8.86	14.95	14.63	29.81	6.93	8.32	4.12	289.8	1.418
11	13.32	8.98	4.63	6.18	4.26	89.21	20.65	24.89	22.34	8.28	15.11	14.58	28.86	6.71	8.77	4.38	281.15	1.365
12	10.98	7.15	3.29	6.03	4.49	62.11	21.57	20.15	22.07	7.93	14.99	14.47	28.92	6.72	8.96	4.93	244.8	1.04
13	9.43	6.24	2.96	5.11	5.1	54.36	22.3	20.12	21.36	7.59	14.55	13.68	27.89	5.89	8.43	5.21	230.22	0.943
14	8.12	6.32	2.64	4.29	4.65	53.23	23	19.41	20.97	8.04	14.2	13.16	27.65	5.45	7.07	4.92	223.12	0.918
15	8.42	7.2	1.46	4.47	5.28	49.87	23.5	17.18	20.09	7.97	13.6	12.76	27.43	5.11	7.89	5.08	217.31	0.909
16	7.68	6.16	1.62	3.45	6.98	51.36	21.4	17.65	21.12	7.49	13.6	11.31	27.86	5.34	8.06	4.76	215.84	0.929
Mean	9.83	7.03	3.05	5.07	4.35	56.05	20.63	20.22	20.48	8.1	13.2	12.44	27.68	5.07	6.98	4.41	224.59	1.002
SD	2.43	1.6	1.42	0.95	1.13	24.09	1.72	3.26	1.99	0.7	1.85	2.12	2.29	1.48	1.42	0.51	40.138	0.249
TEL	34.57	5.87	6.71	21.2	469	86.7	112.82	153	74.8	108	–	–	88.8	6.22	–	–	–	–
PEL	391	128	88.9	144	254	544	1494	1398	693	846	–	–	763	137	–	–	–	–
ERL	16	44	16	19	85.3	240	600	665	261	384	320	280	430	63.4	–	430	3912.2	–
ERM	2100	640	500	540	1100	1500	5100	2500	1600	2800	1880	1620	1600	260	–	1600	25,430	–

**Table 2** Concentrations of 16 US-EPA important and priority in sediment samples, PAHs (ng g<sup>-1</sup>-dw) from the PSEEZ of Persian Gulf in summer

Station	NAP	ACL	ACE	FLA	ANT	PHE	Flu	PY	BaA	CHR	BbFA	BkFA	BaP	DBahA	IP	BghiP	ΣPAHs	LWM/HWM
1	9.8	6.72	3.8	5.06	6.9	52	21.19	19.28	21	10.06	14.4	14.4	28.43	3.43	5.6	4.27	226.34	0.989
2	9.21	7.01	3.8	5.39	7.2	50.1	24	20.87	20.7	10.52	15.1	14	28.93	3.97	6.58	4.27	231.65	0.926
3	8.56	8	3.25	5.86	6.25	47.2	23.22	18.7	21	10.15	15.9	13.6	30.8	4.57	5.67	4.84	227.57	0.888
4	7.9	6.72	1.92	5.71	4.3	29.3	19.34	15.98	19.3	9.05	10.4	11.1	24.7	3.6	5.46	4.44	179.22	0.755
5	10.2	8.64	3.25	6.16	5	49.1	20.29	18.2	20.5	9.69	12.2	12.2	28	5.05	6.27	4.27	219.02	1.004
6	10.6	8.93	3.84	6.87	5.55	42.5	24.38	21.5	21.6	9.6	12.5	13.4	28.6	5.35	6	4.4	225.62	0.886
7	12.5	9.7	3.84	7.12	5.94	49.1	24.87	21.5	23	9.78	13.9	15.1	29.7	5.95	7.6	5.29	244.89	0.938
8	14.3	9.89	5.7	7.29	5.6	96	21.4	26.3	25.6	10	15	16.6	31	6.68	8.06	3.91	303.33	1.406
9	15.5	10.8	7.36	7.96	5.8	105.8	21.6	27.5	25.8	10	16.6	17.1	30.8	7.46	8.15	3.94	322.17	1.511
10	14.4	10.8	6.66	7.84	5.8	110.6	23.3	27	27	10	16.3	17.1	30.73	7.6	8.69	4.46	328.28	1.511
11	14.8	10.66	5.5	7.32	5	103	22.6	25.6	25.8	9.78	17.3	16.6	31.6	7.28	9.13	4.56	316.53	1.432
12	13.2	10.08	5.38	7.41	5.86	72.8	23.3	22.6	26.3	10.71	18.2	17.6	34.2	7.52	10.3	5.64	291.1	1.084
13	11.3	9.7	4.74	7.42	7.31	68	25.29	22.6	25.3	10.4	19	17	32.3	7.16	9.76	5.44	282.72	1.037
14	9.8	8.26	4	7.4	7.5	64	26.19	20.76	24.9	10.06	17.3	16.5	30	6.8	8	5.29	266.76	1.015
15	10.3	8.4	3.01	7.35	7.78	56	25.3	18.46	23.5	9.32	16	15	28	5.95	8.12	5.44	247.93	0.998
16	8.98	8.16	2.91	7.38	8.4	58.6	24.29	18.5	24	8.95	15.3	14.5	29	5.89	8.75	5.04	248.65	1.021
Mean	11.33	8.9	4.31	6.85	6.26	65.88	23.16	21.58	23.46	9.88	15.34	15.11	29.8	5.89	7.63	4.72	260.1	1.087
SD	2.44	1.39	1.46	0.9	1.14	24.93	1.95	3.48	2.47	0.49	2.28	1.93	2.17	1.43	1.54	0.57	43.21	0.231
TEL	34.57	5.87	6.71	21.2	469	86.7	112.82	153	74.8	108	—	—	88.8	6.22	—	—	—	—
PEL	391	128	88.9	144	254	544	1494	1398	693	846	—	—	763	137	—	—	—	—
ERL	16	44	16	19	85.3	240	600	665	261	384	320	280	430	63.4	—	430	3912.2	—
ERM	2100	640	500	540	1100	1500	5100	2500	1600	2800	1880	1620	1600	260	—	1600	25,430	—



**Table 3** Concentrations of 16 US-EPA important and priority in sediment samples, PAHs (ng g<sup>-1</sup>-dw) from the PSEEZ of Persian Gulf in autumn

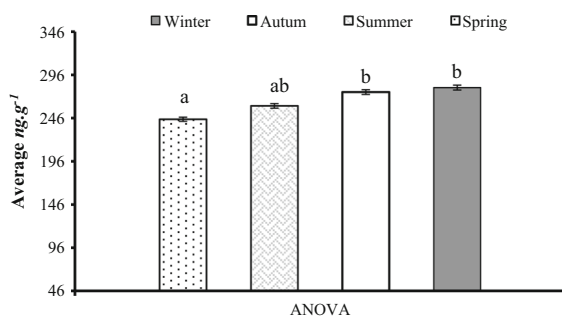
Station	NAP	ACL	AC	FLA	ANT	PHE	Flu	PY	BaA	CHR	BbFA	BkFA	BaP	DBahA	IP	BghiP	ΣPAHs	LWM/HWM
1	9.5	6.95	4.1	5.49	8.11	53.22	22.51	20.71	22.46	11.52	15.82	16.42	29.22	4.12	6.18	4.86	241.19	0.947
2	9.45	6.82	4.22	5.42	8.41	51.31	25.31	22.3	22.19	11.98	16.53	15.7	29.54	4.56	7.24	4.72	245.7	0.892
3	9.32	7.46	4.64	5.84	7.46	48.4	24.53	20.13	22.47	11.61	17.34	15.32	31.39	5.32	6.26	5.43	242.92	0.867
4	9.85	8.51	4.95	5.62	5.51	30.52	20.65	17.41	20.75	10.51	11.85	12.74	25.25	4.21	6.05	5.24	199.62	0.804
5	10.34	8.92	5.24	6.23	6.21	50.33	21.6	19.63	21.92	11.15	13.68	13.96	28.58	5.64	6.86	4.86	235.15	0.984
6	10.62	9.49	5.62	6.58	6.76	43.72	25.69	22.93	23.09	11.06	13.99	15.13	29.29	5.94	6.59	4.93	241.43	0.87
7	11.98	9.97	6.16	6.95	7.15	50.34	26.18	22.93	24.43	11.24	15.37	16.82	30.25	6.54	8.21	5.88	260.4	0.919
8	13.45	10.15	6.79	7.91	6.81	97.25	22.71	27.73	27.07	11.46	16.49	18.43	31.53	7.27	8.65	4.5	318.2	1.349
9	15.1	10.48	7.13	8.24	7.01	107.01	22.91	28.93	27.26	12.48	18.14	18.87	31.37	8.05	8.74	4.53	336.25	1.425
10	14.51	10.56	7.27	8.16	7.01	111.82	24.61	28.43	28.42	12.42	17.72	18.85	31.33	8.19	9.28	5.05	343.63	1.441
11	14.2	10.06	7.31	8.21	6.21	104.23	23.91	27.03	27.27	12.24	18.73	18.35	32.19	7.87	9.73	5.15	332.69	1.372
12	13.21	9.13	6.87	8.2	7.07	74.03	24.61	24.03	27.71	12.17	19.61	19.37	34.75	8.11	10.87	6.25	305.9	1.054
13	12.4	8.54	6.27	8.06	8.52	69.24	26.6	24.03	26.74	11.83	20.4	18.75	32.84	7.75	10.35	6.03	298.35	1.017
14	12.13	8.38	5.46	8.02	8.71	65.22	27.5	22.19	26.33	11.52	18.78	18.23	30.55	7.39	8.55	5.88	284.84	1.017
15	11.87	8.2	5.1	7.92	8.99	57.23	26.61	19.89	24.96	10.78	17.46	16.43	28.59	6.54	8.71	6.31	265.59	0.995
16	11.14	8.17	4.43	7.94	9.61	59.82	25.6	19.93	25.21	10.41	16.79	16.38	29.37	6.28	9.13	5.51	265.72	1.024
Mean	11.82	8.86	5.72	7.17	7.47	67.11	24.47	23.01	24.89	11.52	16.79	16.86	30.38	6.49	8.21	5.32	276.1	1.06
SD	1.9	1.2	1.12	1.12	1.14	24.93	1.95	3.48	2.46	0.64	2.27	1.95	2.16	1.41	1.52	0.6	39.88	0.19
TEL	34.57	5.87	6.71	21.2	469	86.7	112.82	153	74.8	108	–	–	88.8	6.22	–	–	–	–
PEL	391	128	88.9	144	254	544	1494	1398	693	846	–	–	763	137	–	–	–	–
ERL	16	44	16	19	85.3	240	600	665	261	384	320	280	430	63.4	–	430	3912.2	–
ERM	2100	640	500	540	1100	1500	5100	2500	1600	2800	1880	1620	1600	260	–	1600	25,430	–

**Table 4** Concentrations of 16 US-EPA important and priority in sediment samples, PAHs (ng g<sup>-1</sup>-dw) from the PSEEZ of Persian Gulf in winter

Station	NAP	ACL	AC	FLA	ANT	PHE	Flu	PY	BaA	CHR	BbFA	BkFA	BaP	DBahA	IP	BghiP	ΣPAHs	LWM/HWM
1	5.45	6.77	4.71	29.81	9.62	9.62	7.12	4.2	6.7	54.43	8.41	22.62	20.81	22.54	11.55	16.73	241.09	0.628
2	5.31	7.83	5.15	30.13	9.3	16.2	7.34	4.3	6.63	52.52	8.51	25.43	22.4	22.27	12.01	17.44	252.18	0.689
3	6.02	6.85	6.85	31.98	9.45	15.8	7.52	4.8	7.05	49.61	7.56	24.65	20.23	22.59	11.64	18.25	250.85	0.737
4	5.83	6.64	4.8	25.84	9.46	13.3	7.95	4.8	6.83	31.73	5.61	20.77	17.51	20.85	10.54	12.76	205.22	0.788
5	5.45	7.45	6.23	29.17	10.58	14.44	8.46	5.45	7.44	51.54	6.31	21.72	19.73	22.03	11.18	14.59	241.77	0.725
6	5.52	7.18	6.53	29.88	11.12	15.67	8.92	5.62	7.79	44.93	6.86	25.81	23.3	23.17	11.09	14.6	247.99	0.735
7	6.47	8.8	7.13	30.84	11.94	17.53	9.37	6.47	8.16	51.55	7.25	26.3	23.3	24.58	11.27	16.28	267.24	0.747
8	5.28	9.24	7.86	32.12	12.38	18.8	9.93	7.12	9.12	98.46	6.91	22.83	27.83	27.16	11.49	17.4	323.93	0.599
9	5.19	9.33	8.64	31.96	12.97	19.33	10.43	7.64	9.45	108.22	7.11	23.03	29.03	27.35	11.49	19.05	340.22	0.576
10	5.64	9.87	8.78	32.92	13.5	19.32	10.34	7.61	9.37	113.03	7.11	24.73	28.53	28.55	11.49	18.63	348.42	0.572
11	5.74	10.32	8.46	32.78	13.11	18.79	10.32	7.59	9.42	105.44	6.31	24.03	27.13	27.38	11.27	19.64	337.73	0.598
12	6.86	11.46	8.7	34.34	12.41	19.8	10.33	7.15	9.41	75.24	7.17	24.13	24.73	27.13	12.2	20.52	312.6	0.723
13	6.86	10.94	8.34	33.43	12.24	19.2	10.08	6.19	9.23	66.43	8.81	17.6	24.13	26.89	11.89	21.31	293.57	0.749
14	6.47	9.14	7.98	31.14	11.71	18.75	8.54	6.19	9.23	66.43	8.81	27.62	22.2	26.45	11.55	19.69	291.9	0.687
15	6.76	9.3	7.13	29.18	11.24	17.2	8.14	6.05	9.13	58.44	9.09	26.73	19.99	25.18	10.81	18.37	272.74	0.702
16	5.82	9.52	6.67	29.76	11.09	16.74	7.98	6.12	9.15	61.03	9.71	25.72	20.03	25.46	10.44	17.7	272.94	0.686
Mean	5.92	8.79	7.12	30.96	11.38	16.91	8.92	6.08	8.38	68.06	7.58	23.98	23.18	24.97	11.37	17.69	–	–
SD	0.58	1.52	1.38	2.16	1.39	2.73	1.19	1.16	1.12	24.93	1.15	2.53	3.5	2.42	0.49	2.29	–	–
TEL	34.57	5.87	6.71	21.2	469	86.7	112.82	153	74.8	108	–	–	88.8	6.22	–	–	–	–
PEL	391	128	88.9	144	254	544	1494	1398	693	846	–	–	763	137	–	–	–	–
ERL	16	44	16	19	85.3	240	600	665	261	384	320	280	430	63.4	–	430	3912.2	–
ERM	2100	640	500	540	1100	1500	5100	2500	1600	2800	1880	1620	1600	260	–	1600	25,430	–

were 296 ng g<sup>-1</sup>. Therefore, the total concentrations in winter were higher compared with other seasons. Significant differences in  $\sum$ PAH content were observed between winter, summer, and spring (per ANOVA tests) (Fig. 2). In winter, the samples presented the highest PAH concentrations (Fig. 2). As a result, the risk of PAHs on the coast of South Pars in winter is higher than in other seasons. These studies suggest that the winter concentration of contaminants would be higher due to increased combustion due to fuels as a result of domestic and industrial activities.

The results of Pearson correlation coefficient between total of sediments PAH concentrations in spring was (sig = 1); therefore, there is no significant relationship between sediment concentrations in spring. As given in Table 5, Pearson correlation coefficient between total of sediments PAH concentrations in summer was (sig = 0.987\*\*); thus, there is a significant relationship between sediment concentrations in spring at the level of 0.001. Furthermore, the Pearson correlation coefficient in autumn was (sig = 0.986\*\*), and in winter, it was (sig = 0.987\*\*). Thus, there is a significant relationship between sediment concentrations in autumn and winter at the level of 0.001. Investigating every season exclusively, no significant correlation was found in all seasons. On the other hand, the  $\sum$ PAH values were not correlated with sediment's total organic carbon concentration across all seasons ( $r = 0.230$ ;  $P < 0.05$ ). However, analyzing the correlations among total PAH concentrations in different seasons showed positive results in each season ( $P < 0.05$ ) (Table 5).



**Fig. 2** Comparison of cumulative sediment PAH concentrations in various seasons using ANOVA studies

**Table 5** Pearson correlation coefficient between PAH concentrations of total sediments in various seasons

	Spring	Summer	Autumn	Winter
Spring	1			
Summer	0.987**	1		
Autumn	0.986**	0.999**	1	
Winter	0.987**	0.996**	0.997**	1

\*\* Correlation is significant at level 0.01 (two-tailed)

## PAH composition and source identification

The isomeric ratios Ant/(Ant + Phe), BaA/(BaA + Chry), IP/(IP + BghiP), and Flua/(Flua + Pyr) were calculated as diagnostic indicators to identify the potential PAH origin. For instance, the common ratio FLA/(FLA + PYR) < 0.4 typically (indicates predominantly petrogenic sources) defines petroleum contamination; a ratio between 0.40 and 0.50 (indicates predominantly petrogenic sources) shows petroleum fuel combustion; a ratio > 0.50 indicates coal or biomass combustion (combustion origin of biomass) (e.g., coal, wood, grass); a BaA/(BaA + Chr) ratio < 0.20 is commonly taken as an indication of a predominant petroleum origin; a ratio between 0.20 and 0.35 indicates either combustion or petroleum; and  $N > 0.35$  shows only combustion (Katsoyiannis & Breivik, 2014; Tobiszewski & Namieśnik, 2012). The ratio An/(An + Phe) < 0.10 demonstrates the predominance of the petroleum source, and  $N > 0.10$  shows the dominance of combustion (Baumard et al. 1999).

Based on results, a precise reconstruction of the biogeochemical characteristics of pollutants at various periods and spatial scales is needed to understand the real influence of polluted sediments on the aquatic environment and their possible effects on the ecosystem. Moreover, a stagnant understanding of the effect of contaminated sediments on the coastal marine environment is questioned by normal and personal interactions between land-to-coast and coast-to-deep-sea systems. In addition, approaches to monitoring aquatic sediments that have historically been limited to unique and restricted areas (where pollutants simply tend to be more abundant in surface-to-surface sediments) hinder an accurate understanding of their effects on the marine ecosystem. Indeed for the

aquatic ecology and the atmosphere, polluted sediments must be treated as dynamic time- and space-modulated active sources of contaminants. For holistic and appropriate management of contaminated aquatic sediments, a current view must take into account the chemical and physical behavior and the complex dynamics of pollutants in various ecosystems. The authors therefore suggest and propose that a larger buffer zone be established as an external and systemic aspect of the surveillance action committed to the heavily contaminated marine coastal regions.

## Discussion

The concentrations of the 16 PAHs in samples of surface sediments from various stations in different seasons are represented in Tables 1, 2, 3, and 4. The  $\Sigma$ PAH concentration ranged from 145.7 to 348.42 ng g<sup>-1</sup>dw, and the mean concentration is 260.52 ng g<sup>-1</sup>. The maximum levels of PAH content of 348.42 ng g<sup>-1</sup> — dw were found at station 10. This station is located near the first and second phases of the petrochemical industrial zone, which had the highest industrial activity and was the major discharging area for the petrochemical industrial influent stream to the coastal region. In addition, this zone was affected by the effluent discharge from ballast water due to ship traffic, petroleum transfer, etc. Specifically, this station demonstrated the highest range of  $\Sigma$ PAHs in all seasons. In addition, this zone was affected by the effluent discharge from ballast water due to ship traffic, petroleum transfer, etc. Specifically, this station demonstrated the highest range of  $\Sigma$ PAHs in all seasons. CHR was detected to have the highest concentration (113.03 ng g<sup>-1</sup>) in the winter season, followed by Phe, FL, and Bap, with concentrations of 111.82, 34.75, and 34.34 ng g<sup>-1</sup>, in autumn and winter, respectively. In addition, sediment properties play a key role in the distribution of PAHs. Grain size is modified for different seasons and this is crucial for PAHs fixation. Under all conditions, i.e., seasons, time, temperature, and pH, the distribution of the slopes of the combined leaching curves is larger for some metals than for others. In addition to the environmental driving factors, it can be postulated that the release activities of any metal are contingent on the metal/sediment mix. The aggregates are then dissolved and the framework in an extremely acidic

environment is broken. Free and soluble metal compounds and the difference in concentration between the sample matrix and the leachate induce the gradual degradation of metals in the washing liquids in the early phases under acidic and alkaline conditions. Diffusion (0.35 < slope < 0.65) and depletion (slope < 0.35) are the prevailing processes of the metals over the entire leaching cycle under acidic and alkaline conditions.

This outcome is in line with the results of Vijayan et al. (2019). Their findings showed that laboratory snowmelt experiments with actual snow samples in late winter, just before the final snowmelt, in two similar cities in northern Sweden, were related to CHR with the highest concentrations. The  $\Sigma$ PAH concentrations in sediments can be grouped into four categories:  $\Sigma$ PAHs < 100 ng g<sup>-1</sup> indicated low contamination, 100–1000 ng g<sup>-1</sup> showed moderate pollution, 1000–5000 ng g<sup>-1</sup> represented heavy pollution, and  $\Sigma$ PAHs > 5000 ng g<sup>-1</sup> referred to very heavy pollution (Chen et al., 2013; Neşer et al., 2012). Therefore, according to the amount of  $\Sigma$ PAH concentration, the sediments in the PSEEZ area indicated moderate to heavy pollution. Tables 2 and 5 show that all stations were moderately polluted. Therefore, the sedimentary surface ecosystems of the Persian Gulf were considered as moderately polluted, compared with other ecosystems worldwide (Table 6). The higher concentrations at stations 8, 9, 10, and 11 can be explained by different input sources in winter, such as discharge of wastewater and runoffs, petrochemical industrial effluent, and oil spills from different vehicles like boats and ships. This is because these sites are located in a more industrialized and urbanized region and near the Asaluyeh phases. In addition, there were lots of ships and boats traveling in this area, which might contribute to the higher amount of PAH concentrations. In fact, the PAHs might be caused by the local sources of petroleum hydrocarbons, such as ferries and fishing boats. These results are consistent with the findings of Gune et al. (2019), Sarti et al. (2017), and Liu et al., (2012). Urban wastewater and runoff also tend to have different energies during the seasons, resulting in significant changes in grain size, organic matter, and mineralogy. Therefore, these factors are also very important in creating seasonal changes and assessing the aromatic polycyclic hydrocarbons in the aquatic resource center's coastal sediments. In order to forecast PAH

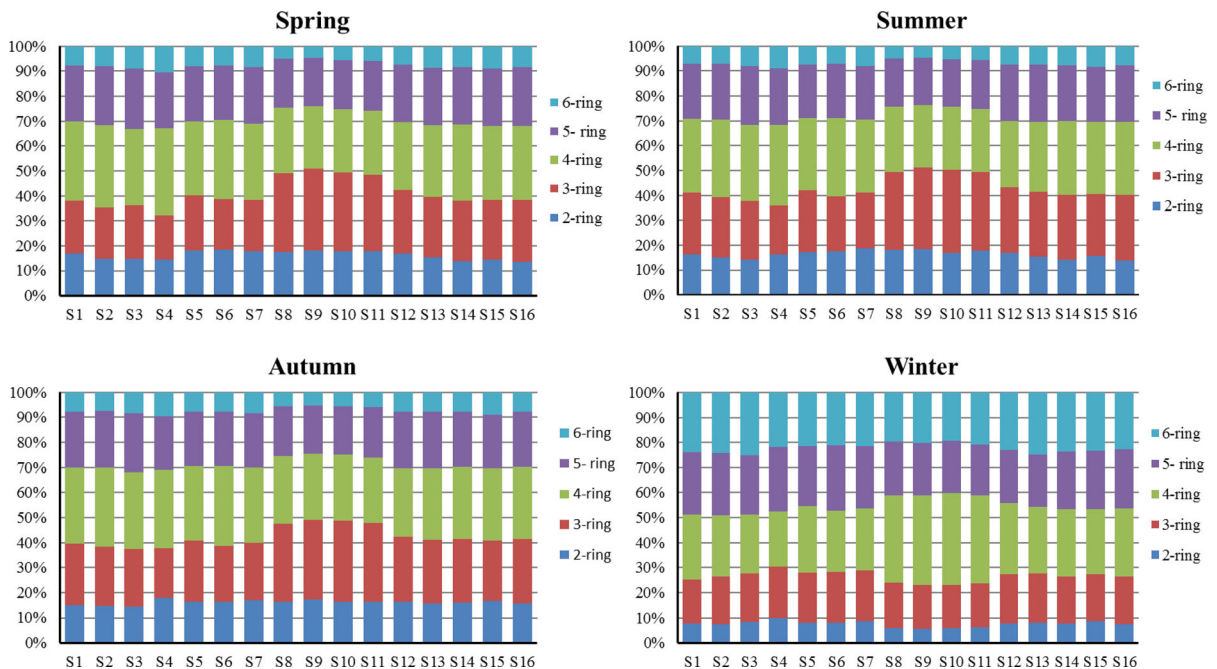
**Table 6** Comparison of the sum of 16 PAH concentrations in sediment samples with other aquatic ecosystems worldwide

Case study	Number of PAHs studied	PAH concentrations (ng g <sup>-1</sup> )	Pollution level	References
PSEEZ, Iran	16	145.7–348.43	Moderate	This study
Cochin estuary, India	16	194–14,149	Moderate–heavy pollution	(1)
Intertidal zone of Bohai Bay, China	16	37.2–206.6	Low–moderate	(2)
Mediterranean coastal zone, Spain	14	5.3–2627.4	Low–heavy	(3)
Liaodong Bay, China	16	144.5–291.7	Moderate	(4)
Western Taiwan coastal plain	16	2.41–218.54	Low–moderate	(5)

emissions, knowledge of the PAHs partitioning activities and their differences are critically significant in gaseous and particulate phases and in size-segregated phases. Previous studies have shown that the partitioning of PAHs depends primarily on their volatility between the gaseous and particulate phases and that the particulate partitioning of PAHs depends to a large degree on the scale, the unburned carbon content, and the species of metal particulates.

The average concentration of PAH for spring (145.7–289.8 ng g<sup>-1</sup>) was calculated to be 244.59 ng g<sup>-1</sup>; for summer, 260.11 ng g<sup>-1</sup> (179.22–328.28 ng g<sup>-1</sup>); for autumn, 276.10 ng g<sup>-1</sup> (199.62–343.63 ng g<sup>-1</sup>); and for winter, 281.3 (205.85–348.43 ng g<sup>-1</sup>) (Figs. 1, 2, 3, and 4). Total concentrations in winter were higher compared with other seasons, followed by fall, and the lowest levels of  $\sum$ 16 PAHs occurred in spring (Tables 1, 2, 3, and 4). The highest  $\sum$ PAH concentration was observed in winter (348.42 ng g<sup>-1</sup>) because of the increase in fuel usage and the high rate of precipitation in winter. Furthermore, enhanced photodegradation in summer is proposed as a probable explanation for higher PAHs in winter (Dimashki et al., 2001; Kim and Chae, 2016). It could be explained as a result of the intensified use of oil products and could carry the petroleum and its derivatives by urban runoff and rivers, industrial and municipal effluent discharge, and atmospheric deposition in winter (Guo et al., 2007; Keshavarzifard et al., 2014). Kim and Chae (2016) indicated that PAHs have a negative temperature dependency and that temperature and atmospheric PAHs have a negative relationship. In addition, at a warm period of time, photolysis, volatilization, and the lower PAH number can be

explained by algal blooming, along with sorption into suspended particle matter. Sorption of gases from air to water, accompanied by elevated levels of PAHs in the air, may be another cause for increasing levels of PAH in seawater during cold times. In other beach habitats such as the Khambhat Gulf, India (Reddy et al., 2005) and European coastal mussel waters, higher concentrations of PAH were also found in winter relative to warm periods of time (Olenycz et al., 2015). The maximum PAHs were found in the monsoon by Ramzi et al. (2017), Szopińska et al. (2019), and Zhang, Wang, et al. (2020), Zhang, Pan, et al. (2020)) (Ramzi et al., 2017). For all seasons, the highest amounts of  $\sum$ PAH concentrations were, respectively, observed at stations 10, 9, and 11. These stations were the major discharging zones for the oil and petroleum industry wastewaters outflowing to coastal areas. Among all tested PAHs in sediments, PHE and BaP were the predominant species of all stations. BaP is considered as an indicator for combustion-originated PAHs and intensely supports the predominantly incomplete combustion derived (pyrogenic) from PAH in sediments (Magi et al., 2002; Neira et al., 2017). The results of the parametric Pearson correlation analysis showed no significant relationship between TOC and PAHs ( $P > 0.05$ ,  $n = 16$ ), and this was in compliance with the literature where the TOC content of sediments had no correlation with PAH concentration as a main controlling factor (Barhoumi et al., 2014; Chen et al., 2013; Sany et al., 2014).



**Fig. 3** Distribution of 2–6-ring  $\Sigma$ 16PAH compounds in sediments from the PSEEZ (Persian Gulf)

### Seasonal deviations in PAH composition

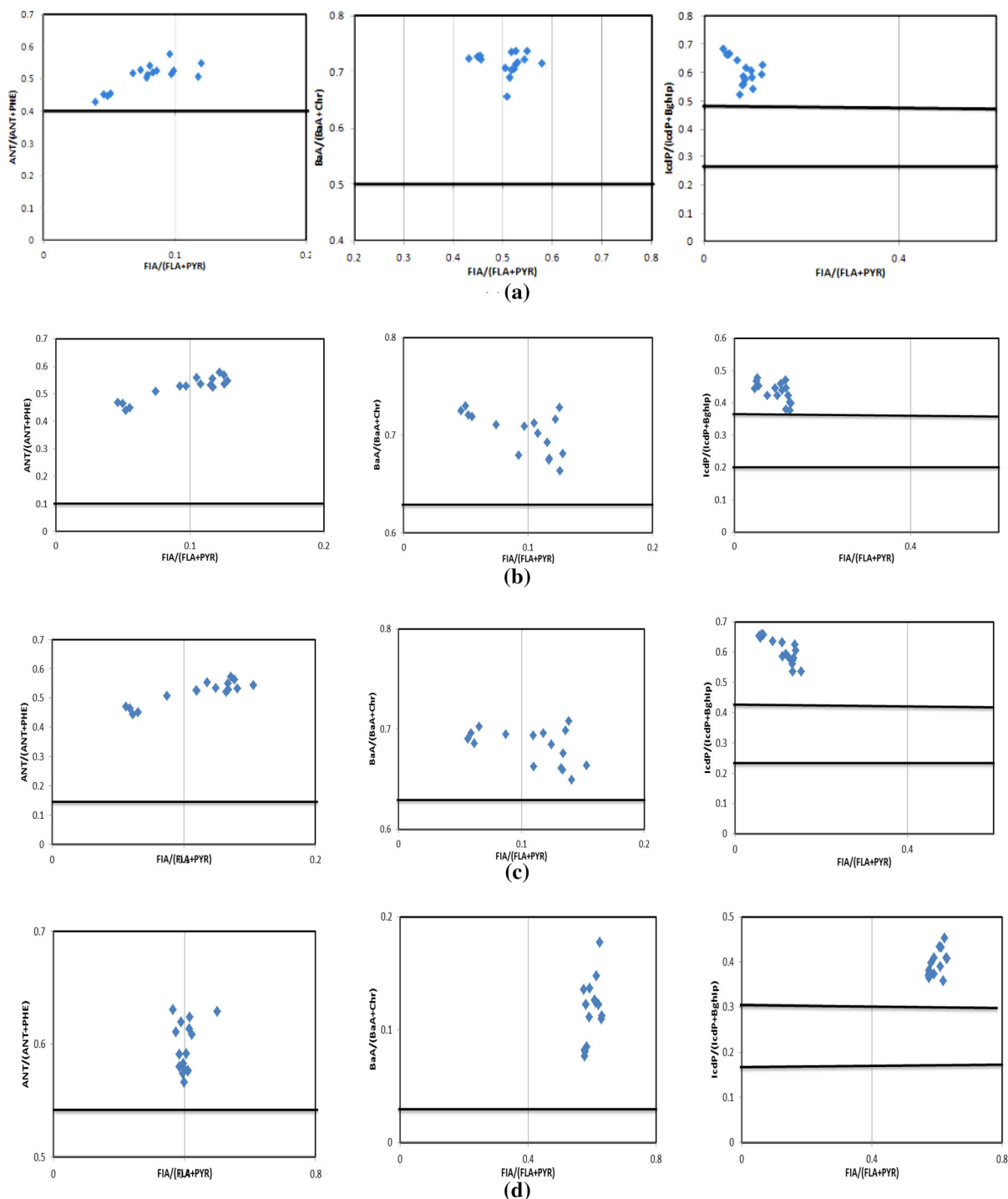
In spring, the four-ring PAHs were the most prevailing compounds, with an average value. The 2- and 3-ring PAHs were the second dominant compounds (Fig. 2). The 4-ring PAHs were the most prevailing compounds in all seasons (Fig. 2). The principal compound in the winter was inhabited by medium molecular weight (MMW, 4-ring) PAHs. The compounds of the 5-ring PAHs became the second dominant PAHs. The key compound was occupied in autumn by the 4-ring PAHs.

The four-ring PAHs dominated the primary compound of PAHs throughout the autumn. The second prevalent PAH compounds were the 3-ring PAHs. In conclusion, the four-ring PAHs in both seasons were the most prevalent PAH compounds. These results are consistent with the findings of Khaksar et al. (2019), and Škrbić et al. (2019).

### PAH source diagnosis

The PAHs of the 2–3 (low molecular weight)- and 4–6 (high molecular weight)-ring diagnostic ratio fractions can be applied as a marker to determine the PAH

origins; LMW/HMW ratio  $> 1$  is usually considered as a petrogenic origin, and a ratio  $< 1$  represents a pyrolytic origination (Wang et al., 2011). Petrogenic mechanism is responsible for PAH pollution distinguished by 2–3-ring PAHs, while pyrogenic mechanism is responsible for PAH contaminations distinguished by 4–6-ring PAHs (Baumard et al., 1998; Ramzi et al., 2017). The number of PAHs ratio in the present analysis (2–3 rings/4–6 rings) in different seasons in stations 8, 9, 10, 11, and 13 was higher than one, indicating the petrogenic origin of pollution. On the other hand, in stations 2 and 3, 4–6-ring PAHs (LMW/HMW) ratios were lower than one (values were between 0.78 and 0.99), showing a pyrolytic origin. These results confirmed the petrogenic source of pollution for most stations. The existence of 2 and 3 rings in PAHs represents that the source of PAHs is from shipping discharges and oil spills. Since some sediment samples contained LMW PAHs, engine missions from port transportation and vehicles exhausts were also a source of contamination. Among all tested PAHs in sediments, PHE and BaP were the predominant species in all stations. Moreover, BaP can be used as an indicator for combustion-originated PAHs because the value of BaP is



**Fig. 4** Scattering plots of PAH diagnostic ratios,  $FLA/(FLA + PYR)$  versus.  $(BaA + CHR)$ ,  $ANT/(ANT + PHE)$ , and  $(IcdP + BghiP)$ , for (a) spring, (b) summer, (c) autumn, and (d) winter

approximately insignificant in petroleum (Magi et al., 2002). Engine vehicles and their exhaust systems were

considered as the main origin of potential carcinogenic PAHs, including BaA, BbkF, and BaP, exclusively in



urban zones (Neira et al., 2017; Jakovljević et al., 2018; Ciarkowska et al., 2019; Siemering and Thiboldeaux, 2020).

In order to realize the transport and fate of PAHs in the ecosystem, the assessment of the dominant origin of them is important. Thus, diagnostic ratio values of various groups of PAH contents have been widely calculated for identifying the potential origins of PAHs. Some diagnostic ratios, such as Ant/Ant + Phen, Flua/Flua + Pyr, IP/qIP + BghiP, and BaA/(BaA + Chry), were widely chosen as an accepted index to distinguish between PAHs from different sources (such as pyrolytic and petrogenic origins). Pyrogenic compounds are typically a class of composition resulting from fossil fuel, and petrogenic combustion compounds are the product of petroleum operations. Higher molecular weight PAHs (4–6 benzene rings) are compounds from pyrogenic backgrounds, whereas those from petrogenic sources have an LMW of 2 or 3 rings of benzene (Neff, 1979; Zakaria et al., 2002). It is proved that the prevalent source is petrogenic when the Ant/Ant + Phen diagnostic ratio is less than 0.1, whereas a higher ratio ( $> 0.1$ ) suggests that the root of PAHs is pyrolytic. Based on the Fluo/(Fluo + Pyr), the values lower than 0.4 suggest a petrogenic source of contamination of PAHs, and those between 0.4 and 0.5 suggest petroleum combustion sources, whereas ratios higher than 0.5 suggest that the contamination is caused by grass, wood, and coal burning. BaA/BaA + Chry values less than 2 demonstrate petrogenic sources; values between 2 and 0.35 show either a petroleum or combustion origin. Finally, (BaA/BaA + Chry) values higher than 0.35 imply a pyrogenic origin (Li et al., 2016; Yunker et al., 2002). Meanwhile, for IP/(IP + BghiP), the ratios less than 0.2 and 0.4 imply that the main source is petroleum, and if the ratios are both over 0.5, coal, wood, and grass combustion origins are suggested. Other internal ratios are seemed to correspond to petroleum combustion (particularly crude oil, vehicle, and liquid fossil fuel) (Yunker et al., 2002).

In order to assess the source of PAHs in all seasons, these distinguished ratios as indicators of PAH compound's origin were plotted in Fig. 2. In winter, diagnostic values of Ant/(Ant + Phe) versus Flu/(Flu + Pyr) implied that the ratios of Flu/(Flu + Pyr) were 0.56–0.63, all of which are higher than 0.4; the proportions of Ant/(Ant + Phe) is greater than 0.1,

which means that pyrogenic sources tainted the sediment samples. The values of BaA/(BaA + CHR) in sediment samples ranged from 0.07 to 0.17, all of which are lower than 0.4, demonstrating that the coastal sediments were contaminated by petroleum input. The values of IP/(IP + BghiP) were 0.35–0.45 (between 0.4 and 0.5), suggesting that the origins of PAHs in sediments can be associated with pyrogenic and petrogenic sources. Considering the combined diagnostic ratios in other seasons, including spring, summer, and autumn, the PAHs were mainly originated alike (Fig. 2). In autumn, the distinguish ratios of Ant/(Ant + Phe) were 0.056–0.15, and the Ant/Ant + Phen levels were greater than 0.1 at stations 1, 2, 3, 4, 6, 7, 13, 14, 15, and 16, suggesting ignition origins at these sites. The ratios of Flu/(Flu + Pyr) in sediment samples were between 0.44 and 0.57, and most varied between 0.40 and 0.50. These findings revealed that both pyrogenic and petrogenic origins were correlated with the primary source of PAHs. The values of IP/(IP + BghiP) and BaA/(BaA + CHR) were 0.53–0.66 and 0.44–0.5, respectively, both greater than 0.5 and 0.355 (Fig. 3). These data suggest that PAHs are produced from the combustion of grass, petroleum, tar, and wood in coastal sediments. In summer, the quantity of Ant/(Ant + Phe) was 0.04–0.13, denoting that the PAH compositions in this season were associated with pyrogenic and petrogenic sources. The quantities of Flu/(Flu + Pyr) and BaA/(BaA + CHR) were 0.44–0.57 and 0.67–0.72, respectively, denoting that the PAH compositions in these stations were mainly derived from incomplete burning of organic matter sources (pyrogenic). Finally, the quantity of IP/(IP + BghiP) was 0.37–0.47, all of which were lower than 0.5, which indicated that the coastal sediments were contaminated by petroleum input. However, in spring, the quantity of Ant/(Ant + Phe) was 0.04–0.12, indicating that the PAH compositions in this season were derived from pyrogenic and petrogenic sources. The quantity of Fluresul/(Flu + Pyr) was 0.43–0.57, and the values of BaA/(BaA + CHR) were 0.65–0.74, indicating that in these stations, the PAH compositions were primarily extracted from incomplete burning of organic matter sources (pyrogenic). Finally, the distinguished ratios of IP/(IP + BghiP) were 0.52–0.68, all of which were higher than 0.5, which indicated that the coastal sediments were polluted by pyrogenic input.

## PAH risk assessment

The rate of threshold effects, medium impact range, and median effect range are efficient methods for assessing the ecological risk effects of inorganic and organic pollution in aquatic sediments (CCME, Long et al., 1995; Macdonald et al., 1996; 2002). SQGs are clarified by comparing the multiple sediment toxicity responses of aquatic species with the sediment content of PAHs considered. Thus, it is recommended as an efficient method for assessment of risk levels and protection of water ecosystems (Hu et al., 2011; Long et al., 1995). Sediment quality guidelines (SQGs) are classified into three categories according to PAH concentrations ranges. The values of PAH contaminants can be less than ERL and TEL (biological effects are rare), between ERM/PEL and ERL/TEL ratios (occasional), and finally, greater than ERM and PEL (representing that biological effects are expected frequently). The  $\sum$ PAH concentrations in all seasons were importantly less than ERL and ERM quantities (Tables 2–5). These results indicated that biological effects would seldom happen. These results were in agreement with other studies such as those by Manneh et al. (2016) and Kucuksezgin et al., (2013) (Kucuksezgin et al., 2013; Manneh et al., 2016; Wang et al., 2017). According to our findings, in spring, the amount of PHE (Wu et al., 2020) at stations 8, 9, 10, and 11; the amount of Acl at stations 3 and 5–16; and the amount of DBahA at stations 9, 10, 11, and 12 were more than TEL, but less than PEL, ERL, and ERM. In summer, the concentrations of PHE at stations 8, 9, 10, and 11; Acl at all sixteen stations; Ace at station 9; and DBahA at stations 8 to 14 were lower than PEL, ERL, and ERM, but greater than TEL. In autumn, the concentrations of PHE at stations 8, 9, 10, and 11; Acl at all sixteen stations; Ace at stations 8, 9, 10, 11, and 12; and DBahA at stations 7–16 were less than PEL, ERL, and ERM, but greater than TEL. In winter, the concentrations of AC at stations 3 and 7–16, as well as those of FLA, ACL, and DBahA at all sixteen stations, were greater than TEL, but less than PEL, ERL, and ERM. This indicated that adverse toxicological effects resulting from these compounds might occasionally happen in each season, and potential risks were relatively low. These stations were located near the first and second phases of the petrochemical industrial zone, which had a large number of industrial activities, and were the main zones for depletion of

petrochemical industrial wastewater effluent into the coastal areas. In addition, this zone was affected by the effluent depletion of ballast water due to the heavy traffic of national and foreign ships.

## Conclusion

One of the world's strategic regions is the Persian Gulf. In the countries that lay on the Persian Gulf coast, about 60% of the world's oil reserves are located. In addition, in this area, about 25 percent of the world's total soil is created. During the war between Iran and Iraq and the war between Iraq and Kuwait, in the Persian Gulf, roughly 2–4 and 6–8 million barrels of crude oil were poured, respectively, and a large portion of the coastline in this area was severely polluted.

Dramatic industrial development and population growth have been triggered by the abundance in the Persian Gulf and its coastal areas, along with rich capital, resulting in intensive emissions of toxins into marine environments. In addition, through residential and industrial towns in the area, the rivers of Karun, Zohreh, and other large rivers snake and bring industrial, agricultural, and domestic waste to the Persian Gulf. The site of the Pars Special Energy Economic Zone is Asaluyeh, on the northern side of the Persian Gulf (APSEZ). Therefore, in this study, seasonal variations, source identification, and potential ecological assessment of sixteen PAH compounds were investigated based on a quantitative investigation of  $\sum$ 16PAH compounds in intertidal sediments in PSEEZ, Iran. Qualitative and quantitative evaluations found that PAHs with mild pyrogenic superiority came from both petrogenic and pyrogenic origins. Source distribution using key component analysis found that the key causes of PAHs have been fossil fuel combustion, traffic-related PAHs, biomass and coal combustion, and petrogenic PAHs. Hence, it seems the consumption of energy is the main reason for PAH contamination in the intertidal zone of Asaluyeh. Intertidal environments play an important role in the earth's biogeochemical processes. In order to prevent/reduce the detrimental health impacts on the environment and public health, more controls and changes can also be made in the intertidal ecosystem of Asaluyeh. This study has in fact investigated the adverse effects on water bioavailability. Therefore, this study will

provide a scientific background, planning, and policies for PAH pollution control and environmental protection in Iran and similar regions around the world. Finally, in order to support the understanding of seasonal and spatial distribution of PAHs in sediments, physical, chemical and mineralogical properties of the sediments should be investigated in future studies. It is also proposed that future studies should rely on the use of statistical methods in addition to the Pearson correlations of principal component analysis (PCA)/factor analysis (FA). This approach can support some novel and relevant conclusions regarding sediment complex matrix and geochemistry of PAHs.

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## Declaration

**Conflict of interest** The authors declare no conflict of interest.

**Consent to participate** All authors have read and agreed with their participating.

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